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HEAVY METALS AND ORGANIC COMPOUNDS FROM WASTES USED AS ORGANIC FERTILISERS

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LIST OF ABBREVIATIONS

AOX	sum of adsorbable organic halogen compounds
BaP	benzo-[a]-pyrene
BBP	Butylbenzyl Phtalate
CEC	cation exchange capacity
CV	coefficient of variation
d.m.	dry matter
DBP	Dibutyl Phatalate
DDT	1,1,1-trichlor-2,2-bis(p-chlorphenyl)ethan
DDX	metabolites of DDT
DEHP	di 2-(ethylhexyl) phthalate
f.m.	fresh matter
FQAC	Federal Quality Assurance Association Compost Germany (Bundesgütegemeinschaft Kompost e.V., Deutschland)
Hc	Henry constant
HCB	hexachlorbenzene
HCH	hexachlorcyclohexane
JRC	Joint Research Centre of the EU Commission, Institute for Environment and Sustainability, Soil and Waste
Kow	Octanol/water coefficient
LAS	linear alkylbenzene sulphonates
MBT	mechanical-biological treatment (pre-treatment) of the organic fraction of MSW
MBTC	compost from mechanical biological treatment of waste
MC	manure compost
mio.	million
MRLs	maximum residues limit
MSt	Member States
MSW	municipal solid waste
MSWC	municipal solid waste compost
n.i.	not investigated
NPE	nonylphenol
OM	organic matter
OPs	organic pollutants
PAH	polychlorinated hydrocarbons
PCB	polychlorinated biphenyls
PCDD	polychlorinated dibenzodioxins
PCDF	polychlorinated dibenzofurans
PTEs	potential toxic elements *(mainly used synonymously with heavy metals)*
SOM	soil organic matter
SSC	sewage sludge compost

Used Acronyms for EU Member States

AT	Austria	FI	Finland	NL	Netherlands
BE	Belgium	FR	France	NO	Norway
DE	Germany	GR	Greece	PT	Portugal
IE	Ireland	IT	Italy	SE	Sweden
DK	Denmark	LU	Luxembourg	UK	United Kingdom
ES	Spain				

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SUMMARY

Objectives and methods

This report is the result of a study carried out on behalf of the Directorate-General for the Environment of the European Commission in the context of the European waste management policy and work on the biological treatment of biodegradable waste.[1]

The objectives of the study were as follows:

1.) Identification of the pollutants (heavy metals, organic compounds) likely to be found in foodstuffs for human consumption.

2.) Evaluation of the concentration of most representative pollutants in compost produced from biowaste that is separately collected and from unsorted waste and/or residual municipal waste across the Community.

3.) Assessment of the influence of the different sampling techniques and analytical methods that are used in Member States for determining the concentration of pollutants in compost and stabilised biowaste. Identification of the applied ISO, CEN or other international or national standards.

4.) Elaboration of a scientific concept for limiting the amount of pollutants in compost and stabilised biowaste with a view to long term soil protection in the Community.

The study aimed to evaluate background concentrations of relevant pollutants (potential toxic elements (PTEs); organic pollutants (OPs)) in the areas of (in brackets, the relevant section of the report were the reader will find detailed information)[2]:

1.) Food and feeding stuff (chapter 2)

2.) Soils as receptors of waste derived organic fertilisers (chapter 3)

3.) Specific source materials for composting and other biological treatments (chapter 4)

4.) Materials intended as organic fertiliser or soil amendment (chapters 4 and 5)

   i. Compost from source separated collection schemes and sludge
      - Biowaste compost (BWC) from organic household waste including kitchen waste
      - Green compost (GC) from garden and park waste materials (grass clippings, bush and tree cuttings, leaves, flowers etc.)

   ii. Mixed municipal solid waste-derived compost (MSWC) or the stabilised organic waste fraction from mechanical biological treatment plants (MBTC)

5.) Other organic fertilisers or soil amendments such as manure, organic soil amendments and growing media, mineral fertilisers (chapter 6)

This study examined the literature (scientific publications, reports as well as original datasets provided by research institutes or local authorities). The data found include regional and temporal variations and the comparison with existing threshold and limit values.

The study conducted a comparative survey of *sampling and analytical methods* of composts, the variability of analytical results on the basis of inter-laboratory trials and specific investigations, as well

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[1] The content of this report does not necessarily reflect an official position of the European Commission.

[2] In this context PTEs are defined as chemical elements that have the potential to cause toxicity to humans, flora and/or fauna, depending on concentration, bio-availability and bio-accumulation. The definition of "PTEs" commonly refers to heavy metals; in contrast to OPs they are not of organic nature, and they may not be degraded by microbial activity or chemico-physical impacts. The latter can – in general terms – be addressed as "persistent" and have to be considered when assessing toxicological effects in time.

as statistical rules for admissible deviations within national standards (chapter 7). The review of existing proposals for the definition of the quality of organic fertilisers and composts (chapter 8) resulted in the presentation of a possible *framework for the use of waste derived composts as organic fertilisers in agriculture* with the goal of ensuring the long term multifunctional productivity of agricultural soils (chapter 9).

Potential toxic elements (heavy metals) in food and feeding stuff (chapter 2.3.1)

The variation in reported concentrations of PTEs for each type of foodstuff was considerable. Such variability may result from:

i. *Spatial variation:* the overall uptake of heavy metals may be affected by the nature and type of particular soils and the pattern of industrial sources in different districts.
ii. *Temporal variation*: analyses have been carried out at different times and this may be linked, in turn, to all other affecting factors, such as new test methods, development of local industrial activities, etc.
iii. *Heterogeneity of sampling and test* methods (in most cases surveys specify neither the sampling nor the test method).
iv. *Intra- and inter-laboratory variations*.

A comprehensive assessment of the literature relating to PTEs concentrations in foodstuffs leads to the conclusion that these elements are found in the food chain to various extents, in particular:

- Leafy vegetables and vegetables (one of the basic components in daily diet throughout Europe) tend to show a higher concentration of PTEs than cereals.
- Lower concentrations found in cereals are affected by plant physiology, which tends to concentrate PTEs more in the shoot and leaves than in the seeds.
- Fish tend to accumulate relatively high concentrations of PTEs such as As, Cd, Cr, Pb. Arsenic, in particular, has been reported by some authors at high concentrations, though it may be affected by one or more of the aforementioned factors causing variation

Data were lacking for many types of foodstuff, particularly for milk products and eggs.

Organic pollutants in food and feeding stuff (chapter 2.3.4)

The most investigated compounds are *dioxins, polychlorinated biphenyls (PCBs), polychlorinated hydrocarbons (PAHs),* and the pesticides *hexachlorbenzene (HCB), DDT, Aldrin and Dieldrin.* Factors affecting organic pollutant distribution included spatial variation, i.e., climatic and soil variations, proximity to local sources of pollution, and concentration in foodstuffs during preparation.

It is likely that the ongoing improvements in environmental legislation, and particularly new threshold values for air emissions, (e.g. as set for incineration in Directive 2000/76/EC), may reduce concentrations of organic pollutants in foodstuffs produced at farm sites near point sources. Arguably, this may also have positive implications on the future quality trends of organic waste materials used as fertilisers. A similar trend is likely to occur for sludge, if Programmes for the Prevention of Pollution were to be introduced in the Sewage Sludge Directive 86/278/EEC.

Spatial variation: OP background concentrations were compared at the following levels:

i. Member States level (*geographic scale*) and
ii. The *local level* addressing the OP content in food commodities in rural areas compared to industrial districts. However, the data collected was not sufficient to build a scenario covering the whole of Europe. Some case studies representing experiences and investigations carried out at local level are provided in this report.

Geographic scale: There is a general lack of information on several countries - in particular in Southern Europe - and, therefore, an analysis of *geographical trends* is not possible. Geographical variations can become blurred through the transportation of goods around Europe or imports of goods from outside the region. Care should, therefore, be taken in the identification of sources of these products before conclusions are drawn concerning sources of contamination.

Proximity to local pollution sources: Incinerators, petrochemical and metallurgical activities were generally identified as potential sources of organic pollutants. Many parameters influence the effect of the sources of organic pollutants on foodstuffs, the main ones being the type of industry, chemical identity and path through which the contaminant gets in touch with the foodstuff, distance from the industrial site, etc.

Temporal variation: The numbers reported show in most cases a decrease in detected concentrations over time. Arguably, these trends mainly depend on the implementation of technological improvements with particular reference to equipment to prevent formation and/or avoid dispersion of organic compounds dangerous for human health. As already remarked, enforcement of specific environmental legislation (as, for instance, Directive 2000/76/EC on the incineration of waste) may arguably be seen as a main driver for such technological improvements.

Effect of cooking on the OP concentration in food: Some studies indicate the general increase of concentrations of benzo(a)pyrene or polychlorinated hydrocarbons (PAHs) when food was cooked compared to uncooked foodstuffs. Those findings do not apply for PCBs, DDT or polychlorinated dibenzodioxins/furans (PCDD/F). Benzo(a)pyrene is formed through incomplete combustion of organic matter. Incomplete combustion mainly occurs during heating processes like grilling, barbecuing and roasting where foods are close in contact with the heat source (over an open flame). This includes drying with smoke, or by absorption during the smoking. The degree to which this process contributes to an undesirable increase in PAH concentrations in compost feedstock, and consequently in compost, depends on the proportion of cooked, grilled or smoked food residues in the overall feedstock composition.

Referring to the microbial degradation of PAH and the actually detected PAH concentration in compost, the report concludes that the effect of increased PAH concentration in cooked food residues is not likely to constitute a threat to the recycling of kitchen waste and its safe application via composting (see report, chapter 2.3.5).

Inorganic and organic pollutants in soil and factors influencing the content of pollutants in foodstuffs (chapter 3)

A number of interactive biotic and abiotic processes govern the behaviour of ***heavy metals*** in soils and define chemical speciation and bioavailability. The major factors driving the biogeochemical processes in soils are pH, cation-exchange-capacity (CEC) and redox potential.

Assessment of "total" PTE concentrations detected in soils after digestion with strong acids is the commonly agreed basis for the evaluation of conditions for long-term soil conservation. Aqua Regia ($HCl_{conc.}$: HNO_3 = 3 : 1) is still the first choice and was adopted as the preferred extracting agent for sludge, soil improver and growing media as well as soil.

Problems dealing with ***background concentrations*** arise from

i. their great variability due to spatial variation of soil parent material and pedogenetic (weathering, decomposition, sedimentation, translocation and leaching processes) conditions,
ii. the fact that “natural” soils (i.e. without any anthropogenic influence) are scarce due to historical land use patterns and diffuse emissions and
iii. different sampling and analytical methods employed.

Ranges of so-called background concentrations from different authors, and a recently conducted study on behalf of the Joint Research Centre (Ispra), emphasise the great variance due to site and soil forming conditions and the obvious need for the harmonisation of sampling, analyses and interpretation.

The term "background" concentrations of ***organic pollutants*** refers more or less to anthropogenic pollution, since these compounds typically do not occur naturally in soils or soil parent materials. Consequently it differs from land use patterns related to soil management practice. Moreover, the varying susceptibility of the different organic compounds to bio-degradation under different soil conditions makes it difficult to identify reference concentrations. This could only be done with organic pollutants, which show a high persistency (such as PCBs, PCDD/F, PAH).

Potential toxic elements (PTEs) in compost (chapter 4)

In compost produced from separately collected organic household waste the PTE concentrations decreased by a factor 2 to 10 as compared to *municipal solid waste compost* or a stabilised organic waste fraction. Quality related regulations for composts have already reflected this development within the last 10 to 15 years in many EU Member States.

Potential source materials showed variability in PTE concentrations similar to that identified in foodstuffs. Cadmium, Copper and Zinc showed the highest concentrations in residues from food and semi-luxury food production as well as separately collected biowaste. Because of these pre-existing concentrations, it would seem difficult to comply with existing limit values for compost given the mineralisation of organic matter in the course of aerobic degradation. However, these particular high concentrations in some of the source materials did not seem to exert a major impact on the overall compost quality, as long as compost mixtures were coming anyway from source selected materials.

The proportion of physical impurities in the input wastes, did, however, have a measurable effect on their concentrations in the resultant compost. Final screening at the end of the composting process did not fully remove the pollutant input introduced by impurities in the raw material. For this reason collection at the doorstep (*door to door collection*) showed distinct advantages compared with collections using road-side containers, where the proportion of impurities was increased.

National investigations indicated that ***green waste compost*** *(GC)* tended to have slightly lower PTE concentrations than ***biowaste compost*** *(BWC)*. This difference showed a fairly marked pattern following the order: Cu > Zn >> Hg > Pb with differences between 20 and 50 % under well defined standard collection conditions. Cr, Cd and Ni gave an indifferent or heterogeneous picture with no clear trend observed. However, these differences should not lead to the exclusion of biowaste composts from the use as organic fertiliser and soil improver, as long as final PTE concentrations would still not lead to an unacceptable pressure to soil quality under good practice application schemes (see concept of chapter 9 in the report and below).

The difference between ***urban and rural biowaste composts*** was not as evident as could have been expected. Typical *pedogenic* metals like Cr and Ni tended to be higher in composts from rural areas compared to urban areas. Elements typically associated with *human (industrial)* activities such as Cu, Pb, Zn and Hg show relatively higher levels in urban than in rural areas. This was identified both in local and national surveys in Germany and in Austria.

Temporary and seasonal variation for Hg, Ni and Zn was reported to be lower than for Pb, Cd, and Cu in a German investigation in Baden-Württemberg. No evidence for a systematic differentiation of *winter* and *summer* compost was found. The comparison of mean PTE concentrations in biowaste compost and green compost investigated over a period of eleven and ten years in Germany and Switzerland respectively, show a similar outcome. The copper content increased (+ 45 % and + 30 % respectively), Zn remained on the same level or increased slightly (11 %), whereas all the other metals indicated a relative decrease of concentrations of -17 % to -38 %.

With the exception of copper, in many countries PTE concentrations in sewage sludge have constantly decreased within the last 20 years. This development increases the possibility to use sewage sludge as a feedstock for the production of compost complying with precautionary quality standards. Notwithstanding, on average, sewage sludge composts (SSC) still contained distinctly higher concentrations of Cd, Cu, Hg and Zn than biowaste compost and green compost.

It can be further noted that

- regional and seasonal variations of PTEs in biowaste compost and green compost were based on unavoidable background concentrations in soils and caused by atmospheric deposition and the individual fluctuation even at similar collection schemes and purity level;
- countries in the early phases on implementing separate collection systems showed 10 to 50 % higher PTE concentrations in biowaste compost and green compost than it is found in long established schemes. But this has to be considered in the light of (i) the still limited data set and (ii) in some cases the difficulty of distinguishing between facilities producing compost only from bio and greenwaste and those also including sludge;
- when sewage sludge or animal manure is used as source material, Cu and Zn usually reach higher concentrations. However, the choice of high quality manure and sludge would also match quality standards set in member states for biowaste and green waste compost. This would be dependent on the source of the material: for example, in the case of sewage sludge the district served by the wastewater treatment plant and for manure on the use of feed additives containing Cu and Zn.

Organic pollutants in compost (chapter 5.2)

Selection criteria for the evaluation of organic pollutants were set based on their potential occurrence in compost, the availability of published data, knowledge of physico-chemical properties and feasibility of chemical analysis. The compounds considered were: Polychlorinated biphenyls (PCB), polychlorinated dibenzodioxins and dibenzofurans (PCDD/F), polycyclic aromatic hydrocarbons (PAHs), ·chlorinated pesticides and adsorbable organic halogen (AOX) *(Aldrin, Biphenyl, o-Phenylphenol Chlordane, Dieldrin, Endrin, Heptachlor, DDT [1,1,1-trichlor-2,2-bis(p-chlorphenyl)ethan], lindane, HCH-isomers [hexachlorcyclohexan], hexachlorobenzene, hexachlorobenzol, heptachlor, pentachlorophenol pyrethroides, Thiabendazole)*, linear alkylbenzene sulphonates (LAS), nonylphenol (NPE), di (2-ethylhexyl) phthalate (DEHP), butylbenzyl phtalate (BBP), dibutyl phtalate (DBP).

Accumulation scenarios were only performed for PCBs, PCDD/F and PAHs, since these compounds are the best documented and show comparatively high persistency.

Conclusions for the key substances covered by reliable literature data in the area of organic waste and its recycling via composting were:

PCB: Since PCBs have been excluded from industrial processes since the 1980s a continuous reduction in their occurrence in the environment can be expected. In general, they were detected in higher concentrations in composts stemming from urban areas. Some, but not all investigations reviewed showed higher concentrations in biowaste than in green compost. PCB content in composts from mixed municipal solid waste is approximately 50 to 100-fold higher than that found in compost from source separated bio and green waste.

The observed reduction of PCBs during composting was up to a maximum of 45 % through either bio-degradation or volatilisation. However, there are considerable uncertainties since, given the concurrent mineralisation/volatilisation of part of the organic substrate, generally higher concentrations attended to be found in compost than in feedstock. Degradation occurs mainly for congeners with lower chlorination.

Input of PCBs to soil from compost was considerably less than atmospheric deposition rates. Assuming a half-life ($t_{1/2}$) of 12 years reported in literature, and average concentrations in composts, no accumulation in soils can be expected. Even in a worst-case scenario ($t_{1/2}$=50 years) the soil precautionary reference value of the German Soil Protection Ordinance (0.05 mg kg^{-1}) would not be reached within 100 years also assuming a comparatively high PCB concentration of 0.1 mg kg^{-1} d.m. in compost.

PCDD/F: The level of PCDD/F in composts mainly depended on the background concentrations in the soil and the source material following diffuse emissions in the catchment area of the composting plant. A differentiation between urban and rural areas could not be clearly identified. Mean values of several investigations demonstrated a trend towards lower values in green compost than in biowaste compost. A trend of continuous decrease of PCDD/F levels over time in composts was observed. PCDD/F content in composts from mixed municipal solid waste was typically 50 to 100 times higher than in compost from source separated bio- and green waste.

In general PCDD/F tended to concentrate during the degradation process, mostly due to the mass loss during mineralisation of organic matter. Biowaste and green waste feedstocks showed generally lower concentrations than the finished composts. The reported initial generation during the rotting process only contributes to a negligible degree to dioxin content in composts and only occurs with temperatures > 70°C and in the presence of primary substances such as *trichlorophenol* and *pentachlorophenol*. Here it is important to say that properly managed processes keep temperatures in the range of 45 to 60°C. Higher temperatures are sought only for a short period to ensure sanitisation, although many regulations just mandate 55 to 60 °C to be reached for hygienisation. Most tests conducted during the composting process of biowaste showed an increase of hepta- and octa- PCDD. On the other hand, the content of low chlorinated PCDD/PCDF decreased during the composting process (thereby leading to a decrease in overall toxicity). Furans also generally diminished.

PCDD/F input by regular compost application can be estimated to be in the same range as or slightly higher than mean atmospheric deposition rates. However, in none of the scenarios when modelling the long term change of soil PCDD/F status accumulation was thought to occur (this holds true even if no further decay in soil is assumed). In most of the cases a considerable decrease in soil concentration was found, due to the half-life of these substances, which overcompensates for input through deposition and compost application.

PAHs: The relevant spatial and temporal variability of PAH may be explained by seasonal differences in the composition of the raw material, by different collection capture areas or individual contamination with extraneous materials (e.g. ashes). From the literature considered, higher concentrations for urban areas than for rural areas can be assumed. There was only a slight trend indicating higher values in biowaste compost than in green compost. PAH concentrations in organic wastes were higher than in compost. PAH content in composts from mixed municipal solid waste was typically 1 to 10 times higher than in compost from source separated bio- and green waste.

Though PAHs show a high persistency in soils they undergo an effective reduction under aerobic degradation conditions by up to 70 %. This includes volatilisation as well as bio-degradation and performs more effectively during the maturation stage of composting.

Though a certain accumulation of PAHs may occur in soils with low background concentrations (ca. < 0.5 mg kg^{-1} d.m.) precautionary soil threshold values would not be touched even in the long term.

Conclusions for PCBs, PCDD/F and PAHs:

Concentrations of PCBs, PCDD/F and PAHs in BWC and GC were similar to background concentration in soils. This lead to the conclusion that regular measurements of PCBs, PCDD/F and PAHs and the establishment of limit values for these compounds are not required for the safe use of compost derived from source separated organic waste materials.

Due to the comparatively much higher concentrations found in mixed waste compost they should be measured, instead, when mixed waste compost is used as amendment. The authors recommend to restrict the use of mixed waste compost to limited non-food areas such as land reclamation of brown fields and surface layers on landfill sites or on noise protecting walls aside roads or railways.

AOX and Pesticides: Even though most of the chlorinated pesticides are now banned in the EU they may be found in very low amounts in composts. Generally biowaste compost tended to show higher concentrations than green compost. Organochlorine pesticides, Pyrethroides and Thiabendazole were found to be below related limit values for fertiliser regulations or even close to the lowest detection limit.

The variety of compounds within the group of *chlorinated pesticides* and *absorbable organic halogens* gives a wide range of properties and related behaviour during composting.

Comparisons of the pesticide concentrations in feedstock versus the end composted products showed that composting tends to substantially decrease the concentrations of most compounds. Frequently, thermophilic process conditions were favourable to breakdown the investigated substances.

Specific case studies on *chlorophenols* consistently indicated their high degradability during composting. Therefore they are not considered as substances with priority for analysis in compost or digestate from anaerobic treatment.

In general, the patterns of pesticide degradation in composting parallel the patterns found in soil, but in several studies, pesticide compounds disappeared faster during composting than they typically do in soils, as indicated by their soil half-life values.

From the literature reviewed it can be concluded that there is no indication for the need for regular analyses of AOX or specific pesticides. This is mainly based on general low background concentrations as well as degradation behaviour during composting.

However, given the high persistence/toxicity of compounds used and by means of precaution, pesticide treated wood should be excluded from the production of marketable compost products or any recycling in agriculture.

LAS, NPE, DEHP: The Danish Decree for the agricultural use of sewage sludge and waste derived composts was the only regulation that specifies limit values for LAS, NPE, DEHP. Those compounds are all rapidly degraded under aerobic composting conditions. Extremely low concentrations were found in the literature examined. Thus there is no evidence of a need for general threshold or limit values.

Sampling and analytical methods examined for PTEs (chapter 7)

The study examined sampling frequencies, sampling methods and analytical procedures together with statistical variability in compost testing on the basis of existing compost standards, case studies and inter-laboratory trials.

Random sampling is commonly agreed to be a suitable procedure for testing compost quality approval processes. In line with existing national standards, one sample per 1,000 to 2,000 tonnes of treated biowaste with a maximum of 12 analyses per year can be considered as reliable sampling frequency. However, higher sampling frequencies are required in the commissioning phase applied by external quality assurance systems (QAS).

A minimum of ten consecutive analyses at one composting facility has been recognised as a reliable forecaster of the range and mean values of PTE concentrations, which can be estimated with statistical methods. This figure can then also be used as a reference for possible reduction in the number of normally required analyses. This case-by-case reduction of standard sampling frequencies can be granted by the competent authority or a recognised quality assurance organisation in the frame of a product certification scheme. By statistical evaluation at facility level, expected upper values at a given confidence interval (e.g. 95 %) may give the margin for the identification of facility specific outliers. If an outlier was identified, repeated sampling should be carried out, and if confirmed, specific measures of quality management and a cause study should be applied.

Sample taking: The *portions sampled have to be in the same condition* as the compost batches used or marketed under the same specification. The minimum *number of increments* in national regulations and standards range from 4 to 30 from heaps to stocks, depending on the size of the sampled batch. Based on a statistical evaluation of standard errors, a minimum number of 9 incremental samples independent of the batch size has been singled out as needed. *Sampling points must be distributed evenly* over the volume of the sampled stock.

Repeated sampling and measurements – system of tolerances: In principle we have to distinguish between two types of tolerances for an analytical result:

1.) tolerance which refers to the standard error of the analytical procedure in order to estimate the *true value* of a single measurement

2.) tolerance which refers to an analytical result *relative to given quality classes* or limit values

In the case of ***sampling on behalf of the compost producer*** the experience of investigations on parallel sampling of batches led to the conclusion that it may be advisable to carry out parallel, distinct sampling from one sampled batch. This would tackle the lack of homogeneity of compost materials better than a repeated testing of one single combined laboratory sample.

As a consequence, coefficients of variability of parallel sampling procedures have to be considered. On the basis of the examined studies, as well as of the repeatability factors laid down in analytical standards, for composts a coefficient of variability of ± 30 % would be a reliable maximum value for all elements. In this case the number of parallel samples shall be limited to six and after exclusion of statistically identified outliers, the mean value of the repeated measurements has to meet a given threshold value.

Furthermore, a system of tolerances for an individual measurement result in a series of compost batches should be considered. Making reference to the German Biowaste Ordinance one sample out of four (25 %) may exceed the limit value by 25 %, but the sliding mean value of the most recent four samples must still be below the limit value.

When it comes to a ***control measurement on behalf of the competent authority*** the overall variability of composting processes as well as the sampling and analytical procedure and the coefficient of variation between individual laboratories have to be considered.

Therefore it is recommended to establish an additional tolerance relative to the set limit values regarding the recognition of a *control sample*. The evaluation coefficients of variation (CVs) of inter-laboratory trials (ILTs) and the batch related CV led to the conclusion that a tolerance of 50 % is accepted in the case of an external controlling procedure on the market place.

Extraction method for PTEs: Since the *aqua regia* method was adopted by the CEN Technical Committees[3] and as the majority of experience with compost is based on this extraction method, it seems to be a reasonable way forward to refer to EN 13650 for greater consistency.

Concepts of quality definitions for waste derived organic fertilisers (chapter 8)

Three basic options are available in order to determine "safe" limit values for potential toxic elements (PTEs). They vary according to the viewpoint adopted:

1.) Risk based assessment such as the No Observable Adverse Effect Levels (NOAEL) concept

2.) Mass balance or No Net Accumulation (NNA) in relation to the concentration of contaminants in the soil (precautionary approach)

There are two options for a NNA concept:

i. Limiting **PTE/OP concentrations** in fertilisers and soil amendments to the same level of soil background concentrations ("*same to same*" or "*similar to similar*")

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[3] "Sewage Sludge" (CEN TC 308; EN 13346 "Characterisation of sludges – Determination of trace elements and phosphorus – Aqua regia extraction methods") and "Soil improvers and growing media" (CEN/TC 223; EN 13650 "Soil improvers and growing media - Extraction of aqua regia soluble elements").

   ii. Limiting the **PTE/OP load** so that it matches the amount of tolerable exports from soil via harvested crops, leaching or erosion ("*import = export*")

3.) Between those two polarities manifold hybrid systems and indicators such as the assessment of predicted environmental concentration (PEC) in comparison with the predicted no effect concentration (PNEC) are discussed.

A long-term sustainable strategy will always seek for a combination of measures, which fulfil both:

i. the maximisation of benefits of recovering organic matter (and nutrients thereto linked), and
ii. the minimisation of inputs of contaminants that may adversely affect soil functions and human, animal and plant health.

The ***Risk assessment based on No Observable Adverse Effect Levels (NOAEL)*** is discussed using the example of the *US EPA Part 503 Risk Assessment* for the use of sewage sludge where 14 different pathways were assessed for risks associated with each of the pathways. The key arguments of published criticisms relate to the *non-protective aspects* of the assumptions and the underlying approach. For example, this may be underlined by the fact that *Part 503* allows a yearly Cd load through the use of *bag sludge* (this is the higher quality of sludges defined under Part 503) of 1,900 g ha^{-1}, resulting in an estimated increase of the soil Cd concentration of 0.63 mg Cd kg^{-1} soil d.m. year^{-1}. As a consequence soil is considered as a sink for PTEs as long as *no harm occurs within the food chain*, but long-term sustainability of soil management tends to be overlooked. This is why Europe has historically adopted and enforced an approach aimed at *preserving soil safety* in many regulatory provisions. Such an approach is reflected in both of the following concepts.

In the ***mass balance / no net accumulation concepts*** soil itself is the first subject of protection. Precautionary limit values are designed to safeguard soils against future impacts. In its pure interpretation, the precautionary approach may be regarded as avoiding any increases of soil PTE concentrations above current values. This means that additions of PTEs from soil improvers, fertilizers and the atmosphere should equal the losses via leaching, plant uptake and erosion. Two general options for integrating this concept at the operational level have been formulated: (i) avoidance and/or decrease of pollutant input to zero; no change of given background concentrations and (ii) limiting inputs to (provisionally) tolerable outputs (zero balance; critical-load-concept). A restrictive limitation following a "no net accumulation" policy may, for instance, lead to a maximum compost application of about 2-3 tonnes ha^{-1}y^{-1} and would therefore preclude the benefits of humus management even with high quality composts in agriculture.

The report discusses in detail different *critical load* or *no net accumulation* concepts designed for an unique evaluation of all types of fertilisers, which have mainly been brought up on the background of the German Soil Protection Ordinance. Here the key disadvantages and critical aspects of those concepts are indicated:

- Maximum PTE loads are non science based arbitrary settings.
- Results often depend on the assumed phosphorus content in organic fertilisers, which shows a considerable variation even for composts (0.4 – 1.2 % P_2O_5 d.m.).
- The organic matter supply especially with composts is underestimated.
- The rigour of some of the concepts would exclude 100 to 50 % of produced compost based on qualities achievable under the condition of source separation of organic waste.
- The concepts are not based on the reality of achievable PTE concentrations, even with proper strategies for source separation of clean feedstocks, considering (i) the background contamination from diffuse sources (this makes the approach typically an "end-of-pipe" solution) and (ii) necessary tolerances due to local, seasonal and analytical deviations.
- None of the concepts questions the factual effect of the application system to the soil at least on a mid term scale. But this would be the key element in order to estimate the environmental impact and the maintenance of soil functions in the agro-eco system.

Hybrid systems have therefore been developed in order to overcome the shortcomings of both previous approaches. Similarly to concepts focused on "no net accumulation", hybrid systems focus on the long-term preservation of quality of soils; anyway, in order to avoid any "end-of-pipe" solution, these systems consider the quality achievable through source separation (which tends to exclude from the composted stream the "avoidable" load of pollutants) and check the profile of accumulation in time. The definition of threshold limits therefore allows for slow, provisionally tolerable accumulation, if this is kept well within "safe" thresholds for the soil itself, in order to benefit from organic matter and nutrients vectored by compost onto soils, whilst allowing for a time window long enough to work effectively on further reduction of pollution at industrial and other sources. However the key element of such a concept is the proper and science based derivation of so-called *soil precautionary values*. This would have to take into account the following:

i. Proportions for different pathways [soil-plant-groundwater-food chain, etc.],

ii. the balance of additional inputs in the range between background and precautionary values from different sources based on total contents within a long term time frame (e.g. 200 years) and a soil use specific depth,

iii. mobile fractions and threshold concentrations which would lead to a measurable effect on the environment and

iv. site-specific differentiation regarding current pollutant status.

Based on this it has to be taken into account that organic matter cannot be given up, and related hazards have to be tackled at the source (improving industrial processes, decreasing diffuse contamination). The application is therefore beneficial and environmentally sustainable as long as the quality of materials is high (primarily due to separate collection schemes for feedstocks) and as a consequence, the accumulation profile allows for a long enough time frame to solve the problem of diffuse contamination at source. Any soil managed within such framework will therefore fulfil the requirement of multifunctionality if

i. ecotoxicological effect thresholds (balanced with data on background values) are not exceeded, even once a certain "safety factor" is considered to reduce the threshold,

ii. there is no evidence of unwanted or hazardous effects on plants and groundwater at this concentration level,

iii. there is an adequate distance (safety factor) to the trigger values proposed for the pathway "soil to humans".

Recommendation for a step-wise concept for the sustainable use of compost (chapter 9)

An important element of a comprehensive concept is that the assessment of benefits derived from Good Agricultural Practice (GAP) requirements and the potential adverse effects of contaminants are performed independently in a step by step manner, rather than linking or weighing the desirable and undesirable effects through arbitrary balance equations (e.g. the relation between P and Cd content). This approach guarantees the acknowledgement of the full range of beneficial effects intended through the application of organic fertilisers.

The authors have therefore adopted a crosscutting step-wise approach to check the effects of the accumulation of PTEs in soil; according to this approach, unit loads (tonnes ha^{-1} dry matter) assumed as the input load in scenario calculations have been singled out after a thorough evaluation of possible limiting factors or, conversely, agronomic needs for the application of compost.

The following steps have been followed:

I. Identify the benefits of the fertiliser/soil amendment to the agro system (e.g. relevant amounts of organic matter or plant nutrients);

II. Identify potential pollutants which may have any adverse effects to the agro-ecosystem, to the environment or the food chain;

III. Define the limiting factor – in terms of maximum applicable loads;

IV. Calculate the response – in terms of accumulation of polltants - to the outcome of step III. Incidently, we have to remark that assessing the accumulation at maximum applicable loads depicts a "precautionary" approach, since any lower load would factually translate into a slower profile of acumulation.

The assumptions made for the calculated scenarios are based on the following:

- ***Soil threshold values***:
  - The "precautionary" values of the German Soil Protection Ordinance for sandy and clayey soils; these values are regarded as one of the most stringent approaches to soil safety in Europe;
  - The proposal of the Joint Research Centre (JRC) for acidic to neutral soils ($6 < pH < 7$), which were suggested for a revision of the EC Sewage Sludge Directive on the basis of European soil surveys on PTEs.
- ***Compost PTE concentrations***
  - A statistically weighted average of seven country-median (or mean) values and the 90th percentile values
- ***PTE outputs from the system***
  - Average total export of heavy metals via harvest (cereals, maize, sugar beet, potatoes) and leaching taken from figures in Germany.
- ***Scope***
  - 100 and 200 years as reference time frames for the accumulation of PTEs against the assumed soil threshold values in order to allow for a sufficiently long time-frame to address the issue of diffuse contamination at the source through improved technology in industrial processes, transport etc.
- ***Soil depths and density***
  - 20 and 30 cm at 1.5 g cm^{-3} resulting in 3,000 and 4,500 t ha^{-1}respectively.
- ***Soil background concentrations for PTEs***
  - Differentiated for sandy and clayey soils taken from average values of national surveys of three European countries (DK, FR, DE).
- ***Yearly application of compost***
  - This is derived from a lower (30 kg $ha^{-1}y^{-1}$) and higher (60 kg $ha^{-1}y^{-1}$) phosphorus supply on the basis of P_2O_5 contents in biowaste compost (0.65 % d.m.). It resulted in 4.6 and 9.2 t d.m. compost $ha^{-1}y^{-1}$ respectively
- ***Mineralisation rate of compost incorporated into the soil***
  - Some authors calculated that up to 8 % of the organic matter applied as compost would remain in the soil on a mid to long-term basis. In this model a more precautious approach was taken, and the retained OM was assumed to be 6 %;
  - The level of OM in composts was assumed to be 36 % d.m., which equals a mineralisation rate of 30 % relative to the total dry matter mass. Accordingly, the increase of soil mass was set to 70 % of the annually added compost dry matter mass. The calculated concentrations of PTEs are related to the soil mass at the assumed soil depth (20 or 30 cm) plus 70 % of the yearly applied compost mass. This equals 6.44 t in case of a yearly compost application of 9.2 t $ha^{-1}y^{-1}$.

Further, this study presents a ***possible approach towards threshold values for composts used in agriculture*** after considering both general accumulation scenarios and actual compost qualities. The approach takes into account the following:

- Temporal and regional variations as well as the variation within an individual composting plant, which goes beyond any effective control strategy (as for example effective source separation).
- The possibility that 90th percentile values may occur in individual facilities across Europe due to regional background concentrations in feedstock materials.
- Countries which are in the starting phase of source separation showed PTE concentrations 7 % (Cr) to 56 % (Ni) higher than EU average concentrations. Possible explanations for this result – probably more important than only the efficiency of source separation – are discussed in detail.
- Even allowing for 10 years of regular compost application on soil at the highest allowable PTE concentrations, increases caused by PTE accumulation would hardly be detectable by current analytical methods.

[A] Findings of the general accumulation scenario <u>on the basis of actual compost qualities</u> in Europe:

- Chromium will not reach the soil limits under any of the assumed scenarios.
- In the scenario for *clay soils* assumed soil thresholds will – with the exception of Zn (150 mg kg^{-1} after 137 years) – not be exceeded up to 200 years of compost application.
- On sandy soils Cu, Hg (only at 20 cm soil depth) and Zn exceed the soil threshold values within 100 years (mainly with application rates of 9.2 t ha^{-1}y^{-1} and at 90th percentile compost concentrations).
- In the case of sandy soils, Cu at high application rates (9.2 t ha^{-1}y^{-1}) and Zn even at the low application rates (4.6 t ha^{-1}y^{-1}) must be considered the most critical elements compared with achievable BWC quality. However, their function as trace elements essential for plant nutrition has to be taken into account in the overall fertiliser regime.
- Significantly higher concentrations in composts for the elements Cu and Zn (and to a lesser degree for Ni, Pb and Cd) than the 90th percentile assumed in the model calculation might require a site specific evaluation if such composts are to be used continuously on sensitive sandy soils in the long run.

[B] Scenario findings and justification of the proposed approach towards <u>threshold values for composts used in agriculture</u>:

- The regional and seasonal variations of Biowaste and Green Composts are based on unavoidable background concentrations, which fluctuate even in similar collection schemes and purity levels.
- It has to be recognised that due to specific regional background concentrations, the 90th percentile of country/Europe-wide produced composts may actually represent the average quality of composts achieved at a single composting plant in a certain region.
- Countries in the starting phase of separate collection systems show 10 to 50 % higher PTE concentrations in Biowaste Compost and Green Compost. This has to be considered in the light of (i) the still limited data set and the comparatively low reliability of sampling/testing methods (ii) the inability to distinguish between facilities producing compost from biowaste only and those also including sludge (frequently considered jointly in available statistics as "source selected materials").
- Based on the evaluation of material specific, regional and temporal variability a 50 % tolerance was added to the 90th percentile value of European BWC [level 1] and the averaged 90th percentile of ES and UK composts as representatives for *starting phase countries* mentioned above [level 2]. Table S1 gives a comparison of the resulting concentrations with the proposed limit values of the Working Document "Biological treatment of biowaste" (2nd draft) and the averaged limit values for BWC in European countries.

Table S1. Possible approach for compost limit values derived from the accumulation scenarios for sandy soils (mg kg^{-1} d.m.)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
90-percentile EU all		0.89	37.4	79.5	0.35	29.7	105.2	284.2
90-percentile UK & ES		1.27	42.8	113.1	0.43	42.1	133.4	314.0
Difference [(UK&ES)-EU]/EU (%)		+47	+7	+53	+43	+56	+52	+14
90-percentile EU + 50 %	**[level 1]**	**1.3**	**60**	**110**	**0.45**	**40**	**130**	**400**
90-percentile UK & ES + 50 %	**[level 2]**	**1.9**	**64**	**170**	**0.65**	**63**	**200**	**470**
Averaged limit values of EU countries		1.4	93	143	1.0	47	121	416
WD 2nd Draft 1)	class 1	0.7	100	100	0.5	50	100	200
	class 2	1.5	150	150	1	75	150	400

1) EU Commission, DG Environment. Working Document (WD), 2nd draft: “Biological treatment of biowaste”

- [level 1] can be recommended as general threshold for sustainable, regular use of compost in food and feeding stuff production as well as hobby gardening.
- In order to guarantee comparability between well “stabilised” or mineralised composts and “young” ones (in which concentration of PTEs due to the loss of *organic matter* has not fully occurred, yet) measured PTE concentrations should be standardised to a fixed organic matter content (e.g. 35 or 40 % d.m.). This would be a fair proof of compliance with set limit values and balances the sometimes highly varying degree of mineralisation in composted materials.
- In any case, the calculated increase of PTE concentrations in the soil due to annual compost applications is hardly detectable by analytical means prior to 10 years of application.
- The allowance of a transition period of about 10 to 15 years for the use of [level 2] composts (see Table S1) in food and feeding stuff production may be acceptable from the viewpoint of environmental and soil protection. This would be a viable way in order to
  - allow countries in the starting phase of separate collection systems to improve collection schemes and quality management during the implementation phase;
  - give enough time to improve sludge qualities from industrial and urban wastewater treatment for the use as feedstock of composting.
- However, when sewage sludge or animal manure (specifically pig manure) is used as source material, PTE concentrations of [level 2] could in some cases not be met in the case of Cu and Zn. This requires further efforts for improvement of quality at the source.
- In terms of practicability and a flexible application system, applied quantities may be seen as average yearly dosage within a time span of 20 years. In other words for land reclamation projects for the restoration of degraded soils a maximum incorporation of approximately 200 tonnes per hectare should be granted under consideration of all relevant environmental aspects (i.e. water conservation etc.)
- The graphs presented in chapter 9 of the study show that even at high application rates (9.2 t d.m. ha^{-1}) [level 1; see Table S1] composts would not lead to an accumulation beyond soil reference concentrations for clayey soils within 200 years. The JRC proposal would only be exceeded within 130 years in the case of Zn.

- Thus, for clayey soils, there is still a comparatively wide scope and flexibility for the use of compost. This might be of special importance where a distinct need for the amelioration of degraded soils is recognised (for example, in Mediterranean soils).

- The time span after which reference values for sandy soils would be exceeded when using composts of both proposed qualities [level 1 and 2] is shown in Table S2.

Table S2. Years until precautionary threshold values for sandy soils* are exceeded**

PTE concentration according to:	**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
				years			
[Level 1]	103	237	60	74	122	97	44
[Level 2]	70	218	39	53	73	61	37

* taken from the German Soil Protection Ordinance

** at 9.2 t compost $ha^{-1}y^{-1}$

- It is worth noting that at the proposed levels of threshold concentrations, the limiting factor for compost application is still the phosphorus supply rather than the PTE loading. This is particularly true in Central and Northern European Countries, where specific regulatory provisions are being implemented on P supply.
- Some open issues for research and monitoring may be defined, along the following lines:

  i. Soil concentrations of Cu, Hg and Zn should be regularly monitored on sandy soils, which show *high background concentrations* every 100 to 150 tons of compost application. The definition of high soil background concentration should be made by local authorities based on national soil surveys.

  ii. Monitoring and research of PTE availability/solubility/mobilisation within pilot schemes with plots fertilised with compost is an important tool for further evaluation of potential impacts due to the input of contaminants by fertilisation systems. It would be of great advantage to establish a cooperative research and monitoring network which integrates existing as well as new field trials covering the most important climate, soil and management conditions for a better understanding of the dynamics of the elements and substances concerned in the soil-plant-groundwater system. In any case, the slow accumulation profile caused by the (comparatively) low thresholds made possible by source separation, allow for the possibility of even long-term research programmes, before any risk can be envisaged.

1 INTRODUCTIONAL NOTES AND OBJECTIVES OF THE STUDY

1.1 Objectives and methods

This report is the result of a study carried out on behalf of the Directorate-General for the Environment of the European Commission in the context of the European waste management policy and work on the biological treatment of biodegradable waste.4

The objectives of the study were as follows:

1.) Identification of the pollutants (heavy metals, organic compounds) likely to be found in foodstuffs for human consumption.
2.) Evaluation of the concentration of most representative pollutants in compost produced from biowaste that is separately collected and from unsorted waste and/or residual municipal waste across the Community.
3.) Assessment of the influence of the different sampling techniques and analytical methods that are used in Member States for determining the concentration of pollutants in compost and stabilised biowaste. Identification of the applied ISO, CEN or other international or national standards.
4.) Elaboration of a scientific concept for limiting the amount of pollutants in compost and stabilised biowaste with a view to long term soil protection in the Community.

The study aimed to evaluate background concentrations of relevant pollutants (potential toxic elements (PTEs); organic pollutants (OPs)) in the areas of (in brackets, the relevant section of the report were the reader will find detailed information)[5]:

1.) Food and feeding stuff (chapter 2)
2.) Soils as receptors of waste derived organic fertilisers (chapter 3)
3.) Specific source materials for composting and other biological treatments (chapter 4)
4.) Materials intended as organic fertiliser or soil amendment (chapters 4 and 5)
    i. Compost from source separated collection schemes and sludge
    - Biowaste compost (BWC) from organic household waste including kitchen waste
    - Green compost (GC) from garden and park waste materials (grass clippings, bush and tree cuttings, leaves, flowers etc.)
    ii. Mixed municipal solid waste-derived compost (MSWC) or the stabilised organic waste fraction from mechanical biological treatment plants (MBTC)
5.) Other organic fertilisers or soil amendments such as manure, organic soil amendments and growing media, mineral fertilisers (chapter 6)

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[4] The content of this report does not necessarily reflect an official position of the European Commission.

[5] In this context a PTEs are defined as chemical elements that have the potential to cause toxicity to humans, flora and/or fauna, depending on concentration, bio-availability and bio-accumulation. The definition of "PTEs" commonly refers to heavy metals; in contrast to OPs they are not of organic nature, and they may not be degraded by microbial activity or chemico-physical impacts. The latter can – in general terms – be addressed as "persistency" and has to be considered when assessing toxicological effects in time.

The contamination of foodstuffs with PTEs and OPs has been investigated for the typical diets as far as available in the literature with a view on regional and temporal variations. The latter are based on national surveys from Finland, France, Germany, Italy, Sweden and the United Kingdom.

A detailed evaluation of some national data sets of PTE concentrations in biowaste composts (BWC) and green waste composts (GC) is made, based on actual limit and guide values or on investigations for composts (chapter 4.3.3).

The study also presents a comparative survey of sampling and analytical methods, the variability of analytical results on the basis of inter-laboratory trials and specific investigations as well as statistical rules for admissible deviations within national regulations and standards. From the outcomes of the quoted investigations a general scheme for sampling and analytical procedure including a reliable system of tolerances for analytical results is presented (chapter 7).

Actual approaches for the definition of the quality of organic fertilisers and composts are discussed in chapter 8.

Finally, Chapter 9 presents an overall concept for the use of waste derived composts used as organic fertiliser in agriculture designed for the long-term maintenance of multifunctional productivity of agricultural soils.

The main sources used for the collection of quality data, especially for foodstuffs and composts were:

i. literature research beginning with approximately1990
ii. a review of unpublished studies and surveys
iii. a questionnaire sent out to experts of the national administration and research institutes in the EU Member States (see Annex 6)
iv. personal communication with national experts

Tables with quality data from national surveys as well as a chapter on national classification systems, material and use related standards, and limit values for composts may be found in Annex 1 - 5.

Since the majority of data on compost, manure, feed stocks and food and feeding stuff are available as "aqua regia" values, all further presentations and considerations of limit values, load restrictions and calculations of soil accumulation-scenarios are based on this extraction method. Where other extraction methods were used this is indicated in the concerned tables or figures (see also chapter 7.5)

1.2 General framework of compost production

Beginning in 1984 pilot projects were initiated in Germany, Austria, Switzerland and the Netherlands implementing the separate collection and composting of the organic waste fraction of municipal waste. These activities followed the newly set up waste management plans and the following hierarchy

- reduction and prevention
- recovery/recycling/reuse
- energy recovery/incineration and final disposal.

The main challenge facing the managers of waste is to develop strategies and techniques as alternatives to landfill for biodegradable municipal wastes in order to comply with the Article 5 targets set in the EC Council Directive on the Landfill of Waste (1999/31/EC) (the Landfill Directive targets).

The Directive places strict limits on the amount of biodegradable municipal waste that can be disposed of via landfill, and requires landfill operators to collect, treat and utilise landfill gas. In addition, the Directive introduces a requirement for pre-treatment of all waste prior to landfill.

Under the Directive the amount of biodegradable municipal solid waste that can be disposed of via landfill must be reduced to:

- 75% of the amount produced in 1995, by 2006
- 50% of the amount produced in 1995, by 2009
- 35% of the amount produced in 1995, by 2016

In general there are two strategies that may be pursued for reaching these targets:

- Recycling of source separated organic waste by aerobic (composting) or anaerobic (digestion in biogas plants) treatment
- Pre-treatment of residual waste before landfill by incineration, or mechanical-biological pre-treatment of residual waste

All strategies aim at a low emission landfill technology. The mechanical-biological pre-treatment of residual waste predominantly aims at volume reduction and stabilisation as well as the mechanical separation of partial material flows. In Austria and Germany, for instance, stability criteria with corresponding limit values were recently elaborated (*TOC, upper caloric value, gas generation potential*) in order to assure the general objectives set out in the EU Landfill Directive. Italy also drafted a decree concerning bio-stabilised materials.

The recycling of source separated organic waste results in high quality composts, which serve as an organic fertiliser and as carbon sink (Hogg et al., 2002). In addition, for anaerobic digestion the gain of methane (*bio-gas*) contributes positively to the green house effect by saving non-renewable energy resources.

In 1996, Europe made a second significant step towards an environmentally sound management of the organic waste when the Commission in the *Strategy paper for reducing methane emissions (COM, 1996)* identified composting as a first choice in terms of the aims of the Landfill Directive. The possible objectives of a European compost measure were explained in the framework of a European Compost symposium held in Vienna 1998 (BMUJF, 1999b):

- reduction of biodegradable waste destined for landfill or incineration;
- incentives to collect biodegradable waste separately;
- fix Community-wide standards for compost;
- ensure free circulation of (some qualities of) compost;
- standardise test methods for compost;
- increase the use of compost as soil improver

Enormous efforts have been made within the last 15 years covering all areas concerned such as

- logistics of separate collection systems
- technique of composting
- quality requirements, standardisation and legal framework
- public relations and information services
- quality management systems for composting plants
- marketing and product diversification
- research on the utilisation of compost in the broad areas of agriculture, gardening and landscaping

1.2.1 Organic waste production in EU15

Fig. 1-1 gives an abbreviated survey on the status of legal and structural implementation of source separation and composting systems in European countries. One can distinguish between four groups among European countries (Amlinger, 1999; Barth, 1999)

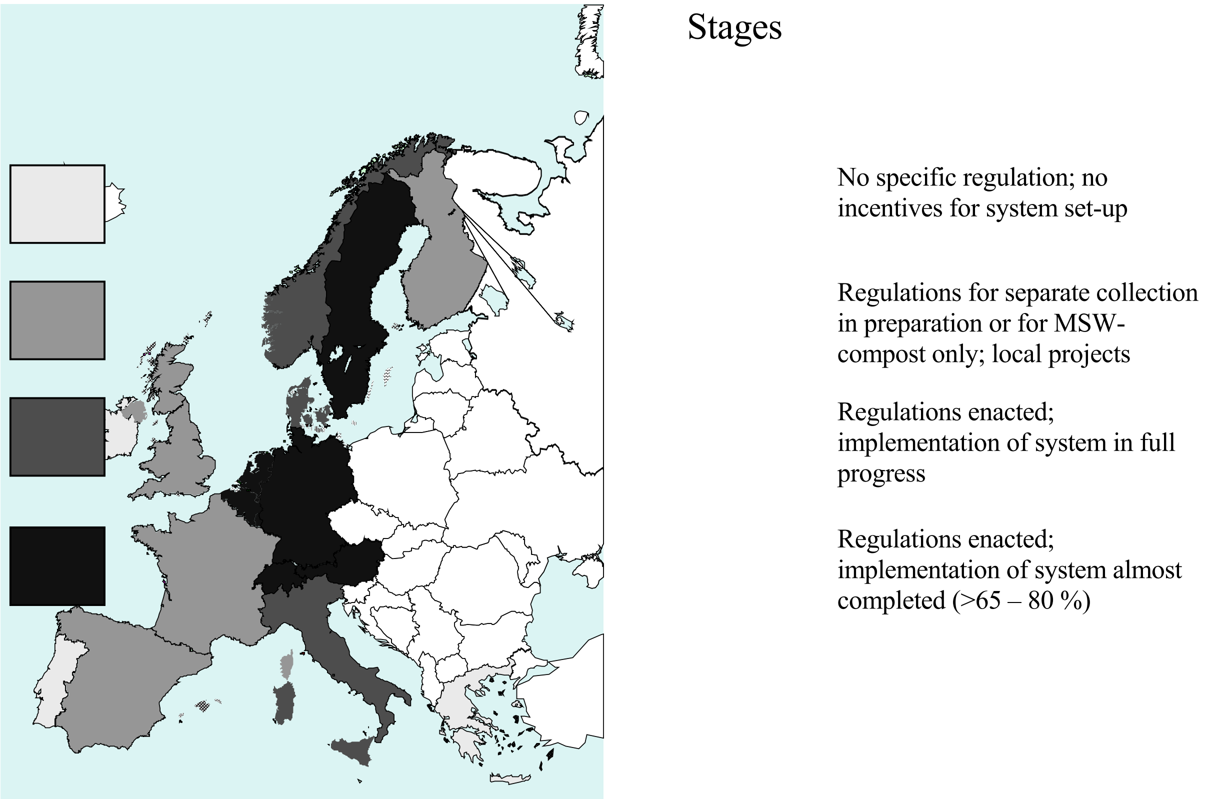

Fig. 1-1: Implementation of source separation for organic waste and legal framework thereto in 2002

Table 1-1 lists the figures on waste production in the EU and shows an estimation of the proportion of the organic waste fraction in (residual) municipal waste. The estimated 35 % putrescibles in MSW equal a total amount of ca. 70 million. tons. From feasibility studies and practical experiences in areas where source segregation is well established it is known that a residual part of organic waste of about 10 – 20 % will remain uncollected. Thus the 50 mio. tons of organic waste potential which might be collected separately at the source seems to be a reliable figure.

Table 1-1: Waste generation in the EU and organic fraction therein 1)

Member State	MSW (1,000 t)	MSW per capita (kg)	Organic fraction in MSW 2) % (m/m)
Austria (1999) a)	4,431	550	29% (1995) 17 - 35% [!] (1998) 3)
Belgium (1999) b)	5,462	538	Flanders: 48% (1996) Wallonia: 45% (1991
Denmark (1999) b)	3,329	636	37% (1994)
Finland (1997) b)	2,510	494	35% (1993)
France (1998) b)	37,800	631	29%
Germany (1996) b)	44,390	550	32%
Greece (1997) c)	3,900	372	49%
Ireland (1998) b)	2,057	579	29%
Italy (1998) b)	26,846	469	33% 3)
Luxembourg (1998) b)	184	447	44%
Netherlands (1999) b)	9,359	601	38%
Norway (1999) b)	2,650	609	(?)
Portugal (1999) b)	4,364	542	44%
Spain (1997) b)	24,470	617	44%
Sweden (1998) b)	4,000	457	25%
UK (1999) b)	33,200	571	21%
EU	**208,952**	**554**	**35%**

1) Source: a) Federal Environment Agency, Austria; b) Eurostat/OECD Joint Questionnaire 2000; c) Ministry of Environment, Greece; http://waste.eionet.eu.int/wastebase/quantities ;

2) does not include paper and cardboard

3) depending on settlement structure and investigation 4) without paper, board, industrial by products

Today 16.7 mio. tons biowaste (source separated organic household waste) and green waste are processed in biological treatment facilities. The total potential of organic waste would produce about 24 mio. tons of compost (f.m.). Assuming an average application rate of 10 – 15 t fresh matter (f.m.) per ha and year, the area demand would be about 2.5 mio. ha or 1 – 2 % of the total agricultural land (EU 15).

Table 1-2: Recovered biodegradable waste (including organic household waste and green waste) in the Community

Member State	**Bio/Green Waste potentially collectable** (1,000 t y^{-1})	**Bio/Green Waste actually collected** (1,000 t y^{-1})	**In % of potential**	**Compost actually produced (Bio+Green waste)** (1,000 t y^{-1})	**Compost actually produced (from unsorted MSW)** (1,000 t y^{-1})	**Compost potentially producible (Bio+Greenwaste)** (1,000 t y^{-1})
Austria (2000)	1,220/970	450/200	30%	250	100	400
Belgium (1999)					n.i.	
Flanders (1999)	1,300	340/390	56%	220		360
Wallonia (1996)	160	120	75%	40		64
total	1,460	850	58%	260		424
Denmark (1999)	100/660	37/650	90%	250	n.i.	360
Finland (2002)[2]	600	142	24%	60	n.i.	280
France (2000)	5,250/3,500	50/1,500	18%	25.7 (**) 500 (***)	532.9 (1998)	3,098 (*) 2,080 (****)
Germany (1999)	9,000	7,000	78%	2,500	1,500	3,600
Greece (1995)	1,800	0			n.i.	700
Ireland (1998)	440	0			n.i.	170
Italy (1998/99)	9,000	1,500	17%	600	550	3,600
Luxembourg (2002)[3]	124	40	32%	15	n.i.	40
Netherlands (2002)[4]	4,500	2,900	64%	700	n.i.	1,000
Portugal (1995)	1,300	10	1%		n.i.	500
Spain (2000)	6,600				428	2,500
Catalonia (2000)	1,270	30 / 20	4%	16		450
Sweden (2002)[5]	1,700	265	16%	100	n.i.	600
UK (2000)	3,200 (2006)	39/1,000	32%	405	n.i.	4,240
Total (rounded)	**49,700**	**16,700**	**34%**	**5,700**		**24,000**

Sources: www.composting.net/biowaste/biowaste.htm or as indicated below:
[1] Danish EPA (2002); [2] Puolanne (2002) ; [3] Mathieu (2002); [4] Brethouwer (2002); [5] Nilsson (2002)

(*) household wastes + trade, commercial, manufacturing & institutions wastes (collected by municipalities) => biodegradable wastes (50% of the amount produced is collectable = 3,744 kt y^{-1}) + paper (80% of the amount produced is collectable = 5,262.4 kt y^{-1}) (1995).
(**) household source separated biowastes (only) (1998)
(***) green wastes from household + public gardens (1998)
(****) 5,900 kt = 900 kt green wastes from public gardens (all are collected) + 5,000 kt household green wastes (there is 8,000 kt household green wastes but only 5,000 kt are potentially collectable) (1995).

1.2.2 Sustainable recovery measures

A central issue in many Member States for applying waste materials on land is the assessment of potential benefits from waste derived soil amendments versus the risks due to potential pollutants and hazardous compounds that may accumulate in the soil system as well as risks of transformation and transfer to ground and surface water, plants, animals and humans.

As a result, regulatory measures throughout the EU have taken into account the potential risk to the environment of various compost application scenarios. Nevertheless, these regulatory approaches for limiting pollutants, application rates, and sensible sites for the use of composts vary within a wide range in the European Union.

This was well documented by several studies and at two European Symposia organised in 1998 and 1999 by the Austrian Ministry for Agriculture and Forestry, Environment and Water Management in co-operation with the European Commission (DHV, 1996; BMUJF, 1999a; Hogg et al., 2002).

On the EU level, Article 4 of Directive 75/442 on Waste (1995) specifies the following principles for the recovery and deposition:

Member States shall take the necessary measures to ensure that waste is disposed of without endangering human health and without harming the environment, and in particular:

- without risk to water, air, soil and plants and animals,
- without causing a nuisance through noise or odours,
- without adversely affecting the countryside or places of special interest.

Furthermore the recovery and deposition methods are defined in this Directive (Annex II). For the production and application of compost the following processes have to be considered:

Recovery methods according to annex IIB:

R3: <u>Recycling/reclamation of organic substances</u>, which are not used as solvents (<u>including composting</u> and other biological transformation processes)

R5: Recycling/reclamation of other inorganic materials

R10: <u>Land treatment resulting in benefit to agriculture or ecological improvement</u>

R11: Use of wastes obtained from any of the operations numbered R 1 to R 10

The precondition for waste recovery therefore is that it is useful and not disadvantageous when regarded from a comprehensive ecological point of view.

1.3 Some basic considerations about the reuse of organic waste resources

Following the Technical Annex of the contract ,the main objective for this study was to elaborate a scheme for maximum allowable concentrations of pollutants in order ***to maintain soil quality in the long term***.

Consequently, the soil, as receptor and potential sink as well as source of *'risk compounds'*, has to be the centre of focus as the essential entity for the evaluation of application of waste derived soil amendments or organic fertilisers (see Fig. 1-2).

Whether a potential pollutant compound or element may perform an adverse impact on the environment depends on the concentration level in the relevant soil horizon. The critical concentration marks the threshold from where, when exceeded the soil pass from a sink into a potential source of 'risk compounds' to a receptor. This central element depends upon soil and site specific properties such as pH, cation exchange capacity (CEC) and cation distribution and must be based on experimental and field experience derived from eco-toxicological effects over defined pathways. With this static value still nothing is said, if the released contaminant would have a toxicological relevance for the final receptor media or entity concerned.

The final definition of a guide value for the multifunctional (predominantly agronomic) use of soil depends very much on the traditional perception of the value of soil as such and the qualitative and quantitative assessment of soil functions. This can then lead to a more or less precautionary approach (introducing a wider or smaller safety buffer).

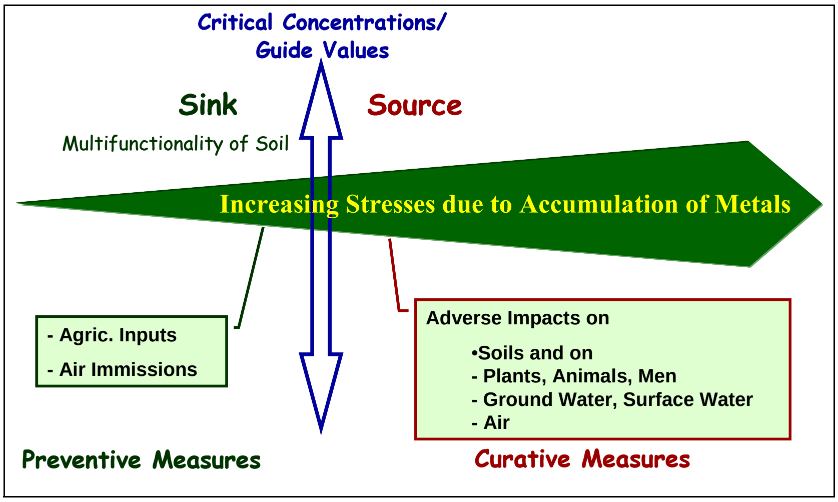

Fig. 1-2: Critical concentration decides whether the soil serves as sink or source of potential pollutants (Gupta, 1999)

The threshold value for the multi-functionality of the soil is the critical point of any sustainable soil management. Therefore, accumulation scenarios for persistent pollutants show that the critical concentration of the soil, above and beyond the quality and the amount of applied compost, plays the decisive role for the assessment of compost management options.

In order to assess whether an element concentration may become “critical” can only be judged if the following processes are understood

- Sorption and mobilisation dynamic under the relevant site and soil specific conditions
- critical charges with respect to the transfer rates and doses-(no)effect concentrations (e.g. PNEC) of the receptor concerned (e.g. soil biota, plant and transfer to plant organs, percolation and folter potential of pathway soil-sediment-groundwater etc.)
- and with special view on the nitrogen fluxes:
- critical balances related to potential N_2O emissions
- critical N balances inducing unbalanced luxury N consumption resulting in increased susceptibility for parasites and plant diseases

It has to be pointed out that critical thresholds might not be the same for all soil types, climatic and land use conditions. But key factors influencing this process might be identified together with conceptual approaches for establishment of sustainable management of potential diffuse sources.

Another crucial factor are the analytical methods, providing the empiric baseline for further estimations or models derived thereof. In direct measuring elements or compounds either ‘totals’ or certain extractable pools, the result is always an approximation of the reality. The correct interpretation very much depends on the knowledge about the coincidence of ‘concentration’ and effects. This is a major factor for an efficient monitoring program.

As a result all empirically or scientifically based elements of an overall strategy have to balance common rules which pertaining to a sustainable use of land within a broad range of applications and individual (‘*site specific*’) conditions which allow a certain variation for more (or even less) stringent rules.

Soil is the basis of life. Soil is the living space for men, animals, plants and soil organisms. It is a compartment of nature and its processes particularly with respect to water and nutrient cycles. Due to its capability to filter, buffer and transform substances, soil represents a natural medium of degradation, dynamic balancing and build-up processes.

The resources of the world are limited. Therefore the material recycling of waste in the agro-system is a rational measure if soil and water protection are considered as a priority. Once the need for certain constituents of organic waste (and products gained thereof by mechanical or biological treatment or transformation) is defined, their use may be addressed basically as an ecologically and economically reasonable measure. Negative attitudes towards waste products like sewage sludge or waste derived compost originate from

- Low quality (esp. in the past)
- Lack of knowledge

- Misleading information
- Prioritisation of disposal

However two entirely different concepts for sewage sludge and compost from source separation have to be distinguished:

Table 1-3: Concepts for the definition of sewage sludge and organic waste compost as potential organic fertilizer or soil amendment

	Sewage sludge	**Compost**
Principal nature of material	RESIDUE of waste water treatment	PRODUCT derived from the selective recovery of source separated organic waste
Main result of processing	(+/-) stabilised; (+/-) sanitised → SINK for potential pollutants [trace elements and organic pollutants] → Potential resources (plant nutrients = N, P, trace elements) are NOT THE TARGET but a SIDE-EFFECT of the process	(+) Stabilised (+) sanitised → by concentration RESIDUAL RISK of polluting the resources (organic matter, plant nutrients) → TARGETED MEASURE: reduction of landfill space and creation of a product providing • soil amelioration aiming at the maintenance and improvement of soil functions (buffer, filter, sorption, transformation, productivity) • constituent of manufactured soils and growing media
Risk management in processing and use of final product	• Entry of pollutants from diffuse sources. Wide spectrum of potential organic contaminant loads • Limited options of exclusion at the source • Degradation, concentration and subsequent separation of contaminants via selective processing techniques • Restriction of use mainly derived from maximum PTE load	• Entry of pollutants from diffuse sources is minimised by source separation • Residual contamination with potential pollutants as a result of the background concentration of the source materials • Additional decay and sorption respectively of organic pollutants via mineralisation and humification processes • Restriction of use mainly derived from *good agricultural practice* → BENEFITS (organic carbon, NPK supply)

For both compost and sewage sludge, the principle of not endangering the environment by unbalanced input of nutrients or pollutants may be regarded as a broadly accepted approach.

1.4 Inorganic and organic pollutants discussed in this report

Annex 1 provides a fact file with basic information on potential toxic elements (mainly heavy metals and other elements) and organic pollutants.

1.4.1 Potential toxic elements (PTEs)

The production and use of potential toxic elements has increased significantly in the course of the last century (Table 1-4). Thus the input of heavy metals into the environment from anthropogenic sources exceeds the geogenic one. As a result anthropogenic inputs now are the primary contributor to the accumulation of heavy metals in the environment.

Table 1-4: Production of heavy metals worldwide in 1000 t y^{-1} (Senesi et al., 1999 quoted in Herter et al., 2001))

Element	Production [1000 t y^{-1}]			
	1930	1950	1970	1985
Ag	-	6.2	9.4	13
As	-	47	49.6	45
Bi	-	1.4	3.7	8.3
Cd	1.3	6.0	17	19
Cr	560	2,270	6,057	9,940
Cu	1,611	2,650	6,062	8,114
Mn	3,491	5,800	18,240	-
Mo	-	15	82	98
Ni	22	144	629	778
Pb	1,696	1,670	3,395	3,077
Sn	179	72	32	94
V	-	1.8	18	34
W	17	39	32	47
Zn	1,394	1,970	5,465	6,024
Zr	-	-	399	775

Based on national and European standards and regulations the following elements are covered in this study:

Standard potential toxic elements (PTEs):

Cadmium (Cd)
Chromium (Cr)
Copper (Cu)
Mercury (Hg)
Nickel (Ni)
Lead (Pb)
Zinc (Zn)

Further trace elements (as far as available in literature):

Aluminium (Al)
Arsenic (As)
Boron (B)
Beryllium (Be)
Cobalt (Co)
Iron (Fe)
Manganese (Mn)
Molybdenum (Mo)
Sodium(Na)
Antimony (Sb)
Selenium (Se)
Tin (Sn)
Tellurium(Tl)
Vanadium (V)

1.4.2 Organic pollutants (OPs)

Organic pollutants are produced by human activities: industry, transportation or the use of pesticides. Some leave residues that may persist in the environment, either intact or as metabolites that may be more toxic than the original product.

From the huge number of organic pollutants which may be found in environment compartments, the persistent ones (POPs) are of major concern. POPs are persistent in the environment, may have long half-lives in soils, sediments, air or biota (half-life of years or decades in soil/sediment and several days in the atmosphere). POPs are typically ‘water-hating' and ‘fat-loving' chemicals, i.e. hydrophobic and lipophilic. In aquatic systems and soils they partition strongly to solids, notably organic matter, avoiding the aqueous phase and in biota since metabolism therein is slow. POPs may therefore accumulate in food chains (Jones & de Voogt, 1999).

The most prominent POPs are the substances regulated in the Stockholm Convention on Persistent Organic Pollutants. Parties signing the Convention have committed to eliminate the production and import/export of the substances listed in Appendix A of the Convention and to restrict production and use of chemicals listed in Appendix B of the Convention. Currently aldrin, chlordane, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene and PCBs are listed in Appendix A and DDT in Appendix B.

Some are emitted accidentally and others are - or were in the past - released into the environment deliberately. Many on the list of potential problem chemicals are better known by their abbreviations: PCBs, PAHs, PCDD/F and DDT for example. They are widespread and some are inevitably present in raw materials for composting. Very few of the regulations examined set limits for organic pollutants for compost from organic waste. Similarly there are many different chemicals that would need selective testing, which is difficult and expensive. Some countries regulate levels of certain chemicals in sewage sludge, e.g. Denmark (Nonylphenols, PAH, DEHP and LAS), Sweden (Nonylphenols, PAH, PCB and Toluene) and Germany (PCB, PCDD/F and AOX). A comparison of typical levels in compost and sewage sludge, as given in the AFR report (Johanson, C., et al. 1997), show that contamination in compost is generally much lower than that in sewage sludge. The German RAL GZ-251 standard set a voluntary limit for pesticide content at <0.5 mg L^{-1} of fresh mass and discussions are being carried out of limits for other pollutants such as PCBs and Dioxins. Through careful selection of feedstock it should be possible to reduce some of the potential contamination. This implies knowledge of the sources of contamination, such as road sweepings, and the origins of feedstock.

Lindane is sometimes found in bark and timber, especially in countries where no regulations exist, i.e. outside the EC. Residues from medical treatments such as antibiotics may also find their way into compost. CEN/TC 223 WG2 reported that residues from the treatment of fish were found in Norway, where fish waste is included in compost feedstock. This should be easy to eliminate at the source if it is considered a problem. Little information is available, however, on the behaviour of animal veterinary medicaments when introduced to the environment. (Bywater, 1998)

From the large number of potentially dangerous and damaging compounds the chlorinated pesticides, the polychlorinated biphenyls (PCB), the polycyclic aromatic hydrocarbons (PAH) as well as the polychlorinated dibenzodioxins (PCDD) and the dibenzofurans (PCDF) are considered to be ecologically significant due to their high stability and toxicity. Set free into the environment by direct application or indirectly by transport with waste water, solid waste and air, these chemicals contaminate soils and growing plants mainly by air deposition and thus, potentially, also the raw materials for composting (Fricke & Vogtmann, 1993).

Recently, particular interest has been addressed to substances occurring ubiquitously in the environment similar to POPs, which are not persistent. Their presence in the environment is explained by high production volumes, a large range of application and thus a continuous release to the environment. Examples for this class of substances are phtalates or some fragrances such as polycyclic musks. Those are mainly considered to be found in municipal sludges (Brändli et al., 2003; Dreher, 2003)

To be certain to reduce contamination the best course of action, after up-stream measures as the reducing or excluding compounds at the source of industrial production, would seem to be to select feedstock carefully with a view to excluding potentially problematic chemicals, and to manage the process so as to maintain aerobic conditions throughout composting. If deemed appropriate, tests done at intervals can be used as a fail-safe mechanism if contamination is suspected to increase confidence in the end product.

The options for minimising organic pollutants could thus be summarised as:

- Segregate organic raw material is at source and refuse raw material as necessary
- Operate an aerobic composting process to provide effective microbiological decay where ever possible
- Test for suspected contaminants only as a last resort

A selection could be done as follows (Brändli et al., 2003):

- Ubiquitous compounds with ongoing release from primary sources mainly due to anthropogenic production such as emissions from incineration of fuel (e.g. polycyclic aromatic hydrocarbons, PAHs) or from volatilisation or leaching of high volume chemicals (e.g. flame retardants),
- Ubiquitous compounds banned for production but still present in anthropogenic repositories (e.g. building parts) and in natural sinks like sediments, soils or water bodies due to their high persistence (e.g. POPs such as polychlorinated biphenyls, PCBs),
- Pesticides or biocides, which in contrast to the former two categories are released to the environment deliberately due to application (e.g. on plants).

The Decision Making Scheme – factors considered to single out investigated compounds

According to the terms of reference of the tender, the Study Team had to propose a series of compounds to be investigated, some of which (PCBs, PAHs, Dioxins and Furans) were explicitly listed in the proposal – albeit as a "for instance".

Given the extremely broad variety of organic chemicals occurring in the "technosphere" (thousands HPVC = high production volume chemical are registered, which are defined, according to Directive 793/93, as those chemicals for which there is at least one producer/importer in volumes exceeding 1000 tonnes per year), the team proposed "to focus on those compounds for which guide or threshold values may be identified either for foodstuff or for soils and soil amendments" (Bidding Proposal, Section 3.1 "Identification of pollutants in foodstuffs – Methods and concepts")

The rationale for such an approach was to keep consistent with the current ecotoxicological concern; as a matter of fact, it was not the scope of the tendered study to investigate ecotoxicological effects of various compounds, nor were the required skills fit to this scope. Rather, the survey had to investigate presence (in organic materials to be used as fertilisers) of elements and compounds for which toxicologists have stressed hazardousness, which called for specific legislation to restrain their diffusion onto farmlands and into the food chain.

Otherwise speaking, the lack of certain compounds in standards/regulations for food and soil provides for an indicator that, so far, toxicologists have not expressed concern on such compounds, or they have not achieved consensus on such concern.

Of course, implementation of new regulations or an update of existing ones may reach a wider coverage, including new compounds, but this is basically a task for toxicologists. As an effect of setting threshold values for new compounds, standards may be derived for soil (and subsequently for compost) by scientists in the field of land management. The concept for the definition of threshold concentrations in materials to be used as organic fertilisers may be applied, mutatis mutandis, also to new elements and compounds to be controlled by legislation or by the guidance measures.

Many of ubiquitous compounds like PCBs, PAHs and PCDD/F exhibit a low acute toxicity however. It is generally accepted, that these substances induce chronic effects on reproduction in mammals and aquatic organisms. Moreover, immunotoxic, genotoxic and endocrine effects are known. Knowledge related to these effects for terrestrial organisms is limited.

It must be stated that in general, knowledge in terrestrial ecotoxicity is incomplete and often insufficient for adequate risk assessments (Aldrich & Daniel, 2003).

Table 1-5 was taken from Arthur Andersen on behalf of DG Environment (2001) summarising, occurrence and effect of the most cited organic contaminant groups found sludge. We inserted a column for compost and added the data were available.

Table 1-5: Properties, occurrence, degradation and transfer of organic contaminant groups found in sewage sludge (Smith, 1996, quoted from Arthur Andersen on behalf of DG Environment, 2001) compared to data of biowaste and green compost

Compound Group	Physico-chemical properties	Conc. in compost	Conc. in sludge	Degradation	Leaching Potent.	Plant uptake	Transfer to animals
Polynuclear aromatic hydrocarbons (PAHs)	Water soluble/volatile to lipophilic	BWC: 0.8-4.5 mg kg^{-1} GC: 1.7-3.8 mg kg^{-1}	1-10 mg kg^{-1}	Weeks → 16 years Strongly adsorbed by soil O.M.	None	Very poor	Possible but rapidly metabolised
Polychlorinated Biphenyls (PCBs)	Complex, > 200 congeners low water solubility, highly lipophilic and semi-volatile	BWC: 0.03-0.86 mg kg^{-1} GC: 0.015-0.48 mg kg^{-1} mixed BWC/MC: 0.02-1.68 mg kg^{-1}	1-20 mg kg^{-1}	Very persistent Half-life ca. 12 years Strongly adsorbed by soil O.M.	None	Root retention Foliar absorption Minimal root uptake and translocation	Possible into milk/tissues via soil ingestion Long half-life
Polychlorinated dibenzo-P-dioxins and Furans (PCDD/Fs)	Complex, 75 PCDD congeners, 135 PCDF congeners, Low water solubility, highly lipophilic and semi-volatile	BWC: 4.3-17.5 ng I-Teq kg^{-1} GC: 3.6-13 ng I-Teq kg^{-1}	Very low <few µg kg^{-1}	Very persistent Half life several years Strongly adsorbed by soil O.M.	None	Root retention Foliar absorption Minimal root uptake and translocation	Possible into milk/tissues Via soil ingestion Long half-life
Phtalate acid esters	Generally lipophilic, hydrophobic and non-volatile	DEHP: 0–69.6 mg kg^{-1} Means: BWC: 1.4–9.6 mg kg^{-1} GC: 0.4–2.9 mg kg^{-1}	High 1-100 mg kg^{-1}	Rapid Half-life <50 days	None	Root retention Not translocated	Very limited
Linear alkybenzene-solphonates (LAS)	Lipophilic	Only one investigation > 0: 9–396 mg kg^{-1} Mean: 41 mg kg^{-1}	Very high 50-15000 mg kg^{-1}	Very rapid in aerobic environment	None	None	None
Alkylphenols	Lipophilic	~ 200 µg kg^{-1}	100-3,000 mg kg^{-1}	Rapid < 10 days	None	Minimal	Minimal
Organoclorine pesticides	Varied, lipophilic to hydrophilic, some volatile	0.6 – 100 µg kg^{-1}	<Few mg kg^{-1}	Slow> 1 year Loss by volatilisation	None	Root retention Translocation not important Foliar absorption	Via soil ingestion persistent in tissues
Monocyclic aromatics	Water soluble and volatile	n.i.	<1-10 mg kg^{-1}	Rapid	Moderate	Limited due to low persistence Rapidly metabolised	Rapidly metabolised
Chlorobenzenes	Water soluble/volatile to lipophilic	HCB 0-156 µg kg^{-1}	<0.1-50 mg kg^{-1}	Lower mol wt types lost by volatilisation Higher mol wt types persistent	High to low	Possible via roots and foliage Maybe metabolised	Important for persistent compounds
Short-chained halogenated aliphatics	Water soluble and volatile	n.i.	0-5 mg kg^{-1}	Lower mol wt types lost by volatilisation Higher mol wt types persistent	Moderate	Foliar absorption Possible translocation	Low
Aromatic and alkyl amines	Water soluble and low volatility	n.i.	0-1 mg kg^{-1}	Slow	High	Possible	Low
Phenol	Varied, lipophilic high water solubility and volatile	n.i.	0-5 mg kg^{-1}	Rapid	Moderate to low	Possible via roots and foliage	None

BWC…Biowaste Compost (including source separated organic household/kitchen waste); GC…Green Compost (from garden and park waste only); n.i. … not identified in literature

2 IDENTIFICATION OF POLLUTANTS IN FOODSTUFFS

The scope of this section is to supply a comprehensive overview of average concentration values (and, whenever available, standard deviations) of most relevant inorganic and organic pollutants in foodstuffs deemed for human and animal consumption.

Numbers herewith reported have been taken from various surveys on foodstuffs carried out throughout last 20 years.

2.1 Adopted methods

To carry out the survey, the following steps have been taken:

[A] Main categories of foodstuffs for human and animal consumption have been singled out according to the following list:

- Cereals
- Potato
- Rice
- Fruit
- Vegetables (roots, tubers, fruits)
- Vegetables (leafy)
- Pulse (legumes)
- Milk
- Milk products
- Eggs
- Meat
- Fish
- Animal feed

[B] Organic and inorganic pollutants of main concern, which may be detected in foodstuffs destined for human and animal consumption, have also been listed.

The following have been considered:

- Potentially Toxic Elements (PTEs): Arsenic, Cadmium, Chromium, Copper, Lead, Mercury, Nickel, and Zinc.
- Persistent organic pollutants (POPs): PCB, PAH, toxaphene, dioxin, aldrin/dieldrin, PCCD/F

[C] An overview of concentrations of the PTEs and POPs in various categories of foodstuffs has been carried out (expressed either as dry or wet matter), based on worldwide surveys.

International literature on features of foodstuffs is admittedly very far-reaching. Nevertheless, it mainly refers to those mineral and organic compounds that give a certain nutrient value to the foodstuff. There is no comprehensive survey on concentrations of polluting/toxic elements. Sometimes, some PTEs (as for instance, Zinc) are simply mentioned as elements that may be somewhat beneficial to human health.

To pull together some relevant figures we have investigated various sources reporting on surveys carried out in several countries. One main problem is, *the testing methods are either sometimes misleadingly mentioned, or entirely omitted.* This explains to some extent the extreme variation of numbers that have been reported.

[D] Numbers reported in such surveys, have been collected in detailed tables, where we sought a certain homogeneity of figures by grouping numbers referring to the same pollutant, and to same type of foodstuff. As numbers are given in various surveys in an extremely varied way, tables have been structured in different columns, which report on concentrations referred to dry or fresh matter, the maximum, minimum, mean and median values, the standard deviation, depending on available data.

[E] We have then organised some summary tables where we have reported on mean and range values, whenever available, grouping types of foodstuffs into their main categories.

2.1.1 Sources and related problems

The first step to collect as much information as possible, was to draw a questionnaire about Heavy Metals (Potentially Toxic Elements, PTEs) and Organic Pollutants (OPs) in food and feeding stuffs; this was sent to Ministries in various Member States, Research Centres, Universities, Agencies for the Protection of the Environment, public health, alimentation, food quality Institutions and Agencies.

References, addresses and contacts were found both in our address books and through an internet search. Contacted Countries were: Belgium, Greece, Spain, Ireland, Luxemburg, France, Netherlands, Portugal, Finland, Sweden, United Kingdom, Austria, Island, Norway, Denmark, Italy, Germany, Switzerland.

Unfortunately, we got very few acknowledgments, probably due to the following reasons:

- Lack of motivation of experts in MSs, as far as the concentration in materials other than commodities is concerned; arguably, the effect on the food chain by concentrations in food scraps is not so straightforward.
- the experiences on investigation of PTEs and OPs in foodstuffs are not so widespread (detection is mostly available only in quite narrow food categories, i.e. “fish”, or only for a few heavy metals).

The majority of the suggestions to better finalise our search were collected through the internet. Websites and URLs were found through many search engines (as altavista, google, yahoo, lycos, excite,…).

The research was carried out with different methods:

- general queries on heavy metal and organic pollutants in food and feedstuffs;
- particular queries on specific metals or organic compounds;
- contacts to institutions working on the topics to find surveys, scientific documents and/or legislation.

In parallel national and European institutions have been contacted and the following sources have been used:

A.) National Ministries for Nutrition in the Member States

B.) Swedish EPA

C.) France : INRA (Institut National de recherche agronomique)

D.) Codex Alimentarius of the FAO and WHO

E.) Commission Regulation (EC) No 466/2001 from 8 March 2001, OJ Nr. L77/1 (Maximum Levels for certain Contaminants in Foodstuffs)

F.) Council Directive 2001/102/CE of 27 November 2001 amending 1999/29/CE

G.) Council Regulation 2001/2375/CE of 29 November 2001 amending 2001/466/CE

H.) Commission Regulation 2002/221/CE of a February amending 2001/466/CE

I.) Commission Recommendation 2002/201/CE of 4 March 2002: dioxin, furans and PCBs reduction in feed and foodstuffs

J.) Council and Parliament Directive 2002/32/EC of 7 May 2002 on undesirable substance in animal feed

K.) Minutes of the SCF meeting of 10-11 July 2001: PAH in pomace oils

L.) Minutes of the SCF meeting of 24 October 2001: PCBs in foodstuffs, aflatoxins in raw cereals, heavy metals, 3-MCPD, nitrates in lettuce, spinach, musks

M.) Codex Alimentarius Austriacus with the Decree of the Ministry for Women Affairs and Consumer Protection from 10 March 1997 (EZ 32.007/2-Vl/B/1b/96); containing guide values for Pb, Cd and Hg in several foodstuffs

N.) Germany: Guide Values for Foodstuffs (Anonymous, 1989) and for fodder (Hecht, 1982).

2.2 Relevant pollutants in regulations and standards

2.2.1 Introduction

Contaminants are substances that have not been intentionally added to food. These substances may be present in food as a result of the various stages of its production, packaging, transport or holding. They also might result from environmental contamination.

2.2.2 Existing Legislation

The basic principles of EU legislation on contaminants contained in food are in Council Regulation (EEC) 315/93 of 8 February 1993:

- food containing a contaminant to an amount unacceptable from the public health viewpoint and in particular at a toxicological level, shall not be placed on the market
- contaminant levels shall be kept as low as can reasonably be achieved following recommended good working practices
- maximum levels must be set for certain contaminants in order to protect public health.

Maximum levels are set for certain contaminants in foodstuffs in Commission Regulation 466/2001 of 8 March 2001 and in the subsequent amendments as follows:

- nitrate in lettuce and spinach (Commission Regulation 563/2002)
- aflatoxins in nuts, dried fruit, cereals, spices and milk (Commission Regulation 257/2002 and Commission Regulation 472/2002)
- the heavy metals lead, cadmium and mercury in a range of foods (Commission Regulation 221/2002
- 3-monochloropropane diol (3-MCPD) in soy sauce and hydrolysed vegetable protein dioxins in a range of foods (Council Regulation 2375/2001
- ochratoxin A in cereals and cereal products and dried vine fruit (Commission Regulation 472/2002)

Proposals for maximum levels of other contaminants, such as patulin and fusarium toxins (deoxynivalenol and other trichothecenes, zeaalenone, fumonisins) are being considered. Also, work is ongoing to reduce levels in food and to clarify the possible risks from substances such as polycyclic aromatic hydrocarbons (PAHs), organotins and acrylamide.

In addition, provisions for the sampling and analysis for the official control of the abovementioned levels have been set

- for aflatoxin levels: Commission Directive 1998/53/EC amended by Commission Directive 2002/27/EC
- for levels of heavy metal and 3-MCPD: Commission Directive 2001/22/EC
- for dioxin levels: Commission Directive 2002/69/EC
- for ochratoxin A levels: Commission Directive 2002/26/EC

Annex 3 summarises the collection of regulation on PTEs and organic pollutants issued by the European Union and by some Member States following the Communitarian dispositions. The evolution of the integrations and amendments is reported.

Potential toxic elements [PTEs] – heavy metals in Foodstuffs

Section 1 in Annex 3 presents a data collection of the EU regulations on PTEs in foodstuffs and animal feed, namely Cadmium, Arsenic, Lead and Mercury.

In addition regulations of some Member States (Spain, Austria and Germany) are listed which, after having assimilated the EU regulation, have issued national laws governing other contaminants or other food commodities.

Pesticides in Foodstuffs

The Community has in place a regime permitting the setting, on a scientific basis, of maximum residues limits (MRLs) in order to adequately protect the whole consumer population, including infants, and based on shared responsibility between the Community and the Member States. The work is under constant review. Legislation for pesticide residues, including the setting of MRLs in food commodities is a shared responsibility of the Commission and the Member States. To date, more than 17,000 Community MRLs have been set for various commodities for 133 pesticide active substances. It is important to note that these MRLs are not maximum toxicological limits. They are based on good agricultural practice and they represent the maximum amount of residue that might be expected on a commodity if good agricultural practice was adhered to during the use of a pesticide. Nonetheless, when MRLs are set, care is taken to ensure that the maximum levels do not give rise to toxicological concerns. For those pesticide/commodity combinations where no Community MRL exists, the situation is not harmonized and the Member States may (subject to satisfying their obligations under the Treaty) set MRLs at national level to protect the health of consumers.

Legislation at Community level dates back to November 1976 when Council Directive 76/895/EEC fixed MRLs for 43 active substances in selected fruits and vegetables. The MRLs that were set in the Directive were based on the best data available at that time. These older MRLs are gradually being reviewed and, where appropriate, being replaced with newer MRLs based on the newer information and the higher standards of today.

Current pesticide MRL legislation is derived from/based on four Council Directives:

1.) Council Directive 76/895/EEC establishing MRLs for selected fruits and vegetables
2.) Council Directive 86/362/EEC establishing MRLs for cereals and cereal products;
3.) Council Directive 86/363/EEC establishing MRLs in products of animal origin; and
4.) Council Directive 90/642/EEC establishing MRLs in products of plant origin, including fruits and vegetables.

Lists of these and all relevant amending legislation in this area and of all foodstuff commodities for which Community MRLs have been set for some 150 pesticides are given in Annex 3

2.3 Survey on PTEs and OPs in food and feeding stuff

2.3.1 PTE (heavy metal) levels in food and feeding stuff

In the following summary tables, various types of foodstuffs have been grouped into main categories. Mean and range concentration values of various PTEs are reported either on d.m. or f.m. (depending on availability)

Sources for Tables 2-1 to 2-8: Genevini et al. (1983), Swedish environmental protection agency survey, Storelli & Marcotrignano (2001a), Storelli & Marcotrignano (2000a), Bruggemann et al.. (199?), Page et al (1988), Centemero (1991), Storelli & Marcotrignano (2001b), Storelli & Marcotrignano (1999), Storelli & Storelli & Marcotrignano (2000), Santoprete (1997), Mordenti & Piva (1997), Storelli & Marcotrignano & Giacomelli. (1998), Storelli & Marcotrignano (2000b), Storelli & Marcotrignano (2001c)

Table 2-1: Arsenic (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	**MEAN**		**RANGE**	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS	0.019			0.016-0.058
POTATO				
RICE				
FRUIT		0.7		
VEGETABLES (ROOTS, TUBERS, FRUITS)		0.234		
VEGETABLES (LEAFY)		1.40		
PULSES		0.12		
MILK				
MILK PRODUCTS				
EGGS				
MEAT				
FISH	34.05	29.66	10.45-69.15	6.15-58.8
FEEDING STUFF (vegetal raw materials)		0.7		0.13-2.15
FEEDING STUFF (mineral raw materials				
FEEDING STUFF (proteinic raw materials)				
FEEDING STUFF (whole product feedstuff)				

Table 2-2: Cadmium (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	**MEAN**		**RANGE**	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS	0.048	0.019		
POTATO	0.108		0.009-1.0	
RICE			<0.001-0.25	
FRUIT		0.0019		
VEGETABLES (ROOTS, TUBERS, FRUITS)	0.019	1		
VEGETABLES (LEAFY)	0.38	18.48	0.034-3.8	
PULSES	0.61	0.25	0.001-2.17	
MILK	< 0.001			
MILK PRODUCTS				
EGGS				
MEAT				
FISH	0.55	0.59	0.04-1.04	<0.002-0.83
FEEDING STUFF (vegetal raw materials)				
FEEDING STUFF (mineral raw materials	0.57		0.34-0.79	
FEEDING STUFF (proteinic raw materials)	0.64		0.28-0.99	
***FEEDING STUFF** (whole product feedstuff)*	0.16		0.08-0.24	

Table 2-3: Chromium (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	f.m.	d.m.	f.m.
CEREALS	0.069	0.102		0.0011-0.018
POTATO	0.40	0.0675	0.55-0.57	0.012-0.060
RICE	0.07	0.055	0.12-0.23	
FRUIT	0.355	0.029		
VEGETABLES (ROOTS, TUBERS, FRUITS)	0.698	0.065		0.01-0.160
VEGETABLES (LEAFY)	0.852	0.0607	0.042-2.4	
PULSES	0.552	0.206		0.024-0.8
MILK				
MILK PRODUCTS				
EGGS		0.02		0.005-0.02
MEAT	0.003	0.14		
FISH	2.251	0.471	0.04-4.4	0.001-1.10
FEEDING STUFF *(vegetal raw materials)*	3.93		1.2-10.0	
FEEDING STUFF *(mineral raw materials*	110		46-538	
FEEDING STUFF *(proteinic raw materials)*	8.25		2.5-14	
FEEDING STUFF *(whole product feedstuff)*	8.07		1-25	

Table 2-4: Copper (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	f.m.	d.m.	f.m.
CEREALS	5.37	5.10	4-16.1	2.6-11
POTATO	4.9			
RICE				
FRUIT				
VEGETABLES (ROOTS, TUBERS, FRUITS)	9	9.23		
VEGETABLES (LEAFY)	19.97	18.75	8.51-41.13	
PULSES	17	8.55		
MILK	0.3			
MILK PRODUCTS				
EGGS				
MEAT	2.1			
FISH				
FEEDING STUFF *(vegetal raw materials)*	9.42		3-24.2	
FEEDING STUFF *(mineral raw materials*	991		332-1650	
FEEDING STUFF *(proteinic raw materials)*	222.5		214-231	
FEEDING STUFF *(whole product feedstuff)*	43.67		14-75	

Table 2-5: Mercury (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	f.m.	d.m.	f.m.
CEREALS		< 0.01		
POTATO				
RICE				
FRUIT				
VEGETABLES (ROOTS, TUBERS, FRUITS)				
VEGETABLES (LEAFY)				
PULSES				
MILK				
MILK PRODUCTS				
EGGS				
MEAT				
FISH	0.17			0.05-4.26
FEEDING STUFF *(vegetal raw materials)*				
FEEDING STUFF *(mineral raw materials*	0.03		0.02-0.03	
FEEDING STUFF *(proteinic raw materials)*	0.09		0.08-0.09	
FEEDING STUFF *(whole product feedstuff)*	0.21		0.05-0.38	

Table 2-6: Nickel (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	f.m.	d.m.	f.m.
CEREALS	0.283	1.407		0.03-2.69
POTATO	0.187			
RICE				
FRUIT		0.4		
VEGETABLES (ROOTS, TUBERS, FRUITS)		1.859		0.11-0.6
VEGETABLES (LEAFY)		3.66		
PULSES		2.723		
MILK	< 0.001			
MILK PRODUCTS				
EGGS				
MEAT	0.014			
FISH				
FEEDING STUFF *(vegetal raw materials)*		17.8		3-44
FEEDING STUFF *(mineral raw materials*	9.55		5.10-14	
FEEDING STUFF *(proteinic raw materials)*	6.9		6.7-7.1	
FEEDING STUFF *(whole product feedstuff)*	7.53		4.5-10	

Table 2-7: Lead (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS	0.04	0.0099	<0.001-3.6	0.0048-0.028
POTATO		0.04	0.001-2.2	0.01-0.14
RICE			< 0.001-0.08	
FRUIT		0.04		0.01-0.37
VEGETABLES (ROOTS, TUBERS, FRUITS)	0.004	0.035	0.002-0.46	0.01-0.38
VEGETABLES (LEAFY)		0.083	0.036-2.3	0.01-0.51
PULSES	0.005		<0.001-0.35	
MILK	0.052			
MILK PRODUCTS				
EGGS				
MEAT	0.012			
FISH		0.76		0.04-5.26
FEEDING STUFF *(vegetal raw materials)*				
FEEDING STUFF *(mineral raw materials*	1.95		1.2-2.7	
FEEDING STUFF *(proteinic raw materials)*	0.98		0.56-1.4	
FEEDING STUFF *(whole product feedstuff)*	0.74		0.58-0.87	

Table 2-8: Zinc (mg kg^{-1}) concentrations in food and feeding stuff

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS	29	25.	11-76	4-26
POTATO	14		5.1-35	
RICE		10	7.7-23	
FRUIT				7-44
VEGETABLES (ROOTS, TUBERS, FRUITS)		41	3.8-61	31-100
VEGETABLES (LEAFY)	190.19	127.25	13-334.4	
PULSES		40.4	17-70	
MILK	32			
MILK PRODUCTS				
EGGS				
MEAT	79.83			
FISH		5.225		
FEEDING STUFF (vegetal raw materials)	21.57		11-57.1	
FEEDING STUFF *(mineral raw materials*	2,006.5		843-3,170	
FEEDING STUFF *(proteinic raw materials)*	1,162		844-1,480	
FEEDING STUFF *(whole product feedstuff)*	172.33		54-266	

2.3.2 Remarks on spatial and temporal variation

The variation in reported concentrations of PTEs for each type of foodstuff was considerable. In some cases, the maximum value has been reported (see for instance Zinc in leafy vegetables, Ni in cereals) at levels, which are some orders of magnitude higher than the minimum value.

Such variability may result from:

- *spatial variation*: the overall uptake of heavy metals may be affected by geogenetic factors (presence of any such metal in the parent rocks and therefore in soils) and by the diffusion of anthropogenic sources in different districts (with specific reference to industrial sources)
- *temporal variation*: analyses have been carried out at different times and this may be linked, in turn, to all other affecting factors, such as new test methods, development of local industrial activities, etc.
- *heterogeneity of sampling and test methods*: in most cases surveys where numbers have been reported specify neither the sampling nor the test method. Needless to say, a very important factor is that concentrations detected for heavy metals are deeply affected by the extraction method (aqua regia; sulphuric acid + hydrogen peroxide; nitric acid + perchloric acid, etc)
- *intra- and inter-laboratory variations*: for any given test method, results may be affected in different labs – and to a certain extent, in the same lab - when samples are managed by different researchers, by different pre-treatment methods and/or different analytical instruments.

The following tables compare PTEs threshold values set by EU legislation with values found in food commodities and feeding stuff.

The following should be remarked:

- some countries have actually issued specific regulation, whose structure partly differs from the tables below, in that they either cover different contaminants or refer to other types of commodities.
- particular care should be there assumed when comparing threshold values, which are expressed on fresh matter basis to actual concentrations, which sometimes are expressed on dry matter or lipid basis;
- very often reported concentrations refer to experimental conditions where a particularly high concentration of pollutants have been induced in the soil on purpose in order to detect effects on the food chain

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Table 2-9: CADMIUM - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	EU THRESHOLD mg/kg f.m.	REF.	VALUE/RANGE ASSESSED	REF.
Meat of bovine and poultry	**0.05**	**a**		
Horsemeat	**0.2**	**a**		
Liver of cattle, sheep, pig and poultry	**0.5**	**a**		
Kidney of cattle, sheep, pig and poultry	**1.0**	**a**		
Bivalve molluscs	**1.0**	**a**	**< 0.002 – 0.83 f.m.**	Storelli & Marcotrignano (2001b)
Cephalopods (without viscera)	**1.0**	**a**	**0.14 – 1.04 d.m.**	Storelli & Marcotrignano (1999[p42])
Cereals excluding bran, germ, wheat grain and rice	**0.1**	**a**	**0.01 – 1.7 d.m** **0.0005 – 0.35 d.m.** **0.0022 – 0.011 f.m.**	Genevini et al. (1983) Page et al (1988) Swedish environmental protection agency survey
Bran, germ, wheat grain and rice	**0.2**	**a**	**0.005 – 0.22 d.m.**	Genevini et al. (1983) Page et al (1988) Swedish environmental protection agency survey
Soybeans	**0.2**	**a**	**0.001 – 1.20 d.m.**	Genevini et al. (1983) Page et al (1988)
Vegetables excluding leafy vegetables, fresh herbs, all fungi, stem vegetables, root vegetables and potatoes	**0.05**	**a**	**0.009 – 1.0 d.m.** **0.2 - 2.0 f.m.**	Genevini et al. (1983) Page et al (1988)
Leafy vegetables, fresh herbs, celeriac and all cultivated fungi	**0.2**	**a**	**0.034 – 3.8 d.m.** **1.3 – 10.3 f.m.**	Genevini et al. (1983) Page et al (1988)
Stem vegetables, root vegetables and potatoes, excluding celeriac. For potatoes the maximum level applies to peeled potatoes	**0.1**	**a**	**0.011 – 3.6 d.m.**	Genevini et al. (1983) Page et al (1988)
Muscle meat of fish excluding species listed below	**0.05**	**b**		
Muscle meat of wedge sole, eel, spotted sea-bass, horse mackerel or scads, grey mullet, common two-banded sea bream, sardine, tuna, sardinops, bonito, European Anchovy, louvar or luvar	**0.1**	**b**		
Crustacean, excluding brown meat of crab and excluding head and thorax meat of lobster and similar large crustaceans	**0.5**	**b**	**0.04 – 0.1 d.m.**	Storelli & Marcotrignano (1999)
Feed materials of vegetable origin	**1**	**c**		
Feed materials of animal origin except: feeding stuffs for pets	**2**	**c**		

FOOD COMMODITY	EU THRESHOLD mg/kg f.m.	REF.	VALUE/RANGE ASSESSED	REF.
Phosphates	**10**[6]	**c**		
Complete feeding stuffs for cattle, sheep and goats except: complete feeding stuffs for calves, lambs, kids	**1**	**c**	**0.08 – 0.24 d.m.**	Genevini et al. (1983[p56])
Other complete feeding stuffs except: feeding stuffs for pets	**0.5**	**c**	**0.17 - 0.99 d.m.**	Genevini et al. (1983[p57])
Mineral feeling stuffs	**5**[7]	**c**	**0. 79 d.m.**	Genevini et al. (1983[p58])
Other complementary feeding stuffs for cattle, sheep and goats	**0.5**	**c**		

a. COMM. REG. 2001/466/CE Annex I Section 3 ; b. COMM. REG. 2002/221/CE Annex I; c. COUN. DIR. 1999/29/CE Annex I

Table 2-10: LEAD - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	EU THRESHOLD mg/kg f.m.	REF.	VALUE/RANGE ASSESSED	REF.
Cows' milk (raw milk, milk for the manufacture of milk based products and heat treated milk as defined by Dir. 92/46/EEC)	**0.02**	a	**0.052 d.m.**	Bruggemann et al.. (199[p59]?)
Infant formulae and follow-on formulae	**0.02**	a		
Meat of poultry	**0.1**	a	**0.008 – 0.009 d.m.**	Bruggemann et al.. (199[p60]?)
Meat of bovine	**0.1**	a	**0.016 – 0.019 d.m.**	Bruggemann et al.. (199[p61]?)
Edible offal of cattle, sheep, pig and poultry	**0.5**	a		
Crustacean, excluding brown meat of crab	**0.5**	a		
Cephalopods (without viscera)	**1.0**	a		
Cereals (including buckwheat), legumes and pulses	**0.2**	a	**0.01 – 3.6 d.m.** **0.0068 – 0.013 f.m.**	Genevini et al. (1983) Page et al (1988) Swedish environmental protection agency survey
Vegetables excluding brassica, leafy vegetables, fresh herbs and all fungi. For potatoes the maximum level applies to peeled potatoes	**0.1**	a	**0.01 – 1.1 d.m.** **0.01 – 0.14 f.m.**	Genevini et al. (1983) Page et al (1988)
Brassica, leafy vegetables and all cultivated fungi	**0.3**	a	**0.036 – 2.3 d.m.** **0.01 – 0.51 f.m.**	Genevini et al. (1983) Page et al (1988)
Fruit excluding berries and small fruits	**0.1**	a	**0.01 – 0.37 f.m.**	Genevini et al. (1983)

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[6] Member States may also prescribe a maximum cadmium content of 0.5 mg per 1% phosphorus
[7] Member States may also prescribe a maximum cadmium content of 0.75 mg per 1% phosphorus

FOOD COMMODITY	**EU THRESHOLD mg/kg f.m.**	**REF.**	**VALUE/RANGE ASSESSED**	REF.
Berries and small fruits	**0.2**	a		
Fats and oils, including milk fat	**0.1**	a		
Fruit juices, concentrated fruit juices and fruit nectars	**0.05**	a		
Wines (including sparkling wines and excluding liqueur wines), aromatised wines, aromatised wine-products, cocktails and ciders, perry and fruit wines. Maximum level applies to products produced from the 2001 fruit harvest onwards	**0.2**	a		
Muscle meat of fish excluding fish species listed below	**0.2**	b		
Muscle meat of wedge sole, eel, spotted sea bass, horse mackerel or scads, grey mullet, common two-banded sea bream, grunt, sardine, bonito, sardinops, tuna	**0.4**	b	**0.04 – 5.26 f.m.**	Storelli & Marcotrignano (2001b)
Bivalve molluscs	**1.5**	b	**0.96 – 1.19 f.m.**	Storelli & Storelli & Marcotrignano (2000)
Feed materials with exception of :	**10**	c[8]		
- green fodder	**40**	c		
- phosphates	**30**	c		
- yeast	**5**	c		
Complete feeding stuffs	**5**	c	**0.56 – 1.4 d.m.**	Genevini et al. (1983[p72])
Complementary feeding stuffs with the exception of:	**10**	c		
- mineral feeding stuffs	**30**	c	**1.2 – 2.7d.m.**	Genevini et al. (1983[p73])

a. COMM. REG. 2001/466/CE Annex I Section 3 ; b. COMM. REG. 2002/221/CE Annex I; c. COUN. DIR. 1999/29/CE Annex I (Data are expressed in mg/kg relative to feeding stuff and to feed materials with a moisture content of 12%)

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[8] Data referred to source c are expressed in mg/kg relative to feedingstuff and to feed materials with a moisture content of 12%

Table 2-11: MERCURY - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	**EU THRESHOLD mg/kg f.m.**	**REF.**	**VALUE/RANGE ASSESSED mg/kg**	**REF.**
Fishery products except those listed in Commission regulation n. 221/2002 (amending the present)	**0.5**	**a**	**0.15 – 0.30 f.m.**	Storelli & Storelli & Marcotrignano (2000)
Anglerfish, Atlantic catfish, bass, blue ling, bonito, eel, halibut marlin, pike, plain bonito, portuguese dogfish, rays, redfish, sail fish, scabbard fish, shark, snake mackerel, sturgeon, swordfish, tuna, emperor or orange roughtly grenadier	**1.0**	**b**	**0.05 – 4.26 f.m.**	Storelli & Marcotrignano & Giacomelli. (1998) Storelli & Marcotrignano (2000b) Storelli & Marcotrignano (2001c)
Feed materials with exception of :	**0.1**	**c**		
- feeding stuffs by the processing of fish or other marine animals	**0.5**	**c**		
Complete feeding stuffs with exception of:	**0.1**	**c**	**0.02 – 0.38 d.m.**	Genevini et al. (1983)
- complete feeding stuffs for dogs and cats	**0.4**	**c**		
Complementary feeding stuffs with the exception of: complementary feeding stuffs for dogs and cats	**0.2**	**c**		

a. COMM. REG. 2001/466/CE Annex I Section 3 ; b. COMM. REG. 2002/221/CE Annex I; c. COUN. DIR. 1999/29/CE Annex I

Table 2-12: ARSENIC - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	**EU THRESHOLD mg/kg f.m.**	**REF.**	**VALUE/RANGE ASSESSED**	**REF.**
Feed materials with exception of :	**2**	**c**		
- meal made from grass, from dried Lucerne and from dried clover, and dried sugar beet pulp and dried molasses sugar beet pulp	**4**	**c**	**0.13 – 2.15 f.m.**	Genevini et al. (1983[p79])
- phosphates and feeding stuffs obtained from the processing of fish or other marine animals	**10**	**c**		
Complete feeding stuffs with exception of:	**2**	**c**		
- complete feeding stuffs for fish	**4**	**c**		
Complementary feeding stuffs with the exception of:	**4**	**c**		
- mineral feeding stuffs	**12**	**c**		

c. COUN. DIR. 1999/29/CE Annex I

2.3.3 Conclusions PTEs in food and feeding stuff

A comprehensive assessment of the tables herewith reported that refer to PTEs in foodstuffs, leads to the following conclusions:

- Numbers are lacking for many types of foodstuff, particularly for milk products and eggs
- some numbers refer to dry matter, whereas others consider fresh matter. We draw the attention of the reader to the fact that, considering the scope of this survey, and lacking values concerning moisture, only the values referred to dry matter should be considered
- The variation of concentrations for each type of foodstuff is remarkable, indeed. In some cases, the maximum value has been reported (see for instance Zinc in leafy vegetables, Ni in cereals) at levels which are some orders of magnitude higher than the minimum value. Details have been given elsewhere in this section, about main factors likely to affect such variation (*spatial variation, temporal variation, heterogeneity of sampling and test methods, intra- and inter-laboratory variations*)
- however, the preliminary assessment of numbers reported already leads to some remarkable conclusions:
    - foodstuffs such as leafy vegetables and vegetables (one of basic components in daily diet throughout Europe) tend to show a higher concentration of PTEs than cereals;
    - lower concentrations found in cereals are affected by plant physiology, which tends to concentrate PTEs in the edible part (seeds) rather than in the shoot and in the leaves
    - fish tends to accumulate relatively high concentrations of such PTEs as As, Cd, Cr, Pb. Arsenic, in particular, has been reported by some authors at high concentrations, though it may be affected by one or more of the aforementioned factors causing variation.

2.3.4 Organic pollutants (OPs) in food and feeding stuff

Table 2-13 shows figures of PCDD/PDCF in food commodities. Data are expressed according to each case in pg I-TEQ g^{-1} on dry matter basis or lipid basis. Unfortunately, when data on lipid basis are given, the corresponding content of lipids in specific food stuffs is not included, nor is any reference given to dry matter content in the case of numbers on dry matter basis. This arguably makes comparisons very difficult.

The mean value and the range listed are calculated using the whole set of data collected that originate from different literature sources. This is why the ranges are very wide and include more than one order of magnitude. It also need to be stressed that the mean value referred to milk is calculated on a greater number of data than other food commodities, i.e. cereals, due to the higher availability of data from literature.

In general, the low availability of numbers, their heterogeneity and the difficult comparison of concentrations expressed on dry matter basis or on lipid basis makes the assessment of PCDD/PDCF background concentration in foodstuff quite flawed.

Remarkably, our survey therefore discloses some scope for further in depth analyses to detect PCDD/PDCF for all food typologies in a standardised way across Europe.

Table 2-13: PCDD/PCDF concentrations in food and feeding stuff-[pg I-TEQ g^{-1} ; pg WHO-TEQ g^{-1} for feeding stuff]

CATEGORY	MEAN		RANGE	
	d.m.	**on lipid basis (g fat)**	**d.m.**	**on lipid basis (g fat)**
CEREALS	0.05	0.084		
POTATO	0.1	0.016		0.0036-0.039
RICE				
FRUIT	0.065	0.015		
VEGETABLES (ROOTS, TUBERS, FRUITS)	0.0042	0.09	0.019-0.45	
VEGETABLES (LEAFY)	0.059	0.01	0.036-0.054	
PULSES				
MILK	0.06	1.64		0.2-47.5
MILK PRODUCTS	1.93	1.35	1.1-8.4	0.35-3.8
EGGS	0.69	1.94		0.19-23.4
MEAT	0.3	5.72	0.08-0.59	0.03-16.7
FISH	0.21	30.81		1.0-214.3
FEEDING STUFFS ***(pg WHO-TEQ g-1)(fat industrial waste)***		5.22		0.39-25
FEEDING STUFFS ***(pg WHO-TEQ*** g^{-1}***)(food industrial waste)***		14.67		0.036-60

Sources: Hietaniemi & Kumpulainen (1994), AEA Technology, 1999), AA. VV. (1996), Storelli & Marcotrignano (2001d), Pancirov & Braun (1977), Storelli & Storelli & Marcotrignano (2001), UK Committee on toxicity of chemicals in food, consumer products and the environment (2001), Penisch (2002), BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)

In Table 2-14 mean values and/or range values of PCBs concentration in food commodities are given. In the given bibliography from which data derive PCBs are specified as follows:

(1) Hietaniemi & Kumpulainen (1994): investigated congeners: PCB 8, 15, 20, 28, 35, 52, 53, 77, 81, 101, 106, 104, 114, 118, 126, 128, 138, 149, 153, 156, 169, 170 and 180 according to the IUPAC classification and numeration; outcomes were expressed as sum of the congeners.

(2) AA. VV. (1996): congeners are not specified

(3) Storelli & Storelli & Marcotrignano (2001): congeners are not specified

(4) Penisch (2002): the following congeners were investigated: 2,4,4'-Trichlorbiphenyl (28), 2,2',5,5'-Tetrachlor-biphenyl (52), 2,2',4,5,5'-Pentachlor-biphenyl (101), 2,2',3,4,4',5'-Hexachlor-biphenyl (138), 2,2',4,4',5,5'-Hexachlor-biphenyl (153), 2,2',3,4,4',5,5'-Hepta-chlorbiphenyl (180)

Table 2-14: PCB concentrations in food and feeding stuff [µg kg^{-1}]

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS				
POTATO	28			
RICE				
FRUIT				
VEGETABLES (ROOTS, TUBERS, FRUITS)	7		7,000-16,000	
VEGETABLES (LEAFY)				
PULSES				
MILK		33 on lipid basis		
MILK PRODUCTS		< 8		
EGGS		< 4		
MEAT		< 4		
FISH		112.67		0.2-199
FEEDINGSTUFFS *(vegetal raw materials)*	88		12.0-150	

In Table 2-15 mean values and/or ranges of PAHs concentration in food commodities are given. In the given sources PAHs are specified as follows:

(1) Hietaniemi & Kumpulainen (1994): phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)antracene, triphenylene, chrysene, benzo(a)pyrene, benzo(e)pyrene, benzo(ghi)perylene, benzo(b)fluoranthene, benzo(j)fluorantene, benzo(k)fluoranthene, dibenz(a,h)antracene, indeno(1,2,3-cd)pyrene and b,b'-binapthyl were investigated; outcomes are expressed as sum of PAH cogeners.

One important sentence of this survey reads: *"An interesting result was that carcinogenic compounds such as benzo(a)antracene, chrysene, benzofluoranthenes, benzo(a)pyrene, and indeno(1,2,3-cd)pyrene were found in traditionally dried bread and corresponding flour, and in the meat group. However, most of the compounds detected were not carcinogenic, such as phenanthrene, anthracene, fluoranthene and pyrene. According to previous studies and official tolerance limits established by many countries, only smoked and grilled products together with benzo(a)pyrene have been considered important. Finland has issued guidelines for benzo(a)pyrene and a tolerance limit of 1 µg kg^{-1} in smoke aroma preparation. Conversely, in Austria, PAH compounds in smoke aroma preparations are prohibited and the tolerance limit for benzo(a)pyrene in smoked foods is 1µg.kg^{-1}. In addition, Germany has established tolerance limits of 1µg kg^{-1} for benzo(a)pyrene in meat and cheese."*

(2) UK Committee on toxicity of chemicals in food, consumer products and the environment (2001): benzo(a)anthracene, benzo(a)pyrene and dibenz(a,h)antracene were investigated, as specified in Annex 3.

(3) Storelli & Storelli & Marcotrignano (2001): phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, benzo(a)pyrene, benzo(ghi)perylene, and dibenz(a,h)antracene were investigated, as specified in Annex 3.

(4) Pancirov & Braun (1977) : anthracene, fluoranthene, pyrene, benzo(a)anthracene, benzo(a)pyrene, benzo(ghi)perylene, and dibenz(a,h)antracene were investigated, as specified in Annex 3.

Table 2-15: PAH concentrations in food and feeding stuff [μg kg^{-1}]

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS			4-291	
POTATO				
RICE				
FRUIT				
VEGETABLES (ROOTS, TUBERS, FRUITS)				
VEGETABLES (LEAFY)				0.01-3.17
PULSES				
MILK				
MILK PRODUCTS				1.0-4.0
EGGS		38		
MEAT				0.01-0.26
FISH		11.36		0.03-64.7
FEEDINGSTUFFS				

Table 2-16: HCB concentrations in food and feeding stuff [μg kg^{-1}]

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS		10		
POTATO		< 10		
RICE		< 10		
FRUIT		10		
VEGETABLES (ROOTS, TUBERS, FRUITS)		< 10		
VEGETABLES (LEAFY)		< 10		
PULSES				
MILK		9.26 [on lipid basis]		0.2-25 [on lipid basis]
MILK PRODUCTS		< 3		
EGGS		< 10		
MEAT		< 3		
FISH				
FEEDINGSTUFFS				

Table 2-17: Aldrin/Dieldrin concentrations in food and feeding stuff [mg kg^{-1}]

CATEGORY	MEAN		RANGE	
	d.m.	**f.m.**	**d.m.**	**f.m.**
CEREALS	< 0.01	< 0.01		
POTATO		< 0.01		
RICE		< 0.01		
FRUIT		< 0.01		
VEGETABLES (ROOTS, TUBERS, FRUITS)		< 0.01		
VEGETABLES (LEAFY)		< 0.01		
PULSES				
MILK		< 0.0005		
MILK PRODUCTS		< 0.0004		
EGGS		< 0.01		
MEAT		< 0.004		
FISH				
FEEDINGSTUFFS *(vegetal raw materials)*	0.016	/	0.003-1.0	

Table 2-18: DDT and congeners concentrations in food and feeding stuff [µg kg^{-1}]

CATEGORY	MEAN		RANGE	
	d.m.	f.m.	d.m.	f.m.
CEREALS				
POTATO				
RICE				
FRUIT				
VEGETABLES (ROOTS, TUBERS, FRUITS)				
VEGETABLES (LEAFY)				
PULSES				
MILK		20 on lipid basis		5 -28 on lipid basis
MILK PRODUCTS				
EGGS				
MEAT				
FISH		94.3		0.2-147
FEEDINGSTUFFS				

2.3.5 Remarks on the effects of cooking on some organic pollutants

The table below recaps on some surveys about the effect of cooking on concentrations of OPs in food. Symbols (+) and (–) indicate an increase or decrease of concentrations in cooked/uncooked food.

Table 2-19: The effect of cooking on concentrations of OPs in foodstuffs

Food commodity	**OPs investigated**	**Type of Cooking**	**Presence in cooked food**	**Presence in uncooked food**	Reference
Meat (hamburger, steak and chicken with skin)	Benzo(a)pyrene (PAH)	Grill/barbecue	+	–	Kazerouni et al., 2002
Meat (sausages, steak, pork and chicken)	Benzo(a)pyrene (PAH)	Charcoal or log broiling, smoked, oven	+	–	Lijinsky, 1991
Not specified	PAHs	Not specified	+	–	Phillips, 1999
Leafy vegetables (komatsuna, lettuce and spinach)	PCDDs, PCDFs and Co-PCB	Washing and boiling	–	+	Tsutsumi et al., 2002
Hamburger, bacon and catfish	PCDDs, PCDFs and Co-PCBs	Broiling	– (amount) Variable effects on concentrations*	+ (amount) Variable effects on concentrations*	Schecter et al., 1998
Fish	tDDT and PCBs	Baking, broiling, boiling, smoking and microwaving	–	+	Wilson et al., 1998
Ready-to-eat foods	PCBs, PBBs*** and TCDD****	not specified	–	+	Zabik et al., 1999

* mean concentrations on wet weight show variable responses in hamburger, bacon and catfish due to different effects of cooking on the water content and possible mineralisation of some volatile solids.

** tDDT: total DDT

*** PBB: polybrominated biphenyls

***** TCDD: Tetrachlorodibenzo-p-Dioxin

The outcomes seem to show a general decrease of PCBs and dioxin in foodstuffs after cooking while for PAH an opposite trend has been detected.

Benzo(a)pyrene is formed by the way of incomplete combustion of organic matter Incomplete combustion mainly occurs during heating processes like grilling, barbecuing and roasting where foods are close in contact with the heat source (over an open flame). This includes drying with smoke, or by absorption during the smoking.

Average PCBs/dioxin reductions depend on the previous washing (especially for vegetables), the cooking method, water content and lipid content loss after cooking and skin removal (especially for poultry meat).

As a matter of fact, cooking shows to have some effects on presence of OPs, although this shows no general trend. However, the effect of cooking on OPs should be considered in the light of the magnitude of related effects on the production of organic fertiliser such as compost.

In such respect, it is important to consider the percentage of cooked food in biowaste sent for composting; although there exists no systematic and widespread information on this, cooked materials seem to cover a minor share of total composted biowaste. We sought information from our overseas contacts, but we could not get any split data for cooked/uncooked materials in biowaste; rather, the common typology of "food waste" is normally singled out in sorting analyses.

We may anyway move our steps from an Italian survey which was carried out in order to define the food waste fractions on which to focus efforts for collection. The survey *("Composizione merceologica del rifiuto organico da raccolta differenziata" – Milan, Dec. 1996*), carried out by AMSA (the public company for waste management in Milan) focused on food waste collected in Milan area, with split figures for households and big producers (canteens, bars, restaurants, etc). Noteworthy, Milan and generally Italian collection schemes tackle food waste separately from garden waste, which enhances meaningfulness of the outcomes. Summary outcomes are reported in the table below:

Material	from households (%)	from big producers (%)
Fraction < 20 mm	15,92	15,68
Uncooked foodwaste	61,56	51,17
Cooked foodwaste	6,51	21,82
Glass and inerts	0,90	0,48
Metals	0,79	0,45
Paper and cardboard	14,32	10,40
TOTAL	100	100

The under screen fraction (< 20 mm) may be considered as mainly composed of cooked food scraps (leftovers coming from preparation of meals are usually bigger, i.e. salad leaves, peelings, skins, etc.). Adding the fraction < 20 mm to cooked food waste, we can assume the cooked fraction being around 22% at households (which account for the vast majority of composted materials) and 37% at big producers.

Remarkably, unlike Italian schemes, schemes diffused in Central and Northern Europe focus much less on the cooked part of food scraps, given some issues related to management of the waste by households where collection frequencies are much lower and no other tool is adopted to make the system "user-friendly" (e.g. biogas, widely adopted in Italy and Spain). As a matter of fact, very frequently collection schemes in such Countries are usually worded as "VGF" ("Vegetable, Garden and Fruit") or "GFT" ("Groen, Fruit, Tuinavfal", in the Netherlands) or "Vor dem Teller" ("before the pot" in Germany), all of which remark the very uncommon presence of cooked materials in compostable waste, which makes its percentage much lower than the already relatively low percentage in Italian schemes.

Further it is important to remark that for the sake of proper aerobic process management kitchen waste constitutes only 15 to 50 % (w/w) in the initial feedstock mix for composting. This gives total figures for cooked food residues in feedstock mixtures for composting between 0 % and app. 18 % (w/w) in common biowaste collection and processing schemes.

Therefore, also referring to the microbial degradation of PAH (see chapter 5.2.10)and the actually detected PAH concentration in compost, we may conclude that the effect of increased PAH concentration in cooked food residues is not likely to constitute a threat to the recycling of kitchen waste and its safe application via composting.

The following tables compare OPs threshold values set by EU legislation with values found in food commodities and feeding stuff.

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Table 2-20: PCBs[9]- concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	EU THRESHOLD ng/g fat (ppb)	REF.	VALUE/RANGE ASSESSED	REF.
Eggs, eggs products, fresh poultry meat and derived products	**200**	**d**	**< 4 f.m.**	Penisch (2002)
Fresh pig meat and derived products	**200**	**d**	**< 4 f.m.**	Penisch (2002)

d. COMM. DEC. 1999/640/ EC Annex A

Table 2-21: HCBs - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	EU THRESHOLD mg/kg[10]	REF.	VALUE/RANGE ASSESSED	REF.
All feeding stuffs with exception of:	**0.01**	**e**	**n.f.**	
- Fats	**0.2**	**e**	**n.f.**	

n.f. not found; e. COUN. DIR. 1999/29/ EC Annex I

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[9] Sum of the following PCBs (IUCPA): 28, 52, 101, 118, 138, 153, 180.
[10] relative to a feedingstuff with a moisture content of 12%

Table 2-22: Dioxins and Furans - concentrations in food and feeding stuff as compared to EU threshold values

FOOD COMMODITY	EU THRESHOLD	REF.	VALUE/RANGE ASSESSED	REF.
	pg WHO-PCDD/F-TEQ/g fat or product[11]		pg I-PCDD/F-TEQ/g fat	
Meat and meat products originating from: - Ruminants (bovine animals, sheep)	**3**	**f**	**0.03 – 16.7**	AEA Technology, (1999[p106])
- Poultry and farmed game	**2**	**f**	**0.4 - 17**	AEA Technology, (1999[p107])
- Pigs	**1**	**f**	**<0.1 – 0.90**	AEA Technology, (1999[p108])
Liver and derived products	**6**	**f**	**< 1.1 – 61**	AEA Technology, (1999[p109])
Muscle meat of fish and fishery products and products thereof	**4**	**f**	**1.0 – 214.3**	AEA Technology, (1999[p110])
Milk and milk products, including butter fat	**3**	**f**	**0.2 – 47.5**	AEA Technology, (1999[p111])
Hen eggs and eggs products	**3**	**f**	**0.19 – 23.4**	AEA Technology, (1999[p112])
Animal fats	**3**	**f**		
- from ruminants	**2**	**f**		
- from poultry and farmed game	**1**	**f**		
- from pigs	**2**	**f**	**0.06 – 1.2**	AEA Technology, (1999[p113])
- mixed animal fat	**0.75**	**f**		
Vegetal oils	**2**	**f**		
	ng WHO -PCDD/F-TEQ/Kg[12][13]		pg WHO-TEQ/g of fat	
All feed materials of plant origin including vegetable oils and by-products	**0.75**	**g**	**0.036 – 1.7**	BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)
Minerals	**1.0**	**g**		

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[11] sum of polycholrinated dibenzo-para-dioxin (PCDDs) and polyclorinated dibenzo furans (OCDFs) expressed in World Health Organization (WHO) toxic equivalents, using the WHO-TEFs (toxic equivalency factors, 1997) PCDD/F

[12] These maximum limits shall be reviewed for the first time before 31 December 2004 in the light of new data on the presence of dioxins and dioxin-like PCBs, in particular with a view to the inclusion of dioxin-like PCBs in the levels to be set and will be further reviewed before 31 December 2006 with the aim of significantly reducing of the maximum levels. Fresh fish directly delivered and used without intermediate processing for the production of feedingstuffs for fur animals is exempted from the maximum limit. The products, processed animal proteins produced from these fur animals cannot enter the food chain and the feeding thereof is prohibited to farmed animals which are kept, fattened or bred for the production of food.'

[13] relative to a feedingstuff with a moisture content of 12%

FOOD COMMODITY	EU THRESHOLD	REF.	VALUE/RANGE ASSESSED	REF.
Animal fat, including milk fat and egg fat	**2.0**	**g**	**0.9 – 2.16**	BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)
Other land animal products including milk and milk products and eggs and egg products	**0.75**	**g**	**1.9**	BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)
Fish oil	**6**	**g**	**12**	BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)
Fish, other aquatic animals, their products and by- products with the exception of fish oil	**1.25**	**g**	**10-60**	BiPRO, IRCE, Ökometric, AFC Consultant, European POPs Expert Team (2002)
Compound feeding stuffs, with the exception of feeding stuffs for fur animals, pet foods and feeding stuffs for fish	**0.75**	**g**		
Feeding stuffs for fish and pet foods	**2.25**	**g**		

f. COUN. REG. 2001/2375/EC Annex I ; g. COUN. DIR. 2001/102/EC

2.3.6 Key remarks on distribution of OPs and PTEs between edible and non edible parts of food

An investigation on different distributional patterns of OPs between edible and non edible parts could be of some interest, since food waste may be assumed to be mainly consisting of non edible parts of vegetables.

The table below recaps on available data as to concentration of lead and three OPs (PCBs, Aldrin and Dieldrin) in some food commodities, comparing the content in the edible and non edible parts; concentrations detected in soils are also given.

CATEGORY	**in soil** µg/kg d.m.	**Σ PBCs[1]** µg/kg d.m.		**Aldrin (a) dieldrin (d)[1]** µg/kg d.m.		**Pb[2]** µg/kg f.m.	
		edible	non edible	edible	non edible	edible	non edible
CEREALS							
Maize	550 (d)			< 10			
Maize	780 (a)			< 10			
Maize (seeds)	400-3000 (a)(d)				3-8		
Maize (plant)	1200 (d)				20		
Maize (leaves)*	2500-250000	0	3220-12450				
Maize (leaves)	92-144**		45-81				
Maize (plant)	111-453		27-53				
Maize (top)	20000		2				
Sorghum (top)	20000		58				
Oats (plant)	13		26				
POTATO	380-830	28	40-61				
VEGETABLES (ROOTS, TUBER, FRUITS)							
Beet	1200 (d)			70	30		
Carrots	100000	7000-16000					
Carrots	50-500		0				
Carrots	5000		81				
Beet	240	4	7				
Beet	20000		815				
Beet	300		10-150				
Radish	20	10					
Radish	50-500		0				
Radish	5000		25				
Apple	n.a.					0,01-0,05	0,01-0,024
Pear	n.a.					0,01-0,03	0,01-0,023
VEGETABLES (LEAFY)							
PULSES							
Peanut (seeds)	400-3000 (a)(d)				100-1000		
Peanut (hull)	80-200 (a)(d)				80-130		
Peanut (top)	20000		473				

** µg l^{-1} in the sludge used for fertilisation
n.a. not available
Sources: [1] AA. VV. (1996); [2] Genevini P.L., Vismara R. Mezzanotte V. (1983)

Some remarks need to be drawn from data shown above and their cross-comparison:

- Number of available data on which it is possible to compare the PTE and/or OP content in foodstuffs is quite poor and lacks comprehensiveness,

- Moreover, figures cannot be assumed as “typical” or “background” concentrations in any given situation, for the following reasons:
  a) Remarkably, data drawn from sources herein quoted [AA. VV. (1996); Genevini P.L., Vismara R. Mezzanotte V. (1983)] have been detected in a wide range of situations. Such surveys actually investigate the influence of contamination of soils, and related effects; it may be doubtful, whether effects in a “contaminated” site may be assumed to draw conclusions on “background” concentrations. By way of an example, the content of PCBs detected in maize are recorded in association with the content of PCBs in soils at different levels of contamination;. the absorption curve is thereby deduced and the concentration in different plant parts measured. Data reported therefore refer to widely different levels of contamination, at which the plant response show different concentrations of PCBs. This explains such different values and related ranges even for the same species and same part of the plant.

  b) PTEs and OPs concentrations in the leafy system strongly depend on the atmospheric deposition, so that the typical background concentration can’t be traced back to a vegetal species or to a group, but mainly depends on the boundary conditions (i.e. rural or industrial area) in which the plant is grown.

  c) Concerning PCBs, the authors (AA. VV., 1996) remark that these are semi-volatile compounds, insoluble in water and prominently subject to diffusion in a gaseous phase in the air in the pore space in the soil; thereby they may reach the aerial part of plants, given the gaseous exchange between soil and air. So, even if the PCBs peak concentration is higher than 200 mg kg^{-1} in the soil, their volatility promotes assimilation mainly through the aerial parts.

2.4 Spatial variation: case studies

As far as spatial variation is concerned, we consider it in this section at two levels :

- geographic scale: intended as comparison between Member States of OP background concentrations in similar contexts
- local scale: detection of similarities/differences in OP content in food commodities in rural areas compared to industrial districts (this will refer in particular to milk, as this has been the most investigated issue).

However, the data collected did not allow us to construct an organic scenario covering all Europe, as little data is available. In the next sections we have provided, some case studies representing experiences and investigations carried out at the local level.

2.4.1 Geographic scale

An Italian survey on concentration of OPs in Italian milk (Storelli, 2001) is reported below. This study also compares the findings with some data from literature from other countries. From this standpoint, no significant differences in OP concentration were found.

Four hundred fifty milk samples (cow, buffalo, goat, sheep) from Apulian farms were investigated. The parameters were: polychlorinated biphenyls (PCBs), hexachlorobenzene (HCB) and organochlorine pesticides (OCPs). Results showed that PCBs, *p,p’-DDE*, HCB and hexachlorocyclohexane α isomer (α-HCH) were present in almost all of the samples analysed, while lindane (γ-HCH) was found only in cows' milk.

As far as geographical variation is concerned, the results of OP residues in cows' milk obtained from the survey were compared to those found in other countries (various sources), where DDT and HCH are still used on various crops and comparable/similar ranges were found.

Table 2-23: Presence of some Organic Pollutants residues in cows' milk from different countries (µg kg^{-1}) (source Storelli 2001 mod.)

Country	France 1989	Spain 1991	Italy 1991	Italy 1992	Italy 1994	Italy 2000
	(Venant et al., 1991)	(de la Riva & Anadon, 1991)	(Cantoni et al., 1994)	(Cantoni et al., 1994)	(Cantoni et al., 1994)	(Storelli et al., 2001)
α-HCH	3	18	6	7	5 – 20	1.03
γ-HCH	24	24	5 –25	7	5	5
HCB	-	8	6	-	-	7
P,p'-DDE	-	21	5 –14	5 – 15	5 – 10	18

Conclusion

The study of the environmental concentrations of dioxins across the European Union (AEA Technology, 1999) has tried to consider the influence of geographical variations; in particular, climate differences which might affect environmental processes, different agricultural practices, industrial development and dietary regimes. However, it was found that there is a general lack of information on several countries - in particular in Southern Europe - and, therefore, an analysis of geographical trends is not possible.

By comparing geographical variations in the background level of pollution the definition of geographical units must be considered. For example, a rural location in Ireland or Northern Sweden is likely to show much lower concentrations of background contamination than a rural location in Germany or the Netherlands, because of the overall density of population and industrialisation. This causes problems when trying to define the range of background concentrations across the EU.

Geographical variations can become blurred through the transportation of goods around Europe or imports of goods from outside the region. This is particularly important for foods and feeding stuff. Care should, therefore, be taken to identify the sources of these products before conclusions are drawn concerning sources of contamination.

2.4.2 Local scale

The section below contains some information on variation at local scale. Concentrations of organic pollutants in samples coming from rural sites are compared to those detected in areas near potential (point) sources of contamination.

Belgium (AEA Technology, 1999)

There is little published data available on dioxins in foodstuffs in Belgium. However, the recent incidence of food contamination by PCBs and dioxins in Belgium has raised concerns on the issue, and a programme for detection , whose results will be published shortly, has been promoted.

Van Cleuvenbergen et al (1993) provided data on cows' milk in Flanders . Cows' milk was sampled in 1993 at seven farms across Flanders, at sites within a few kilometres of industrial locations such as Incinerators for MSW and power stations. The results are shown in the table below.

Table 2-24: Dioxins Concentrations in cows' milk from industrial locations in Flanders (AEA Technology, 1999)

Location	Potential source of contamination	Concentration (pg I-TEQ g^{-1} milk fat)
Mol	Coal-fired power station and some nuclear industry	3.9
Moerkerk	Large MSWI	2.4
Berendrecht	Petrochemical industry	2.7
Zelzate	Metallurgical and chemical industry	5.1
Ham	Close to a highway and chemical industry	3.7
Vilvoorde	Coal-fired power station ad MSWI	3.1
Menen	MSWI	12.6

These concentrations are relatively high in comparison to analogous point sources found across Europe,

Another survey on milk samples taken in Belgium in 1991 (Belgische Kamer van Volksvertegenwoordigers 1992), concerning milk from bulked tankers showed a range of concentrations 1.1-3.1 pg I-TEQ g^{-1} milk fat and a mean of 2.1 pg I-TEQ g^{-1} milk fat.

These latter numbers may be assumed as more representative of background concentrations. In 1998, the mean level for dioxins in milk was 2.0 pg TEQ g^{-1} fat (Vinkx, *pers comm.*).

Three areas of contamination continue to be monitored: Wachtebeke (metal industry), Aiseau (incinerator) and Menen (incinerator, in France). The maximum detected concentration (winter 1998/99) was 23 pg TEQ g^{-1} fat, at Aiseau. This milk was not sold for consumption. The median concentration across all sample sites was 7.4 pg TEQ g^{-1} fat.

France (AEA Technology, 1999)

Limited data are available on dioxins in foodstuffs in France. Cows' milk and dairy products have been analysed in sites near potential sources of contamination, and some findings have resulted in action to reduce emissions.

Surveillance programmes to analyse concentrations of dioxins in cows' milk in such areas were undertaken by the French Ministry of Agriculture and Fisheries in 1994-1995 and 1997, and dairy products were sampled in 1996 (MAPAAR, 1997, 1998).

The first survey covered 14 Departments. Milk was collected from farms within 5 km of sources of pollution and also from Department blending points to represent regional averages (Table 2-25).

Table 2-25: Concentrations of dioxins in French cows' milk (1994-1995) (AEA Technology, 1999)

Department	number of farm samples	Mean concentration in farm samples (pg I-TEQ g^{-1} fat)	Concentration in Department blend (pg I-TEQ g^{-1} fat)	Overall average (pg I-TEQ g^{-1} fat)
Seine-Maritime	4	4.53	3.12	4.25
Pas-de-Calais	2	4.03		4.03
Sarthe	1	4.54	1.36	2.95
Ille-et-Vilaine	4	2.57	1.95	2.44
Somme	4	1.63	3.88	2.08
Doubs	4	1.63	0.87	1.47
Vendee	1	1.26	1.35	1.31
Cotes-d'Amor	4	1.32	1.30	1.31
Meuse	4	1.26	1.48	1.30
Bas-Rhin	4	0.99	1.03	0.99
Mayenne	4	0.98	0.84	0.95
Rhone	4	0.95	0.93	0.94
Cantal	1	0.79	0.88	0.84
Manche	4	0.81	0.91	0.83
AVERAGES		**1.8**	**1.53**	**1.81**

Germany (AEA Technology, 1999)

Various monitoring programs were performed in the different federal states of Germany. In Bavaria, 160 samples were taken from individual farms in autumn 1989 and 1990 (Lassek et al 1993). Data are shown in the table below. In 1992, the 27 existing collection points of Bavarian cows' milk were sampled and analysed for dioxins. The limit value for the sale of cows' milk of 5 pg I-TEQ g^{-1} fat was not exceeded in any of the samples, neither was the guideline concentration of 3 pg I-TEQ g^{-1} fat

Table 2-26: Concentrations of dioxins in Bavarian cow's' milk in 1989/90 and 1992 (pg I-TEQ g^{-1} fat) (AEA Technology, 1999)

	Year	N	Min	Max	Mean	Median
Rural samples (no point source nearby)	1989/90	143	0.60	1.54	1.00	0.96
Samples close to potential point sources	1989/90	17	0.48	5.6	1.85	1.39
All samples	1989/90	160	0.48	5.62	1.76	1.30
All samples	1992	27	0.69	1.12	0.87	0.89

Findings seem to confirm a higher concentration near point sources, and particularly high peak values in such cases.

The table below compares samples from rural areas to samples from areas with potential pollution sources . The samples from farms near sources have a wider range of concentrations, of 0.48-24 pg I-TEQ g^{-1} fat.

Table 2-27: Concentrations of dioxins in cows' milk from individual farms in Germany (pg I-TEQ g^{-1} fat) (AEA Technology, 1999 mod)

Location	Mean	Range	Source
	Rural areas		
Lower Saxony, rural	1.5	0.9-3.24	CUA Oldenburg 1991
Bavaria, rural	1	0.6-1.54	Lassek et al 1993
Baden-Württemberg, rural	0.76	0.27-3.1	Malisch 1995
Augsburg, rural	0.52	0.34-0.8	Hippelein et al 1996
	Contaminated areas		
Close to cable smelter		5-24	Beck et al 1991
Close to industrial areas		1.6-6.6	Beck et al 1991
Close to hazardous waste dump Münchehagen	2.12	0.81-7.06	CUA Oldenburg 1991
Impact area of open cable burning	2.05	0.85-5.15	CLUA Münster 1991, 1992
Close to point sources	1.85	0.48-5.62	Lassek et al 1993
Close to former copper plant	1.3	1.08-1.62	Krause et al 1993
Close to PVC fire, Lengerich		0.76-5.86	Documentation Lengerich 1994

United Kingdom (AEA Technology, 1999)

According to numbers reported in a first MAFF survey on dioxins in cows' milk , two dairy farms near Bolsover in Derbyshire, close to a chemical waste incinerator and smokeless fuel works, showed high concentrations, above the Maximum Tolerable Concentration. Milk from these farms was not accepted onto the market until the concentrations fell below the MTC in 1992.

In the period 1993-1995, milk was sampled at 93 farms in the proximity of 29 industrial sites.

Further samples were also taken in 1996 and 1997 (MAFF 1997d; 1997f; 1997g; 1997h; 1997i; 1998a). Findings have been summarised below, grouping them by type of source.

Overall the milk samples showed a range of 0.81 to 20 pg I-TEQ g^{-1} milk fat, but this includes two very high values. A value of 20 pg I-TEQ g^{-1} milk fat was detected near the chemical waste incinerator and smokeless fuel works in Bolsover, and the value of 11 near a municipal solid waste incinerator in Huddersfield. For the purpose of comparison, the expected range for milk from individual farms was considered to be 1.1-7.1 pg I-TEQ g^{-1} milk fat (MAFF 1992); many of the samples in this survey fell into this range. All samples except the one near Bolsover were below the MTC for dioxins in milk.

PCBs have also been analysed in these samples, in order to assess their contribution to the overall toxicity in TEQ. Again, in all samples except one the concentration was below the MTC 16.6 pg I-TEQ g^{-1} milk fat (MAFF 1997d).

Table 2-28: Toxicity in cows' milk from farms near potential sources (AEA Technology, 1999)

Type of potential source	year	N	Equivalent Toxicity Range pg I-TEQ g^{-1} milk fat	Reference
Incinerator of MSW	1992	4	2.3-5.0 * (3.0 *)	Ball et al (1993)
	1994	15	1.2-3.8	MAFF (1997d)
	1995	22	2.1-11	MAFF(1997d; 1997i)
	1996	12	1.9-8.6 (4.6)	MAFF (1997i)
	1997	8	1.1-3.4 (1.9)	MAFF (1997i)
Other incinerator	1993	4	0.87-1.7	MAFF(1997d)
Chemical waste incinerator and smokeless fuel works	1997	19	1-20 (2.6)	MAFF (1997h)
Chemical waste incinerator	1993	9	1.2-2.4	MAFF(1997d)
	1994	12	1.5-3.4	MAFF(1997d)
Sludge incinerator	1995	6	1.9-4.1	MAFF(1997d)
Cement kiln	1994	6	1.5-4	MAFF(1997d)
Chemical works	1993	6	1.2-5.5	MAFF(1997d)
Chemical works and disposal site	1995	3	1.5-2.7	MAFF(1997d)
Metal reclamation plant	1993	1	2.2	MAFF(1997d)
Power station, and Metal reclamation works	1994	1	1.6	MAFF(1997d)
Secondary metals refiners	1996	5	1.1-3.6	MAFF (1997f)
Precious metals refiner	1996	5	1.4-2.1	MAFF (1997f)
Steel works	1996	2	2.2-8.1	MAFF (1997f)
	1997	9	1.1-5.6 (2.2)	MAFF (1997g)
Waste disposal site	1995	8	1.5-6.2	MAFF(1997d)
Recycling plant	1996	2	0.81-1.4	MAFF (1997f)

* = raw data as whole milk; conversion to milk fat basis done assuming 4% fat; n = number of samples; mean values are in brackets if available

Italy

A survey (Green peace Italia, 2002) was carried out on four cows' milk samples from farms at different distances from MSW incinerators.

In order to have a control, a sample from a farm site far from incinerators has been analysed.

The study was designed to test the concentration of dioxins in cows' milk and to investigate a potential correlation between the distance from MSWI's and the concentration of such organic pollutants (Table 2-29).

Table 2-29: Dioxins and furans in 4 cow's milk samples in comparison with the control samples (in bold) (Green peace Italia, 2002 mod.)

Location	Distance farm-incinerator (m)	Incinerator capacity (ton y^{-1})	Dioxins (ng I-TEQ kg^{-1} fat)
Como	4,000	75,000	2.74
Ferrara	5,500	45,000	2.89
Bologna	1,500	140,000	1.23
Bologna	/	/	**0.83**

Finland

An investigation (Hietaniemi, 1995) was carried out to determine the amount of PCBs, OCPs and PAHs in representative samples of Finnish milk and dairy products, cereals and meat products, fish and vegetables.

We hereby report on the findings, highlighting spatial variations, where detected. Regional representative samples of Finnish low-fat (1.9%) milk and cheese were obtained from main dairies in 18 Regions.

PCBs and OCPs (Organochlorine Pesticides). The results showed that organochlorine pesticides were below the limit of detection (0,05 µg kg^{-1} wet weight) in the Finnish milk. Low concentrations of some PCB congeners were found in these samples.

Regional differences in PCB or OCP concentrations could not be assessed because of the low level of PCBs detected in cows' milk (Lang V., 1992).

Higher values have been identified due to contaminations from leaking capacitors and milk silo towers during transportation and storage and from recycled mineral oils. PCBs are absorbed by cows from fodder; the original PCB pattern of the technical product (Aroclor, Clophen, etc.) is then metabolised and partially deposited into the fatty tissue. The lower chlorinated PCB compounds are usually degraded, whereas the higher chlorinated congeners are resistant to metabolisation.

PAHs. Although the level was slight, regional differences in PAH concentrations in milk samples were found (Hietaniemi V., 1995). The highest PAH contents (33 µg kg^{-1}) were found in samples from industrialized areas where fossil fuels are used by large industries. The lowest PAH contents (4 µg kg^{-1}) were detected in milk samples from Lapland and Ahvenanmaa, which are the most sparsely populated non-industrialized regions.

PAH concentrations in cheese were low (1-4 µg kg^{-1}) and regional differences were not detected.

2.4.3 Conclusion

Incinerators and petrochemical and metallurgical activities are generally identified as potential sources of contamination by organic compounds. There are a lot of parameters that can influence the effect of the sources on the content of organic pollutants in foodstuffs, the main ones being: type of industry, chemical identity and path through which the contaminant gets in touch with the foodstuff, distance from the industrial site, etc.

In most cases reported in the literature (France, Italy, UK, Finland) data collected near potential source of contamination show remarkably higher concentrations of organic pollutants than the ones referred to rural areas. However it is not possible to quantify and clearly describe the spatial variation detected, or to define an accurate correlation model between the concentration detected and the types/distances from the source of contamination.

One key remark, as far as a strategic assessment is concerned, is that the ongoing improvements in environmental legislation, and particularly new threshold values for emissions, (e.g. as set for incineration in Directive 2000/76/EC), are likely to reduce concentrations of organic pollutants in foodstuffs produced at farm sites near point sources. Arguably, this may also have positive implications – besides on the safety of commodities - on the future quality trends of organic waste materials used as fertilisers. A similar trend may likely occur for sludge, if Programmes for the Prevention of Pollution (as outlined in the previous Working Documents for the Revision of the Directive 86/278/EEC) were to be adopted.

2.5 Temporal variation: case studies

As far as temporal variation is concerned, some nationwide examples are reported in which OP concentration in foodstuffs (milk, dairy products, meat and fish) have been monitored over several years.

The temporal variations trends collected refer in most of cases to fatty foodstuffs. This is due to the chemical affinity that makes fats privileged carriers for accumulation of OPs.

2.5.1 Germany (AEA Technology, 1999)

In North Rhine Westphalia, sample of dairy products have been monitored for dioxins in 1990, 1994 and 1998. In cases where dairies did not produce milk, butter, cheese or cream products were collected and analysed. The results from these surveys are shown in (Fürst et al 1992, Fürst & Wilmers 1995, Fürst *pers comm.* 1998). The mean decrease in the average concentration in each four-year period was 24%.

Table 2-30: Concentrations of dioxins in cows' milk in North Rhine Westphalia in 1990, 1994 and 1998 (pg I-TEQ g^{-1} fat)

Year	Number of Dairies	Number of samples	Min	Max	Mean	Median	95% Percentile
1990	43	168	0.76	2.62	1.35	1.27	2.04
1994	30	120	0.61	1.75	1.02	1.02	1.45
1998	29	111	0.47	1.78	0.78	0.70	1.29

Another overview of the dioxin concentrations in dairy products from Germany is shown in Table 2-31. The table, which compares various nationwide surveys carried out at different times, shows that there has been a general decrease in concentrations over time.

Table 2-31: Concentrations of dioxins in German milk and dairy products (pg I-TEQ g^{-1} fat) (AEA Technology, 1999 mod.)

Matrix	Min	Max	Mean	Reference
Milk	1.0	2.8	**1.8**	Beck et al 1991
Milk/dairy products	0.76	2.62	**1.35**	Fürst et al 1992
Milk/dairy	0.61	1.75	**1.02**	Fürst et al 1995
Milk	0.6	1.12	**0.87**	Mayer 1995
Milk	0.45	1.12	**0.71**	Malisch 1995

Compared with the large number of milk samples analysed for dioxin in Germany, relatively few data exist for meat and meat products. The existing data were summarised by Fürst (1998) and are shown in Table 2-32. Numbers seem to show a decrease of dioxin concentration in different kinds of meat, although the fairly limited time covered (three years) suggests a certain caution in drawing conclusions on the general trend in a longer time frame. Relatively low concentrations were found in pork, which may be due to the kind of food fed to the animals, the age of slaughter, which affects the built-up of fatty tissues in animals, and the relatively high body fat of the animals, which results in a dilution of the pollutants.

Table 2-32: Concentrations in German meat and meat products (pg I-TEQ g^{-1} fat) (AEA Technology, 1999)

Type of meat	**Min**	**Max**	***Mean***	***Reference***
Pork			**0.5**	Fürst et al 1990
			0.28	Beck et al 1991
	0.22	0.61	**0.41**	CVUA Münster 1992
Suckling pig			**0.13**	CLUA Freiburg 1995
Beef			**3.5**	Fürst et al 1990
			2.6	Beck et al 1991
	0.56	4.31	**1.44**	CVUA Münster 1992
Veal			**7.4**	Fürst et al 1990
	0.03	1.27	**0.70**	CVUA Münster 1992
	0.35	1.13	**0.70**	CLUA Freiburg 1995
Sheep			**2.0**	Fürst et al 1990
			1.65	Beck et al 1991
	0.03	0.59	**0.23**	CVUA Münster 1992
Poultry			**2.3**	Fürst et al 1990
			2.25	Beck et al 1991
	0.50	1.07	**0.70**	CVUA Münster 1992
Meat products			**1.7**	Fürst et al 1990
Lard			**0.8**	Fürst et al 1990

2.5.2 United Kingdom (AEA Technology, 1999)

There have been two surveys on retail milk in the UK, in 1990 and 1995, undertaken by the Ministry of Agriculture, Fisheries and Food (MAFF 1997j). The results of these surveys are shown in Table 2-33. The milk was collected from retail outlets in 12 locations in 1995, and 6 locations in 1990. Range and average (where available) concentrations given in this table show that there has been a decrease over this time frame, from 1.88 to 0.93 pg I-TEQ g^{-1} milk fat.

Table 2-33: Concentrations in cow's milk from background locations and retail sources; (pg I-TEQ g^{-1} milk fat) (AEA Technology, 1999 mod.)

Location	1990 *	1995
Bristol	2	0.9
Cambridge	1.4	0.8
Carlisle	1.1-1.4	0.7
Central London	1.4-3.3	1.0
Commuter London	1.7-2.1	0.9
Exeter	1.1-3.3	1.3
Average	**1.88**	**0.93**

* = raw data as whole milk; conversion to milk fat basis done assuming 4% fat

In comparison with these results the UK Total Diet Survey analysis found that in 1982 average milk concentrations were 4.5 pg I-TEQ g^{-1} milk fat whereas in 1992 this had reduced to 2 pg I-TEQ g^{-1} milk fat (MAFF 1997b).

Table 2-34: Average concentrations in milk (pg I-TEQ g^{-1} milk fat) (AEA Technology, 1999 adapt.)

Year	Concentration
1982	4.5
1992	2.0

Table 2-35 includes the trend for cows' milk from Huddersfield. It can be seen that, in general, the concentrations of dioxins have declined over time. A MSWI was the largest point source of dioxins in this area when the first samples were taken in 1995, but operation ceased in 1996, as it was unable to meet the emissions standard for dioxins. Concentrations of dioxins in milk samples from this area in 1997 were found to have declined to below the Maximum Tolerable Concentration (MAFF 1997i).

Table 2-35: Time trends in cows' milk data for Huddersfield (AEA Technology, 1999)

Year	n	Range of concentrations (pg I-TEQ g^{-1} milk fat)	Mean (pg I-TEQ g^{-1} milk fat)
1995	6	3.1-11	6.3
1996	12	1.9-8.6	4.6
1997	8	1.1-3.4	1.9

Assessment based on Total Daily Intake calculations

Some useful data for drawing up time trends can be extrapolated from the Total Daily Intake calculation.

To get the Total Daily Intake it's necessary to start evaluating the dioxins concentration in the food commodities. This kind of data is mostly found in Total Diet Surveys (TDS). TDS food samples are composites of similar food types, the individual components being purchased at retail outlets.

The first TDS analyses for dioxins were conducted in 1988, at two locations: Stonehaven and Port Talbot (MAFF 1992). A small number of samples was analysed because of the very high costs of test methods to detect dioxins at that time. This means that the results were limited in use because the samples were not fully representative of the UK. The food groups covered were carcass meat, offal, poultry, meat products, fish, fats and oils, eggs, dairy products, green vegetables, potatoes and fresh fruit. Milk samples from a separate retail milk survey were also used in the calculations of total dietary intake and further data regarding fish were also included. (The latter samples - retail milk and fish - had been collected for an earlier study for MAFF (Startin et al 1990), from retail outlets in Norwich). Cereals and cereal products were not analysed directly for the TDS study. However, concentrations had been estimated on the basis of the fat content of the 'Miscellaneous cereals' food group, and assigned the equivalent dioxin concentration based on the 'Fats and oils' group.

In 1994-5, TDS numbers from 1982 and 1992 were compared. Samples were collected from 24 locations across the UK, and eleven food groups were covered giving a true representation of an average UK diet. Fruit and vegetables were not analysed in this survey because they were assumed to contain negligible amounts of dioxins (MAFF, 1995b).

The following data referring to 1982, 1988 and 1992 give an overview of the time trends in several food commodities.

Table 2-36: Concentration of dioxins in some food commodities in the UK in 1982, 1988 and 1992 (AEA Technology, 1999 mod.)

	Concentration in Food groups (pg I-TEQ g^{-1} fresh weight)		
	1982	**1988**	**1992**
Miscellaneous cereals	0.13	0.05	0.17
Bread	0.02	-	0.03
Carcass meat	0.49	0.68	0.13
Offal	1.57	0.46	0.59
Poultry	0.5	0.33	0.13
Meat products	0.32	0.21	0.08
Fish	0.41	0.48	0.21
Fats and oils	1.26	0.65	0.2
Eggs	0.92	0.19	0.17
Milk Products	1.2	0.21	0.16
Milk	0.16	0.08	0.06

A decrease in dioxin concentration is observed in most of food types (meat, meat products, fats and oil, milk and milk products).

2.5.3 Sweden (AEA Technology, 1999)

In 1968 a Swedish program was started to monitor pollutants in the Baltic environment. Samples were taken annually to evaluate temporal trends of contamination in various aquatic species. Target locations were areas with little or no known local discharges. 10-25 specimens represented each annual sample at a site. The selection of samples maintained consistency in sex, age, size and sampling season.

The survey was actually considering various bio-monitors. Only a few of them can be considered as relevant to foodstuff. We hereby report on findings on fish at Lake Storvindeln.

Lake Storvindeln is a forest lake near the Swedish Alps, an Arctic sampling site. Muscle samples from 20 pike collected in spring were analysed each year since 1968. Fig. 2-1 shows the results for PCB and PCDD/PCDF reported in the AMAP (Arctic Monitoring and Assessment Programme) report (AMAP 1998) for these samples. The Figures show the correlation between the various congeners, but also the variation of concentrations over time.

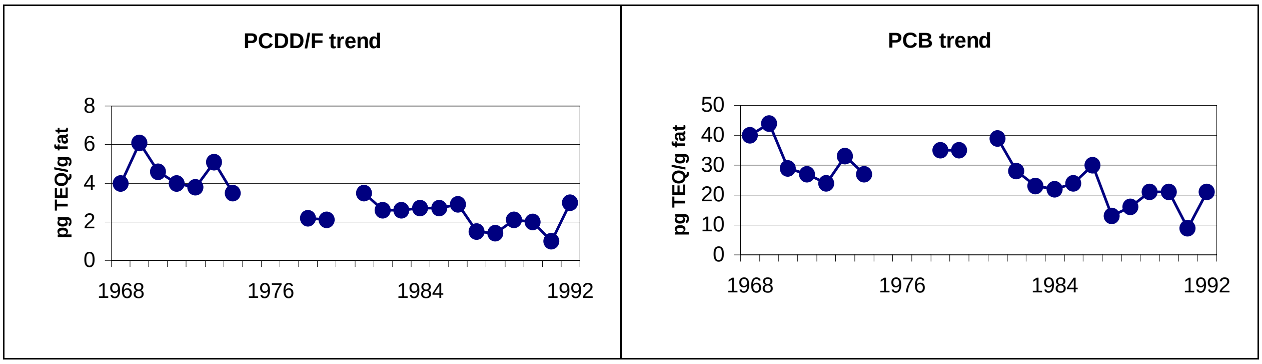


Fig. 2-1: Concentration of PCDD/F and PCB CONGENERS IN lake Storvindeln, Sweden, pike muscle between 1968 and 1992 (AMAP, 1998 mod.)

2.5.4 Germany (AEA Technology, 1999)

A total of 222 butter samples were analysed for dioxin in Baden-Württemberg, Germany. The sampling periods were from 1993 through 1996. As can be seen from Table 2-37, the mean concentrations show a decreasing trend, although a wide variation is detected. None of the samples exceeded the guideline concentration of 3 pg I-TEQ g^{-1} fat, and none of the samples exceeded the limit concentration for retail sale of 5 pg I-TQ g^{-1} fat (Malisch, 1999).

Table 2-37: Germany/ Baden-Württemberg: Dioxin in butter samples (pg I-TEQ g^{-1} fat) (AEA Technology, 1999)

Year	n	Mean	Minimum	Maximum
1993	27	0.83	0.19	1.52
1994	37	0.68	0.46	1.38
1995	92	0.64	0.27	2.00
1996	66	0.55	0.19	0.87

A summary of the dioxin concentrations detected in 667 cows' milk samples collected in Germany between 1993 and 1998 is shown in Table 2-38. The concentrations in the samples did not show the same decreasing trend as the butter samples, although they were coming from the same geographic area. From Table 2-38, it can be seen that, towards the end of 1997, the dioxin concentrations in cows' milk had started to increase. The reason for this was that in Baden-Württemberg the first evidence was detected of the Europe-wide contamination of dairy products, due to high concentrations of dioxins in citrus pellets added to the cattle feed.

Table 2-38: Germany: Dioxin in milk samples (pg I-TEQ g^{-1} fat) (AEA Technology, 1999)

Year	n	Median	Mean
1993	97	0.63	0.68
1994	222	0.67	0.79
1995	104	0.62	0.69
1996	115	0.59	0.60
1997	112	0.62	0.71
01/97-08/97	76	0.59	0.62
09/97-12/97	36	0.82	0.89
01/98-02/98	27	1.06	1.41

2.5.5 Conclusions

The numbers reported show a decrease in detected concentrations in most cases over the years.

Arguably, these trends mainly depend on the implementation of technological improvements with particular reference to equipment to prevent formation and/or avoid dispersion of organic compounds dangerous for human health.

Enforcement of specific environmental legislation (as for instance the EC Directive 2000/76 on Incineration) may arguably be seen as a main driver for such technological improvements.

3 SOIL QUALITY FACTORS INFLUENCING THE CONTENT OF POLLUTANTS IN FOODSTUFFS

3.1 General aspects of mobility and behaviour of pollutants in soils

Soils and plants are major factors influencing pollutant content of foodstuff under given exposure conditions. In this context "soils" stands for a number of specific properties summarised as filter, buffering and transforming capacity and "plants" represent different cultivars or genotypes.

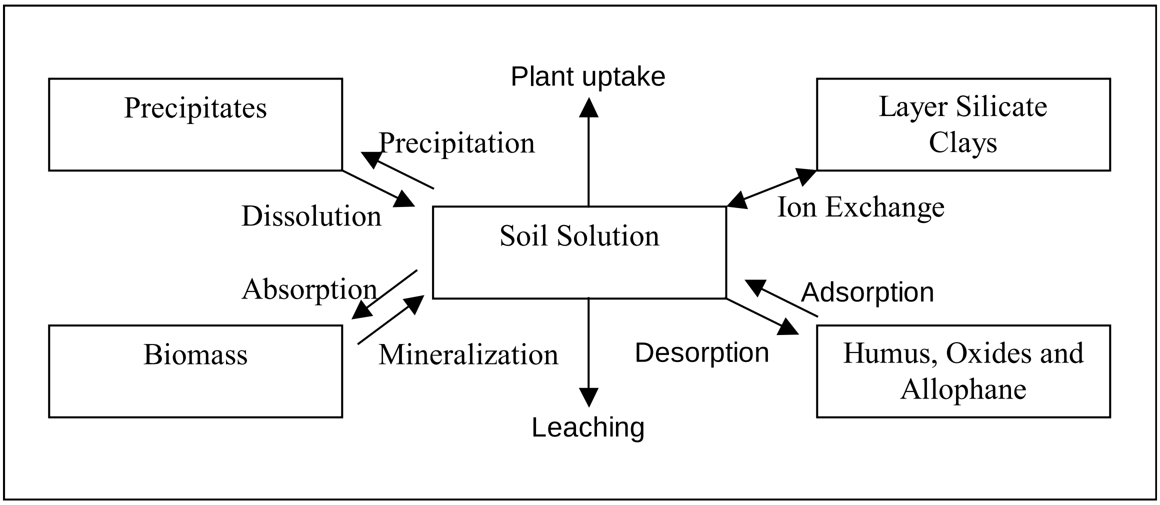


Fig. 3-1: Key interactive processes in the soil system affecting the partitioning of trace metals between the aqueous and solid phases (Adriano, 2001)

In general any contaminant has to be mobile and transported and be "bio available" to be of any consequence to the assimilating organism.

Bioavailability is a dynamic process comprising a physico-chemically driven desorption process and physiologically driven uptake process (de GROOT et al., 1998) expressing

- the fraction present in soil available for intake into plant roots or soil micro-organisms
- the fraction in soil, dust or diet available for uptake in the gastro-intestinal tract of animals and humans (Thornton, 1999)

A number of interactive biotic and abiotic processes govern the behaviour of heavy metals in soils (see Fig. 3-1) and define chemical speciation and bioavailability (Adriano, 2001).

The major factors driving the biogeochemical processes in soils are pH, Cation-Exchange-Capacity and redox potential.

According to their response or sensitivity to heavy metal concentrations in soils plants can be divided in three major groups (Adriano, 2001):

- Excluders: insensitive to metals over a wide range of soil concentrations (e.g. sudan grass, bromegrass, fescue).
- Indicators: plant species responding correspondingly to metal concentrations in soils (e.g. corn, soybean, wheat, oats).
- (Hyper)accumulators: plant species accumulating heavy metals (e.g. lettuce, spinach, tobacco).

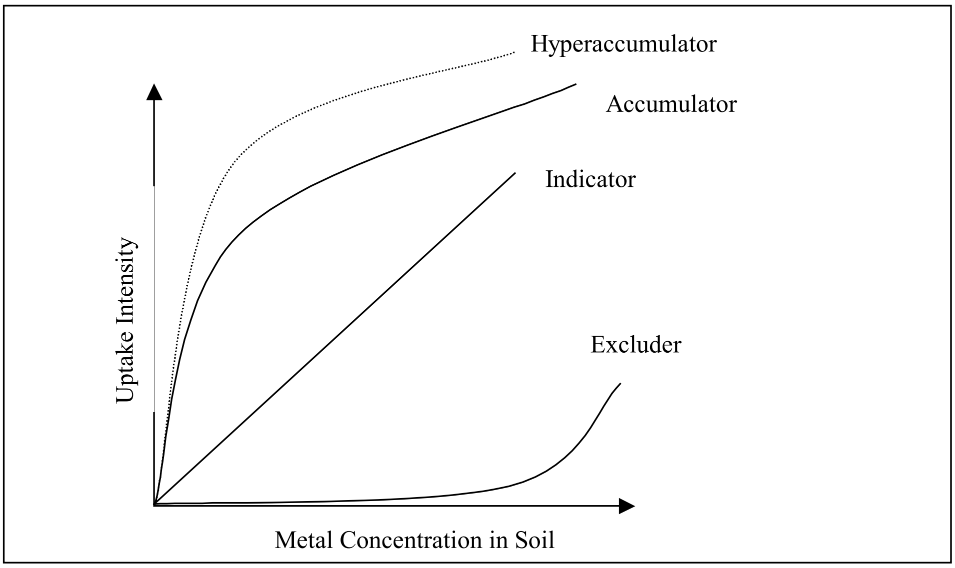


Fig. 3-2: Relative uptake and bioaccumulation potential among plant species (Adriano, 2001)

Indicators of bioavailability for heavy metals in the soil-plant pathway are the determination of soluble metal-fractions by sampling of soil solution (via suction cups, lysimeter or centrifugation) or using "weak" extractants (e.g. $CaCl_2$, NH_4NO_3 etc.). The so-called extractable fraction generally correlates closely with plant uptake, but varies in time and space due to seasonal and climate fluctuations or soil management practices.

"Total" PTE concentration detected after digestion with strong acids is the commonly agreed basis for long term soil conservation. (Grün et al. 1990 quoted in Wilcke & Döhler, 1995). Moreover, the assessment of mobilisation dynamics is hardly possible, since additional information on pH, clay content, SOM etc. is needed, and this is widely site-specific. The assessment of the "bioavailable only" fraction looks less agreeable and come short of a precautionary approach, as the availability of PTEs may change over time, depending on variations in the physic and chemical state of the soil. . In the framework of soil protection and research programmes the conditions for PTE mobilisation were simulated with several extracting agents such as weak acids $NaNO_3$, NH_4NO_3, DTPA, EDTA, $CaCl_2$, water saturated extract, DTPA–$CaCl_2$ solvents.

With these digestions, methods for better correlation with transfer rates into the plant and percolated water respectively were found. The ratio of extracted element depends on the strength of the solvent, the capability of exchanging metals adsorbed to clay minerals, organic complexes or pedogenetic oxides and the strength of sorption of the individual element.

Following the German Soil Protection Ordinance (BBSchV, 1999) the 1 M NH_4NO_3 extract is used for the determination of potentially available heavy metal fractions in the soil. Only 0.5 to 2.5 % of the "total" (aqua regia extractable) Cd and Zn concentration is detected in compost with this method. For Cu the maximum value is 5 % of the total content (Gäth, 2001)

Though there is an agreement that an available fraction would fulfil the demand for a understanding of the potential eco-toxicological impact of heavy metals by far better than "total" figures there is no consensus on the specific method up to now.

Following the discussion on the need of harmonisation of analytical methods in the area of environmental assessment (soil – sludge – organic fertiliser) there is a clear tendency not to follow the strategy of site specific evaluation by measuring plant available fractions (Langenkamp & Marmo, 2001; Langenkamp et al., 2001).

Aqua Regia ($HCl_{conc.}$: $HN0_{3\ conc.}$ = 3 : 1) is still the first choice and was adopted as preferred extracting agent for sludge, soil improver and growing media as well as soil.

Other indicators used are:

$$\underline{\textit{soil plant transfer factors}} = \frac{\textit{metal concentration in plants}}{\textit{metal concentration in soil}} \qquad \textit{(see Table 3-1)}$$

$$\underline{\textit{transfer coefficients}} = \frac{\textit{increase of metal concentration in plants}}{\textit{increase of metal concentration in soil}} \qquad \textit{(see Table 3-2)}$$

Table 3-1: Transfer factors of trace elements in the soil-plant pathway (Adriano, 2001*)

	soil (mg kg^{-1})			Transfer factor			
Element	Range		Average	Grain	Vegetable	Root/tuber	Fruit
As	0.1	40	6	0.004	0.037	0.004	0.003
Cd	0.01	2	0.35	0.036	0.223	0.008	0.09
Pb	<1	300	19	0.002	0.0016	2E-5	14E-5
Hg	0.01	0.5	0.06	0.085	0.009	0.002	0.009
Se	0.01	1.2	0.4	0.002	0.015	0.042	0.021

*Data derived from Kabata-Pendias & Pendias (1984) and Fergusson (1990).

Table 3-2: Transfer coefficients of trace elements in the soil-plant (edible parts) pathway (Lübben & Sauerbeck, 1991)

	range	plants	range	plants	range	plants	range	plants
Cd	0.01-0.1	maize pea	0.1-0.5	field salad oats, wheat carrot dwarf bean	0.5-1.0	leek	1.0-3.0	little radish curled lettuce spinach celery
Cr	0.001-0.005	maize, pea oats, wheat leek dwarf bean carrot, celery	0.005-0.01	curled lettuce field salad	0.01-0.02	spinach	0.02-0.03	little radish
Cu	0.01-0.05	maize celery, pea leek, carrot	0.05-0.1	dwarf bean oats, curled lettuce, wheat spinach	0.1-0.2	little radish field salad		
Ni	0.01-0.1	maize curled lettuce	0.1-0.2	wheat	0.2-0.5	celery leek carrot pea	0.5-1.0	little radish oats field salad dwarf bean spinach
Pb	0.001-0.005	maize, oats Wheat. pea dwarf bean	0.005-0.01	carrot celery	0.01-0.02	field salad leek curled lettuce little radish	0.02-0.05	spinach
Zn	0.1-0.2	maize pea	0.2-0.5	Oats, carrot wheat dwarf bean leek, curled lettuce	0.5-1.0	celery	1.0-3.0	field salad

The occurrence of organic contaminants in plants is more a result of atmospheric deposition than absorption via contaminated soils, especially regarding hydrophobic substances like PCB, PCDD/F and PAHs.

With organic pollutants deposited on soils the main focus is set on mineralisation or transformation capacity in the soil compartment, determined through soil properties like organic matter content, pore volume and water content (Blume & Brümmer, 1987; Litz & Blume, 1989).

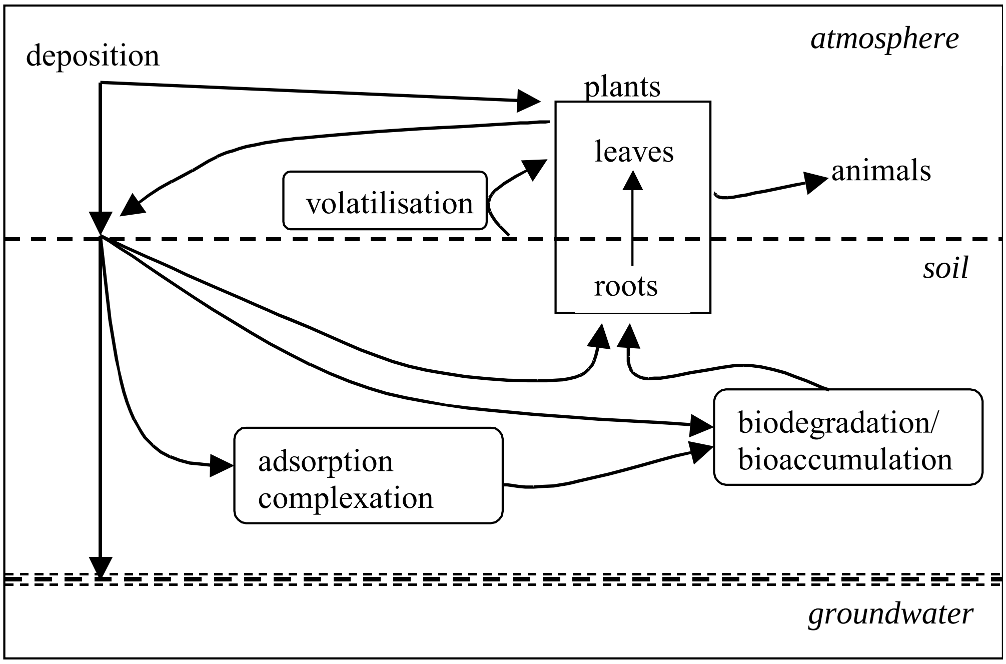


Fig. 3-3: Relevant processes in the soil-plant pathway of organic compounds (Becker-van Slooten & Tarradellas, 2000; modified)

In general compound specific properties like water-solubility, octanol/water-coefficients (logK_{ow}), volatilisation (H_c) and persistence (half-life-time) are used to describe their environmental fate. Extent of plant uptake due to absorption on the surfaces of roots and tubers is depending on the contents of lipophillic substances within this organs. For leaves volatilisation from soils followed by foliar uptake is considered the most important pathway (Rommel et al., 1998).

Soil and plant concentrations of organic pollutants vary in a great range and do not show clear correlations. This is mainly caused by the fact that the different pathways (soil-plant; soil-air-plant; air-plant) lead to overlapping effects. Transfer-factors (plant/soil) for organic pollutants cannot be used in a general manner for risk assessment (Rommel et al., 1998)

3.2 Background concentrations of inorganic and organic pollutants in European soils

PTEs (Cd, Cr, Cu, Hg, Ni, Pb, Zn)

Background concentrations of heavy metals in soils stemming from soil parent material (rocks, sediments) serve as reference values. Their exceeding is used for identification of anthropogenic soil pollution and they serve as a basis for soil protection regulations in many European countries.

Problems dealing with background concentrations (at least on a supra national level) arise from

- their great variability due to spatial variation of soil parent material and soil forming conditions,
- the matter of fact that "natural" soils (i.e. without any anthropogenic influence) are scarce due to historical land use patterns at least in central Europe and diffuse immissions,
- different sampling and analytical methods

There is a difference between background concentrations and soil limits (or threshold values). The variability of background concentrations stems from different parent material and soil forming conditions whereas soil limits are set from an (eco) toxicological point of view taking into account different protection goals. In most cases the most sensitive protection goal is used to finally determine limit values. As a matter of fact it happens quite often that "global" soil limits are exceeded by background concentrations (e.g. As or Ni) and site-specific limit values have to be derived regionally.

The German Federal Environment Agency and DG ENVIRONMENT of the European Union commissioned a feasibility study on „Trace element and organic matter contents of European soils“. The project is not finished yet. Preliminary results stemming from the provisional report (Gawlik et al., 2003) are shown below. Data on heavy metal contents in soils were harmonised to aqua-regia extractable amounts by using conversion algorithms, variabilities due to different sampling strategies have not been taken into account yet. Although data compilation and harmonization on an European level has made a reasonable step forward with this study, the authors claim that *"the bigger part of the standardisation problems still has to be solved"*.

Data shown in the table below represent the *"usual content"* of heavy metals in top soils of all land use types regarded (i.e. arable land, grassland, forest, other land use; main focus lies on arable land and grassland). *"Usual content"* reflects the *"concentration of a substance in soils resulting from both the natural pedo-geochemical content and moderate diffuse source input into the soil"* (ISO/CD 19258, 2002).

Table 3-3: Range of heavy metal contents (median) according to soil parent material* and with regard to all land use types [mgkg^{-1}] (Gawlik et al., 2003)

Soil parent material	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Undifferentiated alluvial deposits (or glacial deposits)	0,07-0,76	7 – 40	4 – 30	0,02-0,24	4 – 34	6 – 64	13 – 128
Calcareous rocks	0,26-1,23	10 – 56	12 – 24	0,06-0,23	13 – 39	16 – 63	49 – 100
Clayey materials	0,14-0,90	9 – 50	7 – 22	0,05-0,13	7 – 36	9 – 37	25 – 78
Sandy materials	0,09-0,30	5 – 37	2 – 21	0,04-0,18	3 – 22	9 – 39	6 – 62
Loamy materials	0,10-0,85	10 – 62	8 – 21	0,04-0,18	9 – 29	11 – 54	35 – 86
Detrital formations	0,18-1,23	16 – 49	12 – 24	0,10-0,13	21 – 32	17 – 46	52 – 107
Crystalline rocks and migmatites	0,11-0,94	8 – 43	11 – 31	0,04-0,20	10 – 38	15 – 73	40 – 101
Volcanic rocks	0,25-1,48	10 – 68	10 – 32	0,11-0,29	8 – 48	26 – 58	55 – 130
Other rocks	0,10-1,00	5 – 40	4 – 32	0,07-0,24	3 – 37	14 – 88	19 – 125

* highest aggregation level (MAT 11) taken from the European Soil Data Base

As background value for the scenario in chapter 9 we took the mean background values on sand and clay soils in DE, FR and DK. The precautionary guide values for sand and clay soils of the German Soil Protection Ordinance should reflect an example of reference values for multifunctional use of agricultural soils.

Table 3-4: Background-concentrations in soils in mg kg^{-1} d.m. (Arthur Andersen on behalf of DG Environment, 2001)

	Cd	Cu	Cr	Hg	Ni	Pb	Zn
				sandy soils			
DK	0.1	5.6	6.4	0.03	2.9	10.5	18.4
FR**	0.2	9.1	n.a.	0.04	10.5	18.2	35
	0.2	9	n.a.	0.04	12.3	25.1	43
DE	0.1	7.1	8.9	0.05	6.4	24	27.3
UK	1	23	59	n.a.	38.2	35	106
				clay soils			
DK	0.2	9	17.1	0.05	9.6	12.1	43.3
FR**	0.3	15.8	n.a.	0.04	24.7	26	65
	0.3	15	n.a.	0.05	29.5	29.9	73
DE	0.3	23.8	27.8	0.08	33.3	36.4	76.4
UK	<0.2	7.4	13.2	n.a.	7.5	22	35

** results of two different extraction methods

3.3 Limits and threshold values for PTEs in European soils

On European level a derivation of PTE threshold values differentiated for according to for instance soil parent materials or particle size distribution (sand, loam, clay) as established in Germany is still missing. Nevertheless following the EU Sludge Directive (86/278/EEC) most countries have set threshold limits for heavy metal concentrations in agricultural soils together with ordinances and directives regulating the reuse of sewage sludge and/or composts derived from municipal waste (see Table 3-6).

Only Germany and the Netherlands have chosen a differentiated approach considering soil organic matter, clay content or particle size distribution for the establishment of soil guide or threshold values.

The German so-called precautionary threshold values are established in the Federal Soil Protection Ordinance (BBSchV, 1999) and provide three concentration lavels depending on the soil particle sice caharcteristics (sand, loam/silt, clay). The concept is explained in detail in chapter 8.3.3.

The Netherlands have established soil target values, indicating if not being exceeded, a multifunctional use of the soil.

The target values for soil/sediment for metals depend on the concentration of organic matter (*humus*) and clay. In assessing the quality of the soil the values for a standard soil are converted to values applying to the actual soil concerned on the basis of the measured organic material (measured as volatile solids / loss of ignition), and clay content (percentage of particles with a diameter of less than 2 µm). The converted values can then be compared with the measured concentrations in the soil. The conversion formulas are given in Table 3-5:

Table 3-5: Substance dependent constants and resulting convertion formulas for the derivation of heavy metal target values

	Constants				convertion formulas
	A	B	C		Mg kg^{-1}
As	15	0.4	0.4	→	15+0.4*(CC+OM)
Cd	0.4	0.007	0.021	→	0.4+0.007*(CC+3OM)
Cr	50	2	0	→	50+2CC
Cu	15	0.6	0.6	→	15+0.6*(CC+OM)
Hg	0.2	0.0034	0.0017	→	0.2+0.0017*(2CC+OM)
Ni	10	1	0	→	10+CC
Pb	50	1	1	→	50+CC+OM
Zn	50	3	1.5	→	50+1.5*(2CC+OM)

CC … Clay Content (grain size < 2 µm); OM … Organic Matter (percentage in the soil)

Table 3-6: Limits of heavy metal concentrations in soil in mg kg^{-1} d.m. (EU15)

	86/278/EEC	**Austria[1]**	**Belgium**		**Germany**				**Denmark**	**Spain**		**France**	**Finland**
	6<pH<7		**Flanders**	**Wallonia**	**Sludge directive**	**Soil Protection Ordinance**				**pH<7**	**pH>7**		
						sand	**loam (ph<6)**	**Clay (ph<6)**					
Cd	1 – 3	1 – 2	1.2	2	1.5	0.4	1 (0.4)	1.5 (1)	0.5	1	3	2	0.5
Cr	-	100	78	100	100	30	60	100	30	100	150	150	200
Cu	50 – 140	60 – 100	109	50	60	20	40	60	40	50	210	100	100
Hg	1 – 1.5	1 – 2	5.3	1	1	0.1	0.5	1	0.5	1	1.5	1	0.2
Ni	30 – 75	50 – 70	55	50	50	15	50	70	15	30	112	50	60
Pb	50 – 300	100	120	100	100	40	70	100	40	50	300	100	60
Zn	150 – 300	150 – 300	330	200	200	60	150 (60)	200 (150)	100	150	450	300	150
		As 20	As 27										
		Co 50											
others		Mo 10											

[1] ranges due to different regulations in Federal Countries

Table 2–8: Limits of heavy metal concentrations in soil in mg kg^{-1} d.m. (EU15) *continued*

	Greece	**Italy**	**Ireland**	**Luxembourg**	**Netherlands**	**Portugal**			**Sweden**	**United Kingdom[3]**			
						pH<5.5	**5.5<pH<7**	**pH>7[2]**		**5<pH<5.5**	**5.5<pH<6**	**6≤pH≤7**	**pH>7**
Cd	1 – 3	1.5	1	1 - 3	0.5	1	3	4	0.4	3			
Cr	-	<1µM Cr (VI)	-	100 - 200	30	50	200	300	60	400[4]			
Cu	50 – 140	100	50	50 - 140	40	50	100	200	40	80	100	135	200
Hg	1 – 1.5	1	1	1 - 1.5	0.5	1	1.5	2	0.3	1			
Ni	30 – 75	75	30	30 - 75	15	30	75	110	30	50	60	75	110
Pb	50 – 300	100	50	50 - 300	40	50	300	450	40	300			
Zn	150 - 300	300	150	150 - 300	100	150	300	450	100 - 150	200	250	300	450
others										Mo 4[4]			
										Se 3[4]			
										As 50[4]			
										F 500[4]			

[2] only fodder production [3] max. permissible concentration in soils after sludge application [4] non statutory

3.4 Organic pollutants (PCB, PCDD/PCDF) in European soils

Different from heavy metals the term “background” concentrations of organic pollutants refers more or less to a determinable extent of anthropogenic pollution, since these compounds do not occur naturally in soils or soil parent material. Consequently a differentiation has to be drawn from land use patterns related to soil management practice. Moreover background concentrations can only be used with persistent organic pollutants.

Organic pollutants might enter soils via the following pathways (Brändli et al., 2003):

- Atmospheric deposition: compounds from incineration such as dioxins, ubiquitous compounds like phthalates or *organochlorine pesticides*, and pesticides not considered as ubiquitous,
- Road spray drift and deposition (mostly particle-bound),
- Application of plant protecting agents (pesticides),
- Agricultural supplies such as fertilizers (mineral fertilizers, farmyard manure, sewage sludge, compost, other waste products), soil improvers, mulching agents, pesticides.

Organic pollutants in soil undergo the following processes:

- Volatilisation
- Solubilisation in water (water is the most important transport medium in soil)
- Sorption (adsorption and absorption) including formation of bound residues,
- Physical, chemical or biological decay or degradation
- Incorporation into organisms and plants (bioaccumulation)

These processes are guided by the following factors:

- Physico-chemical properties of the substances,
- Type and structure of the soil,
- Biological activity of the soil (microorganisms, invertebrates and plants).

The persistence of a substance is characterized by its half-life ($t_{1/2}$), which is defined as the time necessary to degrade 50 % of a substance. It is usually determined in laboratory experiments[14]. Some phthalates, tensides, phenols and benzenes are relatively rapidly degraded with a half-life between 1 week and 1 month. Other compounds like di-(2-ethylhexyl)phthalate (DEHP) or linear alkylbenzyl sulphonates (LAS) may exhibit a higher half-life under certain conditions (Hesselsoe et al., 2001). Half-life of chlorinated hydrocarbons increases proportionally with increasing substitution with chlorine. For PCDD/F, PAHs and PCBs a $t_{1/2}$ is commonly reported between 1 and 10 years. Half-life of substances such as DDT is even higher than 10 years (Jauzein et al., 1995).

---

[14] OECD 304A, 1981, Inherent Biodegradability in Soil; OECD draft 307, 2000, Aerobic and Anaerobic Transformation in Soil; ISO 11266, 1994, Soil quality - Guidance on laboratory testing for biodegradation of organic chemicals in soil under aerobic conditions

Table 3-7: Typical concentrations of organic contaminants in soils in μg kg^{-1} d.m. (others indicated) (data source: UMEG, 1999; Litz, 1998; cit. in Erhardt & Prüeß, 2001)

	area	land use	Median/ 50%ile	90%ile
EOX	rural area		0.3mg kg^{-1}	0.6mg kg^{-1}
	urban area		0.4mg kg^{-1}	0.9mg kg^{-1}
Phthalates **(EPA 606)**		arable land	786	1,262
		grassland	893	1,825
PAH **(EPA 16)**	rural soils		0.82	3.99
	urban soils		2.67	10.92
PCB **(IUPAC 6)**	rural soils		14	98
	urban soils		34	243
		arable land	10	40
		grassland	16	101
		private gardens	30	284
PCDD/F			2.44 ng I-Teq(NATO) kg $^{-1}$ dm n = 132	13.3 ng I-Teq(NATO) kg $^{-1}$ dm n = 132
HCB			<1.0	5.0
HCH$_{sum}$			<1.0	4.3
DDT$_{sum}$			3.6	45.5

Threshold values for DDT, PCB, PAH and PCDD/F related to the soil/plant/animal pathway have been set in Germany on a precautionary basis (see Table 3-8).

Table 3-8: Threshold values for organic pollutants in respect to the contamination pathways soil-plant and soil-animal (Erhardt & Prüeß, 2001)

Substance	unit	threshold value	pathway
HCB. HCH Heptachlor. Endrine	mg kg^{-1} soil	0.05	Soil-plant/-animal
DDT-Sum	mg kg^{-1} soil	0.10	Soil-plant/-animal
PCB (congeners)	mg kg^{-1} soil	0.05	Soil-plant/-animal
PAH 16	mg kg^{-1} soil	10	Soil-plant/-animal
BaP	mg kg^{-1} soil	1	Soil-plant
PCDD/F	ng I-TEq kg^{-1} soil	40	Soil-plant/-animal

4 POTENTIAL TOXIC ELEMENTS [PTEs] IN COMPOST

According to the existing legal standards and requirements for composts, the following pollutants are considered:

PTEs: As, Cd, Cr, Cu, Hg, Ni, Pb, Zn

A two-fold approach is employed for the evaluation of compost contamination with potential pollutants :

i. National surveys on different compost types
ii. Specific investigations on well documented sources of used raw materials

The following compost types were identified as defined by characteristic (dominant) source materials:

i. Municipal solid waste (MSW) / mechanical-biological treated waste (MBT) compost / stabilised organic waste fraction (MSWC; MBTC)
ii. Biowaste compost (BWC): compost of source separated kitchen waste from private households and industrial origin if collected in a common system; mixed with the necessary proportion of structural or bulking material (chopped or shredded tree and bush cuttings)
iii. Green waste compost (GC): compost made of garden and park waste such as grass clippings, shredder material, chopped wood, leaves, flower residues etc.
iv. Manure compost (MC)
v. Sewage sludge compost (SSC)

4.1 MSW / MBT compost / stabilised organic waste fraction (MSWC; MBTC)

Mechanical-biological treatment of the residual waste remaining after source separation of hazardous waste is designed to separate mixed waste from municipal collection systems. The creation of an inert inactive fraction before final disposal is achieved by biological stabilisation of the fine fraction (< 60 – 80 mm) after removal of metals and fractions with high caloriic value. The mechanical pre-treatment includes screening, and ballistic separation (magnetic metal separation, wind separation of the light fraction and separation of fractions with a high specific weight). The basic flow chart of mechanical biological pre-treatment is shown in Fig. 4-1.

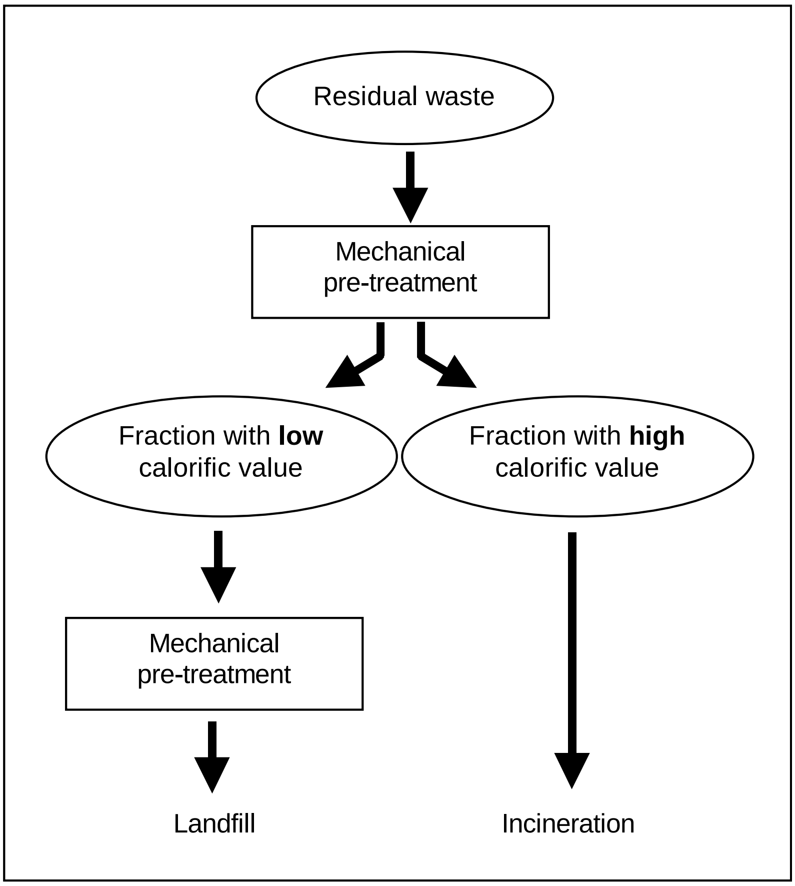


Fig. 4-1: Flowchart of basic steps in mechanical biological pre-treatment of residual waste (Binner, 2002)

After examining the range of mean values and ranges of heavy metal contents in MSWC from several surveys in European countries it becomes evident that diversity and the effect of relevant conditions such as

- collection systems (with or without separate collection of hazardous waste, glass, metals, paper, organic waste),
- settlement characteristics (urban, rural, industrialised areas),
- pre-treatment of mixed MSW (screening, metal separation etc) and
- preparation of the final MSWC fraction (mesh size of final screening, separation of plastics, metals and high weight fraction)

play a significant role in the final quality.

4.1.1 PTEs in MSW and MBT compost

Ranges of mean values derived from national surveys are summarised in Table 4-1.

Table 4-1: Range of PTE concentrations in MSWC and MBTC from national surveys (range of mean values)

Cd	Cr	Cu	Hg	Ni	Pb	Zn	As
------------	------------	------------	*mg kg^{-1} d.m.*	------------	------------	------------	
1.7 – 5.0	70 – 209	114 – 522	1.3 – 2.4	30 – 149	181 – 720	283 – 1,570	(12.7)*

* only one investigation

As one can see from the examples of older and more recent data on MSWC and stabilised material from MB treatment (see Table 4-2 and Table A5–3 in Annex 5), modern pre-treatment techniques and general source segregation for paper, glass, metals and hazardous waste are still no guarantee of a significant reduction of heavy metal levels. Reduction might very much depend on the existence of a full-scale source separation scheme for organic waste in the catchment area, which serves as *qualitative dilution agent,* if left in residual MSW for composting. Another important factor is settlement structure and the exclusion of industrial waste.

In Table 4-2 older data from MSWC are compared with recent investigations following modern pre-treatment techniques and pre-removal of recyclables and hazardous household waste. It is obvious that no systematic quality increase is visible. There is only one example from a mixed waste composting plant with a purely rural catchment area in Scotland (Anderson, 2002) where PTE concentrations are significantly reduced. In this case no older data from the same facility were available for a comparison of quality development.

More detailed figures on mean and median values are listed in Table A5-3 in Annex 5.

Table 4-2: Heavy metal contents of MSW and MBT composts

		MSW-Compost (older data)						
		Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
		----------	----------	----------	*mg kg^{-1} d.m.*	----------	----------	----------
(1) A 1984-86; mean. n = 32		5.0	98	333	-	80	728	1.450
(2) A 1988-90; mean. n = 25		3.3	85	455	2.5	71	461	1.187
(3) D before 1985		5.5	71.4	274	2.4	44.9	513	1.570
(4) D; mean. n = 128		3.0	164	330	2.3	87.6	588	915
(5) F; mean; n = 9-56		4.62	126.34	164.37	1.64	60.35	325.92	554.28
(6) I; mean; n = 14		2.80	78.9	177.8	-	41.8	365.7	1.025
		MBT-Compost						
(7) A 1998-2000; n = 9	range	0.7-6.1	24-344	161-500	0.1-4.1	18-253	64-963	235-990
	median	2.7	209	247	1.3	149	224	769
(8) UK 2001; 1 plant;	range	0.22-1.87	3.7-50.6	25.3-306	0.001-0.93	9.6-93.9	73.4-683	130-560
n = 16; *30 % OM basis*	median	0.41	15.8	91.1	0.15	31.0	166.8	286

(1) Lechner (1989); (2) Amlinger et al. (1990); (3) LAGA (1985); (4) Ulken (1987); (5) Charonnat et al. (2001); (6) Centemero (2002); (7) Amlinger et al. (2000); (8) Anderson (2002)

Table 4-3 compares limit values for MSWC and MB treated residual waste with the averaged threshold values from biowaste and green waste compost.

Table 4-3: Heavy Metal limit values for MBTC (stabilised biowaste from MBT) in Austria, Italy and the proposal of the 2nd draft of the working document *Biological treatment of Biowaste*

		Example Limit values for MBTC						
		Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
		mg kg^{-1} d.m.						
Austria	Compost Ordinance (2002); final landfill cover	3	250	500	3	100	200	1,800
	Landfill Directive waste for disposal	30	5,000	5,000	20	2,000	3,000	5,000
Italy	Draft Decree (MBTC) 1st quality (land reclamation)	3	--- *	300	3	100	280	1000
	DCI 27/07/84 (MSWC)	10	500 **	600	10	200	500	2500
EU, WD 2nd draft	stabilised biowaste 1)	5	600	600	5	150	500	1,500
Averaged limit values for BWC in European countries		1.4	93	143	1.0	47	121	416

* Cr^{VI}: 3 kg^{-1} d.m.; no limit value for Cr_{tot}; ** Cr^{VI}: 10 kg^{-1} d.m.

1) proposed areas of application: final landfill cover, landscape restoration in old and disused quarries and mines, anti-noise barriers, road construction, golf courses, ski slopes, football pitches and the likes

As a result of the increase of quality due to separate collection, also the limit values for BWC and CG have been lowered by the factor 2 to 10.

4.2 Separate collection systems – quality as a result of source materials

This chapter differentiates between potential influences (green waste – biowaste; urban or rural origin; seasonal variations) on compost quality in a more systematic way only for PTEs (heavy metals). Because only a few data sets and publications were available for organic micro pollutants analogous statements were impossible. For this reason organic pollutants are only treated in chapter 5, with the exception of stabilised material from mixed solid waste and sewage sludge,.

4.2.1 Background concentrations in potential source materials

Because the overall goal is a beneficial and environmentally sound recycling policy for decomposable organic waste, it is important to assess what types of potential source materials and which specific origins are suitable for integration into the organic waste recycling stream. Knowing what quantities of pollutants could be expected in waste might be advantageous for identifying potential reduction measures (*cleaner production*) as well as applying the optimum material management by excluding *risk material* at the processing plant.

When examining background concentrations of source materials for the processing of compost the effect on the final product should be the priority. Thus we have to consider how the specific element concerned or compound will behave throughout the biological process until the time when the product is ready for use. The main processes at work here are

- Concentration / accumulation processes (non-degradable or poorly degradable constituents)
- Degradation / decay / decomposition / metabolism
- Volatilisation or leaching and
- Sorption to organic compounds

Since the following section only adresses non degradable PTEs and focusses on the final compost product, only the concentration/accumulation process is discussed. .

The knowledge of the concentration factor for pollutants during rotting is significant for the estimation of the quality to be achieved for known compositions. The actual concentration depends on the input content of organic matter and decomposition rate . The decomposition rate normally ranges between 50 and 70%. The simplest and most used method is to relate the element concentration on a dry matter basis (at the actual organic matter (OM) level) to a standard OM level (e.g. 30 % or 40 %) assumed for the final compost. These figures meet the common range of OM content of ready-made organic waste composts.

This standardisation-method was also introduced in compost standards such as the first compost standard in Austria from 1993 (ÖNORM S2200; withdrawn with April 2002 after the enactment of the Compost Ordinance), the German RAL GZ 251 ‘Quality Standards for Composts’ and was proposed for the definition of quality classes in Annex III of the 2nd draft of the working document *Biological treatment of Biowaste*. In all cases the standard reference value for OM was 30 % d.m.

The calculation follows the formula:

$$PTE_{STD} = \frac{100 - OM_{STD}}{100 - OM_{ACT}} \times PTE_{ACT}$$

where

PTE_{STD} … is the PTE concentration at the standardised OM level

OM_{STD} … is the standardised OM level

OM_{ACT} …is the actual OM concentration of the material concerned

PTE_{ACT} … is the PTE concentration at the actual OM content of the material concerned

Riess et al. (1993) described enrichments of 110 – 230% of the initial heavy metal concentration in a mixture of grass with chopped wood. The authors demonstrate that for the evaluation of raw materials this method might still lead to some uncertainties: it appeared that grass clippings from green strips harvested with a sucking device show a significantly higher contamination than those having been picked up manually. The difference is partly reversed by the conversion of the heavy metal contents to the basis of 30% organic matter as a result of the lower ash content in grass.

Ranges and single data of PTE- concentrations in source materials for composting and anaerobic digestion are shown in Annex 4. Table 4-4 summarises the background levels for the most important categories of feedstock materials, *mixed garden waste, kitchen waste and wood clippings or shredder material* as compared to sewage sludge, the weighted median and weighted 90th percentile values for PTEs of national compost data and background concentration in Austrian arable soils.

Table 4-4: Heavy Metal Content of different feedstock as Value Ranges [mg kg^{-1} d.m.]; not standardised to 30 % OM d.m.

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Garden Waste	0.07-0.65	3-24.6	11-68.8	0.05-0.18	3-17.0	6-30.3	46-100
Kitchen waste	0.059-1.4	1.65-9	10-25	0.029	1.6-30	2-20	20-55
Wood clippings	< 0.1-0.4	6.5-17.6	5-20	0.05< 0.1	4.0-8.4	13-20	20-137
Sludge: EU – range of means [1)]	0.4–3.8	16-275	39-641	0.3-3	9-80	13-221	142-2,000
Median BWC[2)]	0.46	21.00	47.33	0.17	17.00	62.67	181.00
90th percentile BWC[2)]	0.89	37.40	79.50	0.35	29.73	105.17	284.17
Soil 75th percentile [AT] [3)]	0.28	48	28	0.22	30	23	92

[1)] Arthur Andersen on behalf of DG Environment (2001)

[2)] statistically weighted median and 90th percentile values for PTEs of European national compost data

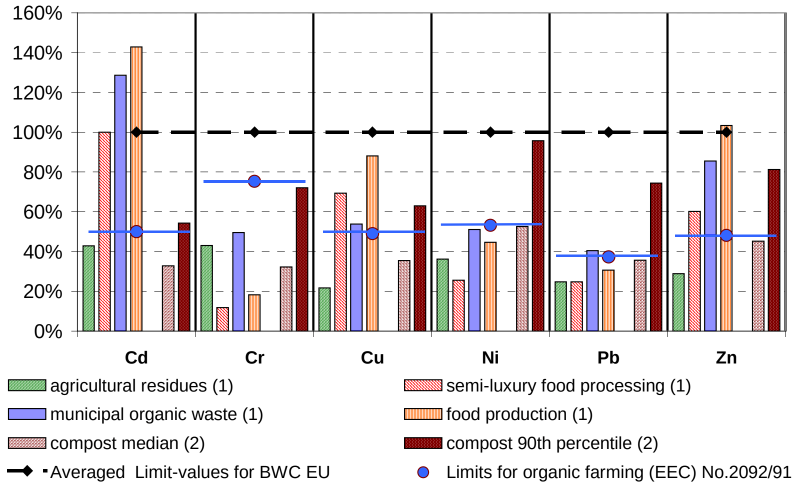
[3)] 0-20 cm arable land in Austria (Amlinger & Peyr, 2001);

Table 4-5 summarises a German investigation (Sihler & Tabasaran, 1996) on feedstock materials for composts and compares the analytical results at the actual OM level of the materials and the concentration after theoretical decomposition to a standardised value of 30 % OM (bold numbers)

Table 4-5: Mean PTE concentration of organic residues and waste materials as initial value [mg kg^{-1} d.m.] and standardised to 30% OM d.m. [bold numbers)

	n	OM [%]	Cd	Cr	Cu	Hg	Ni	Pb	Zn
agricultural residues (1)	16-17	74 **30**	0.4 **0.6**	26 **40**	18.5 **31**	-	9.5 **17**	18 **30**	70 **120**
municipal organic waste (1)	23	85 **30**	0.3 **1.8**	8 **46**	18 **77**	-	8 **24**	16 **49**	70 **356**
food production (1)	17-19	89 **30**	0.2 **2**	1.4 **17**	11.5 **126**	-	2.4 **21**	3 **37**	40 **430**
Semi-luxury food processing (1)	21-23	91 **30**	0.2 **1.4**	2.2 **11**	23 **99**	-	2 **12**	4 **30**	50 **250**
Others (1)	7-10	88 **30**	0.25 **1.4**	1.8 **20**	10 **81**	-	1.2 **9**	7.5 **45**	50 **292**
Max. values of all categories			3.3-8	52-160	260-2,500		34-107	140-640	996-3,050
Averaged Limit-values for BWC in the EU			*1.4*	*93*	*143*	*1.0*	*47*	*121*	*416*
Limits - organic farming (EEC) No. 2092/91			*0.7*	*70*	*70*	*0.4*	*25*	*45*	*200*
BWC: AUT 75th %ile (2)	*582*		*0.51*	*31*	*61*	*0.29*	*26*	*49*	*211*
Soil* AUT 75th %ile (2)			*0.28*	*48*	*28*	*0.22*	*30*	*23*	*92*

(1) Sihler & Tabasaran (1996), bold figures standardised to 30 % organic matter
(2) 0-20 cm arable land in Austria; Amlinger & Peyr (2001)



(1) Sihler & Tabasaran (1996); (2) Median and 90th percentile PTE concentrations in Austrian BWC
All data relative to the averaged limit values of standards and regulations for BWC in European countries.
All material data are calculated (standardised) on the basis of 30 % OM in the dry matter

Fig. 4-2: Comparison of PTE concentrations of 4 groups of potential source materials for composting with BWC quality and European limit values for composts

When one compares these standardised values with the averages from the European limit values, it becomes clear that – particularly for the cadmium content in raw materials with exception of the agricultural residues – it is very difficult to comply with the limit values. In the case of composting of pure wastes from food production zinc may also be expected to exceed the limit value.

A comparison with the stricter threshold values of the EC Regulation (EEC) No. 2092/91 shows that only chrome (when related to 30 % organic dry matter) still would remain distinctly beyond the permitted concentrations in all material types shown in Table 4-5.

With the exception of Cd, kitchen waste usually shows higher levels of PTEs than the other organic waste types. Standardised on an organic matter content of 30 % d.m. the following feedstock materials frequently even exceed the averaged limit-values for BWC in the EU (see also Annex 4 Table A4-2):

- Paper (Cu, Zn)
- Potatoes (Cd, Cu, Zn)
- Tomatoes (Cd)
- Spinach (Cd)
- Funghi (Cd, Cu, Hg, Zn)
- Garden Waste (Cd)
- Kitchen Waste (Cd, Ni)
- Wood Choppings (Pb, Zn)

It is interesting that this particular outcome for specific source materials does not seem to be have an major effect on the overall compost quality which the medians achieve between 30 % and 55 % of averaged limit values in Europe.

There is much speculation as to why composts concentrate PTEs to an extent that would not be expected by rates of background concentrations in foodstuffs. With both vegetable and animal foodstuffs it can be shown that the contamination by residues from food trimmings may be a multiple of the PTE concentrations in food itself (Schiller-Bertz, 1994). This is mainly caused by adhering soil particles (e.g. for root crops) and atmospheric depositon respectively

Table 4-6: Ratio of the heavy metal concentration in food residues relative to the edible part of foodstuffs (= 1), from Schiller-Bertz (1994)

Foodstuff	Cd	Pb	Zn
Apples	2.5:1	6:1	2.7:1
Pears	2:1	8.6:1	2.5:1
Kale	-	0.5:1	-
Cucumbers	0.8:1	2.5:1	-
Carrots	3:1	9.6:1	-
Potatoes	-	7.7:1	-
Cabbage	-	1.9:1	1.6:1
Turnip	-	6.7:1	1.8:1
Lettuce	1.9:1	3.5:1	1.4:1
Leek	2:1	0.3:1	-
Celery	0.4:1	0.7:1	-

Another question of course is to what extent undesired extraneous components and impurities might have a systemic impact on the resulting compost quality. Extremely high levels of Cu, Pb and Zn can be detected in printed media (Bidlingmaier, 1987, cit. in Maier et al. 1993). The paper/cardboard fraction today plays an smaller role in the biowaste collection since waste paper is one of the most commonly recycled materials recovered by source segregation systems throughout Europe.

Table 4-7: Heavy metal content in mg kg^{-1} in different paper-fractions, analysed after separation from the domestic waste, from Kautz (1995)

Element	Paper (1)	Paper	Cardboard	Journals
Zn	102	58	225	107
Pb	63	31	144	34
Cu	37	47	86	39
Cr	10	21	26	33
Ni	7	11	13	23
Cd	0.4	0.4	0.8	0.1
Hg	-	<0.3	<0.3	-

(1) Bidlingmaier (1990)

During the pioneer stage of separate collection of biowaste in Vienna, randomised samples of 120 l bio-bins in four different districts resulted in 0.1 % (two garden district) to 1.7 % (m/m; two districts with dwellings without gardens) paper fraction (Amlinger et. al., 1993). Referring to the figures in Table 4-7 paper and cardboard tend to be somewhat more contaminated than the kitchen waste fraction. Nevertheless no significant quantities of heavy metals are contributed by paper due to the fact that paper is not systematically collected together with biowaste.

Plastics introduce heavy metals into the composting stream in the form of stabilisers and dyestuffs. According to Bidlingmaier (1990) lead, zinc and particularly cadmium are contributing thereto. With these elements the contamination is significantly higher than in kitchen waste.

On the other hand the cadmium content is decreasing decidedly due to bans in industrial production. Again the proportion of plastics in biowaste collected separately is low. The Vienna figures from 1991 in 4 different districts are 0.2 (garden district) to 0.5 % (m/m) (dwellings without garden) of plastics in the separately collected biowaste. (Amlinger et. al., 1993).

Table 4-8: Heavy metal content in mg kg^{-1} in the materials class *Synthetics*, analysed after separation from domestic waste, from Kautz (1995)

	According to (1)	According to Brahms-Eder	According to Brahms-Eder, 705 Polyethylene	DSD Synthetics
Zn	181	428	262	**678**
Pb	30	182	127	62
Cu	64	83	53	3
Cr	39	30	19	12
Ni	33	20	11	4
Cd	36	**46**	**14**	0.14
Hg	-	<0.3	<0.3	-

(1) Bidlingmaier (1990)

A comparison between normal bio-bins and two-chamber-bins shows that the collection scheme may play a significant role in the quality of the biowaste and consequently the final product: two-chamber-bins provoke a considerable increase in impurities resulting in significant higher standard deviation of heavy metal concentrations (Riess et al., 1993).

Another important pre-condition for the purity of the collected material is the general collection scheme. In Italy and Catalonia capture rates increased and impurities decreased when collection was at the doorstep rather than in road container systems (see Table 4-9).

Table 4-9: Specific capture and purity rates in schemes for source segregation of food waste in Catalonia. Doorstep schemes are highlighted

Municipalities / schemes	Performances of schemes for food waste	
	Quantity (g / inhabitant . day)	Quality (% impurities w/w)
Torrelles de Llobregat	139	1.8 %
Molins de Rei	116	2.1 %
Baix Camp	175	5.2 %
Igualada	125	3.8 %
Castelldefels	292	7.2 %
Castelldefels (March 2000)		4.5 %
Gavà	223	4.7 %
Viladecans	128	2.8 %
Viladecans		3.6 %
Castellbisbal	254	2.1 %
Vilanova i la Geltrú	239	---
Sant Cugat del Vallès (April 2000)	213	2.6 %
Barcelona (Major de Gràcia)	52	18.7 %
Barcelona (Gracia Comercial) (January 2000)		5.7 %
Barcelona (38 markets) (January 2000)		3.7 %
AVERAGE road container	***177***	***4.9 %***
Tona (October 2000)	**265**	**0.9 %**
Tiana (August 2000)	**285**	**4.0 %**
Riudecanyes (October 2000)	**298**	**1.9 %**
AVERAGE doorstep	***283***	***2.3 %***

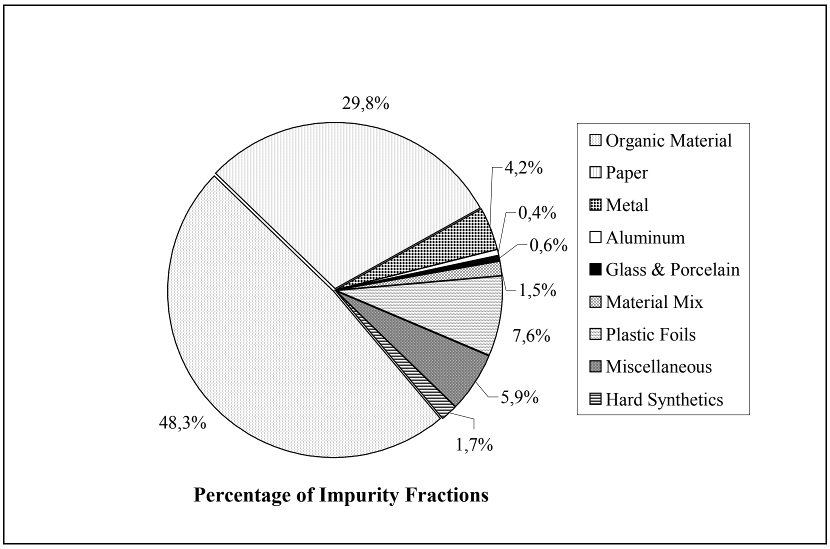


Fig. 4-3: Composition of impurities in biowaste collected with one-chamber bio-bins (Gronauer et al., 1997)

The examination of extraneous constituents in biowaste bins as well as the estimate of the PTE concentration in the single components that were selected may lead to an assessment of the impact of different proportions of impurities on the final compost quality (Gronauer et al., 1997).

The composition of the impurities is shown in Fig. 4-3. The main components is paper (30 %) followed by plastic foils, other materials and metals. 47 % consist of organic materials adhering to the single particles. When analysing the different fractions on their PTE content, the ferrous metal fraction contributes most to the introduction of heavy metals in to the biowaste. Predominantly Cr (max: 52,900 [!] mg kg^{-1}) and Ni (max: 1,100 [!] mg kg^{-1}) might occur in high concentrations in screw caps, spoons or cans. Hg and Zn showed elevated contents in wires and spoons. Cu was found in high concentrations in non-ferric metals. With the exception of Cr and Hg in rigid plastics, all metals were above limit values for compost in Germany and Cd (3.3 mg kg^{-1}) and Zn (1,730 mg kg^{-1}) showed the highest values.

In addition to the contamination of the single waste fractions by impurities, the amount of impurities is decisive for the factual effect on the final compost quality. Fig. 4-4 demonstrates the relative increase of the PTE concentration in compost as a result of 3 different levels of impurities (1 %, 5 % and 10 % m/m).

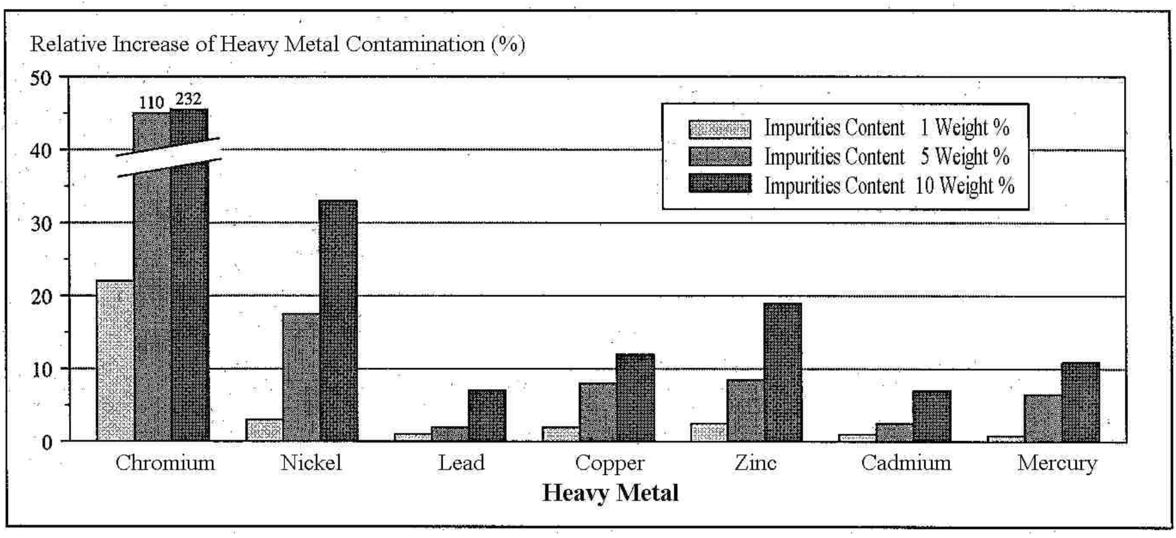


Fig. 4-4: Contribution of Various Impurity Contents to the Heavy Metal Contamination of Compost

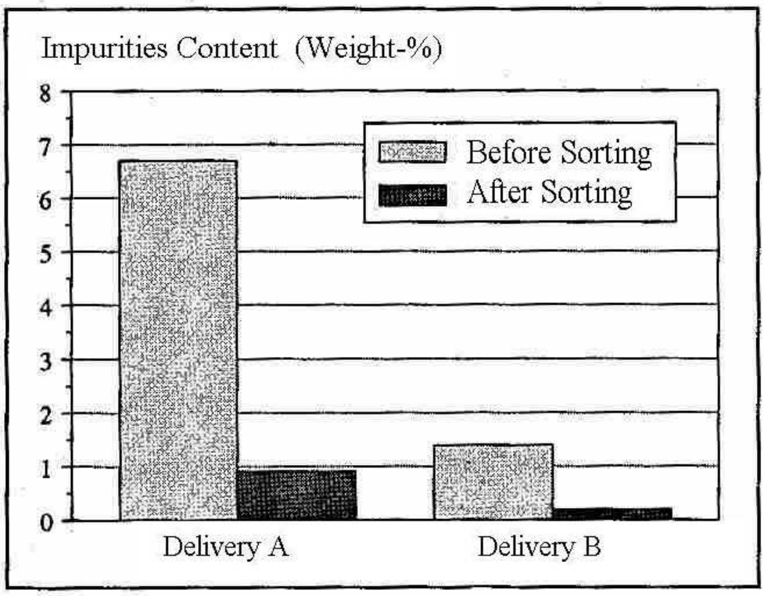


Fig. 4-5: Possibilities of Impurity Reduction by Sorting (Gronauer et al., 1997)

Due to the high concentration level for Cr in the metal fraction the Cr concentration in the final compost is raised by 20 % even if the overall amount of impurities is low (1 % m/m).

The importance of sorting (screening) the input material, which contains (visible) amounts of impurities, is presented in Fig. 4-5. It is clear that the degree of purity after a mechanical sorting step depends primarily on the initial proportion of undesired waste constituents. The reduction of impurities can 95 % when sorting is properly performed.

In a more recent investigation of composts in 376 composting plants Reinhold (2004) investigated the correlations between the heavy metal concentration on the and impurities, self heating test, organic matter and content of nutrients (N, P_2O_5, K_2O).

The occurrence of significant linear relations on facility level emerged in the following order:

content of nutrients > organic matter > impurities > self heating

Table 4-10: Proportion of 376 composting plants with significant linear correlations between the heavy metal concentration and selected compost parameters

Correlation between heavy metal concentrations and …	progressive	degressive
Content of nutrients	31,9 %	1,1 %
Organic matter	5,6 %	14,6 %
Impurities	9,6 %	3,2 %
Self heating	2,7 %	9,6 %

The content of nutrients normally featured a positive correlation to the heavy metal concentrations, whereas higher organic matter content predominantly leads to lower heavy metal concentrations.

As a progressive correlation between impurities and heavy metal concentrations was verified in only about 10 % of the investigated composting plants, no universally valid correlation between these to parameters can be assessed. A better input screening and the use of container detector systems could probably also eliminate these 10 %.

4.3 Biowaste compost vs. green compost

To what extent a systematic differentiation could be identified between Biowaste Compost and Green Compost has been widely discussed. In Table 4-11 national and regional comparisons of the heavy metal concentration of BWC and GC are summarised. Here Only those investigations were included which draw on a comparatively large data base. Where either original data or statistical evaluations were available, significant differences are indicated. Breuer et al. (1997) found for Cu, Pb and Zn and also Hg lower concentrations in GC than in BWC. For Cd, Cr and Ni no difference was identified. The authors concluded from the skewing of the Pb, Cu and Zn distribution it is clear that not only the general level of environmental pollution of green waste but also introduction of extraneous materials (impurities) are responsible for the contamination of BWC with heavy metals.

Table 4-11: Relation of heavy metal contents of BWC to GC composts [mg kg^{-1}] of several national investigations; different letters behind the numbers mean statistically significant differences

Austria (1)		***n***	**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
BWC	mean	*552-582*	0.42 a	26.7 a	50a	0.19 a	21 a	40 a	181 a
GC	mean	*64-65*	0.46 a	23.71 b	40 b	0.14 b	21 a	38 a	168 a
	BWC ›/‹ GC		*-8.5%*	***12.8%***	***25.2%***	***43.1%***	*2.3%*	*5.5%*	*7.5%*
Belgium (2)									
BWC	median	*195*	0.82	22	45	0.15	12	69	229
GC	median	*229*	0.70	17	32	0.12	9	44	169
	BWC ›/‹ GC		*17.1%*	*29.4%*	*40.6%*	*25.0%*	*33.3%*	*56.8%*	*35.5%*
France (3)									
BWC	mean	*12-27*	1.07 a	42.8 a	109.8 a	0.63 a	25.5 a	106.1 a	325.7 a
GC	mean	*22-123*	1.4 a	45.6 a	50.8 b	0.52 a	22.4 a	87.3 a	186.5 b
	BWC ›/‹ GC		*-21.9%*	*-6.1%*	***116%***	*21.2%*	*13.8%*	*21.4%*	***74.6%***
Italy (4)									
BWC	mean	*127*	1.38 a	33.1 a	89.1 a	-	26.3 a	84.4 a	219 a
GC	mean	*70*	0.95 b	33.4 a	62.7 b	-	23.1 a	71.7 a	165.8 b
	BWC ›/‹ GC		***45.2%***	---	***42.1%***		*13.9%*	*17.7%*	***32%***
Luxembourg (5)									
BWC	mean	*175*	0.41	32.0	38.6	0.12	15.8	48.7	218.6
GC	mean	*57*	0.34	23.7	32.4	0.13	12.8	44.5	164.1
	BWC ›/‹ GC		*20.6%*	*35.0%*	*19.1%*	*-7.7%*	*23.4%*	*9.4%*	*33.2%*
Baden-Württ./ DE (6) **									
BWC	median	*179-196*	0.32 a	27.0 a	52.2 a	0.18 a	12.9 a	47.0 a	181 a
GC	median	*28-86*	0.28 a	28.9 a	36.7 b	0.12 b	13.1 a	31.0 b	141 b
	BWC ›/‹ GC		*14.3 %*	*- 6.6 %*	***42.2 %***	***52.5 %***	*- 7.2 %*	***51.6 %***	***28.4 %***
Germany (7) **									
BWC	mean	*for both: 490*	0.78 a	33.73 a	43.24 b	0.33 a	19.13 a	77.64 b	232.8 b
GC	mean		0.70 a	27.04 a	32.67 a	0.27 a	17.53 a	60.8 a	167.8 a
	BWC ›/‹ GC		*11.1 %*	*24.7 %*	***32.4 %***	***22.2 %***	*9.1 %*	***27.7 %***	***38.7 %***
NRW / DE (8)									
BWC	median	*60*	0.46		42.5	0.13		42.5	180
GC	median	*12*	0.71		42.0	0.16		56.0	205
	BWC ›/‹ GC		***-35.2%***		*1.2%*	*-18.8%*		*-24.1%*	*-12.2%*

*data from 2 BWC and 1 GC plants respectively; ** data from 13 BWC and 12 GC plants respectively
(1) own calculation, databasis 1994 –2000; Devliegher (2002); (3) Charonnat et al. (2001) ; (4) Centemero (2002); (5) Mathieu (2002); (6) Breuer et al. (1997); (7) Fricke & Vogtmann (1994); (8) Stock et al. (2002)

In Denmark in two composting plants the Cd concentration was evaluated systematically with increasing proportions of organic household waste (BW) in the total composting blend. No systematic differentiation could be identified (Table 4-12)

Table 4-12: Cadmium in compost with different parts of organic household waste in Denmark (Hogg et al. (2002)

% of household waste in relation to garden waste in raw material	Number of plants	Cadmium in compost (mg kg^{-1} d.m.) (Limit value = 0.4)
85-100	2	0.27
70-80	2	0.40
40-60	2	0.50
10-30	2	0.40
0	2	0.47

National investigations in Table 4-11 indicate the following ranking of significant or trending differences between GC and BWC:

Cu > Zn >> Hg > Pb

with differences between 20 and 50 % under well defined standard collection conditions. Cr, Cd, Ni give a indifferent or heterogenous picture with no clear trend. The Cu, Pb and Zn figures for biowaste compost in France are cinsiderable higher than in other countries and investigations. These numbers can therefore not be considered to representing a standard quality of biowaste compost from source seperated organic household waste.

4.3.1 Further comparison of biowaste and green waste compost on country level

Fig. 4-6 shows the distribution of PTE concentrations in Biowaste Compost and Green Compost in several Member States. The data are compared to two levels of limit values:

1.) the threshold for heavy metals for composted household waste of Annex IIA of the EC Regulation 2092/91 (EEC[FA221]) (Regulation No. 2092/91/EEC, 1991) on Organic Farming (amendment from 29 July 1997, EC regulation 1488/97/EEC) and

2.) the averaged limit values for BWC of European countries.

Table 4-13: Limit Values for BWC as compared to PTE concentration distribution in BWC used in Fig. 4-6

	Cd	Cr	Cu	Hg	Ni	Pb	Zn	As
				mg kg^{-1} d.m.				
Averaged limit values of EU countries	1.4	93	143	1.0	47	121	416	23
Limit values Annex IIA Reg. (EEC) No.9092/91	0.7	70	70	0.4	25	45	200	--

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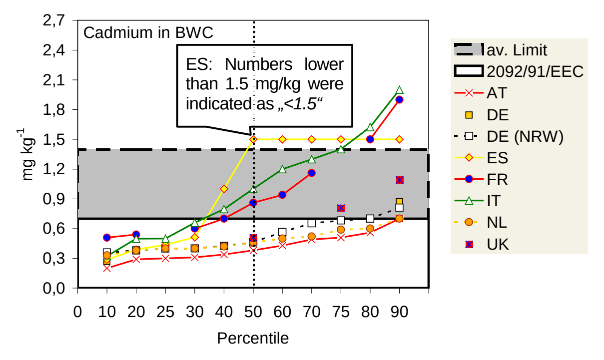
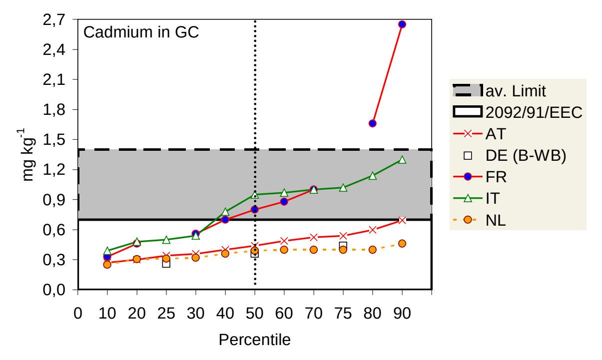
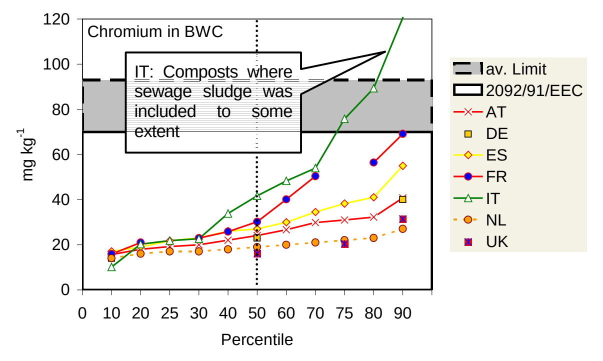
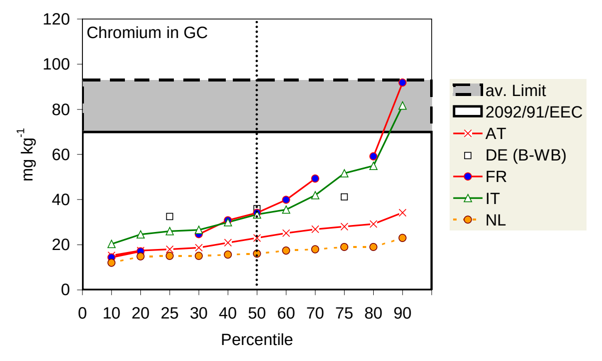


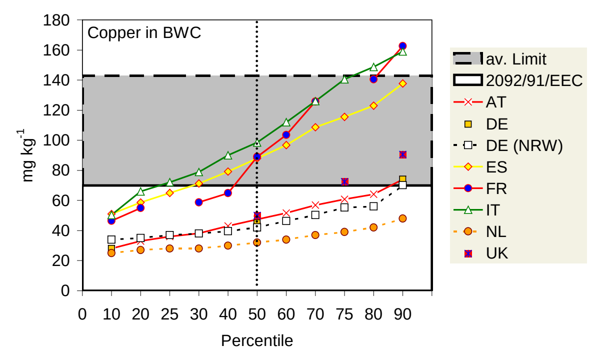
ES: Numbers lower than 1.5 mg/kg were indicated as „<1.5“

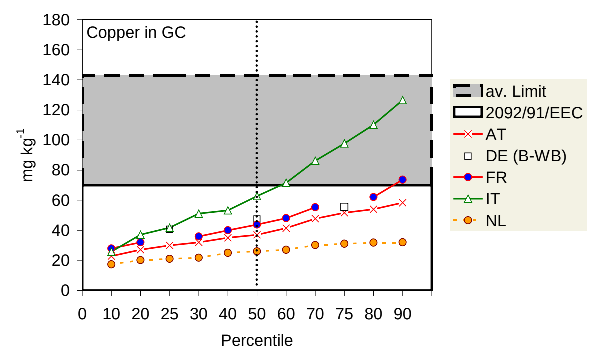


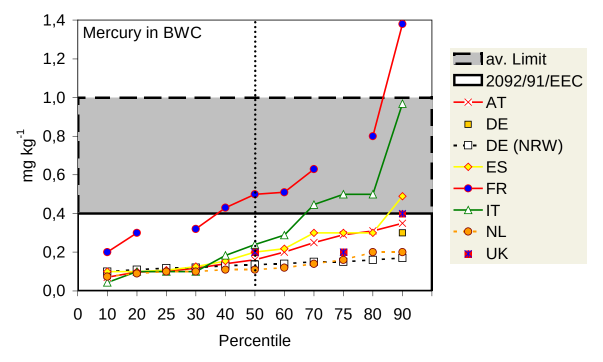


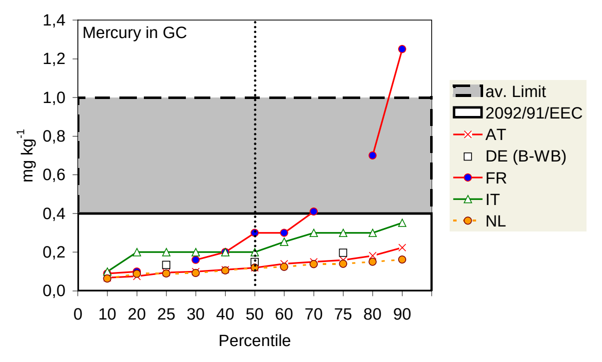
IT: Composts where sewage sludge was included to some extent











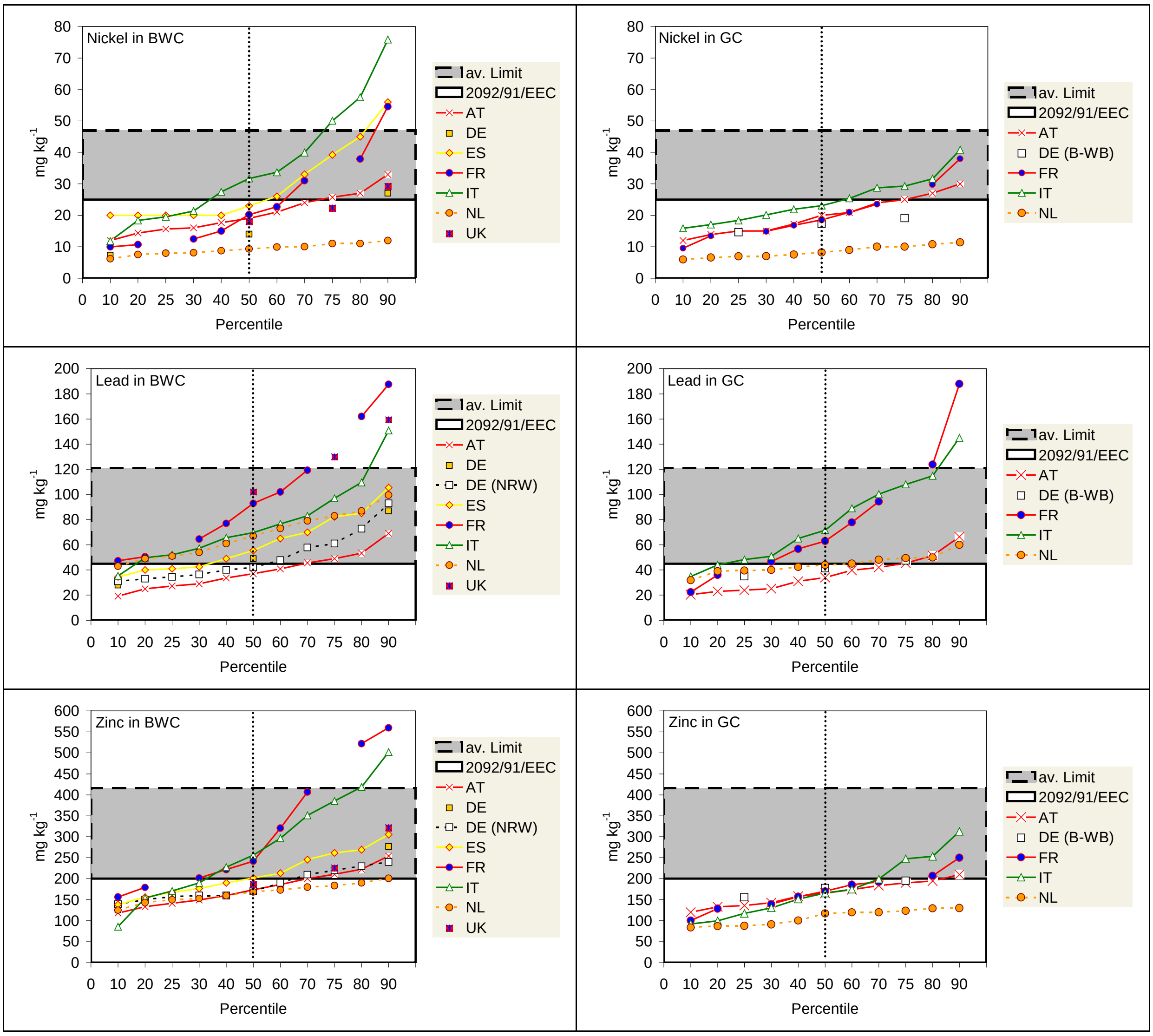


Fig. 4-6: Distribution of PTE concentrations in of BWC and GC in several MSt

From the country comparison of Biowaste Compost and Green Compost it may be concluded:

(1) *Differentiation between* Biowaste Compost and Green Compost

- Concentration levels for BWC tend to increase more significantly above the median than levels for GC. This might be partly due to less effective source separation schemes in some situations. It might be noteworthy here that performances of schemes seems to depend much more on the type of scheme than on the size of the town covered and on the type of dwellings.
- The difference is more significant in countries which are in the starting phase of separate collection systems. Sometimes this might be the effect of a confusion – both from the regulatory and operational standpoint – that it is not always clear if compost that also includes a certain percentage of sludge.
- The comparison of biowaste compost and green compost on country level gives a slightly modified picture as that one gained from median or mean value comparison (Table 4-11), the differentiation follows the row: Pb, Zn > Ni, Hg > Cu > Cd, Cr.

(2) *Differentiation between countries*

- Countries, which are in the starting phase in introducing separate collection systems for green and biowaste (UK, FR, ES, IT), generally show higher PTE concentrations in composts than countries with a fully established source separation in place (10 – 15 years; AT, DE, NL).
- In BWC the difference decreases in the sequence: Cu > Cd, Ni, Pb, Zn > Cr, Hg.

  - In GC the difference decreases in the sequence: Pb, Cd, Cr > Cu > Hg, Ni, Zn. The effect for Pb might be due to a higher level of attention paid to green waste coming from roadsides and high traffic areas in most countries with "mature" schemes.
  - *NL* composts show significantly lower metal concentrations; above all for Ni, Cu, Cr (BWC and GC), Zn (GC) as compared to other high performance countries (AT, DE); this phenomenon is less evident for the metals Cd, Hg and Pb.

    We exclude a significant influence of diverting analytical methods for the figures used in this graph. The information provided (see chapter 7.5) indicates that at least *aqua regia* and similar material:acid ratios are used.
- It has to be noted that the detailed evaluation of the *Italian data* revealed the frequent use of municipal sewage sludge as a constituent of raw materials even in composts indicated as BWC and GC. This was identified as a main reason for the relatively high figures in Italian composts.
- In contrast, the relatively high figures of *France* are taken from the ADEME report (Charonnat et al., 2001), which refers specifically to pure source separated biowaste and green waste. No indications were found that sewage sludge would have been a possible constituent in these composts.

4.3.2 Urban (industrialised) and rural origin of source materials

Only a few specialised investigations were carried out, focussing on the question of regional specification of biowaste or green composts. Nevertheless within some general surveys on compost quality information on the origin from rural, urban or industrialised areas are given.

For example, the Austrian survey on composts from *Vienna (Wien)*, *other urban origins (Stadt)*, cities with more than 100,000 inhabitants) and *rural areas (Land)* is shown in Table 4-14.

Table 4-14: Differentiation of heavy metal concentration in biowaste compost from urban and rural areas

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
		------------------------------------			*mg kg^{-1} d.m.*			---------------------------------
Austria (1)	*Vienna [n=112]*	0.37a*	23.9a	55.9a	0.32a	19.4a	50.1a	200a
	Other cities [n=35]	0.34a	31.9c	52.3ab	0.20b	25.8b	44.1ab	178ab
	rural [n=426]	0.44b	27.0b	48.4b	0.16b	21.3a	37.6b	176b
Germany (2)	*urban*	0.80a	30.64a	46.71a	0.39a	16.76a	88.58a	245.70a
	rural	0.77a	36.00a	42.74a	0.28a	28.51a	71.33a	229.71a

(1) Amlinger & Peyr (2001); (2) Fricke & Vogtmann (1994); *mean values with different letters are significantly different (p=0.05).

Though differences with the exception of Ni may be identified, if similar good collection and quality management strategies are used differentiation between urban and rural biowaste composts is not as large as had been expected. Typically *pedogenic* metals like Cr and Ni might also be higher in composts from rural areas. Typically *anthropogenic* metals such as Cu, Hg and Pb show higher levels in Vienna but still at moderate concentrations. Zethner et al. (2000) only found a differentiation (trend; p=0,1) of urban and rural composts for Mn and Ni with higher levels in rural origins.

A regional distribution of contamination with heavy metals was identified in Baden-Württemberg (Germany) in an investigation on potential source materials of biowaste and green composts (Krauß et al., 1995a). With the exception of Zn, separately sampled leaves showed higher metal levels in urban and industrialised areas than in rural ones. For the city of Stuttgart Cr, Ni and Hg had a uniform distribution over all settlement types whereas Zn and Cd increased with the traffic intensity. Pb as the typical road–side metal shows a clear downward tendency.

It could be argued, that in general (due to increased anonymity) purity of sorted food waste tends inevitably to get much lower in highly populated areas with mainly high rise building dwellings.

A comprehensive evaluation of spatial and seasonal variation of heavy metal concentrations in separately collected biowaste in the city of Vienna demonstrated that, after elimination of extreme values, no differentiation between the different settlement patterns could be observed (Table 4-15; Plahl et al., 2002). Impurities such as plastics and metals were removed before analysis. In this way the basic quality of the decomposable fraction of biowaste is demonstrated.

Table 4-15: Heavy metal contents of biowaste (average of all seasons) in different city areas (data standardized at 30 % organic matter; outliers eliminated); Plahl et al. (2002) and Hauer (1997)

BIOWASTE raw materials		**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
		------------	------------	------------ *mg kg^{-1} d.m.*	------------	------------	------------	------------
Samples after removal of impurities	*High building density [n=410]*	0.02		28.76		10.56	29.96	138.62
	Low building density [n=90]	0.01		25.40		17.12	32.91	204.45
	City outskirts [n=244]	0.05		33.12		14.43	32.28	171.45
	Vienna total	**0.03**		**28.97**		**12.79**	**31.09**	**159.76**
BIOWASTE – compost								
No removal of purities*	*High building density [n=6]*	0.60		143.83	0.43	26.05	171.25	435.17
	Vienna [n=112]	0.37	23.9	55.9	0.32	19.4	50.1	200

* average of 6 compost lots set up every 2 month over a period of 1 year; impurities were not removed before composting.

Compared to the average heavy metal concentration of Vienna composts all *inner city* (high building density) composts had elevated levels, particularly for the elements Pb, Cu and Zn. This was not fully consistent with the fact that partly raw materials showed higher concentrations of heavy metals in the low-density areas than in the densely populated districts. However the number of investigated samples of the densely populated districts was too low to draw a conclusive picture.

In contrast, the Italian and Catalonian experience shows that *the quality of collected biowaste seems to be much more dependant on the system adopted for collection than on the size of towns* and many situations are reported where schemes are also successful in large towns and inner cities. For example, the figures for the purity of separated biowaste (percentage of compostable materials) reported in various sorting analyses performed across Italy are given in Fig. 4-7.

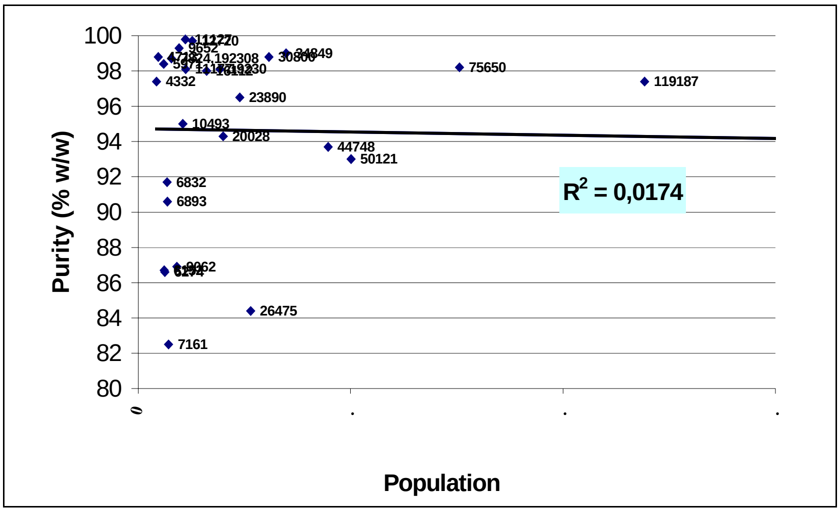


Fig. 4-7: Purity of collected food waste vs. population in the catchment area (Favoino, 2001)

As a result no relationship can actually be detected between the size of the population in the catchment area and purity of compost. This means that other factors are affecting the purity more than the population covered by the scheme, specifically the *type* of scheme adopted. Doorstep schemes generally perform much better than road container schemes. Similar results are also reported from Catalonia, where both types of schemes are currently run.

A spatial evaluation of > 6,400 German biowaste and green waste composts from 1991 – 1997 (Reinhold, 1999) indicated a systematic regional distinction of heavy metal levels following the statistical calculation of the expected range of mean values. Three categories (groups) were distinguished for the expected upper mean values (Table 4-16).

Table 4-16: Ranking of the upper expected mean values of heavy metals in BWC and GC in the federal provinces of Germany (Reinhold, 1998)

Ranking of the upper expected mean values [mg kg^{-1} d.m.]													
Cd		**Cr**		**Cu**		**Hg**		**Ni**		**Pb**		**Zn**	
State	**value**	**State**	**value**	**State**	**value**	**State**	**value**	**State**	**value**	**State**	**value**	**State**	**value**
MV	0.369	B	19.2	MV	36.1	NS	0.129	NS	9.1	BA	47.2	BW	171
NS	0.432	BA	19.9	SL	37.4	NW	0.149	BR	9.1	BW	48.0	NS	172
BW	0.432	SH	20.0	NS	38.5	HE	0.152	SH	9.3	NS	50.1	SH	183
TH	0.452	HH	20.7	HH	46.4	SH	0.160	MV	10.0	SH	52.8	MV	189
RP	0.459	NS	21.4	SH	46.5	BW	0.181	HH	10.4	HE	53.5	B	190
HE	0.475	NW	23.3	NW	47.1	BR	0.205	B	10.6	MV	53.7	BA	194
BA	0.505	RP	23.6	BR	48.8	HH	0.210	BB	13.6	NW	62.5	HE	204
SH	0.513	BR	23.7	BW	48.9	BA	0.225	NW	15.0	RP	66.4	SL	206
BR	0.530	BB	24.5	HE	49.2	B	0.229	BA	15.1	SL	66.6	RP	207
BB	0.665	SX	25.9	RP	51.1	RP	0.237	SX	15.8	TH	70. 4	BR	212
B	0.685	BW	30.1	BB	53.2	MV	0.248	BW	16.2	BR	72.6	NW	213
SX	0.696	SL	32.1	B	53.6	BB	0.251	RP	19.5	HH	77.8	HH	221
SA	0.702	HE	32.5	BA	59.7	SX	0.254	SA	19.5	BB	78.2	BB	234
HH	0.748	TH	40.1	TH	59.2	TH	0.291	SL	20.8	B	86.5	TH	254
NW	0.751	MV	45.8	SX	62.1	SA	0.311	TH	22.4	SX	91.3	SX	271
SL	0.768	SA	46.3	SA	72.9	SL	0.428	HE	29.8	SA	95.2	SA	332

Table 4-17: Considerations on the regional heavy metal distribution in BWC in Germany (Reinhold, 1999)

Element	**Description**
Cd	Indicator of former and current heavily industrialised areas. Lowest values occur in areas without traditional industries. Potentials for the reduction of the Cd contamination of composts in non industrialised areas are not to be transferred to industrialised zones.
Cr	Generally low concentrations (basalt; TH, HE, BW). Agents used for impregnation of construction wood (SA, MV)
Cu	Increased 1991-1997; elevated concentration along with increasing OM content; specific import from other sources (construction wood; SX, SA)
Hg	Mining areas (cinnabar; SL). Elevated concentrations in former GDR due to pesticides and chlorine-potash industries

Ni	Metallurgic industries and batteries play a minor role (SA); predominant geogenic and pedogenic sources (basalt and ultra basic rocks) (HE, RP, BW)
Pb	Time span when lead was removed as additive from petrol; density of traffic; accidental contamination by batteries; highest concentration in former GDR heavily industrialised zones (Sachsen and Sachsen-Anhalt)
Zn	Indicator for general background pollution as antirust agent and galvanising industries. Additive in pig fattening (Zinc-Bacitracine). Relatively uniform distribution.

A similar investigation was made of biowaste compost quality from the nine Provinces of Austria (Amlinger & Peyr, 2001). The provincial capitals were included. The differentiation varies with the individual metals. Cd, Cu and Pb are on a higher level in Salzburg whereas in Vienna the concentration of Hg, Pb and Zn is increased relative to the other provinces. Elevated Cr and Ni contaminations are found in the BWC samples due to geogenic background concentrations, which are of regional importance, especially in Styria (Steiermark).

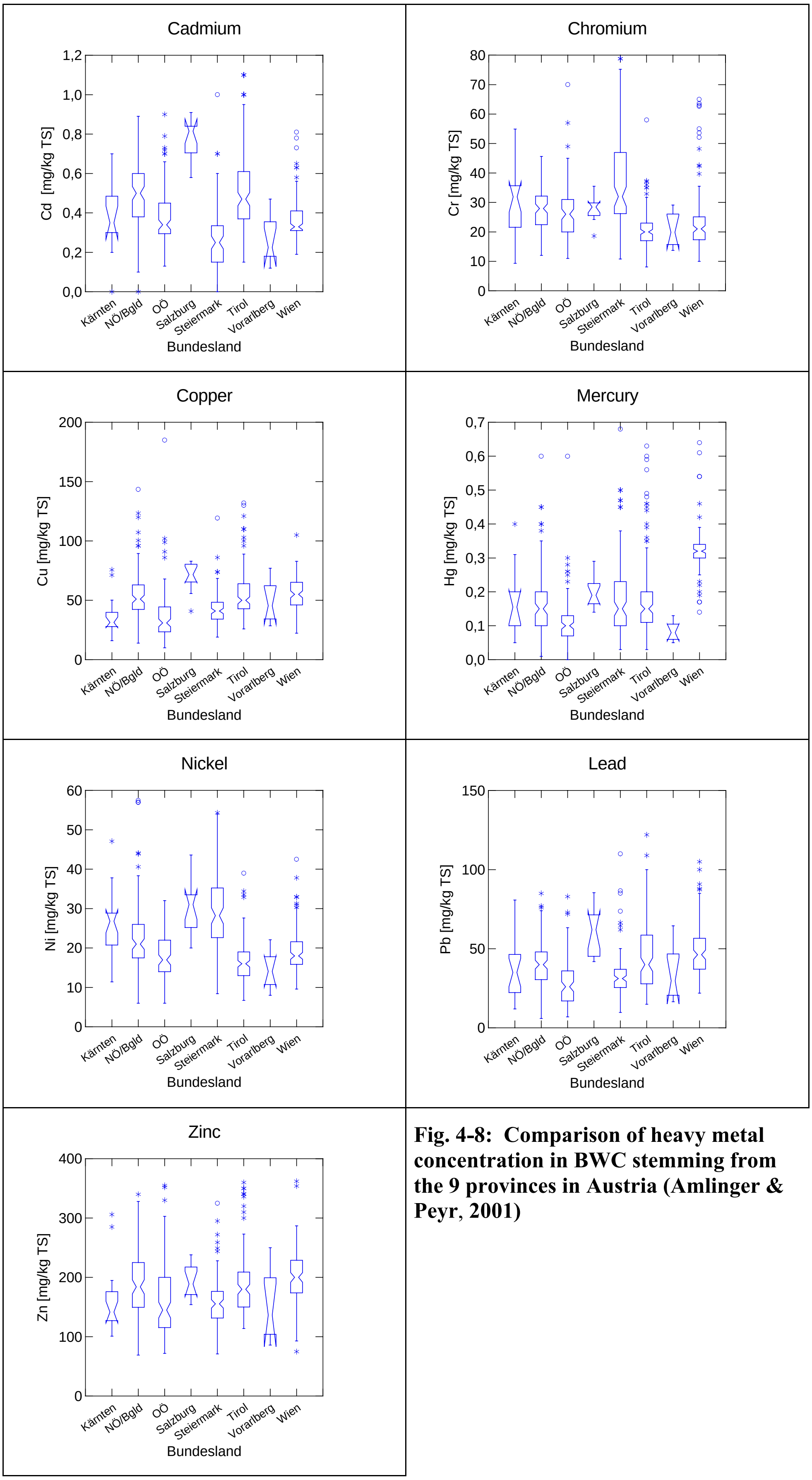


Fig. 4-8: Comparison of heavy metal concentration in BWC stemming from the 9 provinces in Austria (Amlinger & Peyr, 2001)

Rieß et al., 1993 report a distinct increase of metals in compost when soil concentrations in the area concerned are raised (due to industrial activity):

Table 4-18: Heavy Metal Content in BWC from 2 different catchment areas of one district. Catchment area 1 features elevated heavy metal concentrations in the soil [mg kg^{-1} d.m.] (Rieß et al., 1993)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Catchment area 1	0.9	18	41	0.17	17	694	278
Catchment area 2	0.9	22	33	0.11	14	83	172

Shortly biowaste composts from 13 “urban” and 29 “rural” composting plants in Hessen, Germany, produced between 1998 and 2002 were examined for defferences depending on the settlement density. The result is shown in Table 4-19.

Table 4-19: Means of Composts from urban and rural regions in Hessen and the lower and upper confidence limits (Reinhold, 2003a).

Parameter		**statist. difference 1)**	**Compost urban regions**			**Compost rural regions**		
			lower confidence limit	**mean**	**upper confidence limit**	**lower confidence limit**	**mean**	**upper confidence limit**
impurities	% d.m.	0	0.04	**0.14**	0.24	0.08	**0.13**	0.19
Cd	mg kg^{-1} d.m.	0	0.32	**0.42**	0.53	0.44	**0.47**	0.50
Cr		(–)	24.6	**29.3**	34.1	28.5	**36.5**	46.4
Cu		(+)	38.1	**49.6**	61.1	41.8	**47.7**	53.6
Hg		0	0.10	**0.14**	0.18	0.10	**0.13**	0.16
Ni		–	14.0	**15.1**	20.3	17.8	**30.1**	42.5
Pb		(+)	36.9	**48.3**	59.7	38.7	**44.1**	49.5
Zn		(+)	160	**187**	214	163	**178**	193

1) statistical differences: compost of urban areas as compared to compost from rural areas: (+) … trend of increased values ($p<0.1$); 0 … no difference; (–) … trend of reduced values ($p<0.1$); – … sigificantly reduced values ($p<0.05$)

The key results from this evaluation were:

- The means of composting plants in urban regions show a tendency to slightly increased organic matter contents and concentrations of N, Pb, Cu and Zn in comparison to rural regions and a tendency to a higher variation between the plants for impurities, Pb, Cd, Cu and Zn. **(→ anthropogenic influences)**
- The means of composting plants in rural regions show significantly higher concentrations of Ni and a tendency to increased Cr values in comparison to urban regions. In addition the variation between the plants are significantly higher. **(→ geogenic influences)**

In order to classify the heavy metal contents and their distribution in composts with respect to relevant environmental data, a comparison with soil data is shown in Table 4-20.

Table 4-20: Distribution if heavy metal concentrations in Austrian Biowaste composts

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
n	*552*	*582*	*582*	*564*	*577*	*570*	*579*
Percentile	------------------------------------ *mg kg^{-1} d.m.* ------------------------------------						
10%	0.20	15.7	28.0	0.07	12.0	19.2	117.9
25%il	0.30	19.2	36.0	0.10	15.6	27.4	141.1
Median	**0.38**	**24.1**	**47.4**	**0.16**	**19.0**	**37.0**	**173.6**
Mean	***0.42***	***26.7***	***50.1***	***0.20***	***21.2***	***40.4***	***180.7***
75%i	0.51	31.0	60.8	0.29	25.7	49.0	210.5
90%	0.70	40.8	74.0	0.35	33.0	69.1	253.9
Arable land AUT 75th %ile *	**0.28**	**48**	**28**	**0.22**	**30**	**23**	**92**
Pasture AUT 75th %ile **	**0.43**	**47**	**32**	**0.20**	**34**	**43**	**119**
Soil Vienna 75th %ile***	**0.75**	**31**	**53**	**0.67**	**32**	**106**	**188**

* 0-20 cm arable land; ** 0-5 cm pasture land; *** 0-10 cm urban parks and greens

Based on the comparison of soil data of arable soils, pasture land and Vienna park soils, as well as the quoted investigations on rural and urban biowaste compost two groups of PTEs can be distinguished:

(1) predominantly geogenic distribution ➔ Cr, Ni

(2) predominantly anthropogenic distribution ➔ Cd, Cu, Hg, Pb, Zn

This definitely endorses the results gained in Germany and leads to the conclusion that Cr and Ni concentrations in compost are mainly driven by pedigenic and geogenic background situations.

4.3.3 Temporary and seasonal variations

Two approaches may be adopted if temporary variations in compost qualities are to be investigated.

1.) Analysis of seasonal and long term variations on the basis of individual composting facilities in a single continuously monitored catchment area.

2.) Analysing the long term development of quality levels from the beginning of source separation and composting of organic household and garden and park waste.

Existing data and literature were examined in order to search for systematic time-effects on compost quality.

Breuer et al. (1997) found low seasonal variation for Hg, Ni and Zn in Baden-Württemberg. The difference between the maximum and minimum values was in most cases below a factor of two. More significant was the variability of Pb, Cd, and Cu. Extreme values were interpreted as *accidents* caused by metallic particles which found their way from the sampled lot into the test sample. For these 25 composting plants there was no evidence for a systematic differentiation between *winter* and *summer* compost.

Aldag et al. (1995) report low seasonal variation. In winter compost higher levels for Pb and lower levels for Cu and Hg could be detected (see Table 4-21).

Table 4-21: Seasonal variability of heavy metal contents of bio- and greenwaste composts (BW/G-C) and relation to sewage sludge composts (SSW) [mg kg^{-1} d.m.] (Aldag et al., 1995)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				[mg kg^{-1} d.m.]			
Winter mean	0.7	32.9	63.4	0.27	19.94	**152.1**	293
Summer mean	0.7	31.6	**69.5**	**0.38**	22.29	105.4	288

Kehres (2000) calculated the mean and the upper expected coefficient of variation (CV) for 262 composting plants for heavy metals. The mean CV for all metals and composing plants accounts ±28 % relative to the mean value of all analyses of one year.

Table 4-22: Mean and the upper expected coefficient of variation (CV) of heavy metals (262 composting plants associated to the FQAC, Germany)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				[± deviation in % of mean value (per year)]			
Mean CV on plant scale	±27 %	±30 %	±22 %	±46 %	±26 %	±25 %	±15 %
Upper expected CV on plant scale	±61 %	±65 %	±40 %	±63 %	±57 %	±58 %	±64 %

The changes of mean (resp. median) heavy metal values in BWC and GC in Germany from 1991 to 1997 were evaluated on the basis of a trend analyses at a probability limit of 95 % (Reinhold, 1998). A significant increase was shown only for Cu. With the exception of Zn this did not change over the years, all other metals were reduced. The increase of Cu was reported to be caused by direct transfer by copper containing metals such as copper water pipes, as well as by treated wood and pig slurry.

Table 4-23: Heavy metal concentration in German biowaste and green waste composts 1991, 1997 and 2002 (Reinhold, 1998; Reinhold, 2003b)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				[mg kg^{-1} d.m.]			
1991 *[median]*	0.64	34.3	39.9	0.22	20.4	68.2	182.9
1997 *[median]*	0.53	24.7	48.7	0.18	16.1	56.6	196.2
2002 *[mean]**	0,47	25,3	57,7	0,16	16,3	46,4	203,7
1997 rel. to 1991	*83%*	*72%*	***122%***	*82%*	*78%*	*83%*	***107%***
2002 relative to 1991	*73%*	*74%*	***145%***	*73%*	*80%*	*68%*	***111%***
Absolute yearly change 1997 to 2002	*-0,015*	*-0,82*	*+1,62*	*-0,005*	*-0,37*	*-1,98*	*+1,89*

* taken from 346 composting plants; calculated as estimated value for 2002 after statistical trend calculation on facility level based on the data between 1998 and 2002

The comparison of Biowaste Compost and Green Compost mean PTE concentrations investigated in 1988 and 1998 in Switzerland shows an outcome nearly parallel to the results in Germany. Again the Cu content increased whereas all the other metals decreased, indicate a clear improvement of the compost quality.

Table 4-24: Heavy metal concentration in Swiss biowaste and green waste composts 1988 and 1998 (Golder, 1998)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
			mean values [mg kg^{-1} d.m.]				
1988 (n=30)		36.5	36		19.5	69.5	179
1998 (n=135)		22.8	47		15.1	44.5	162
'98 relative to '88		*62 %*	***130 %***		*77 %*	*64 %*	*90 %*

Fig. 4-9 shows the development of PTE concentrations in BWC and GC on a facility level in Luxembourg. Again copper is the only element which shows a distinct and continuous increase over the period 1997 – 2001 for both BWC and GC. A decline can be discerned for Pb > Cr, Zn > Hg, Ni. An irregular trend is shown for Cd.

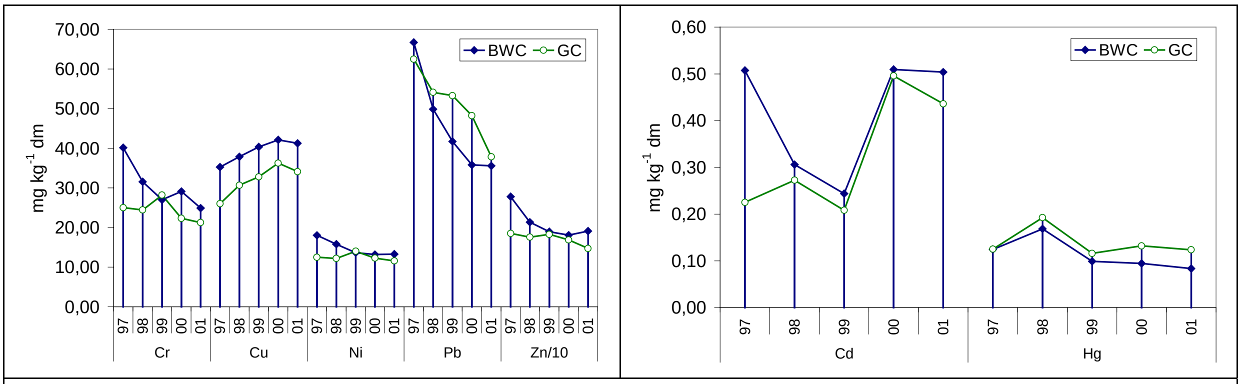


Fig. 4-9: Development of heavy metal concentrations in biowaste (BWC) and green waste (GC) composts in Luxembourg 1997 – 2001 (weighted mean of mean values on facility basis; 2 BWC and 1 GC plants resp.) - Data provided by Mathieu (2002)

Conclusions from long term investigations on compost heavy metal concentrations

The evaluation of the data gained over a period of four to eleven years show a partly significant trend of quality increase. This is the case for Cd, Cr, Hg, Ni and Pb. Zn staid on the same level or does not show a clear trend. Only the Cu concentration is still increasing constantly at rates of 30 and 45 % in Switzerland and Germany respectively over a period of one decade. There is still no clear justification based on scientific investigations available though the following assumption can be made:

- Increased use of copper containing fungicides also in professional and private horticulture
- Increased proportion of copper pipes for water supply. This may increase the copper content in the irrigation water in vegetable production
- Increase of copper as food additive
- Increase of the proportiion of source seperated kitschen waste in the overal biowaste arrisings.

4.4 Compost from sewage sludge

Poor response was given to the questionnaire which was sent out to experts and representatives of the Member States (see Annex 6) regarding composts prepared from municipal or industrial sewage sludge (SSC).

Partly dewatered sludges as feedstock material for composting show dry matter concentrations of 15 to 30 % d.m. The proportion of sewage sludge as constituent in the initial mixture of compost heaps ranges between 20 and max. 50 % by volume. Higher ratios would result in critical pore volume distribution and mechanical stability of the rotting pile inducing insufficient gas exchange.

Table 4-25 summarises the range of PTE concentrations in sewage sludge.

Table 4-25: Ranges of mean concentrations of heavy metals in sewage sludge in different EU countries

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
		Ranges and mean values [mg kg^{-1} d.m.]						
EU (1)	range of means	0.4 – 3.8	16 - 275	39 - 641	0.3 - 3	9 - 80	13 - 221	142 – 2,000
Germany (2)		1.4	46	274	1	23	63	809
Switzerland (3)	mean	1.7	74.1	341.1	1.7	31.9	94.5	929.3
Spain (4)		4	162	258	-	43	212	955
France (5)		2.9	58.8	309	3	31.9	106.7	754.2
Austria (6)	median	1.3	40	200	0.8	25	55	900
	90th percentile	2.2	80	320	2.9	45	110	1,400

(1) Arthur Andersen on behalf of DG Environment (2001); (2) Bannick et al. (2001); (3) Herter et al. (2001); (4) Soliva et al. (1982); (5) Arthur Andersen on behalf of DG Environment (2001); (6) Aichberger at al. (2002)

Fig. 4-10 gives an example of the development of PTE concentrations in sewage sludge from Upper Austria. It demonstrates – with the exception of Cu – a constant decrease of contamination within the last 20 years. Even though in average composts from sewage sludge still show distinctly higher values for Cd, Cu, Hg and Zn than BWC and GC. Due to the differing extraction method, the significant lower figures from the Nordic countries must not be included in this comparison (see Table 4-26).

As a consequence sewage sludge compost would not comply with most of the European threshold values, which have been developed for BWC originating from source separated kitchen and garden waste.

A possible procedure for the use of SSC in agriculture is considered in chapter 9 on the basis of a *Good Agricultural Practice* model related to a reasonable organic matter and plant nutrients supply.

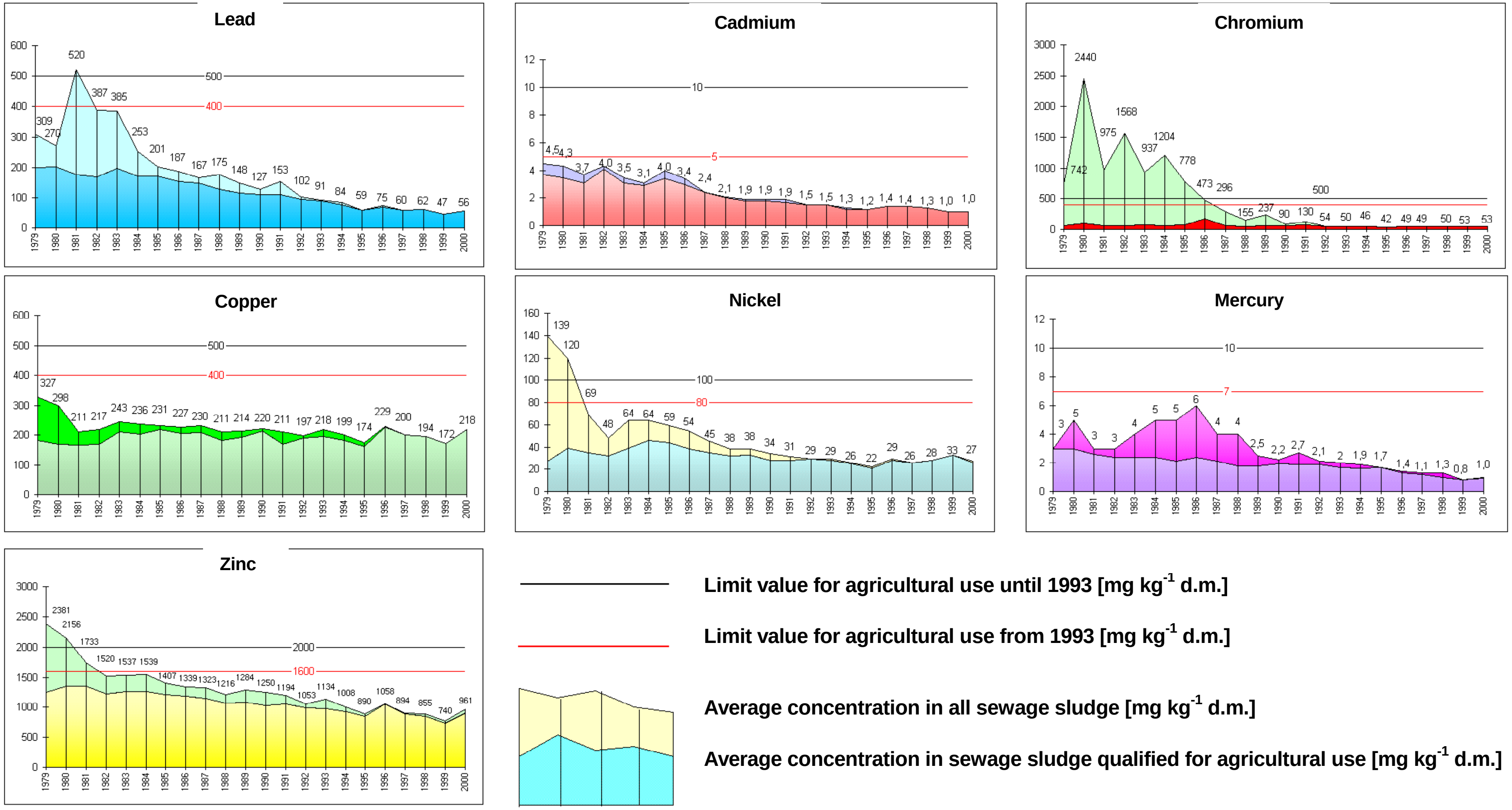


Fig. 4-10: Heavy metal content in sewage sludge (province of Upper Austria) 1979 – 2000; reduction by purification measures at the source (Aichberger at al., 2002)

Table 4-26: Examples for heavy metal concentration in compost made from sewage sludge

Compost with sludge		**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**	**As**
		------------------------------------ *mg kg^{-1} d.m.* ------------------------------------							
Austria	Amlinger (2000) median; n = 19-21	1.18	50	112	0.86	26	59	538	-
Denmark	Petersen (2001b) [mean]; n =?	[0.34]	[8.8]	[28]	[0.07]	[5.7]	[23]	[140]	[2.8]
Finland	Vuorinnen (2002) [mean]; n = 19-33	[0.56]	-	-	[0.31]	[21.11]	[15.81]	-	[2.37]
France	Charonnat et al. (2001) median [mean]; n = 12-99	1.08 [1.62]	38.87 [50.59]	119.03 [152.14]	1.75 [2.2]	18.06 [25.81]	39.00 [61.81]	293.78 [404.92]	3.75 [3.30]
Germany	(Tabasaran & Sihler, 1993	1.46	129.38	185.25	-	40.63	96.00	965.00	-
	Aldag et al. (1995) [mean]; n = 4 (1 plant)	[1.4]	[28.1]	[301]	[0.51]	[19.96]	[55.9]	[700]	-
Italy	Centemero (2002) median [mean]; n = 68	1.4 [1.83]	34.02 [44.68]	175.00 [177.68]	-	23.7 [32.25]	80.00 [93.75]	525.00 [637.26]	[4.8] (n=37)
Norway	Amundsen, Eggen, et al. (2001) [mean]: n= 3 (1 plant)	[0.55]	[8]	[56]	[0.18]	[5.5]	[8.9]	[163]	-
Heavy metal concentration in European compost (statistically weighed figures)									
Weighed median BWC		**0.50**	**23.0**	**45.1**	**0.14**	**14.1**	**49.6**	**183**	-
Weighed 90%ile BWC		**0.87**	**39.9**	**73.9**	**0.30**	**27.0**	**87.6**	**276**	-
Averaged limit values for BWC and GC in European countries									
		1.4	**93**	**143**	**1.0**	**47**	**121**	**416**	**23**

Comparative view on composts of different feedstock materials

The comparison of composts originating from different source materials is shown in Fig. 4-11 and in in Fig. 4-12. The graphs speak a clear language. Fig. 4-12 originates from an Italian evaluation of composts, manure and commercial soil amendments.

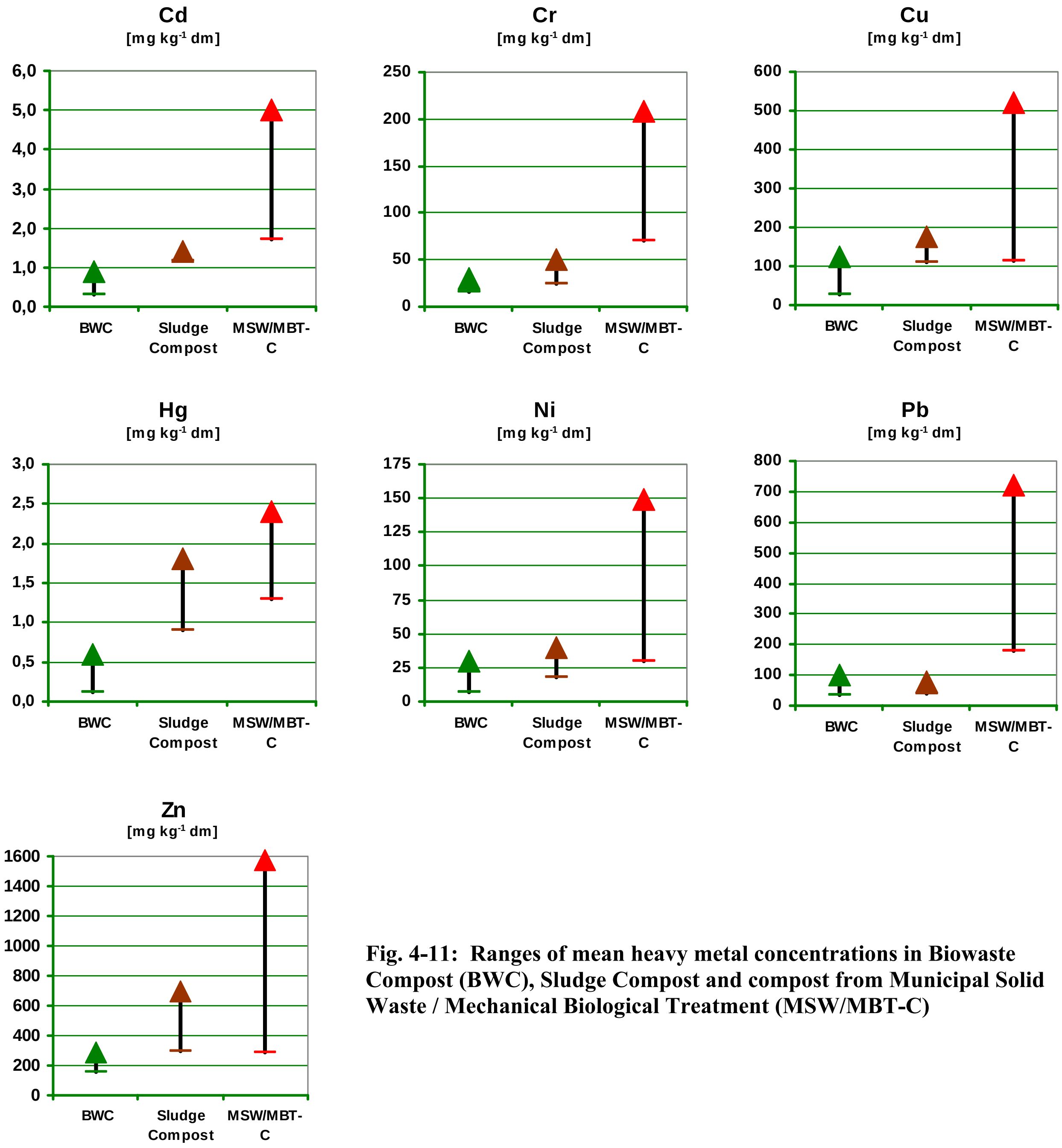
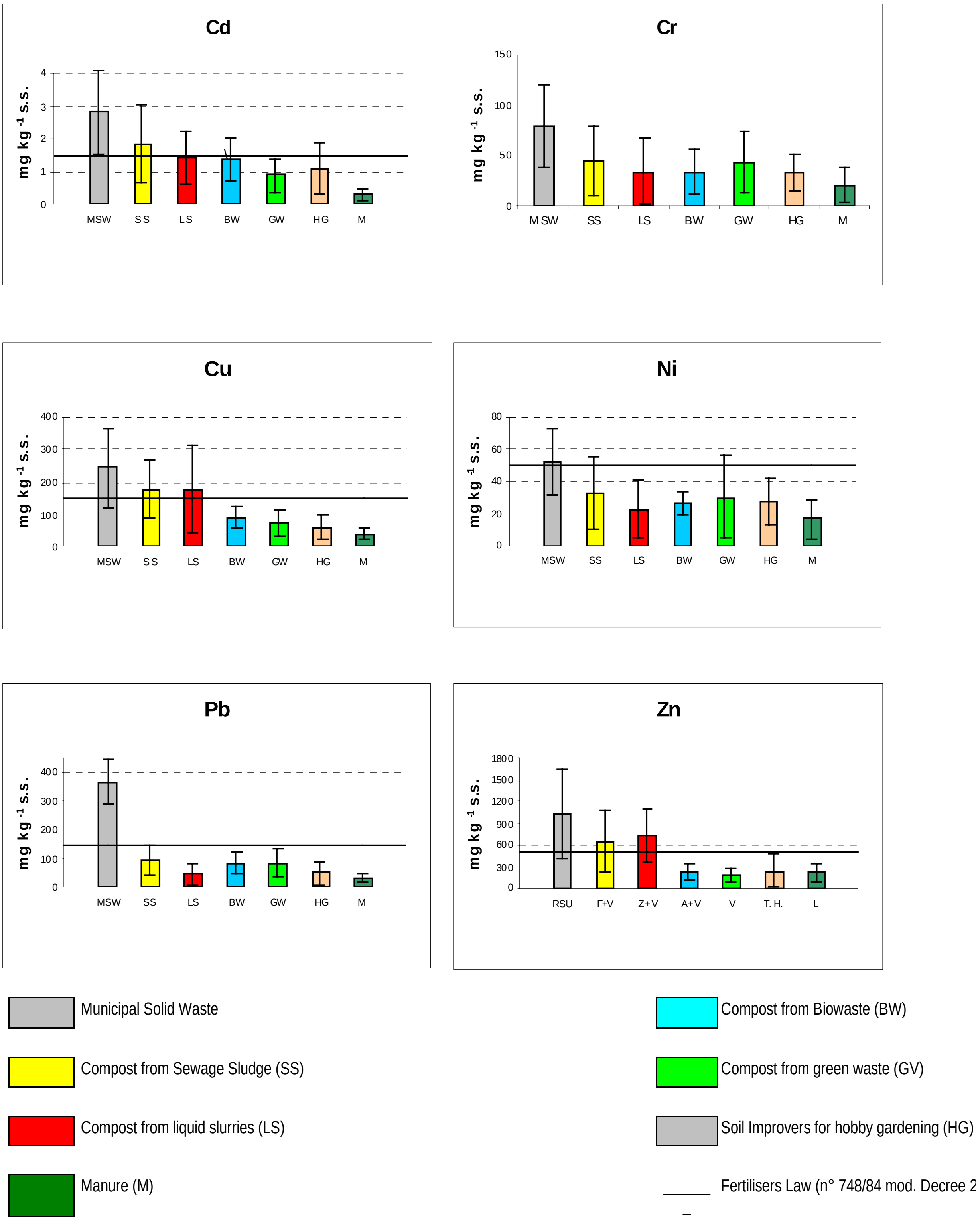


Fig. 4-11: Ranges of mean heavy metal concentrations in Biowaste Compost (BWC), Sludge Compost and compost from Municipal Solid Waste / Mechanical Biological Treatment (MSW/MBT-C)



From: Centemero M. modified, 2000

Fig. 4-12: Heavy Metal levels of Soil Improvers (compost and manure) from different sources compared to Italian Fertiliser Law limit values

5 ORGANIC POLLUTANTS

The main question for both PTEs and organic pollutants in compost is whether the ranges found originate from a general background contamination in the environment and lead to an unavoidable distribution in organic waste, or if technological aspects of collection schemes or introduced impurities are responsible for at least a certain portion of the detected contaminants.

When searching for data of organic pollutants in composts as well as in feedstock materials it was soon evident that regular analyses in the framework of compost quality assessment do not exist. Published studies are based on specific investigations on local scale. They are predominantly found in countries with a long tradition in separate collection and composting of organic waste.

Selection criteria are potential occurrence in compost, relatively good knowledge of physico-chemical properties, feasibility for chemical analysis and availability of original published data. Therefore this list of compounds cannot be exhaustive.

- Polychlorinated biphenyls (PCB)
- Polychlorinated dibenzodioxins and dibenzofurans (PCDD/F)
- Polycyclic aromatic hydrocarbons (PAHs)
- Chlorinated pesticides and Adsorbable Organic Halogen (AOX) *(Aldrin, Biphenyl,* **o-Phenylphenol** *Chlordane, Dieldrin, Endrin, Heptachlor, DDT [1,1,1-trichlor-2,2-bis(p-chlorphenyl)ethan], Lindane, HCH-isomers [Hexachlorcyclohexan], Hexachlorobenzene, Hexachlorobenzol, Heptachlor,* **Pentachlorophenol** *Pyrethroides Thiabendazole*
- Linear alkylbenzene sulphonates (LAS)
- Nonylphenol (NPE)
- Di (2-ethylhexyl) phthalate (DEHP), butylbenzyl phtalate (BBP), dibutyl phtalate (DBP)

Brändli et al. (2003) qualifies further the following substances and compounds with potential importance for which no or very little information is available with respect to compost:

- Polychlorinated Naphthalenes (PCNs) only soil - data available
- Polychlorinated n-Alkanes (PCAs) no data (occurrence in the environment) available
- Brominated flame retardants (BFRs): polybrominated diphenylethers (PBDEs), tetrabromobisphenol A (TBBPA) and hexabromocyclododecane (HBCD) only soil - data available
- Alkylphenol ethoxylates (APEs) only soil - data available
- Bisphenol A (BPA) no data available
- Tributyltin (TBT) no data available

Further substances for which also little information is available and which are unlikely to be relevant for compost:

- Chlorinated Aliphatic Hydrocarbons (CAHs)
- Chlorobenzenes (CBs)
- Chlorophenols (CPs)
- Polydimethylsiloxanes (PDMS)
- Personal care products

Due to the lack of information and the probability to be relevant in organic waste derived composts the compounds listed in the foregoing two paragraphs are not considered in the further investigation in this study.

5.1 Feedstock materials

Numbers on organic contaminants in specified source materials for composting are scarce. In order to get an impression of the most frequently investigated compounds, figures for PCDD/F, PCB and PAH are summarised in Table 5-1.

Table A4-3 and A4–4 in Annex 4 give further data on PCDD/F and PCB in different feedstock materials.

Where available further comparison between feedstock and compost is given in chapter 5.2.

Table 5-1: Undesirable compounds in different source materials (PCDD/F in ng TE kg^{-1}d.m.; PCB and PAH in mg kg^{-1}d.m.; mean (with standard deviation)) (Sihler, 1996)

	n	OM	Sum PCB	PAH	PCDD/F
			PCB 28 –180	6 PAH FL, BbFl, BaP, Bper, IP	(Spring)
		%	mg kg^{-1}d.m.	mg kg^{-1}d.m.	ng TE kg^{-1}d.m.
green waste	1-9	77 30	0.008 0.046	0.566 3.250	4.96 ± 3.56 16.44 ±14.58
Brändli et al. (2003)	see explanations in 1st par. of 5.2		0.231 range: 0.023-0.196	5.874 range: 0.156–63.091	3.6 range: 0.3 - 31.4
bio waste	3-5	78 30	0.010 0.048	0.174 0.685	1.62 ±1.01 5.17 ±2.07
Brändli et al. (2003)	see explanations in 1st par. of 5.2		0.296 range: 0.024-0.529	2.148 range: 0.090-11.331	6.6 range: 0.3 - 31.6
bark	4-9	94 30	0.007 0.103	0.621 7.799	1 ±0.57 13.11 ±10.47
materials from back yard composting	2-6	63 30	0.052 0.096	0.93 1.741	2.24 ±0.99 4.61 ± 2.92

5.2 Organic pollutants in biowaste and green compost

Beside own investigations we also refer here to a recently published literature research carried out by Brändli et al. (2003)[15]. The authors characterised compost and organic waste samples according to certain criteria (input material, collecting area, season). Datasets with five entries or more have been taken into account. For the comparison of compost contamination levels according to the mentioned criteria mean values of the single investigations were judged as the appropriate figures. By experience it can be stated that median values are lower than the mean values by 20-50 % for compost datasets. Moreover it has to be noted that values generated by studies on compost might be higher than average values as highly contaminated composts are often included (e.g. compost produced from roadside grass clippings). Further, the datasets might be not completely coherent due to application of different sampling and analysing methods or to time displacement of the studies.[16]

5.2.1 Polychlorinated biphenyls (PCB)

Levels of PCBs are very low even in compost made from household waste and do not exceed the guide values given in table 5-2. The PCB load of the impurities included in the input material is responsible for the PCB concentration in the end product.

Marb et al. (2001) found higher concentrations of PCB in BWC than in GC, but due to the rather limited number of samples the validity of this ranking is not very high.

In the Baden-Württemberg study, BWC also showed higher PCB contents than GC (Breuer et al., 1997).

This trend was also confirmed by Brändli et al. (2003) when evaluating literature data (concentrations for ∑ 6 PCBs in biowaste compost: mean = 0.04, range = 0-0.579 mg kg^{-1} and green waste compost: mean = 0.0237, range = 0-0.117 mg kg^{-1} d.m.). Compost values are higher than in corresponding input materials (mean: 0.296, range: 0.024-0.529 mg kg^{-1} d.m.; mean: 0.231 mg kg^{-1} d.m.; range: 0.023-0.196 mg kg^{-1} d.m.). The overall mean value of all evaluated datasets of the 6 PCB congeners 28, 52, 101, 138, 153 and 180 was 0.050 mg kg^{-1} d.m. (range: 0–609 µg kg^{-1} d.m.; n≈150).

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[15] The data have been obtained from: Berset and Holzer (1995), Buyuksonmez et al. (2000), Aldag and Bischoff (1995), Strom (2000), Vanni et al. (2000), Zethner et al. (2000), Hund et al. (1999), Tørsløv et al. (1997), Paulsrud et al. (2000), Kuhn et al. (1990), Vergé-Leviel (2001), Wang et al. (2003), Lemmon and Pylypiw (1992), Malloy et al. (1993), Nilsson (2000), Wagman et al. (1999), Lisk et al. (1992b), Harrad et al. (1991), Lisk et al. (1992a), Wilke (1997), Kummer (1996), Kuhn et al. (1990), Houot et al. (2002), Krauss (1994), Marb and Scheithauer (2001), Epstein (1997), Kummer (1990), Bayerisches Landesamt für Umweltschutz (1995), Schleiss (2002), Fischer et al. (2001).

[16] It must be stated that the statistical evaluation of the investigated datasets from literature may be judged as critical. Nonetheless this approach may give an valuable impression of the range of concentration levels when compared with results from specific studies and situations.

Table 5-2: PCB- - Guide values and concentrations in various composts

	Guide values			Types of compost				
PCB [1)] **[mg kg^{-1} d.m.]**	0.2 [A]	0.2 [B]	0.1 [C]	**BWC**	**GC**	**BWC/MC**	**Wet waste**	**MSWC**
Kumer (1992); mean				0.44 3 plants	0.24 5 plants			1.68
Krauß et al. (1992); range of means				0.150 – 0.860 6 plants	0.030 – 0.480 9 plants			0.730 - 1.680 4 plants
Krauß et al. (1992); mean				0.104	0.045			
Fricke et al. (1991); median				0.23 75%il: 0.32	0.15 75%il: 0.24			
Fricke & Vogtmann (1993); mean				0.26* OM: 33.58% ** Difference not significant*	0.178* OM: 37.77%		0.938 OM: 55%	1.493 (“total waste compost”)
Schwadorf et al. (1996); median				0.08	0.07			
Breuer et al. (1997); median				0.056	0.051			
Amlinger (1997[SP277]); mean				0.03 n=6	0.03 n=3	0.02 n=14		
Zethner et al. (2001); median				0.0116 n=42 plants				
Marb et al. (2001); mean				0.077 n=15; OM:30%	0.036 n=5; OM:30%			
Stock & Friedrich (2001); median				0.025 n=30				
Stock et al. (2002); mean				0.00979 n = 60	0.01108 n = 12	only 20 of 60 BWC samples and 5 of 12 GC samples were above the detection limit		
Houot et al. (2003); range				0.034 – 0.104 n=4	0.019 – 0.066 n=2			0.041 – 0.293 n=5
Timmermann et al. (2003)				0.051 0.014 – 0.108 n=30				

[1)] Σ 6 congeners; mean values
MC … Manure Compost
(A) Composting Decree Baden-Württemberg (1994)
(B) Austrian Fertiliser Ordinance (1994)
(C) Luxembourg guide value

The influence of "urban" and "rural" and mixed catchment areas were analysed using a cluster analysis (Zethner et al., 2001). At a confidence interval of 90 and 95 % respectively, higher concentrations were found in urban catchment areas for the sum of PCB(6). Fricke & Vogtmann (1993) could not verify a significant difference between urban and rural areas.

Table -5-3: 16 PCB in compost from urban areas and rural areas (Fricke & Vogtmann, 1993)

Sum PCB PCB 28 –180	**catchment area**		
	Germany	**urban**	**rural**
mg kg^{-1}d.m.	0.254 a	0.279 a	0.282 a

A six year lasting statistical plot trial on 6 different sites in Germany lead to the conclusion that very low PCB levels (mean: 0.051 mg kg^{-1}d.m.) at application rates between 6 and 20 t d.m. per ha and year did not affect the concentration in soil or in crops respectively (Timmermann et al., 2003).

It can be concluded that since PCBs are excluded from industrial processing since the 1980ies a continuous reduction of their occurrence in the environment can be expected. They can still be considered as urban pollutants, being detected in higher concentrations in urban composts. There is also some evidence for systematic higher concentrations in biowaste than in green compost.

5.2.2 Polychlorinated dibenzodioxins and dibenzofurans (PCDD/F)

PCDD/F contents of composts depend upon the processed raw material. In both biowaste and green waste composting lower values are attainable by well-designed collection and sorting of the material (Landesanstalt für Umweltschutz Baden-Württemberg, 1995).

Brändli et al. (2003) calculated mean concentrations of 36.9 ng I-TEQ kg^{-1} d.m. (range: 0.06 – 3800 ng I-TEQ kg^{-1} d.m.; n≈150) for compost and 3.2 ng I-TEQ kg^{-1} d.m. (range: 0–108 ng I-TEQ kg^{-1} d.m.; n=176-350) for organic wastes. Differences are significant for almost all congeners (p=0.05). Comparing the feedstock materials (biowaste and green waste). They found generally (not significant) higher PCDD/F concentrations in biowaste (mean: 6.6, range: 0.3 - 31.6 ng I-TEQ kg^{-1} d.m.) than in green waste (mean: 3.6, range: 0.3 - 31.4 ng I-TEQ kg^{-1} d.m.).

Marb et al. (2001) found a reduction of PCDD/F- contents between samplings in the years 1993 and 2000. BWC showed higher concentrations of PCDD/F than GC, but due to the rather limited number of samples the validity of this ranking is not very high.

A 1994 decree in Baden-Württemberg promulgated a guide value for composts of 17 ng I-TEQ kg^{-1} dry matter (+30 %) (Umweltministerium Baden-Württemberg, 1994). This was derived from the limit value in the German Sewage Sludge Ordinance of 100 ng kg^{-1} dry matter, under the assumption of an annual input of 10 tonnes compost dry matter per hectare. The final guide value is 22 ng I-TEQ kg^{-1} dry matter. In case the Austrian sewage sludge regulations were taken as basis for the derivation of an orientation value for compost, this would give a figure of 25 ng I-TEQ kg^{-1} dry matter (Zethner et al., 2001). However this was not considered by the Austrian Compost Ordinance from 2001.

An investigation of 45 Biowaste and Green Composts by Zethner et al. (2001) with a maximum value of 86.8 ng I-TEQ kg^{-1} d.m. in one compost sample and a sample median of 6.43 ng I_TEQ kg^{-1} dry matter showed that it is not necessary to conduct inspections of compost for dioxins and furans in general. However, when treated wood, dust and input materials contaminated with precursors (such as PCP and PCB) are used, they recommend case by case analyses.

Green waste composts appear to be somewhat less contaminated than biowaste composts. Fricke & Einzmann (1995) consider that this may be caused by (dust charged) impurities and pollutants stemming from the kitchen waste fraction in the bio bin. In order to reduce the levels of impurities intensive public relations activities directed at households would be necessary. However, differences between green waste and biowaste composts from German compost plants were not significant (Fricke et al., 1991).

Krauß (1994) attributes the higher mean values of PCDD/F in the province of Hessen as compared to Baden-Württemberg to a higher regional and deposition-caused background contamination. So far he has not been able to establish a seasonal trend, whereas Fricke & Einzmann (1995) report a rise towards summer primarily of the higher chlorinated PCDD/F.

Table 5-4: PCDD/F - - Guide values and concentrations in various composts [ng I-TEQ kg^{-1} d.m.]

Guide value:	[A] 17 / 22*	[B] 50	[C] 20	[D] 20
PCDD/F	**Types of compost**			
[ng I-TEQ kg^{-1} d.m.]	**BWC**	**GC**	**BW/MC**	**MSWC**
Kumer (1992); mean	15.6 3 plants	11.0 5 plants		19.6
Krauß et al. (1991)	15	10		40
Krauß et al. (1992); Range of means	7-18 6 plants	5-16 (9 plants		
Krauß et al. (1992); mean	14 (+/-9)	11(+/-8)		
Krauß (1994)	Hessen: 13.0 BW°: 10.0 Bayern: 12.0	Hessen: 13.0 BW°*: 6.0		
Fricke & Vogtmann (1993); mean	12.10* OM: 33.58% ** difference not significant*	10.58* OM: 37.77%		
Fricke & Einzmann (1995); standardised to 30% OM	mean: 14.8 median: 11.3 75%il: 17.5	mean: 10.6 median: 7.5 75%il: 13.7		
Amlinger (1997[SP306]); mean; n=23	4.3 n=6	3.6 n=3	1.5 n=14	
Zethner et al. (2001); median	9.18 n=42			
Marb et al. (2001); mean	15.0 n=15; OM:30%	10.5 n=5; OM:30%		
Timmermann et al. (2003)	7.2 2.0 – 13.6 n=30			

PCDD/F soil guide values [ng TE kg^{-1} d.m.]	**Guide values for the use and remediation of soils** (working group on Dioxins Germany, 1992) **recommendations/measures (agricultural and horticultural use)**
< 5	Target value; any soil use possible; ubiquitous background concentration
5 - 40	Threshold for control measure and recommendation for precautionary action
> 40	Restriction for certain agricultural and horticultural soil use; unrestricted use if dioxin transfer is very low

[F] Composting Decree Baden-Württemberg (1994), * Tolerance +30%
[G] Austrian Fertiliser Ordinance (1994); obligatory declaration of PCDD/F concentration between 20 and 50 ng I-Teq kg^{-1} d.m.
[H] Luxembourg guide value
[I] Switzerland: guide value of the draft *Swiss Stoffverordnung*
° BW … Baden-Württemberg, Germany

In contrast Fricke & Vogtmann (1993) found a significant difference of PCDD/F contents in compost stemming from urban and rural areas respectively.

Table 5-5: 16 PCDD/F in compost from urban areas and rural areas (Fricke & Vogtmann, 1993)

PCDD/F	**catchment area**		
	Germany	**urban**	**rural**
ng TE kg^{-1}d.m.	12.07 a	10.15 a	12.68 b[1]

[1] significantly different at $p \leq 0.10$

Krauß (1993) considers a general introduction of limit values problematic, as the contamination of composts from separate collection is governed by the background contamination of the region involved. In regions with a higher soil contamination an increase of the dioxin contents of the composts has to be expected (Krauß, 1993).

Timmermann et al. (2003) evaluated 30 biowaste composts of Baden- Württemberg between 1995 and 2002. In contrast to PCBs they observed a significant reduction of PCDD/F concentrations in this time period.

It can be concluded that PCDD/F concentrations in compost predominantly reflect the ubiquitous background concentration caused by diffuse atmospheric deposition with no clear difference between urban and rural areas but a trend of higher concentrations in biowaste than in green compost. Biowaste and green waste feedstock shows generally lower concentrations than the finished composts.

5.2.3 Polycyclic aromatic hydrocarbons (PAHs)

Mean values for the sum of the 16 EPA PAHs in compost samples (1.956 mg kg^{-1} d.m.; range: 0–19.258 mg kg^{-1} d.m.; n≈100) are in general lower compared to the mean concentrations measured in organic waste (4.229 mg kg^{-1} d.m.; range: 0–181.427 mg kg^{-1} d.m.; n≈200) (Brändli et al., 2003). The mean value of ∑16 EPA PAHs in biowaste compost (2.448 mg kg^{-1} d.m., range: 0.080-13.000 mg kg^{-1} d.m.) is higher than in the input material (1.782 mg kg^{-1} d.m., range: 0–31.345 mg kg^{-1} d.m.). Mean concentrations of ∑16 EPA PAHs in green waste compost (2.148 mg kg^{-1} d.m., range: 0.090-11.331 mg kg^{-1} d.m.) are lower than in green waste (5.874 mg kg^{-1} d.m., range: 0.156–63.091 mg kg^{-1} d.m.).

The low aggregated PAHs, such as *Triphenylene* can be found in concentrations from n.d. up to 63.2 µg kg^{-1} dry matter; highly aggregated PAHs, such as *Benzo(e)pyrene*, were measured with concentrations up to 1264.7 µg kg^{-1} dry matter. The presence of ashes in some of the composts is suspected to be the cause of the raised PAH levels at a few plants (Zethner et al., 2001).

PAHs are higher in material contaminated with tar or asphalt. Exclusion of these materials should reduce any risk to acceptable levels (Bywater, 1998).

Table 5-6: PAH + BaP - - Guide values and concentrations in various composts

	Guide values			**Types of compost**				
PAH + BaP [mg kg^{-1} d.m.]	6 [A]	3 [B]	10 [C]	**BWC**	**GC**	**BW/MC**	**Wet waste**	**MSWC**
Kumer (1992); mean; PAH 16				0.8-1.04	n.a.			4.41
Fricke & Vogtmann (1993); mean PAH 6				1.707 OM: 33.58%	1.560 OM: 37.77%		3.370 OM: 55%	4.412
Schwadorf et al. (1996); median				3.9	3.8			
Breuer et al. (1997); median; PAH 16				3.584	3.586			
Amlinger (1997[SP323]); mean				1.2 n=6	1.7 n=3	0.8 n=14		
Zethner et al. (2001); median				0.962 n=42 plants				

Marb et al.(2001); PAH 16; without NWG mean	4.573 n=15; OM:30%	2.674 n=5; OM:30%		
Stock & Friedrich (2001); median	1.9 n=30			
Stock et al. (2002); median; PAH EPA	2.35 n=60	2.16 n=12		
Houot et al. (2003); range	1.4 – 11.19 n=4	1.51 – 1.68 n=2		1.47 – 4.99 n=5

[A] Composting Decree Baden-Württemberg (1994)
[B] Austrian Fertiliser Ordinance (1994[p330]); Danish Ministry of Environment and Energy (2000[p331])
[C] Luxembourg guide value

Marb et al. (2001) found higher concentrations of PAHs(16) in BWC than in GC, but due to the limited number of samples (5 of each) the validity of this ranking is not very high.

In the Baden-Württemberg study (Breuer et al., 1997) PAH showed no differences between BWC and GC. The relevant spatial and temporal variability of the PAH was explained by seasonal differences in the composition of the raw material, by different collection capture areas or occasional contamination with extraneous materials (Breuer et al., 1997).

The influence of "urban" and "rural" and mixed catchment areas was analysed using a cluster analysis by Zethner et al., (2001). Higher concentrations were found for urban areas at a confidence interval of 90 and 95 % (12 PAHs).

Fricke & Vogtmann (1993) also found a significant difference between urban and rural areas (see Table 5-7).

Table 5-7: PAH in compost from urban areas and rural areas (Fricke & Vogtmann, 1993)

PAH (6)		catchment area	
	Germany	**urban**	**rural**
mg kg^{-1}d.m.	1.707 a	3.421 b[1]	0.585 a

[1] significantly different at $p \leq 0.05$

Kuhn et al. (1990) found in their first study higher values of PAHs in compost containing grass clippings from roadsides compared to reference composts by a factor 2.9 for the sum of 10 PAHs. They also detected an reduction of 23% during composting of when grass clippings. Based on the results they suggested that PAHs were partly degraded during composting.

Table 5-8: Mean concentrations of PAHs in normal compost (nCom), compost containing grass clippings from roadsides in the input material (RoCom), and in grass from roadsides (Grass_Ro) in mg kg^{-1} d.m. (Kuhn et al., 1990))

PAH (10)	**mean_nCom**	**mean_RoCom**	**factor**	**Grass_Ro**	**reduction**
mg kg^{-1}d.m.	1.747	5.158	2.95	6.670	23%

Amundsen et al. (2002) found PAH degradation of more than 60% after 28 weeks of windrow composting of sewage sludge.

In a recent study Kuhn & Arnet (2003) found a smaller difference between compost containing grass from roadsides and normal composts compared to their previous study (factor 1.6 for the sum of the 16 PAHs). However, for many compounds and for the sum of the 16 PAHs the difference is still significant. In the same study foliage and grass from road areas were analysed. As an overall conclusion it can be stated that foliage collected along municipal roads is the least contaminated input material, followed by grass from highway areas (with significant differences for the 3-ring PAHs; p=0.1). The highest contaminated input material was foliage collected along national- and cantonal roads.

In summary, it can be stated that PAH concentrations in organic waste is higher than in compost. Conclusively a degradation process is likely to occur. However, formation of none-extractable residues might be considered. Compost containing a considerable amount of biowaste tends to exhibit higher concentrations of PAHs than green waste composts (Brändli et al., 2003). There is no indication for a systematic difference between biowaste and green compost, whereas urban origins in several investigations proved to produce higher levels than rural ones.

5.2.4 Chlorinated Pesticides and Adsorbable Organic Halogen (AOX)

The combined group of chlorinated hydrocarbons is composed of differing chemical compounds. Chlorinated pesticides as *Aldrin, Biphenyl, Chlordan, Dieldrin, Endrin, Heptachlor, DDT [1,1,1-trichlor-2,2-bis(p-chlorphenyl)ethan]* and its metabolites as well as *Lindane* and *HCH*-isomers *(Hexachlorcyclohexan)*, *Hexachlorbenzene (HCB) and Heptachlor* originate from their application in agriculture.

For some compounds (mostly organochlorines) Brändli et al. (2003) report higher concentrations in compost than in organic waste (Table 5-9). Substances such as γ-HCH (Lindane) and thiabendazole exhibit higher concentrations in organic waste, probably due to degradation during composting (p=0,05). Since the number of measurements is limited, it was not possible to assess the different classes of the input materials and their corresponding composts separately.

Table 5-9: Mean concentrations of pesticides in Compost and organic waste in $\mu g\ kg^{-1}$ d.m. (combined evaluation of literature data with datasets of at least 5 in each investigation)

	Mean Compost	Mean org. waste		Mean Compost	Mean org. waste
pp'-DDT	5.17	10.69	β-HCH	0.42	0.20
pp'-DDD	1.39	1.24	γ-HCH	2.59	7.97
pp'-DDE	8.11	3.77	Pentachlorobenzene	1.16	0.27
op'-DDT	2.88	1.36	Thiabendazole	13.59	27.44
op'-DDD	0.65	0.09	o-Phenylphenol	25.38	31.22
op'-DDE	0.06	0.00	Biphenyl	55.24	39.90
α-HCH	0.38	1.32			

Table 5-10 summarises the main findings of the literature on the occurrence of AOX and other pesticides in BWC and GC.

Table 5-10: AOX and further OPs - - Guide values and concentrations in BWC and GC

	Guide values	Types of compost			
		BWC	GC	BW/MC	Wet waste
AOX **[mg kg^{-1} d.m.]**	500 [A]				
Zethner et al. (2001); median		67 n=42			
Stock & Friedrich (2001); median		54 n=30			
Stock et al. (2002); median		54 n=60	45.5 n=12		
Hexachlorobenzol **HCB [µg kg^{-1} d.m.]**					
Zethner et al. (2001); median		0.6 n=42			
Marb et al.(2001); mean		5 n=15; OM:30%	3 n=5; OM:30%		
Pentachlorophenol **[µg kg^{-1} d.m.]**					
Zethner et al. (2001); median		16.8 n=42			
Marb et al.(2001); mean		17 n=15; OM:30%	6 n=5; OM:30%		
Stock et al. (2002)		n.d. n=31	n.d. n=2		
Biphenyl **[µg kg^{-1} d.m.]**					
Marb et al.(2001); mean		100 n=15; OM:30%	59 n=5; OM:30%		
o-Phenylphenol **[µg kg^{-1} d.m.]**					
Marb et al.(2001); mean		40 n=15; OM:30%	30 n=5; OM:30%		

(A) Austrian Compost Ordinance, limit value for mixed municipal solid waste compost. Analyses every 500 t MSWC production; use of MSWC only as landfill reclamation layer or as bio-filter.

The primary pesticides found in raw feedstock were organochlorine insecticides, including chemicals that have long been banned. Prominent examples include *chlordane*, *dieldrin* and *DDT*. Chlordane, in particular, was detected in a number of studies. Apparently, it is difficult to cleanse the environment of these "old" persistent chemicals. The absence of organophosphates, carbamates and most herbicides in composting feedstock implies that these classes of pesticides are well degraded, diluted or lost to the environment before the feedstock are collected for composting.

Although many studies report that pesticides were found in "low" concentrations, only a few publications offer any basis for identifying what constitutes a low or high concentration. Little information is available regarding the effect of pesticides remaining in the compost with regard to its safety for humans, the food system, plants or the environment. Two publications applied food-based standards to evaluate the concentrations of residual pesticides in compost. Miller et al. (1992) used the maximum allowable tolerance for raw agricultural commodities (for hay crops) as a standard and found that only one individual sample failed this strict test. Similarly, Richard & Chadsey (1989ab) compared pesticides levels found in compost to USDA tolerances for food products. Only *chlordane*, with an average concentration of 0.09 mg kg^{-1}, exceeded the food tolerance standard of 0.03 mg kg^{-1}. However, the authors stated that the chlordane levels found in the leaf compost were well within the range of concentrations typically found in suburban soils.

In the absence of other guidelines, such as background soil levels, food-related tolerance levels provide at least some benchmark for safe concentrations of pesticides in compost. Based on the findings of the studies reviewed by Büyüksönmez et al. (2000), in regard to pesticide residues, compost appears to be safe for use on food crops.

Results of a series of analyses carried out on a range of pesticides in compost in the UK have indicated contents below the level of detection. This series included *lindane, chlorinated hydrocarbons* such as *DDT* and organophosphates (Bywater, 1998). However, the author considers pesticides as potentially the most important group of pollutants in compost

Lindane-concentrations found in compost are extremely low, proving negligible for influencing compost quality. The addition of wood chips to bio waste may occasionally lead to increased gamma-*HCH*-values *(lindane)* in compost, because it is still in use for the protection of lumber in some countries. Zethner et al. (2001) found the occurrence of pesticides in garden compost as well as a few representatives of this group in almost all samples of BWC and GC. For instance *lindane* was frequently detected. Concentrations below 3.8 µg kg^{-1} dry matter were only found in a few samples of compost. At the present time, the authors conclude that a routine investigation of the whole range of chlorinated hydrocarbons was not necessary.

Table -5-11: 16 HCH and DDX in compost from urban areas and rural areas (Fricke & Vogtmann, 1993)

		catchment area	
	Germany	**urban**	**rural**
HCH µg kg^{-1}d.m.	6.85 a	9.57 b	3.94 a
DDX µg kg^{-1}d.m.	29.06 a	35.53 a	22.69 a

Further, Fricke & Vogtmann (1993) conclude from their analyses that the contents of all other xenobiotics in BWC and GC are extremely low and give no rise to any questions with regard to compost quality

In their literature review Brändli et al. (2003) found approx. 100 compost samples, where *Pentachlorophenol* was measured, but no values for organic waste. The mean value in compost was 20.1 µg kg^{-1} d.m. (range: 0.67 – 190 µg kg^{-1} d.m). Besides the single measurements in the database only a few aggregated values are found in the literature. Mean concentration of *HCB* was at 3.93 µg kg^{-1} d.m. (range 0-156 µg kg^{-1} d.m.; n=147). Analysis of *HCB* in organic waste revealed a mean of 2.26 µg kg^{-1} d.m. (range 0-20 µg kg^{-1} d.m.; n=11).

Marb et al. (2001) found in 5 BWC and GC higher concentrations of HCB and PCP in BWC

The influence of "urban" and "rural" and mixed catchment areas were analysed using a cluster analysis by Zethner et al. (2001). At a confidence interval of 90 and 95 % higher γ-HCH concentrations were found in GC and urban areas, AOX had higher concentrations in BWC and urban areas. Backyard composts showed higher contamination for γ-HCH, β-Endosulfan, Pentachlorphenol and AOX.

DDT and metabolites (DDX)

Traces of this pesticide are still found in all parts of the environment and in the biosphere. Zethner et al. (2001) report fifteen GC samples which showed *DDX* values of up to 21.2 µg kg^{-1} dry matter. This demonstrates the potential occurrence of these compounds in the environment. In the soils of Upper

Austria an average of 0.6 and 1.0 µg DDX kg^{-1} dry matter was found under grassland and arable land respectively (Amt der Oberösterreichischen Landesregierung, 1993). The DDX value in the humus layer of Austrian forested locations had a median of 7.79 µg kg^{-1} dry matter (Weiss, 1998).

Zethner et al. (2001) found the highest concentrations for DDX in backyard composts and higher concentrations in BWC than in GC.

The Austrian Fertiliser Ordinance (BGBl. 1007/1994) provides for a "total" limit value for all organochlorine pesticides of 1 mg kg^{-1} dry matter. This group contains the following compounds: *aldrin, dieldrin, endrin, heptachlor, total HCH, DDT and DDE, chlordane and hexachlorobenzene*. The loading of compost with chlorine pesticides is within the limit values of the Austrian Fertiliser Ordinance, a finding that was also born out by other investigations. For this reason, it was concluded that routine investigations of compost for these substances are not necessary (Zethner et al., 2001).

Pyrethroides

In the investigation of Zethner et al. (2001) the concentrations of *Pyrethroides* found were not remarkable. Nevertheless noticeable contamination is to be expected where the compostable materials from allotments, kitchen waste and pest control enter the composting process. The maximum concentration of *Cyfluthrine* and *Deltamethrine* detected were 68 and 60 µg kg^{-1} dry matter respectively which lie significantly below the limit value of the 1994 Fertiliser Ordinance (Zethner et al., 2001).

Thiabendazole

Thiabendazole is a systematic fungicide which is absorbed through leaves and roots. It is used both on crops such as bananas, citrus fruits and mangoes and to control pests and fungi in storage (e.g. protection of potatoes against Fusaria infestans). In the environment its most important toxic effects are on fish.

The median of the measured *Thiabendazole* concentrations in composts ranges between detection and determination limits. Higher concentrations (max. 32.7 µg kg^{-1} dry matter) were found in kitchen compost compared to garden compost (Zethner et al., 2001).

5.2.5 Linear alkylbenzene sulphonates (LAS)

Only Denmark has established a limit value for LAS for sewage sludge and waste derived composts. The limit value of 1,300 mg kg-1 d.m. is mainly justified for sludges, in particular when stabilised anaerobically. In compost LAS may be found below the detection limit or at very low concentrations.

The reviewed literature and recent data from Nordrhein-Westfahlen provided by Stock et al. (2002) confirm that LAS may not be expected to constitute an environmental problem in BWC.

Table 5-12: LAS - Guide values and concentrations in various composts

	Guide values	Types of compost			
		BWC	**GC**	**BW/MC**	**Wet waste**
LAS [mg kg^{-1} d.m.]	1,300 [A]				
Paulsrud et al. (2000)		median: 41 range: 9-396 n=9			
Stock & Friedrich (2001)		not detectable n=30			
Zethner et al. (2001)		not detectable n=15			
Stock et al. (2002)		not detectable n=60	not detectable n=12		

(A) Danish Ministry of Environment and Energy (2000)

5.2.6 Nonylphenol (NPE)

The Danish Ministry of Environment and Energy (2000) has set a limit value for NPE of 10 mg kg^{-1} d.m. for sewage sludge and waste derived composts.

Scheithauer & Marb (2002) report NPE “values” in compost only below detection limit.

Similar to Phtalates and LAS, Nonylphenol has a high degree (up to 60%) of breakdown under aerobic composting conditions (Bywater, 1998, Amundsen et al., 2002).

5.2.7 Phtalates - Di (2-ethylhexyl) phthalate (DEHP), Butylbenzyl phtalate (BBP), Dibutyl phtalate (DBP)

There are a few studies where Phthalates (DBP, DEHP, BBP) have been analysed in composts. Brändli et al. (2003)calculated compost mean values from literature datasets. The results are as follows: DEHP: mean: 9.606 mg kg^{-1} d.m. (range: 0–69.58 mg kg^{-1} d.m.); DBP: mean: 1.584 µg kg^{-1} d.m. (range: 0-7.9 mg kg^{-1} d.m.); BBP: mean: 0.0437 mg kg^{-1} d.m. (range: 0-0.141 mg kg^{-1} d.m.) respectively. For DEHP they computed significantly higher concentrations are detected in biowaste compost than in green waste compost (p=0.05).

Table 5-13: DEHP - - Guide value and concentrations in BWC and GC

	Guide values	Types of compost	
		BWC	**GC**
DEHP [mg kg^{-1} d.m.]	50 [A]		
Marb et al.(2001); mean		8.5 n=15; OM:30%	2.9 n=5; OM:30%
Stock et al. (2002); median		1.35 n=60	0.42 n=12

[A] Danish Ministry of Environment and Energy (2000)

DEHP is probably released into the environment by abrasion, evaporation or extracting from plastics such as foils and floor mats, lacquers etc and also introduced into the bio bin by impurities. Correlation between the DEHP load and parameters like impurities in the input material, sorting effort, and catchment area are not significant. Composts with increased DEHP content do not necessarily show increased contents of other pollutants (Scheithauer & Marb, 2002).

In Denmark the contamination with DEHP of biowaste composts with increasing proportions of organic household waste was analysed (Hogg et al., 2002). The investigation, which was carried out on two composting plants, gave elevated numbers in composts with a very high amount of household/kitchen waste. (> 85 %). No problems (as compared to the Danish limit value) occurred in composts originating from mixtures with less than 80 % household/kitchen waste.

Table 5-14: (DEHP) in compost with different parts of organic household waste in Denmark (Hogg et al. 2002)

% of household waste in relation to garden waste in raw material	Number of plants	DEHP in compost (mg kg^{-1} d.m.) (Limit value = 50)
85-100	2	**60**
70-80	2	31
40-60	2	4.8
10-30	2	0.55
0	2	0.55

5.2.8 *Organic pollutants in MSWC/MBTC*

It is obvious that composts derived from mixed waste collection systems (*grey dust bin*) may contain any of the chemical compounds and their potential metabolites that are introduced at the point of collection. It has been proved that for all types of contaminants a segregation of unwanted and not degradable constituents at the waste treatment plant is not likely to achieve qualities comparable to those expected with source separated materials. Waste recovery methods such as material recycling and thermal recovery not only change the waste quantities but also its composition (Table 5-15). If waste fractions low in potential pollutants are withdrawn from the mixed waste stream by separate collection schemes, the proportion of pollutants should increase in the residual waste fraction. On the other hand any concern about this effect should be reduced by separate collection of hazardous waste the contaminants. Considering these two opposing trends Doedens (1997) estimates that the overall contents of potential pollutants in MSW is declining.

Table 5-15: Change of residual waste properties by intensified recycling or separate collection (Doedens, 1997)

	Change of the concentration in residual WASTE of	
Intensified Recycling and/or Separate Collection	Heavy Metals	Organic Pollutants
Biowaste/Green Waste	+ +	+ +
Waste Paper	–	– AOX
Waste Glass	++ (M)	+ + (M)
Waste Plastics	– Cd	– PCB; Cl; F
Waste Wood	O	– –
Waste Metals	– –	++ (M)
Problematic Waste; Small Quantities of Hazardous Waste	– Hg	– – PCB; ??
Electronic Scrap	– Hg	– PCB; PCN
Refrigerating Appliances	-	– – FCKW
Industrial Waste		

+ Concentration Increase; - Concentration Decrease,
(M) Change by the Elimination of Inert Substances

Table 5-16, Table 5-17 and Table 5-18 summarise investigations on organic pollutants in mixed waste compost comparing the results with existing limit values. The concentrations of PCB, PCDD/F and PAH show a comparatively low variation.

The concentrations found for PCBs and PCDD/F would exceed existing limit values for BWC and GC as well as the limit for municipal solid waste compost laid down in the Austrian Compost Ordinance.

PAH concentrations (range between 0.6 and 9.4 mg kg^{-1} d.m.) would not exclude the use of MSWC in Luxemburg. Also most of the material would meet the requirements for the use of MSWC as surface layer on landfill sites in Austria. Only the Danish threshold value for waste products would clearly eliminate stabilised residual waste only by the expected concentration of PAH.

Table 5-16: PCB in MSWC mg kg^{-1}d.m.

	(1) Range of means from 4 plants PCB (6)	(2) Total WC PCB (6)	(2) Wet WC* PCB (6)	(3) MSRW** PCB (5)	Limits **BWC** LU° / BW°°	Limit **MSWC** AT
Sum PCB	0.730 - 1.680	1.493	0.938	0.1 - 10 (1.0)	**0.1/0.2**	**1°°**

° LU … Luxembourg; °° Composting Decree of Baden-Württemberg; * Waste Compost from fraction remaining after separate collection of dry recyclables; ** MSRW municipal solid residual waste **before** biological treatment or composting; (1) Krauß et al. (1992); (2) Fricke & Vogtmann (1993); (3) Doedens (1997)

Table 5-17: PCDD/PCDF in MSWC and MSW ng I Teq kg^{-1}d.m.

	(1) Total WC	(2) Total WC	(2) Wet WC*	(1) MSW	(3) MSRW**	Limits **BWC** LU° / BW°°	Limit **MSWC** AT°°°
Sum PCDD/F	38±22	103	50	57±39	50	**20/17 (22)**	**50**

° LU … Luxembourg; °° Composting Decree of Baden-Württemberg; °°° Limit Value for Dioxins only; * Waste Compost from fraction remaining after separate collection recyclable dry waste; ** MSRW municipal solid residual waste **before** biological treatment or composting; (1) Krauß et al. (1992); (2) Fricke & Vogtmann (1993); (3) Doedens (1997)

Table 5-18: PAH (6) in MSWC and MSW mg kg^{-1}d.m.

	(1) Total WC	(1) Wet WC*	(2) MBTC	(3) MSRW**	Limit **BWC** LU	Limit **BWC** DK	Limit **MSWC** AT
Sum PAH (6)	4.41	3.37	0.64 – 9.4 ***(3.1)***	34	**10**	**3**	**6**

* Waste Compost from fraction remaining after separate collection recyclable dry waste; ** MSRW municipal solid residual waste **before** biological treatment or composting; (1) Fricke & Vogtmann (1993); (2) Nelles et al. (1997) (3) Doedens (1997)

An investigation in 1992 by Krauß et al., (1992) shows the difference to compost quality accomplished by source separated organic waste, see Fig. 5-1 for PCB and Fig. 5-2 PCDD/F. This gives a clear impression to what extent the level of these two ubiquitous compounds may be reduced by source separation systems.

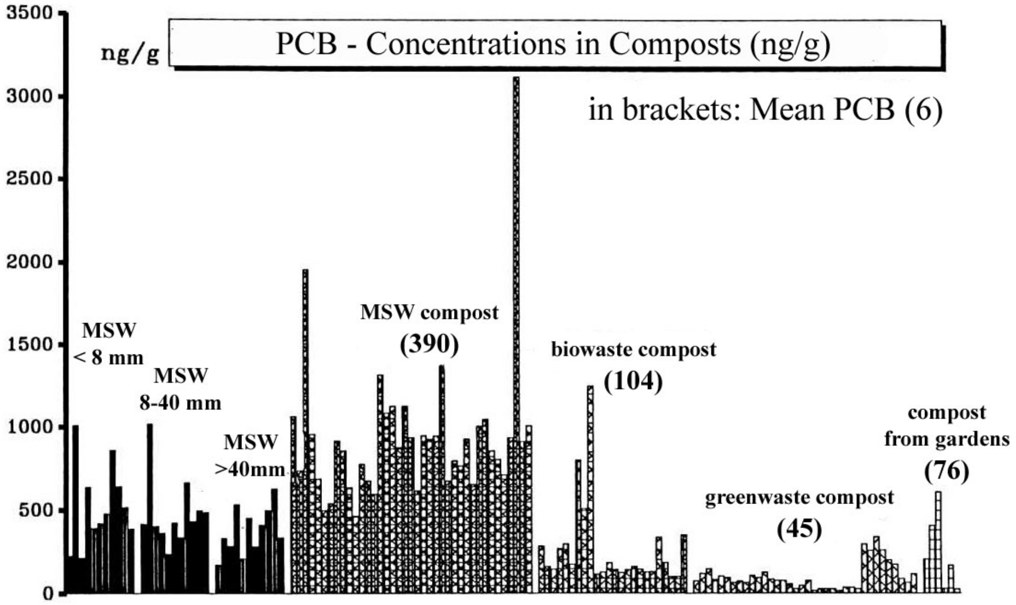

Fig. 5-1: Total PCB concentrations in different compost types and mixed solid waste; in addition the average sums of the congeners 28, 52, 101, 138, 153 and 180 (PCB(6)) are shown in brackets (Krauß et al., 1992)

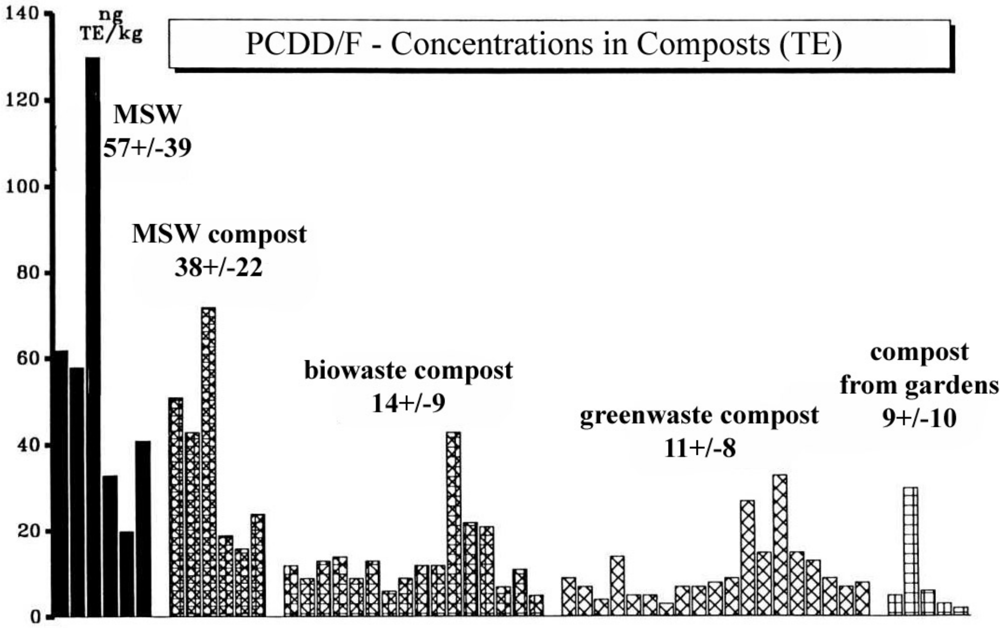

Fig. 5-2: PCDD/F concentrations in different compost types and mixed solid waste; numbers: mean values and standard deviations (Krauß et al., 1992)

5.2.9 *Organic pollutants in sewage sludge*

A wide variety of organic chemicals with diverse physical and chemical properties may be found in sludge. Table 5-19 illustrates the compounds to which the most frequent references are made, and those, which are mentioned in quality standards.

Table 5-19: Average contents of organic micro-pollutants in sewage sludge in 1988/89 compared with data from German publications till 1996

		AOX	PCB	PAH	DEHP	NPE	PCDD/F
		[mg kg^{-1} d.m.]					*[ng TE kg^{-1} d.m.]*
1988/89 (1) range of means		250 – 350	< 0.1	0.25 – 0.75	50 - 130	60 – 120	< 50
1991/96 (1) range of means		140 – 280	0.01 – 0.04	0.1 – 0.6	20 - 60	-	15 - 45
Recent EU survey (2)	*range*	60 – 400	0.001 – 0.232	0.01 – 246	0.2 – 170	0.02 – 824	0.2 – 128
	Range of means / medians	150 – 350	0.04 – 0.113	0.57 – 34	3.16 – 67.3	0.02 – 25.8	4.2 – 20.5
Austria (3)	*range*	13 - 510	0.013- 0.379	0.77 - 8.97	nn*- 47	0.46 - 65	2.87 – 24.68
	mean or median	157°	0.054	3.64°	7.2	25	5.46

(1) Arthur Andersen on behalf of DG Environment (2001); (2) Leschber (2004); (3) Aichberger et al. (2002)
° sum of Acenaphtene, Phenanthrene, Fluorene, Fluoranthene, Pyrene, Benzo(k)fluoranthen, Benzo(a)pyrene, Benzo(ghi)perylene, Indeno(1,2,3-c,d)pyrene;
* detection limit 0.095 mg kg^{-1} d.m.

According to data available in the literature, sludge can contain between 0.018 and 10 mg kg^{-1} d.m. of PAHs in EU Member States (Arthur Andersen on behalf of DG Environment (2001).

PCDD/F data were taken from the compilation of EU dioxin exposure and health data (AEA Technology, 1999). It appears that average concentrations of dioxins are quite similar among Member States, between 15 and 40 ng I-TEQ kg^{-1} d.m. According to the study, this would indicate that the sources of contamination in the Member States are similar. Industrial inputs can also cause important contamination in sludge. In some cases, more than 1,000 I-TEQ kg^{-1} d.m. were reported.

These figures show that after a significant improvement, the level of pollutants is approaching a base level. The improvements result from reducing the sources of the pollution, mostly point sources such as industrial discharge.

For more general information on organic pollutants view the fact sheet in Annex 1 and the summarising table in chapter 1.4.2.

5.2.10 Degradation of organic pollutants via composting [17]

Alterations in the concentration of undesirable organic compounds during the composting process are caused by their volatility and their physical, chemical and biochemical degradation potential.

As for the fate of organic pollutants in soil, the following processes are important during composting (and anaerobic digestion):

- volatilisation,
- solubilisation and consequently leaching from the compost,
- degradation i.e. transformation or mineralisation and sorption.

Incorporation into organisms is less substantial since the organisms involved in the composting process are exclusively micro organisms, for which bioaccumulation plays a minor role (Brändli et al., 2003).

Volatilisation is dependent on the vapour pressure of a compound and since during composting temperature can rise up to 80 °C, this pathway for elimination can be crucial. It has been shown that volatile compounds such as FLU and PCB 77 are partly removed during composting by stripping and since volatilisation is dependent on the temperature, the higher the rotting temperature the more efficiently these compounds are removed (van Raaij et al., 1996).

Leaching is dependent on the water solubility of a specific compound and their metabolites. It is assumed that leaching plays a minor role, since during composting leaching of water is limited if composting is operated properly (e.g. covering outdoor windrows and collecting runoffs). Moreover, organic pollutants have often a high octanol-water partitioning coefficient, which results in a high retention of these compounds in the carbon-rich compost matrix. E.g. Hund et al. (1999) did not observe significant formation of water soluble compounds during composting.

However leaching contains also particles, which may adsorb pollutants, which might induce removal of organic contaminants (Fromme et al., 2002; Buyuksonmez et al., 1999). It was observed that composting favoured the degradation of model substances to polar metabolites and supported their binding to the DOM matrix. It is reported by Hartlieb et al. (2001) that DEHP and simazine were mainly found in the low- to medium-molecular DOM fraction, whereas pyrene and its metabolites had high affinities to high-molecular DOM. It is interesting to see that the binding capacity of DOM changes during composting showing the highest binding capacity for hydrophobic contaminants after 120 days..

The processes of **degradation i.e. transformation** or **mineralisation** and **sorption** are tightly linked. If a compound is sorbed, it is less available to microbial degradation and hence transformed and

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[17] *Parts of this chapter are built on the literature review "Occurrence and Relevance of Organic Pollutants accounting particularly for Compost Digestate and Organic Residues" by Brändli et al., 2003.*

mineralised to lesser extend. Metabolites resulting from degradation processes may contain functional groups that can form covalent bonds to functional groups of the compost humic substance. There are indications that sorption of organic pollutants in compost becomes stronger with time, comparable to the aging process in soil. As explained by Buyuksonmez et al. (1999) abiotic (photolysis, hydrolysis) and biological transformation processes occur simultaneously during composting, however the biological process is assumed to be the most important.

The **microbial degradation** can be enhanced by improvement of necessary factors like moisture, high temperatures and sufficient amounts of organic material. Depending on the degree of the aerobic situation in the composting material, degradation is accompanied by oxidative and also reductive reactions (Haider, 1985; Hurle, 1978; Lester, 1983; Scheffer & Schachtschabel, 1982; all quoted in Fricke & Vogtmann, 1993)

PAHs

Most PAHs are very persistent in soils. Their half-life can reach up to 10 years. They also are slowly biodegraded. PAHs are relatively insoluble in water and are therefore absorbed at the particulate level, especially in organic matter. Loss from soil (mainly for 2 and 3 ringed PAHs) may be due to photodegradation, or volatilisation (Arthur Andersen on behalf of DG Environment, 2001). PAHs are degraded in sunlight by aerobic bacteria and are unlikely to persist through any well-managed composting operation. PAHs have been shown to be partially degraded during composting of organic residues (Vergé-Leviel, 2001; Loser et al., 1999; Martens, 1982), of a mixture of sewage sludge and organic residues (Lazzari et al., 1999) or municipal solid waste in mechanical biological treatment (Drahosch, 1998). Fricke et al., 1989 report a reduction rate for PAHs during a three-month composting period of 56%. Studies with radio-labelled PYR have revealed degradation by 56 % in biowaste compost and by 47 % in sewage sludge compost after 131 days of composting. In biowaste compost degradation was almost completed after 35 days (Hund et al., 1999). Active mineralisation of PHE was observed at mesophilic and thermophilic temperatures (Carlstrom & Tuovinen, 2003). During the thermophilic stage of municipal solid waste composting degradation has been described to be higher than in the following stages (Joyce et al., 1998; Drahosch, 1998). This has been confirmed within a study for remediation of soil using several types of organic wastes as co-composting substrates (Wan et al., 2003). In contrast, Martens (1982) suggests that microbial populations of ripe municipal solid waste compost exhibit higher capacities to mineralise PAHs than micro organisms of fresh compost. The same conclusion was drawn by Vergé-Leviel (2001) for organic waste compost. Houot et al. (2003) report an effective degradation of PAHs (phenanthrene and fluoranthene) to an extent of 50 – 70 % only in mature compost with best performance of BWC for fluoranthene if compared to GC and MSWC respectively. In summary, Brändli et al. (2003) conclude that PAHs undergo degradation during composting, i.e. mineralisation or transformation to metabolites. The latter can be further degraded or persistent. Lighter PAHs may volatilise. Additionally, a fraction is strongly bound to the compost organic matter. The main transformation processes seem to occur during the thermophilic phase of composting. However, some studies suggest the higher potential for degradation during the maturation phase.

In degradation experiments with soil from sites contaminated by PAHs, field trials have revealed half-life values of 6-16 years compared with 0.5-5 years in laboratory scale trials (Knudsen et al., 2001).

PCBs

During the composting process a reduction of the PCB concentrations appears to occur by volatilisation of the volatile congeners of PCBs. It has been considered as negligible (Vergé-Leviel, 2001; Nilsson, 2000; Drahosch, 1998) and accumulation has been observed (Lazzari et al., 1999). No difference was found for degradation between composting and mesophilic thermophilic digestion of organic waste (Nilsson, 2000). An older study revealed elimination of 15 to 74 % (mean: 45%) with volatilisation of the within a three months composting process (Fricke et al., 1989). In a study with radio-labelled PCB 77 formation of $^{14}CO_2$ has been observed after 60 day of composting indicating increasing and complete bio transformation (van Raaij et al., 1996). Degradation of PCBs has been reported in soil composted with yard trimmings (40% in the best case) (Block, 1998). Congener specific PCB analysis indicated that lower chlorinated PCBs (congeners with 1-3 chlorines) were preferentially degraded. The PCB loss observed appeared to be largely due to biodegradation and not volatilisation (Michel et al., 2001). Bywater (1998) found that PCB content in material composted for three months drops to around 25% of its original value. Gibson et al. (2002) found losses of 2% of the original PCB concentration of sewage sludge during 142 days of windrow-composting, whereas approximately 30% losses are reported during a 40-day drying period.

However a systematic degradation during composting bears still considerable uncertainties since generally higher concentrations are found in compost than in feedstock. Nonetheless, lower chlorinated compounds may be degraded to a higher extent than those with a higher chlorination grade.

PCDD/PCDF

Dioxins such as PCDD/F do not easily degrade and in fact tend to become more concentrated as composting proceeds due to the reduction of organic matter. Inputs to compost can be reduced by refusing to accept PCDD/F treated timber (Bywater, 1998). According to some studies formation of PCDD/F cannot be excluded (Fiedler, 1994; Krauss et al., 1994; Oeberg et al., 1994). According to Fricke & Einzmann (1995) a general increase of PCDD/PCDF during the composting of yard waste and biowaste can be observed. Most tests conducted during the composting process of biowaste showed an increase of hepta- and octa- PCDD. On the other hand, the content of low chlorinated PCDD/PCDF decreased during the composting process and generally Furans are diminished.

Initial generation during the rotting process only contributes to a negligible degree to dioxin content in composts (Krauß, 1994). Generation of dioxins during the rotting process only occurs with temperatures > 70°C and in the presence of primary substances as *trichlorophenol* and *pentachlorophenol*. The order of magnitude of the net increase of dioxins is app. 0-2 I-Teq kg^{-1} with a basic contamination with *chlorophenol* of 15-30 mg kg^{-1}.

However, the relevance of PCDD/F-formation during composting is still discussed controversially.

Chlorophenols (CPs)

Laine & Jorgensen (1997) studied the degradation of **CPs** during composting: more than 90% of the CPs were removed during the composting period of 6 months (compost piles with a size of 13 m^3). In a parallel bench scale experiment, an average of 60 % mineralisation of radio labelled *pentachlorophenol* (**PCP)** was obtained in 4 weeks. The authors concluded that a major part of chlorophenols was completely mineralised. Drahosch (1998) reports a degradation and/or formation of bound residues of CPs during composting of mixed municipal solid waste. In other studies composting has been found to enhance degradation (Semple & Fermor, 1997; Valo & Salkinoja-Salonen, 1986) or immobilize CPs by bound-residue formation (Benoit & Barriuso, 1995). Radio labelled PCP was found to degrade almost completely after 131 days (Hund et al. (1999). Considering their degradability during composting, CPs are not considered as substances with priority for analysis in compost or digestate.

Pesticides

Since a vast amount of pesticides exist and their degradation behaviour is very different, final statement about their fate during composting cannot be given. Pesticides may be degraded or accumulated (due to relative low recalcitrance and mass reduction during composting), depending on their properties as on the compost biota. Pesticides have been found to degrade best at temperatures of around 52 °C (Bywater, 1998). Taube (2001) assessed the fate of *chlorothalonil*, *o-phenyl-phenol* and *thiabendazole* during fermentation and following composting. *Chlorothalonil* was found to have a half-life of 0.89d in mesophilic and of 0.44d in thermophilic conditions and was completely degraded during the process. *O-*

phenyl-phenol was found from 65-100% in digestate and *thiabendazole* was stable under mesophilic conditions whereas under thermophilic conditions it was eliminated. This is also supported by Büyüksönmez et al. (2000). The review undertaken showed that thermophilic temperatures enhanced degradation, in most cases, but whether this was due to particular thermophilic organisms or simply the accelerating-effect of higher temperatures is not apparent. Furthermore the high organic matter levels within composting media and compost make adsorption a particularly important mechanism.

Wilke (1997) simulated a composting process of southern fruit peels and studied the degradation of several pesticides. The results are summarized in Table 5-20. Little to almost complete degradation was observed.

Table 5-20: Residues of pesticides after composting in percent of the initial concentration (source: Wilke, 1997 cit. in: Brändli et al., 2003)

residues after composting	(%)	residues after composting	(%)
Thiabendazole	**77**	α-Endosulfan	2
o-Phenyl-phenol	**6.6**	β-Endosulfan	1.7
Biphenyl	**6.1**	Parathion	0.7
Imazalil	**18**	Pirimiphos-methyl	4.5
Metalaxyl	**30.4**	Heptenophos	<0.01
Dichlofluanid	**1**	Triazophos	1.6
Lindane	**6.6**		

The results on the dynamics of concentrations of undesirable organic compounds during a three-month composting period show that HCH-compounds are degraded to a very large extent (86% range: 74 – 93%). This was the case to a lesser degree for the sum of DDX (64%; range: 56 – 83) (Fricke & Vogtmann, 1993)

Michel et al. (1996) found that about 50% of *2,4-D* applied to a laboratory compost system was mineralised, whereas for *pendimethalin* about 14% and for *diazinon* 11 % was mineralised only.

Vergé-Leviel (2001) analysed the fate of *dicamba* and *carbendazime* during composting and found that most of the *dicamba* applied could be extracted after incubation and maturation whereas *carbendazime* resisted the composting process as well but was not extractable by methanol.

Hund et al. (1999) studied *pentachlorophenol* degradation during biowaste composting and found an elimination rate of about 95%. Hartlieb and Klein (2001) found a mineralisation rate of *simazine* of 16% and a ratio of 65% of NER relative to the initially applied pesticide after 370 days of composting.

Frederick et al. (1996) studied the fate of *diazinon* during the composting of yard trimming and found that a relatively small amount (11%) of *diazinon* was mineralised, the major part being extractable. However some none extractable residues were formed. Vandervoort et al. (1997) found a degradation of *isoxaben*, *flurprimidol*, *chlorpyrifos* and *triclopyr* to non detectable levels after a year of composting, 2,4-D seems to be slightly more persistent, whereas clopyralid is degraded very little. Stephenson et al. (1994) found a degradation rate of *2,4-D*, *mecoprop* and *dicamba* of about 80% during turfgrass composting. However it has to be taken into account that considerable amounts of pesticides have been added in the beginning.

Nilsson (2000) analysed several pesticides in organic waste and in the corresponding compost. The input material was processed also by a mesophilic or thermophilic fermentation and pesticide concentrations were measured in the different digestates. No general pattern could be observed which is not surprising since physico-chemical properties pesticides differ considerably.

Vergé-Leviel (2001) found no mineralisation of organic pollutants during the fermentation process. They were just removed by volatilisation. However during the following maturation (at 28ºC) organic pollutants were degraded by microbiological activity and non-extractable residues were formed. The author concludes that maturation after fermentation is crucial for the degradation of organic pollutants.

Vorkamp (2000) simulated a fermentation process in the laboratory and found that methidadion and endosulfan concentration was reduced during the fermentation and the following composting process whereas dodemorph concentration remained stable. It was found that these degradation processes depended on the pH as well as on the temperature.

Büyüksönmez et al. (2000) reviewed research studies concerning the occurrence and fate of pesticides during composting and detected only a few of the target pesticides. Investigations of raw feedstock and compost could not detect many of the target pesticide compounds, and the compounds that were detected occurred at low concentrations.

Comparisons of the pesticide concentrations in feedstock versus the resulting compost showed that composting tends to substantially decrease the concentrations of most pesticides. Pesticides that repeatedly showed little change or even increase in concentration after composting include *dieldrin methoxychlor*, *lindane*, *chlordane*, *toxaphene*, *DDT* and all organochlorines. In the Illinois study (Miller et al., 1992), the herbicides *2,4,5,-T* and *trifluralin* were found to be concentrated. Generally, organophosphate and carbamate insecticides and most herbicides were either not detected in the raw feedstock or their concentration decreased substantially after composting.

In general, the patterns of pesticide degradation in composting parallel the patterns found in soil, but in several studies, pesticide compounds disappeared faster during composting than they typically do in soils, as indicated by their soil half-life values.

Nonylphenols (NP)

Torslov et al. (1997) mention that in laboratory studies and some field studies, it has been found that all, or a large fraction of the nonylphenols (NP) applied through sludge application disappear from soil within 50-100 days. These studies also reported that such an extensive degradation only takes place under aerobic conditions and if microbial activity is not inhibited.

Following several authors, nonylphenols have a high degree of breakdown under aerobic composting conditions (Bywater, 1998, Gibson et al., 2002, Amundsen et al., 2002).

LAS

LAS have also a high degree of breakdown under aerobic conditions. This is supported by Bywater (1998), Brändli et al. (2003), Amundsen et al. (2002). LAS does not degrade under anaerobic conditions, except in particular conditions. Based on the investigation of the HERA project[18] in raw sewage, the LAS concentration was in the range of 1-15 mg/l. When the sewage was properly treated in activated sludge sewage treatment plants (STP), LAS was removed by more than 99%, decreasing its effluent concentration in the 0.009-0.14 mg/l range. The LAS concentration in the sludge was in the range of <1 to 10 g kg^{-1}. During sludge transportation to the farmland, sludge storage, and application on agricultural soil, aerobic conditions are restored and rapid degradation of LAS is resumed. In sludge-amended soils, LAS had a half-life of about one week and levels were around 1 mg kg^{-1} at harvesting time. No accumulation in soil and no bioaccumulation in plants could be detected experimentally.

Amundsen et al. (2002) found degradation rates higher than 60% during 28 weeks of windrow composting of sewage sludge.

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[18] HERA – Human and Environmental Risk Assessment, see: http://www.heraproject.com/ExecutiveSummary.cfm?ID=81

For LAS a wide range of half-life has been reported. It is likely that the observed half-lives are highly dependent on experimental conditions. Jensen (1999) suggests a half life of 7 to 21 days. LAS are readily degraded under aerobic conditions, but no degradation is observed in anaerobic environments. After application of sewage sludge to land LAS have been degraded, thus preventing LAS accumulation in the soil environment (Jensen, 1999).

DEHP

DEHP has a low water solubility and a high octanol-water coefficient. Under aerobic conditions, micro organisms degrade DEHP, explaining the relatively low soil concentration reported in the relevant literature (Torslov et al., 1997). DEHP seems to accumulate in soil after sludge distribution, but only if very large amounts of sludge are applied. At normal doses accumulation does not occur because of the very short half-life of the compound under aerobic conditions. Under anaerobic conditions, DEHP is only very slowly or not degraded at all (Arthur Andersen on behalf of DG Environment, 2001). Also Bywater, 1998) accredits a high degree of breakdown of *Phthalates* in aerobic composting conditions

According to numerous studies phthalates are degraded by a wide range of bacteria and actinomycetes under both aerobic and anaerobic conditions (Staples et al., 1997). Studies with radio labelled DEHP have revealed degradation by 88 % in biowaste compost and by 67 % in sewage sludge compost after 131 days of composting (Hund et al., 1999). In biowaste compost degradation was almost completed after 96 days. In another study 70 % of initial radio labelled DEHP was mineralised (Hartlieb & Klein, 2001). Degradation during thermophilic phase was high (Hartlieb & Klein, 2001; Hund et al., 1999). Formation of non extractable residues (NER) was expected to be low (Hartlieb & Klein, 2001). For describing the fate of phthalates during digestion knowledge from anaerobic stabilization of sewage sludge can be used. Kollotzek et al. (1998) have reported that DEHP was not degraded in both aerobic and anaerobic sludge stabilization. Short chain phthalates were degradable in both processes. Another study showed that under aerobic conditions and a temperature of 63 °C it was possible to achieve up to 70% reduction of the DEHP concentration in sewage sludge (Banat et al., 1999).

5.2.11 Exemplary Accumulation scenarios for PCBs, PCDD/F and PAHs

The assessment of potential accumulation of POPs in the soil as a result of the application of fertilisers is only feasible if persistence is high.

Here we only made the attempt for calculating the change of soil concentration for PCB, PCDD/F. Some phthalates, tensides, phenols and benzenes are relatively rapidly degraded with a half-life between 1 week and 1 month. Due to this fact no accumulation scenarios are computed in these cases.

Without consideration of degradation, Fricke & Einzmann (1995) computed the increase of dioxin in soils through the application of compost: Based on assumptions [compost content = 14 ng Teq kg^{-1} d.m.; application quantity = 10 t d.m. ha^{-1} a^{-1}; soil depth = 30 cm; specific density of soil = 1,4 g $(cm^3)^{-1}$] the dioxin contamination of the soil would increase every 30,3 years by 1 ng Teq kg^{-1} d.m.

In the case of POPs the most critical parameter is the assumed half life time.

The persistence of a substance is characterised by its half-life ($t_{1/2}$), which is defined as the time necessary to degrade 50 % of a substance. It is usually determined in laboratory experiments[19].

For PCBs half life time reported range between several days and 10 years. Biodegradation of PCBs is slow in soils of greater than 10 percent organic content.

Half-life of PAHs and PCDD/F increases proportionally with increasing substitution with chlorine.

Table 5-21: Persistence (as half-life) in soil and sediments of some PCB and PCDD congeners (di Domenico & De Felip, 2000)

Congener	years
PCB (77, 118, 126, 156, 169)	6-10
2,3,7,8-T4CDD	10
1,2,3,7,8-P5CDD	12
1,2,3,4,7,8-, 1,2,3,6,7,8-H6CDD	13

Table 5-22 summarises assumptions taken for the accumulation model of PCB, PAH and PCDD in the soil.

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[19] OECD 304A, 1981, Inherent Biodegradability in Soil; OECD draft 307, 2000, Aerobic and Anaerobic Transformation in Soil; ISO 11266, 1994, Soil quality - Guidance on laboratory testing for biodegradation of organic chemicals in soil under aerobic conditions.

Table 5-22: Assumption for PCB, PAH and PCDD used in accumulation scenarios

		PCB	**PAH**	**PCDD/F**
Half-life in soil				
	(1)	12 & 30 years	16 & 32 years	30 & 100 years
Atmospheric deposition				
	(2)	2 g ha^{-1} y^{-1}	8 g ha^{-1} y^{-1}	29 µg ha^{-1} y^{-1}
Export				
Export (leaching and harvest)		Not assessed; no consistent data available		
Background value soil as starting point for the accumulation in the soil				
LOW	(3)	0.01 mg kg^{-1}dm	0.05 mg kg^{-1}dm	2.44 ng TE kg^{-1}dm
HIGH	(4)	0.04 mg kg^{-1}dm	1 mg kg^{-1}dm	13.30 ng TE kg^{-1}dm
Soil threshold values for multifunctional use				
LOW		0.05* mg kg^{-1}dm	3* mg kg^{-1}dm	5** ng TE kg^{-1}dm
HIGH		0.1* mg kg^{-1}dm	10* mg kg^{-1}dm	40** ng TE kg^{-1}dm
Concentration in compost				
Minimum of means	(5)	0.01 mg kg^{-1}dm	0.6 mg kg^{-1}dm	4 ng TE kg^{-1}dm
Maximum of means	(5)	0.10 mg kg^{-1}dm	4.6 mg kg^{-1}dm	12 ng TE kg^{-1}dm
Assumed high	(5)	0.25 mg kg^{-1}dm	13 mg kg^{-1}dm	18 ng TE kg^{-1}dm
Quantity of compost per ha and year applied				
max. **60 kg P_2O_5 ha^{-1} y^{-1}** (P_2O_5 compost: 0.65 % d.m.)				→ **9.2 t d.m. compost ha^{-1} y^{-1}**
Soil depths and density				30 cm / soil density: 1,5 g cm^{-3} ; → **4,500 t ha^{-1}**
Time frame for the accumulation model				**100 years**

(1) PCB: di Domenico & De Felip (2000); PAH: Knudsen et al. (2001) and PCDD/F: Shatalov et al. (2002)

(2) max. annual load per ha taken from Kupper & Becker van Slooten (2001)

(3) median of background values on arable land or rural soils (PAH) (see Table 3-7).

(4) 90%ile of background values on arable land or rural soils (PAH) (see Table 3-7).

(5) concentrations assessed from Table 5-2, Table 5-4 and Table 5-6

* Precautionary soil threshold values of organic pollutants for soils (German Soil Protection Ordinance, BBodSchV, 1999); low and high values for soils with an organic matter content < and > 8 % respectively.

** Guide values for the use and remediation of soils for agricultural and horticultural use (working group on Dioxins Germany, 1992); 5 ng TE kg^{-1}dm = target value; any soil use is possible; 40 ng TE kg^{-1}dm = threshold for control measure and recommendation for precautionary action.

The accumulation was computed by using the following iterative formula:

$$\boxed{\mathrm{C_n} = \mathrm{C_0} \times \mathrm{x^n} + \frac{I_F + I_D - E_L - E_H}{M_S + M_S} \times \frac{1 - x^{\mathrm{n}}}{1 - x}}$$

where is:

$$x = e^{\frac{-\ln(2)}{t_{1/2}}}$$

C_n … *compound concentration in year n [mg or ngI-Teq* kg^{-1} *d.m.]*

C_0 … *compound concentration in year 0 [mg or ngI-Teq* kg^{-1} *d.m.]*

n … *years for which accumulation is considered*

I_F … *input of compound by fertiliser application [g or µg* $ha^{-1}y^{-1}$*]*

$I_F = C_F. \times M_f$

$= C_F$… *compound concentration in fertilizer applied [mg or ngI-Teq* kg^{-1} *d.m.]*

I_D … *input of compound by atmospheric deposition [g or µg* $ha^{-1}y^{-1}$*]*

E_D … *export of compound by leaching [g or µg* $ha^{-1}y^{-1}$*]*

E_H … *export of compound by harvest [g or µg* $ha^{-1}y^{-1}$*]*

M_s … *Mass of soil layer concerned [t* ha^{-1}*]*

M_f … *Mass of compost addition that remains in the soil after mineralisation [t* $ha^{-1}y^{-1}$*]*

$t_{1/2}$… *half-life time of compound in the soil*

Results

Fig. 5-3, Fig. 5-4 and Fig. 5-5 show the soil change for PCB, PCDD/F and PAH respectively based on the assumptions made given in Table 5-22.

From the computation of accumulation in a certain soil layer it is evident that the graph is not linear and it would approach an asymptotic curve in relation to the concentration level in compost, taking into account the site and management specific long-term mineralisation rate of the compost and the half-life of the contaminant in soil.

For each compound four scenarios with two different half-life times are shown, one with a lower and one with a higher starting point (background concentrations) in soil.

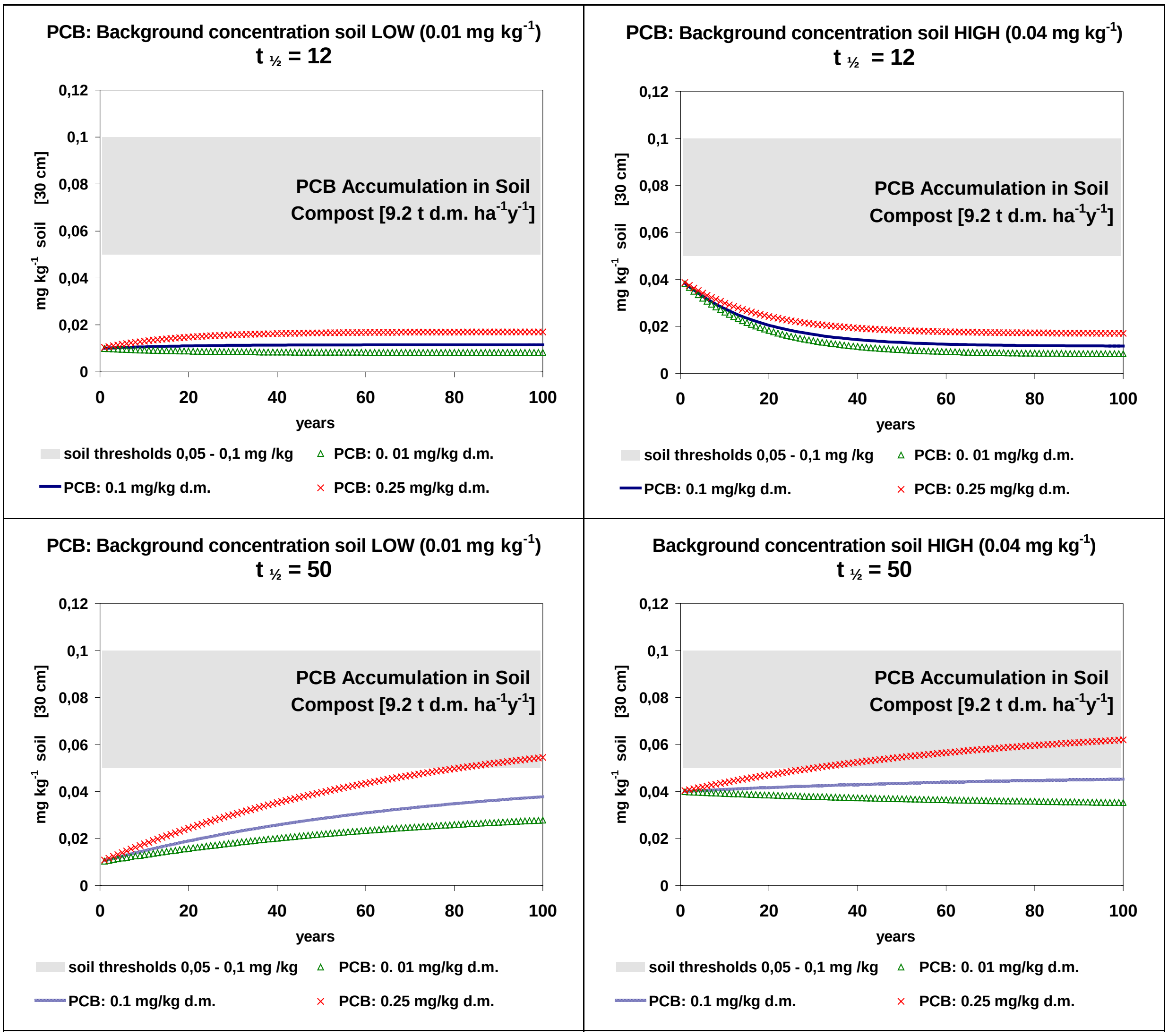


Fig. 5-3: Change of concentrations of PCB due to continuous yearly compost application. *Underlaid grey area … range of threshold values for soils*
top row … $t_{½}$ = 12 years; bottom row … $t_{½}$ = 50 years

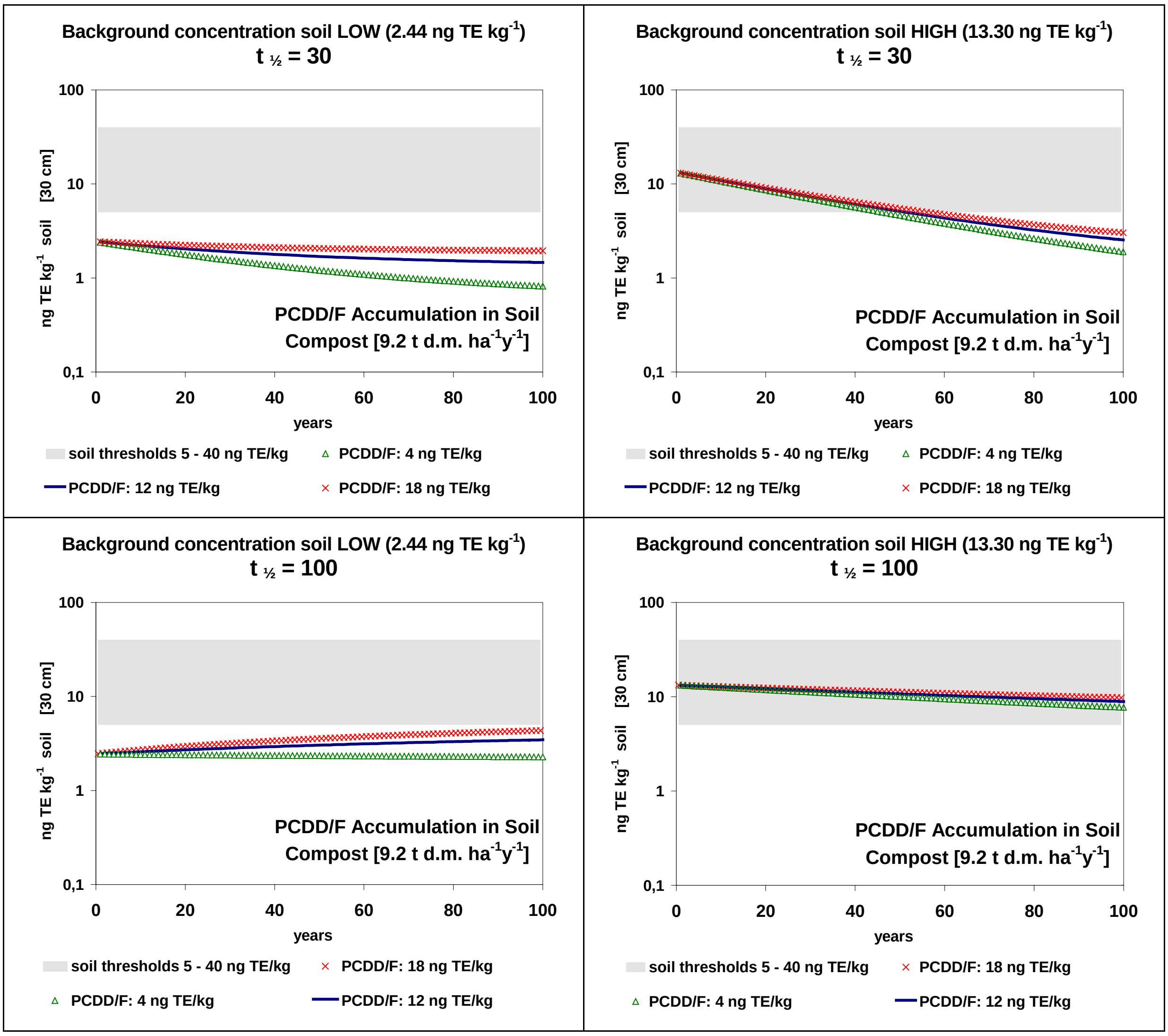


Fig. 5-4: Change of concentrations of *PCDD/F* due to continuous yearly compost application.
Underlaid grey area … range of threshold values for soils;
top row … $t_{½}$ *= 30 years; bottom row …* $t_{½}$ *= 100 years*

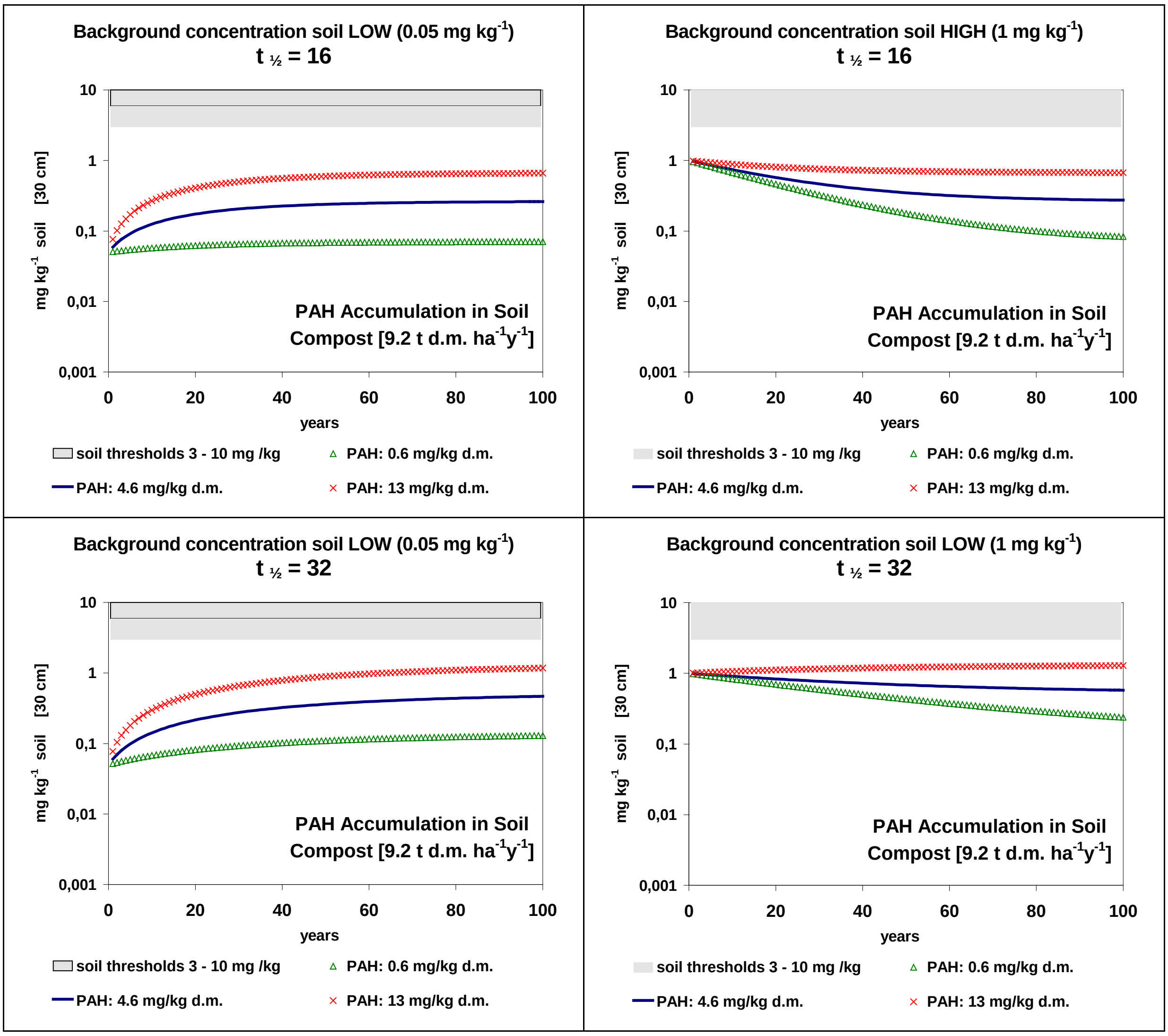


Fig. 5-5: Change of concentrations of *PAH* due to continuous yearly compost application.
Underlaid grey area … range of threshold values for soils;
top row … $t_{1/2}$ = 16 years; bottom row … $t_{1/2}$ = 32 years

The results of Fig. 5-3, Fig. 5-4 and Fig. 5-5 can be summarised as follows:

1.) For **PCB** no accumulation will occur under realistic half-time assumptions ($t_{1/2}$ = 12 years). Half-life degradation would fully compensate inputs by deposition and compost. Only a very moderate increase in soil concentration could be expected when very high compost concentrations are assumed. When we assume a worst case scenario of a half-life of $t_{1/2}$ = 50 years accumulation occurs when starting at low background levels in soil. Even so the lower soil guide value of 0.05 mg kg^{-1} will be exceeded in the case of a high compost concentration (0.25 d.m.) only if combined with a high soil background level (0.04 mg kg^{-1}) within a period of ca. 25 years.

2.) **PCDD/F**: No accumulation occurs in any of the scenarios. In most of the cases we rather find a considerable decrease in soil concentration, even if we assume (unrealistically) nearly no degradation within 100 years ($t_{1/2}$ = 100 years).

3.) Though a certain accumulation of **PAHs** may occur in soils with low background concentrations (ca < 0.5 mg kg^{-1} d.m.) precautionary soil threshold values would not be touched even in the long term.

From these results the need for the establishment of binding limit values for the three compounds for which the most reliable data set can be considered cannot be deduced. The reasons are:

1.) As compared to atmospheric deposition the input by compost can be considered as very low (PCB) at a similar level (PAH) or also slightly higher (PCDD/F)

2.) Commonly a half life of less than 20 years can be estimated → this is much less as assumed in the worts scenarios above

3.) Under the given conditions (very low concentrations in composts) a soil change due to compost application if compared to soil precautionary soil thresholds is of no concern (as long as soil reference values used are acknowledged from an ecotoxicological perception for food and groundwater safety)

For OPs accumulation models must be considered as estimates under the assumed conditions. However they cannot be performed for compounds with even a less comprehensive available data collection than for PCB, PAH and PCDD/F.

Also Aldrich & Daniel (2003) conclude that for PCDD, PAH or PCB sufficient data were available to exclude an unacceptable risk to soil organisms by the recycling of compost in agriculture.

For other substances they judged insufficient data about their concentrations in compost or their ecotoxicological effects would exist to reliably assess the risks. In order to conclusively assess the ecotoxicological relevance of these chemical substances in compost further ecotoxicological, environmental and chemical information would be needed.

Before further considerations are made for researching selected pollutants, the likelihood of there appearance in source separated biowaste must be carefully investigated. In the case of biowaste and green compost it should always be beared in mind that the sources are stemming mainly from our kitchens and gardens.

6 SOIL AMENDMENTS, FERTILISERS, GROWING MEDIA, MANURE

Many expert groups and national authorities have critically discussed the level of potential pollutants in composts in comparison with other fertilisers and manure. Attempts have been made to judge all soil amendments and fertilisers on an individual basis (see chapter 8). In the comparison of organic fertilisers and soil amendments, PTEs and organic pollutants may be standardised to an individual content of organic matter (OM) similar to the former Austrian and German standards, where heavy metal limits and concentrations were calculated at an OM content of 30 % d.m. However, with mineral fertilisers or even liquid agents such as slurry or sewage sludge this comparison on the basis OM would not work as a balanced estimation of the environmental validation of the different materials. In addition, the accompanying mineral substance added to the soil layer has to be taken into account when calculating soil concentration factors (see chapter 9).

The most common approach made is to calculate the ratio between heavy metal and phosphorus level (mg Cd kg^{-1} P or P_2O_5). This method was mainly discussed for the evaluation of the use of sewage sludge as compared to mineral P fertiliser or manure. Austria, for example, set a maximum Cd concentration of 75 mg kg^{-1} P_2O_5 in its Fertiliser Ordinance. The Swedish Act on Tax on Fertilisers (SFS 1984/409) bans P-fertilisers with more than 100 mg Cd kg^{-1} P (=43.6 mg Cd kg^{-1} P_2O_5) and taxes every mg Cd with 30 SEK (~ 3.39 EUR), which exceeds 5 mg kg^{-1} P. In Denmark two alternative systems of limit values have been established for the application of waste products in agriculture: one dry matter and one phosphorus related limitation (Table 6-1). The waste producer may choose either the dry matter or phosphorus related limits.

Table 6-1: Limit values for heavy metals in waste products for agricultural purposes on the basis *dry matter* and *phosphorus* of the Danish statutory order BEK nr. 49.

	Cd	Hg	Ni	Pb
mg kg^{-1} dry matter	0.4	0.8	30	120
mg kg^{-1} phosphorus	100	200	2,500	10,000

It becomes obvious that the heavy metal contents of products with low P concentration like compost will be over estimated as compared to mineral fertiliser or sewage sludge if calculated on a phosphorus basis.

Taking the example of cadmium, the unbalanced situation becomes evident when comparing different fertilisers:

Table 6-2: Cadmium concentration in different fertilisers on dry matter and phosphorus basis respectively

		Cadmium	
		mg kg^{-1} dry matter	mg kg^{-1} phosphorus
BWC (65 composts, A) (1)	mean	0.54	58
	range	0.22-1.34	22-112
BWC (2)		0.56	112
Triple phosphate		28.1	62
Diammon phosphate		28.1	61
Swedish P-fertilisers (3)	mean	---	12
Sewage sludge		1.22	21
Cattle slurry		0.46	24
Pig slurry		0.82	26

(1) Own calculations from 65 Austrian compost analyses
(2) Severin (1999)
(3) Johannesson (2002)

Another approach is to compare the potential risk caused by the level of pollutants and the beneficial effects introduced to the soil-plant system, namely the nutritional (NPK) or soil improving (OM, humus) effect.

Reinhold (1999) found a positive correlation between heavy metal concentration and nutrient level in biowaste composts. If application rates are derived from the efficient supply with plant nutrients it might emerge that heavy metal loads to the soil might be lower with composts rich in both nutrients and metals than when using composts showing a very low heavy metal concentration.

Following this argument it seems justified to compare different fertiliser types on a double basis (acc. to Reinhold, 1999). A proposal for a value-free index has been suggested by the German Quality Assurance Association Compost by weighing potential risks (percentage of PTE concentrations of the limit value) against beneficial effects of the organic fertiliser by OM and nutrient supply (N, P, K, Mg, Ca).

Further discussion see chapter 8 and 9

6.1 Manure

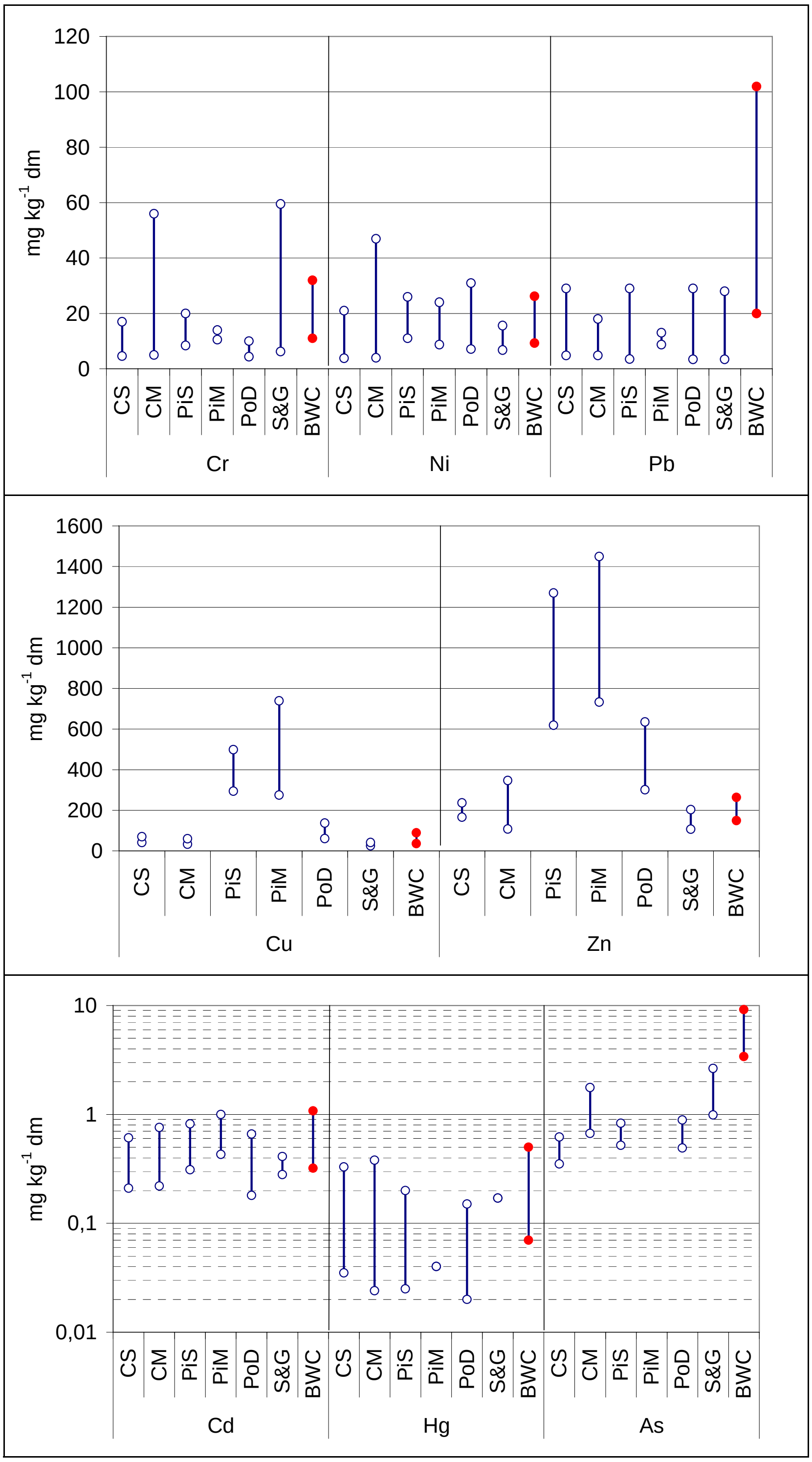
Table 6-3 shows the range of mean values found for different types of liquid and solid manure in comparison with the range of mean PTE concentrations in European BWC.

Only Pb and As occur in higher concentrations in BWC than in most of the manures. Due mineral additives in feedingstuff especially pig manure and slurry is known for its significant elevated concentrations of Cu and Zn (see also Fig. 6-1)

Table 6-3: Ranges of mean heavy metal concentration of several national surveys (without data from Switzerland and Sweden due to not comparable extraction methods)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn	As
		------------	------------	------------	*mg kg^{-1} d.m.*	------------	------------	------------	------------
Cattle slurry		0.21-0.61	4.6-17	42-71	0.04-0.33	3-21	5-29	166-237	0.35-0.62
Cattle manure		0.22-0.76	5-56	33-60	0.04-0.38	4-47	5-18	109-347	0.67-1.77
Pig slurry		0.31-0.82	8.4-20	**294-499**	0.03-0.2	11-26	3.5-29	**619-1270**	0.52-0.83
Pig manure		0.43-1	10.5-14	**276-740**	0.04	8.7-24	8.7-13	**733-1450**	---
Poultry manure/slurry		0.18-2.86	4.4-13	61-137	0.02-0.15	7.1-31	3.4-29	302-636	0.49-0.89
Sheep & goat manure		0.28-0.41	6.3-60	26-41	0.17	6.8-16	3.4-28	107-204	0.99-2.65
BWC*	***low***	***0.32***	***11***	***36.41***	***0.07***	***9.3***	***20***	***150***	***3.4***
	high	***1.08***	***32***	***89***	***0.5***	***26.2***	***102***	***264***	***9.2***

* Range of mean resp. median values of national surveys (AT, BE, DK, FI, FR, DE, IT, LU, NL, NO, CH, UK)



CS…cattle slurry; CM…cattle manure; PiS…pig slurry; PiM…pig manure; PoD…poultry dung; S&G…sheep and goose manure; BWC…biowaste compost

Fig. 6-1: Range of mean values of heavy metals and Arsenic in different animal manures and BWC

6.2 Commercial soil amendments and growing media

Table 6-4 shows range and mean values of several commercial growing media (CGM) and soil improvers. Comparing these figures with the range of average concentrations in biowaste composts one can conclude that on a dry matter basis the qualities lie in a comparable range. If standardised to 30 % OM d.m. the commercial products are in line with the level of highest mean values for compost or in the case of Cd, Cr and Zn exceed this concentration level. When compared to Austrian composts, which have been standardised to 30 % OM, in addition the Pb concentration of CGM exceeds the mean compost value.

Table 6-4: Ranges and mean heavy metal concentration in commercial growing media [CGM] (Tabasaran & Sihler, 1993)

	OM	mg kg^{-1} d.m.						mg kg^{-1} d.m. standardised to 30 % o.d.m.					
CGM	% d.m.	Cd	Cr	Cu	Ni	Pb	Zn	Cd	Cr	Cu	Ni	Pb	Zn
mean	**65.0**	1.0	19.3	38.1	11.2	31.6	111.0	**2.7**	**43.3**	**82.9**	**22.5**	**63.8**	**275.1**
min	45.5	0.2	1.6	7.0	1.0	8.0	17.0	0.3	2.1	9.4	1.3	12.1	22.8
max	87.0	3.4	55.0	210.0	44.0	161.0	330.0	8.1	156.8	297.0	71.7	206.8	914.7
BWC [AT]; median	**35.3**	*0.39*	*30*	*45*	*22*	*41*	*170*	***0.46***	***30***	***51***	***25***	***43***	***188***
BWC*	***low***	*0.32*	*11*	*36.41*	*9.3*	*20*	*150*						
	high	*1.08*	*32*	*89*	*26.2*	*102*	*264*						

* low … high : lowest and highest of means/medians respectively of national surveys (AT, BE, DK, FI, FR, DE, IT, LU, NL, NO, CH, UK)

6.3 Mineral fertilisers

Table 6-5 gives a survey on mean heavy metal concentrations in mineral fertilizers. It shows a wide range of concentrations independent of the heavy metal load derived from the amount of fertiliser applied on land. P-fertiliser may contain distinctly increased levels of Cd and Cr and relatively higher concentrations of Zn as compared to composts.

Table 6-5: Mean heavy metal concentration in mineral fertilizer, quoted from Hackenberg & Wegener (1999)

		Cd	Cr	Cu	Ni	Pb	Zn
Calcium ammonium nitrate		0.31	10.49	4.99	4.71	24.61	55.02
Ammonium nitrate		0.03	1.33	6.33	0.30	0.20	2.30
Urea		0.15	0.68	0.38	0.48	0.36	2.4
other N fertilisers		0.10	6.55	5.15	10.4	1.00	4.00
Super phosphate		20.84	223.50	21.35	31.27	7.20	380.00
Thomas phosphate		0.58	1742.67	30.33	22.00	8.33	56.50
other P-fertilisers		7.51	146.67	15.37	15.37	1.83	225.33
Potash – raw		0.06	10.70	2.35	5.40	0.77	1.57
Potassium chloride		0.10	3.25	3.35	1.25	0.65	4.10
Potassium sulphate		0.09	5.30	3.40	1.90	0.85	2.30
Calcium carbonate		0.50	6.90	8.20	4.60	7.30	58.00
Quicklime		0.10	19.20	11.10	6.00	2.80	15.80
Metallurgical lime		0.10	50.60	4.20	2.45	7.00	8.80
other lime fertilisers		0.33	17.00	19.50	12.50	23.75	35.00
NP-fertilisers		10.23	84.75	24.80	17.10	2.55	115.75
PK-fertilisers		4.80	388.50	22.90	21.40	2.65	153.50
NPK-fertilisers		2.37	32.00	11.80	8.93	11.97	125.33
BWC*	***low***	***0.32***	***11***	***36.41***	***9.3***	***20***	***150***
	high	***1.08***	***32***	***89***	***26.2***	***102***	***264***

* low … high : range of mean resp. median values of national surveys (AT, BE, DK, FI, FR, DE, IT, LU, NL, NO, CH, UK)

A mass balance approach was also taken in order to evaluate the risks posed by cadmium in fertilisers to health and the environment by 10 EU Member States and 2 EEA Member States following the recommendations of the ERM (Environmental Resources Management) study (ERM, 2000).

The existing limit values for cadmium in fertilisers and in soil in the Member States are summarised in Table 6-6.

Table 6-6: Limit values for cadmium in fertilisers and in soil in the Member States (Oosterhuis et al., 2000; adopted from ERM, 2001)

	Cd in fertilisers (mg kg^{-1} P2Os)	Cd input to agricultural soil (g ha^{-1} a^{-1})	Cd in agricultural soils (mg kg^{-1} dry soil)
Austria	75	10$^{(1)}$/5$^{(2)}$	1.0 – 2.0
Belgium/LUX	90	150	1.0 - 3.0
Denmark	47		0.5
Finland	21.5	3	0.5
Germany	40-90$^{(7)}$	16.7$^{(3)}$	0.4 -1.5
Netherlands	$^{(8)}$		0.5
Portugal	40-70$^{(9)}$		1.0 – 4.0
Sweden	43$^{(10)}$	1.75 $^{(4)}$	0.4
UK		0.15$^{(5)}$	3.0$^{(6)}$

Notes:
(^{1}) arable land (^{2}) grassland and vegetables (^{3}) average over a period of 3 years. (^{4}) average for 7 years; will lower to 0.75 g ha^{-1} year^{-1} as from 2000. (^{5}) with sewage sludge only. (^{6}) soils with a pH of 5 and above, treated with sewage sludge. (^{7}) based on a voluntary agreement between government and industry. (^{8}) OECD (1994) mentions a limit of 40 mg kg^{-1} phosphorus (17 mg kg^{-1} P_2O_5). According to a spokesman of VKP, there is no limit for the Cd content in P fertilisers in the Netherlands. Instead a voluntary agreement is in preparation. (^{9}) mentioned in OECD (1994); probably not a legal limit. (^{10}) a voluntary limit of 21.5 mg kg^{-1} P_2O_5 has been introduced by the SLR.

After evaluation of these national risk assessments by ERM (ERM, 2001) the Scientific Committee on Toxicity, Ecotoxicity and the Environment (CSTEE) gave its opinion on the 'Member State Assessments of the Risk to Health and the Environment from Cadmium in Fertilisers' (CSTEE, 2002).

Soil mass balance calculations led to the conclusion that fertiliser Cd concentrations of 20 mg Cd kg^{-1} P_2O_5 yields no increase in soil Cd over a period of 100 years, in most scenarios without additional Cd sources. At 60 mg Cd kg^{-1} P_2O_5 accumulation of Cd is predicted in most of the scenarios (ranging between 40 and 125%) in 100 years. Model parameters were varied over a wide range to cover most agro-climatical conditions in the EU.

Even then considerable uncertainties were identified for the high variability of assumptions made for (i) additional Cd sources (in most cases ignored) (ii) uptake by plants and (iii) leaching.

7 DISCUSSION ON SAMPLING AND ANALYTICAL METHODS

Sampling and analytical methods for pollutants in foodstuffs and compost have a strong impact on the reliability over time of threshold concentrations. Above all, the sampling method must be considered a most important factor. Sampling from bulk, non-homogenous materials (heaps) may contribute to the variance of analytical results by > 1000 % whereas today's validation of laboratory analytical methods lead to negligible standard errors of < 5 – 10 % depending on the absolute level of the parameter concerned. This has been demonstrated by a study on the spatial variability of compost quality in Germany (Breuer et al., 1997) or in inter-laboratory tests conducted in the framework of European standardisation (CEN TC 223 "Growing media and soil improver" – analytical methods)

It is evident that standardised sampling schemes (frequency and method of sampling depending on type of product and capacity of yearly compost production) and analytical methods including tolerances are key issues for any agreement on quality requirements and usability of compost or stabilised biodegradable waste.

This was recognised by the members of the technical committee of CEN TC 223 "Growing media and soil improver" and the related working groups. Thus the problem for the elaboration of European standards for the entire package within the scope of CEN TC223 was not so much finding common and statistically verified analytical methods but conceptual disagreements on safety and specification issues.

In general two steps have to be considered when discussing concepts of analytical tolerances for elements that establish quality classes or limit values:

- Rules for the regular quality approval by the compost producer
  - Repeatability, standard deviation and tolerances of repeated measurements for analytical results for a specified batch within a defined sampling scheme.
  - Tolerances for the maximum deviation of single batches within a certain set of measurements (e.g. tolerance of 25 % of a single batch when the mean value of the last 4 tests meets the limit value).
- Rules for complience tests carried out on behalf of the responsible authority, when taking samples of composts at the market place

Beside standardised sampling methods it has to be made clear how the natural variability of single parameters is considered when randomised control samples are analysed by the authority.

7.1 Comparison of sampling methods

All methods used in the Member States aim at gaining a final and a laboratory sample respectively which represent the entire lot on the one hand, and the material type on the other hand. The latter originates from the general strategy that not every single compost batch produced has to be analysed. This is based on the confidence in a continuous production regime as far as source materials (compost type) and process management (composting technique) are concerned. Therefore the number of analyses to be made follows random sampling systems in most cases in dependence of the total throughput of the entire composting plant. As a matter of course the random sampling scheme has to be applied separately for each substantially specifiable type of compost (e.g. biowaste compost, green compost, sludge compost, stabilised MBT material).

As far as available from national standards the key elements of the sampling systems for composts are described in *7.1.1 Sampling frequency* and *7.1.2 Sample taking*.

7.1.1 Sampling frequency

Table 7-1: Frequency of the external quality inspection

Quantitative classes of composting plants	Number of external quality inspections (sampling)		Remarks
AUSTRIA – Compost Ordinance			
Volume of ready made compost y^{-1}			***Minimum quantity of sampled lot***
under 50 m³	1 single approval		5 m³
>50–300 m³	1 every 3 years		20 m³
>300–1,000 m³	1 every 2 years		50 m³
>1,000–2,000 m³	1 per year		100 m³
>2,000–4,000 m³	2 per year		150 m³
above 4,000 m³	Additional: 1 inspection for every 4,000 m³; 12 per year at maximum		150 m³
MSW Compost	Every 500 m³ ready made compost 1 sampling		200 m³
BELGIUM - VLACO			
Treatment capacity per year			
< 20,000 t	8 per year		
> 20,000 t	12 per year		
DENMARK – Statutory Order			
Treatment capacity per year			
All input capacities	4 per year or every 2,000 m³ compost		
FRANCE – Project proposal for a standard			
Treatment capacity per year			
All input capacities	Every lot minimum: 2 per year		
GERMANY – RAL GZ 251			
Treatment capacity per year	***Maintenance monitoring***		***1st year recognition phase***
≤ 8,000 t	4 per year		≤ 2,000 t 4 per year
> 8,000 t	one analysis per 2,000 t input		> 2,000 t 6 per year
	up to a maximum of 12 analyses per year		> 6,001 t 8 per year
			> 12,001 t 12 per year
LUXEMBURG – Interim Guideline and RAL GZ 251 (from DE)			
Treatment capacity per year	***Internal control***		***External control***
> 20,000 t *heavy metals*	12 per year		4 per year
PAH, PCB	4 per year		4 per year
PCDD/F	2 per year		4 per year
The NETHERLANDS – Internal Quality Control (IKB) of KIWA			
Treatment capacity per year	***Internal control***		***External KIWA control***
All input capacities	6* - 12 per year (1 per 4 weeks)		8 per year
SWEDEN			
Biological treatment per year	***Internal control***		***External control***
	Qualification year	***Maintenance monitoring***	
≤ 5,000 t	2 per year	1 per year	1 per year
> 5,000 t	4 per year	2 per year	1 per year
> 10,000 t	8 per year	4 per year	2 per year
SWITZERLAND – Instructions and recommendations of the FAC**			
Treatment capacity per year	***Internal control***		***Reduced sample numbers***
< 100 t	voluntary		If the results of individual investigations over a longer period do not exceed 50 % of the limit values, the sampling frequency
100 – 500 t	1 per year		
500 – 1,000 t	2 per year		

Quantitative classes of composting plants	Number of external quality inspections (sampling)	Remarks
> 1,000 t	4 per year	values, the sampling frequency can be reduced in agreement with the FAC
UNITED KINGDOM – Composting Association guideline		
Feedstock input per year	***Maintenance monitoring***	***Qualification phase***
< 5,000 t	2 per year	2 per year
5,000 – 10,000 t	2 per year	2 per year
10,000 – 20,000 t	2 per year	3 per year
20,000 – 40,000 t	2 per year	4 per year
> 40,000 t	3 per year	5 per year
Working Document, 2nd Draft " Biological treatment of biowaste"		
Treatment capacity per year	***Internal control***	***Reduced sample numbers***
> 500 – 1,000t	2 per year	When within a period of 2 years results are constantly below 75 % of threshold values the competent authority may on a case-by-case basis allow a reduction of the sampling frequency
1,000 – 10,000 t	At intervals of at least every 1,000 t treated biowaste produced or every 3 months, whichever comes first	
> 10,000 t	12 per year	

* Hotsma (2002); ** Swiss Research Institute for Agro-Chemistry

7.1.2 *Sample taking*

The sampling of materials with low homogeneity is crucial in terms of reducing the total error as related to the analytical results. Special care must be exercised at the individual steps of the entire sampling procedure. Some main principles are summarised from EN 12579, as well as the Austrian, German and Swiss guidelines:

Basic requirements for the taking of representative samples

- The sample portion must be in the same condition with regard to the preparation (sieving, removal of harmful substances etc.) as the associated compost batch, which is intended to be marketed.
- The final, combined sample must represent the whole of the material of the sample portion. The sampling points must be designated at random.
- Compost monitoring must be performed with batches, which are representative of the annual compost production. If the consignment does not appear to be from the same batch (lot) or consists of different materials (products), then the material(s) must be sampled separately.
- Sampling may be preferably undertaken during loading or discharge, if it is done in a way that the combined sample represents the entire lot.

Excursion: Time of sample taking – the effect of organic matter content and the discussed 30 % OM standardisation

The standardisation of heavy metal concentration to a defined OM content of the "resulting" compost is a reliable tool for a rough estimation of the suitability of potential source materials (see chapter 4.2.1). In addition, the first standards for composts in Germany (RAL-GZ 251) and Austria (ÖNORM S2200) introduced this method also for analytical values of PTEs when requiring a standardisation of to 30 % OM d.m.. Due to the fact that mineral components accumulate during continuous decomposition of organic materials, the aim was to guarantee a fair comparison between composts with lower (< 30 %)

and higher (> 30 %) organic matter content. It was also argued that without standardisation to a specific level of organic matter, producers might be encouraged to analyse the composts at early rotting stages in order to meet the limit values if any risk is in sight for exceeding them. Actually the mean OM content of BWC shows higher values. Reinhold (1998) reports that statistical evaluation of over 6400 biowaste composts in Germany shows a median for OM of 36.6 % d.m.. This is in line with the Austrian figure of 35.3 % d.m. (n=540; Peyr, 2000).

It is essential to look at the actual organic matter content of the material at the time of sampling. By this means, for example, the concentration factor of heavy metals in *"composts"* with 60 % OM would be 1.75 if calculated on the basis of 30 % OM, if related to 20 % OM the concentration factor would be 2. It is evident that, when dealing with cut-off values for compost products, the concentration ratio used may cause market problems if the thresholds are set close to the median or 75th percentile level of the compost type concerned. For instance, compost having a Zn concentration of 180 mg kg^{-1} at its original 40% OM level would exceed the threshold value of 200 mg kg^{-1} d.m.if calculated at an OM content of 30 % making it unusable as soil amendment in organic farming (Annex IIA EC Reg. 2092/91/EC). (Table 7-2)

Table 7-2: Relative and absolute increase of concentration values if the values are standardised on 30 % OM basis at different actual levels of OM in compost

		Element in mg kg^{-1} on original OM basis [mg kg^{-1} d.m.]					
		0.5	20	40	80	160	240
Original OM *[% d.m.]*	**Relative increase**	***Element in mg kg^{-1} on 30 % OM Basis [mg kg^{-1} d.m.]***					
35	8 %	0.54	22	43	86	172	258
40	17 %	0.58	23	47	93	187	280
45	27 %	0.64	25	51	102	204	305
50	40 %	0.70	28	56	112	224	252
55	56 %	0.78	31	62	124	249	280
60	75 %	0.88	35	70	140	280	315

Fig. 7-1 shows the continuous mathematical accumulation that occurs with progressing mineralisation.

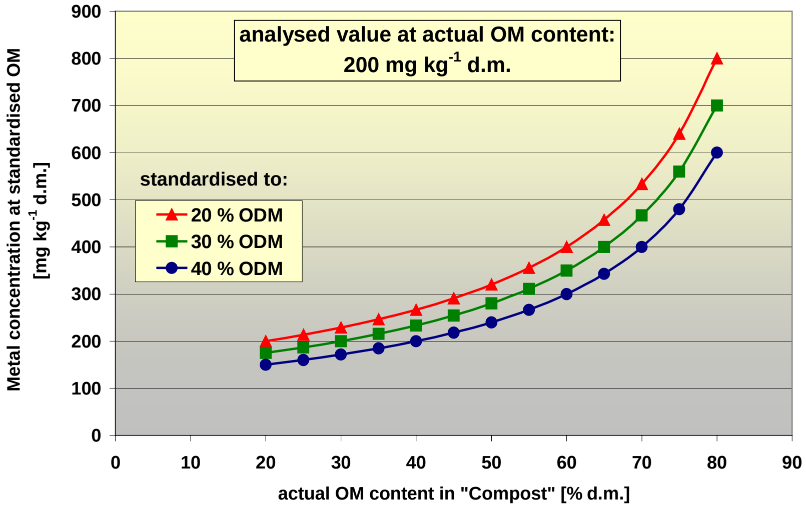


Fig. 7-1: Mathematical accumulation of metal concentration related to a standard organic matter content (20, 30 and 40 % OM d.m.) with varying actual OM levels and metal concentrations of the compost

In contrast to these calculations, Reinhold (1998) draws the conclusion that there is no significant coherence between *organic matter* and *heavy metal* concentration based on the statistical evaluation of the German compost data collected between 1991 and 1997 . Starting from the minimum OM content of 20 % (RAL-GZ251) an increase of 10 % OM leads to a rise of heavy metals by 0.47 % relative to the orientation values. 8 % of the 262 composting plants showed a positive and 13 % a negative correlation between OM and heavy metal levels. The evaluation of 376 between 1998 and 2002 gave a positive correlation between organic matter and the heavy metal concentration in nearly 15 % of the facilities. The author concludes that due to elevated nutrient level of composts rich in OM in general application, rates would be less and the obligatory standardisation to 30 % OM would discriminate those composts.

In addition, the of sample taking should be carried out at the latest possible time before the compost is put on the market or used in order to prevent major deviations due to further biological decomposition at the time the product reaches its final destination.

On European level, due to the still considerable variety of compost definitions it would be advisable to consider a standardisation of compost PTE limits at a defined organic matter content. This would counteract a market distortion with poorly rottet organic materials and allow for a better comparability of compost products or even other organic fertilsers.

Sampling Equipment and Sample Containers

- These have to be
  - clean
  - made of non-contaminating material (carbon steel or another unalloyed type of steel); materials like zinc coated steel, copper or stainless steel are not suitable.
- Sampling drills can only be used in case of favourable properties of the compost (not too dry, not too bulky).
- Drills have to be suitable for the material to be tested:
  - Inner diameter minimum 2.5-times the maximum particle size of the compost (CH)
  - Inner diameter minimum 10 cm and sufficiently wide jaws (e.g. single Edelmann auger, sand type, ∅ 10 cm of the firm Eijkelkamp).
  - Extracted Material should have 1 m minimum length

Beside the detailed description of taking and preparing of incremental samples, combined samples and final or laboratory samples respectively, a *comprehensive documentation* of the sampling is an important tool for the traceability of analytical results. Therefore sampling records where all single steps including a drawing of the location of sampled batch and distribution of incremental samples is an obligatory ingredient of standards.

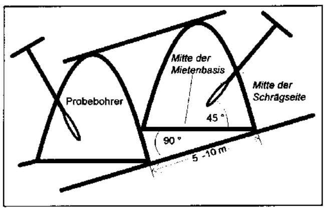
This is of special importance if investigations are carried out via regular random sampling of single lots, which have to represent comparable compost materials within a defined time or throughput related frame.

Table 7-3 outlines the key requirements of sampling procedures of *EN 12579 'Soil Improver and Growing Media – Sampling'*, as well as the *Austrian Compost Ordinance*, the German *RAL GZ 215, the* Swiss *'Instructions and Requirements Compost'* and The Netherlands.

Table 7-3: Schemes for sample taking for composts

<table>
<tr><th colspan="2"><i>Limitations of the size of a sample portion</i></th></tr>
<tr><td>EN 12579</td><td>A sampled portion shall be not more than 5 000 m^3 (bulk) or 10 000 packages (packaged material) of the same material from the same consignment.</td></tr>
<tr><td>Netherlands</td><td>A sampled portion shall be not more than 5 000 m^3</td></tr>
<tr><th colspan="2"><i>Number of final samples</i></th></tr>
<tr><td>EN 12579</td><td>One portion each for the supplier and buyer (receiver) or enforcement officer; one portion for an independent tester if a dispute on the analysis arises.</td></tr>
<tr><td>Netherlands</td><td>Nine individual random incremental samples constitute the 9 final (laboratory) samples, sent to the laboratory.</td></tr>
<tr><th colspan="2"><i>Number of sampling points or incremental samples</i></th></tr>
<tr><td>EN 12579</td><td>One incremental sample from each sampling point.
$n_{sp} = 0{,}5(V^{1/2})$ rounded up to the nearest whole number
where
V is the nominal quantity of the sampled portion in cubic metres
with a minimum $n_{sp} = 12$ and
a maximum $n_{sp} = 30$.</td></tr>
<tr><td><i>Austria</i></td><td>With shovel: Depending on the total volume of the sample portion
<table>
<tr><td>m³ of compost batch investigated</td><td><100</td><td>≥100</td><td>≥200</td><td>≥400</td><td>≥800</td></tr>
<tr><td>Minimum number of sampling points</td><td>4</td><td>5</td><td>6</td><td>7</td><td>8</td></tr>
</table>
A larger number of sampling points is permitted.
With drill: no requirements, depends on the total cubature of the sample portion</td></tr>
<tr><td>Germany</td><td>From heaps or stocks with shovel:
<table>
<tr><th></th><th>≤ 500 m^3</th><th>> 500 m^3</th><th>Quantity / sample point</th></tr>
<tr><td>With shovel</td><td>≥ 2</td><td>≥ 4</td><td>30 – 40 l</td></tr>
<tr><td>With drill</td><td>≥ 10</td><td>≥ 15</td><td>≥ 6 l</td></tr>
<tr><td>During loading</td><td colspan="3">≥ 10 times from the outlet of the conveyer evenly distributed over the entire lot</td></tr>
<tr><td></td><th colspan="3">Number of single packages</th></tr>
<tr><td>Packaged material
[≤ 2l or 2 kg/pack.]</td><td>n ≤ 4</td><td>n = 5 - 16</td><td>n > 400</td></tr>
<tr><td>Nr of sampled pack.</td><td>all</td><td>$\sqrt{n}$</td><td>≥ 20</td></tr>
</table>
</td></tr>
</table>

<table>
<tr><td>Italy (Regione Piemonte*)</td><td colspan="4">Seven incremental samples per 200 m^3</td></tr>
<tr><td>Netherlands</td><td colspan="4">Nine individual random incremental samples constitute the 9 final (laboratory) samples, sent to the laboratory.</td></tr>
<tr><td rowspan="8">Switzerland</td><td>From heaps or stocks:</td><td>< 300 m^3</td><td>> 300 m^3</td><td>< 15 m^3 / 30 m^3</td></tr>
<tr><td></td><td>1 per 15 m^3</td><td>15 total</td><td rowspan="2">3</td></tr>
<tr><td>Sieved compost</td><td>1 per 30 m^3</td><td>10 total</td></tr>
<tr><td rowspan="2">During loading or discharge</td><td rowspan="2">from sieve or reactor output</td><td colspan="2">when loading</td></tr>
<tr><td>from heap</td><td>from stored compost</td></tr>
<tr><td>Unsieved compost</td><td>1 per 15 m^3</td><td>1 per 15 m^3</td><td>1 per 15 m^3</td></tr>
<tr><td>Sieved compost</td><td>1 per 15 m^3</td><td>---</td><td>1 per 30 m^3</td></tr>
<tr><td colspan="4"></td></tr>
<tr><td colspan="5"><i>Distribution of sampling points</i></td></tr>
<tr><td>EN 12579</td><td colspan="4">Bulk: Visually divide the sampled portion into the same number of equal portions as the number of sampling points;
Packaged: Each sampling point shall be in a different randomly selected package.</td></tr>
<tr><td>Austria</td><td colspan="4">Distributed evenly over the total volume of the sample the making of the cuts in the sample must be adapted according to the profile of the pile) (slope, cone, trapezium, sheet).</td></tr>
<tr><td>Germany</td><td colspan="4">Distributed randomly over the lot</td></tr>
<tr><td colspan="5"><i>Volume / mass of incremental samples</i></td></tr>
<tr><td>EN 12579</td><td colspan="4">at least 0.5 l</td></tr>
<tr><td>Austria</td><td colspan="4">With shovel: at least 20 l disregarding at least the top 10 cm
With drill: no requirements; quantity of final sample → <i>at least 30 l.</i></td></tr>
<tr><td>Germany</td><td colspan="4">Each sample has to be of approx. equal weight or volume
particle size < 20 mm → 2 l
particle size > 20 mm → 3 l</td></tr>
<tr><td>Italy (Regione Piemonte*)</td><td colspan="4">1.5 – 2 kg</td></tr>
<tr><td>Netherlands</td><td colspan="4">at least 1 kg</td></tr>
<tr><td>Switzerland</td><td colspan="4">at least 3 l; all samples have to have the same size.</td></tr>
<tr><td colspan="5"><i>Taking the samples</i></td></tr>
<tr><td>EN 12579</td><td colspan="4">Bulk: Incremental samples shall be taken from throughout the depth of the material, ignoring material nearer than 5 cm to any surface;
Packaged: Randomly take incremental samples from throughout the package.</td></tr>
<tr><td>Austria</td><td colspan="4">With shovel: extracted from places distributed over the cut surface disregarding at least the top 10 cm;
To create a parallel sample, the appropriate sample quantity for the individual sample can be extracted per cut in each case from the opposite cut surface.</td></tr>
<tr><td>Netherlands</td><td colspan="4">Method 1: From 9 evenly distributed stratums after making a bore hole in advance the samples are taken from approximately 1 m depth.
Method 2: Two cross-sections of appr. 2 m are made with a loader. From each cross-section 5 and 4 increments respectively of 1 kg each are taken and constitute the 9 final samples.</td></tr>
</table>

Switzerland	Uniform spacing of 5 - 10 m along the pile By opening of the pile cross-section an extraction of 5-6 samples uniformly distributed over the entire cross-section is to be performed by shovel. 
Preparing the final sample	
EN 12579	Combine the incremental samples to form a combined sample. Reduce the combined sample by coning or quartering or with an apparatus for sample division. Prepare the laboratory sample according to EN 13040.
Austria	Reduce the combined sample by coning or quartering or with an apparatus for sample division.
Netherlands	The 9 random incremental samples constitute 9 separate final (laboratory) samples, sent to the laboratory.
Germany	Samples are mixed and transferred to the plastic foil. Clods are to be crushed separately and subsequently reintroduced to the sample. The foil is to be lifted at the corners in such a manner that the compost will roll to the opposite end and is mixed several times until the sample appears to be homogenous. Spread on the foil and 8 sectors are marked by drawing diagonals. Then 2 opposite sectors are removed. This process is repeated until the final sample required for shipping of approx. 20 litres remains.
Quantity of final sample	
EN 12579	5 l → chemical analysis 5 l → physical analysis 5 l → bio assay 15 l → bulk density according to EN 12580
Austria	15 l → chemical, physical analysis, bio assay 1 l → microbiological test (pathogens)
Germany	20 l → all parameters
Italy (Regione Piemonte*)	3 kg
Netherlands	1kg → all parameters
Switzerland	1 l → chemical, physical analysis 4 l → bio assay, impurities

* DiVaPRA Universitá di Torino, IPLA,ARPA Piemonte, 1998. Metodi di analisi dei compost. Regione Piemonte - Assessorato all'Ambiente, Torino. Cfr Regione Piemonte, Metodi di analisi dei compost, 1998

7.2 Systems of tolerances and admissible deviations

Limits are fixed at two different kinds of levels in the various countries. There are cut-off limits, which are absolute levels. Composts, which show contents above these levels, may not be declared as compost or as the defined compost quality to be put on the market or to be used in a certain area. This is valid for any batch produced without any exception.

In addition, some regulations have established a second kind of limit – a system with tolerances or admissible deviations. These systems refer to the specific structure and low homogeneity of compost batches or the regular (not batch wise required) sampling frequency for composts.

As justification the experience of inter-laboratory trials or repeated analyses from single compost batches are cited (see 7.3).

The following systems for admissible tolerances can be distinguished:

Table 7-4: Systems of tolerated deviations of limit values for composts

<table>
<tr><th colspan="6">[I] Product / batch / sample related tolerance
<i>[independent of analytical tolerances of repeated analyses from one laboratory sample]</i></th></tr>
<tr><th>Sample taken on behalf of:</th><th>Tolerance / admissible deviation refer to</th><th>Compost type</th><th>Tolerance factor</th><th>Specification of system</th><th>Country</th></tr>
<tr><td rowspan="7">Producer</td><td rowspan="3">Any sample</td><td rowspan="3">any</td><td>+ 43 %</td><td>Control sample may exceed any limit value for any batch analysed</td><td>NL</td></tr>
<tr><td>+100 %</td><td>To be authorised on a case by case basis by the responsible body for a period of max. 6 months “StoV,” from 09.06.86</td><td rowspan="2">CH</td></tr>
<tr><td>+50 %</td><td>Draft revision of the “StoV” from 01.05.02; See above</td></tr>
<tr><td rowspan="2">Single batch among a series of batches</td><td>any</td><td>+ 25 %</td><td>Value may not exceed limits in the sliding average of the last four tests … and if none of the results of an analysis exceed the limit value by more than 25%.</td><td>DE</td></tr>
<tr><td>any</td><td>+ 50 %</td><td>75% of 5 samples must be below the limit value; one result may exceed the limit value by max. 50%.</td><td>DK*</td></tr>
<tr><td>4 independent samples of 1 batch</td><td>MSWC
MBTC</td><td>+ 30 %</td><td>3 of 4 obligatory independent samples of 1 batch have to meet the limit values. 1 sample may exceed the limit by 30 %</td><td>AT</td></tr>
<tr><td>Any sample</td><td>any</td><td>+ 20 %</td><td><table><tr><th>Samples taken within 12 months</th><th>Max. No. Of samples exceeding any limit</th></tr><tr><td>2 or 4</td><td>1</td></tr><tr><td>12</td><td>3</td></tr></table></td><td>DG Env. WD
2nd draft</td></tr>
<tr><td>Control authority</td><td>Sample from a single declared batch</td><td>any</td><td>+ 50 %
resp.
+ 30 %</td><td>Analytical value of the controlled batch may exceed the limit value by max. 50 % (Cd, Cr, cu, Hg, Ni, Pb) resp. 30 % (Zn, organic pollutants)</td><td>AT</td></tr>
</table>

<table>
<tr><th colspan="3">[II] Limits for repeatability
<i>[tolerances /max. deviations for repeated measurements from one or more samples</i>
<i>for the determination of the concentration of PTEs or organic pollutants in ONE compost batch]</i></th></tr>
<tr><th>Tolerance / admissible deviation refer to</th><th>Specification of system</th><th>Country</th></tr>
<tr><td>Independent random samples of one single batch <i>[any heavy metals and organic pollutants]</i></td><td>• Maximum 6 repeated independent samples from <i>one</i> batch
• Exclusion of max. 2 outliers (extreme values)
• Max. deviation of individual result from mean value: + 30 %</td><td>AT</td></tr>
<tr><td>Repeatability limit for repeated measurements of one laboratory sample</td><td><table>
<tr><th colspan="7">Repeatability limit [% of mean value; at p=95%]</th></tr>
<tr><th>Cd</th><th>Cr</th><th>Cu</th><th>Hg</th><th>Ni</th><th>Pb</th><th>Zn</th></tr>
<tr><td>28 %</td><td>22.4 %</td><td>22.4 %</td><td>36.4 %</td><td>19.6 %</td><td>28 %</td><td>28 %</td></tr>
</table></td><td>DE</td></tr>
<tr><td>Repeatability limit for repeated measurements of one laboratory sample</td><td><table>
<tr><th colspan="7">Repeatability limit [% of mean value; at p=95%]</th></tr>
<tr><th>Cd</th><th>Cr</th><th>Cu</th><th>Hg</th><th>Ni</th><th>Pb</th><th>Zn</th></tr>
<tr><td>19.75 %</td><td>21.55 %</td><td>11.61 %</td><td>---</td><td>14.51 %</td><td>17.94 %</td><td>8.92 %</td></tr>
</table></td><td>WD
2nd draft
EN 13650</td></tr>
<tr><td>Repeatability for repeated measurements of one laboratory sample for PAH</td><td><table>
<tr><th></th><th colspan="3">Repeatability [% of mean value] for PAH</th></tr>
<tr><th></th><th>sample 1</th><th>sample 2</th><th>sample 3</th></tr>
<tr><td>Mean value [mg kg^{-1}]</td><td>13</td><td>62</td><td>2 090</td></tr>
<tr><td>Repeatability [%]</td><td>18.3 %</td><td>11.9 %</td><td>10.7 %</td></tr>
</table></td><td>WD
2nd draft
& ISO
13877</td></tr>
</table>

* In DK tests for chromium, zinc and copper can be omitted where tests are related to concentrations per kilogram of phosphorous. The parameters cadmium, mercury, lead and nickel may be omitted if the waste producer documents that these metals are either not present or only in negligible quantities in the waste product.

7.3 Results from inter-laboratory trials (ILTs) and ring tests (RTs)

Inter-laboratory trials or ring tests are conducted in the framework of

[A] Standardisation work of analytical methods
[B] Admission procedures of laboratories as acknowledged partners for private certification and quality assurance systems (QAS)
[C] Registration procedures for laboratories to be acknowledged in the framework of legal requirements for analytical investigations in order to assess obligatory quality parameters of materials (compost, soil, sludge, waste etc.)

In the first case (A) the aim is to validate the precision and statistical accuracy of an analytical procedure of specified test samples to be implemented in a national or international standard for test methods. Under best laboratory practice conditions samples are taken, subdivided and distributed to a sufficient number of labs (10 - 20) under standardised conditions. Labs get either a number of parallel samples (mostly 2 to 4) or are obliged to prepare a corresponding number of sub-samples for repeated measurements. The aim is to evaluate the standard deviation and *repeatability* (s_r) for a certain type of material as a basis for the determination of repeatability limits at a probability of 95 %. In addition information may be provided about the *reproducibility* (s_R) for a certain parameter. By comparing the mean values of the participating labs with the total mean value, limits for s_R may be set with the same procedure as for s_r. All calculations are made after elimination of outliers following standard statistical methods.

(B) and (C) aim at the performance evaluation of labs in the framework of the acknowledgement, in our case, of compost analyses and certification following legal or internal standards of QAS. In this case, Statistical parameters (*standard deviation [STD], coefficient of variation [CV], empirically set limits such as the 2-fold STD)* are evaluated as criterion for acknowledgment or exclusion of a lab.

The results of ILTs represent a fundamental basis for the assessment of possible systems of limit values, the reliable differentiation of 2 or more quality (PTE) classes and for the establishment of necessary tolerances.

The following summarises key results of ILTs performed with composts.

7.3.1 ILT of the Federal Quality Association Compost in Germany 1993, 1995 and 1999 (Biedlingmaier et al., 1996; Kreft, 1997; Biedlingmaier & Vorreiter, 1999)

Table 7-5: Key results of the 3 German ILTs 1993, 1995 and 1999

General figures			
	1993	1995	1999
Number of labs: *German labs* *Austrian labs*	95 --	127 15	115 19
Number of delivered sub-samples	4 fresh	4 fresh; 1 dry & grinded	4 fresh; 1 dry & grinded
Number of repeated measurements per sub-sample	3	3	3
Validation Parameter	• All calculations after elimination of outliers • Inter-lab variance (variance of lab means) = [CV(MEAN$_{lab}$)] • Intra-lab variance (means of individual lab variance) = [MEAN (CV$_{lab}$)] • Accepted tolerance of individual lab result: ± 2 x STD of total mean value [MEAN$_{tot}$]		
Results for heavy metals (fresh sample) for German labs			
	1993	1995	1999
Cd			
CV(MEAN$_{lab}$)	0.33	0.26	0.15
MEAN (CV$_{lab}$)	0.10	0.08	0.04
CV/MEAN	*3.3*	*3.3*	*3.47*
Tolerance for registr. [2 x STD]	±0.466	±0.52	±0.30
Cr			
CV(MEAN$_{lab}$)	0.38	0.26	0.31
MEAN (CV$_{lab}$)	0.08	0.06	0.04
CV/MEAN	*4.75*	*4.3*	*8.66*
Tolerance for registr. [2 x STD]	±0.76	±0.52	±0.62

	1993	1995	1999
Cu			
CV(MEAN$_{lab}$)	0.15	0.14	0.13
MEAN (CV$_{lab}$)	0.17	0.08	0.03
CV/MEAN	*0.88*	*1.8*	*3.64*
Tolerance for registr. [2 x STD]	±0.30	±0.28	±0.26
Hg			
CV(MEAN$_{lab}$)	0.34	0.30	0.27
MEAN (CV$_{lab}$)	0.19	0.13	0.06
CV/MEAN	*1.79*	*2.3*	*4.47*
Tolerance for registr. [2 x STD]	±0.68	±0.60	±0.54
Ni			
CV(MEAN$_{lab}$)	0.20	0.12	0.19
MEAN (CV$_{lab}$)	0.07	0.05	0.05
CV/MEAN	*2.86*	*2.4*	*4.16*
Tolerance for registr. [2 x STD]	±0.40	±0.24	±0.38
Pb			
CV(MEAN$_{lab}$)	0.20	0.14	0.28
MEAN (CV$_{lab}$)	0.10	0.06	0.04
CV/MEAN	*2.0*	*2.3*	*6.73*
Tolerance for registr. [2 x STD]	±0.40	±0.28	±0.56
Zn			
CV(MEAN$_{lab}$)	0.13	0.10	0.11
MEAN (CV$_{lab}$)	0.10	0.05	0.02
CV/MEAN	*1.3*	*2.0*	*4.59*
Tolerance for registr. [2 x STD%]	±0.26	±0.20	±0.22

7.3.2 Inter-laboratory trial (ILT) of the Austrian Compost quality Society (KGVÖ) 2001 (Binner & Lechner, 2002)

Table 7-6: Key results of the Austrian KGVÖ ILT 2001

General figures	
Number of labs:	14
Number of delivered sub-samples	1 fresh sample; 1 dry and grinded sample
Number of repeated measurements per sub-sample	Up to 3
Validation Parameters	• Calculations after elimination of outliers • Inter-lab variance (variance of lab means) = [CV(MEAN_{lab})] • Accepted tolerance of individual lab result: ± 2 x STD of total mean value [MEAN_{tot}]

CV(MEAN_{lab}) Results for heavy metals

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Fresh sample (KGVÖ, 2001)	0.15	0.15	0.10	0.17	0.22	0.14	0.11
Dry sample (KGVÖ, 2001)	0.29	0.17	0.11	0.40	0.17	0.12	0.12

7.3.3 Ringtest of the Austrian Association of Agricultural Research Institutions (ALVA) 2001 (ALVA, 2001)

Table 7-7: Key results of the ALVA compost Ringtest 2001

General figures	
Number of labs:	19-21
Number of delivered sub-samples	1 fresh sample <10 mm; 1 dry and grinded sample <0.5 mm
Number of repeated measurements per sample	4
Validation Parameters	• All calculations after elimination of outliers • Inter-lab variance (variance of lab means) = [CV(MEAN_{lab})] • Intra-lab variance (means of individual lab variance) = [MEAN (CV_{lab})] • Accepted tolerance of individual lab result: ± 2 x STD of total mean value [MEAN_{tot}]

CV(MEAN_{lab}) Results for heavy metals

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Fresh sample (ALVA, 2001)	0.22	0.12	0.09	0.41	0.22	0.27	0.12
Dry sample (ALVA, 2001)	0.25	0.19	0.09	0.36	0.22	0.28	0.11

7.3.4 Comparison of the evaluated ILT

The essential statistical figure for the evaluation of the variability of the compost sample and the reliability to receive a *true value* from a laboratory is the inter-lab coefficient of variability or *reproducibility* of each individual parameter in the fresh sample.

The relation between [CV(MEAN$_{lab}$)] and [MEAN (CV$_{lab}$)] shows a consistent tendency:

1.) The inter-lab deviation (reproducibility) is always greater than the intra-lab deviation (repeatability). The factors lie between 1.8 and 8.66 for original fresh samples and 2.0 and 5.15 for dried and ground samples respectively (Fig. 7-2; Table 7-8; Fig. 7-3). This relatively constant relation for both fresh and dried sample shows not only that sampling and pre-treatment lead to sufficient homogenous sub-samples but that the inter-lab variation plays the essential role in evaluating a single analytical result.

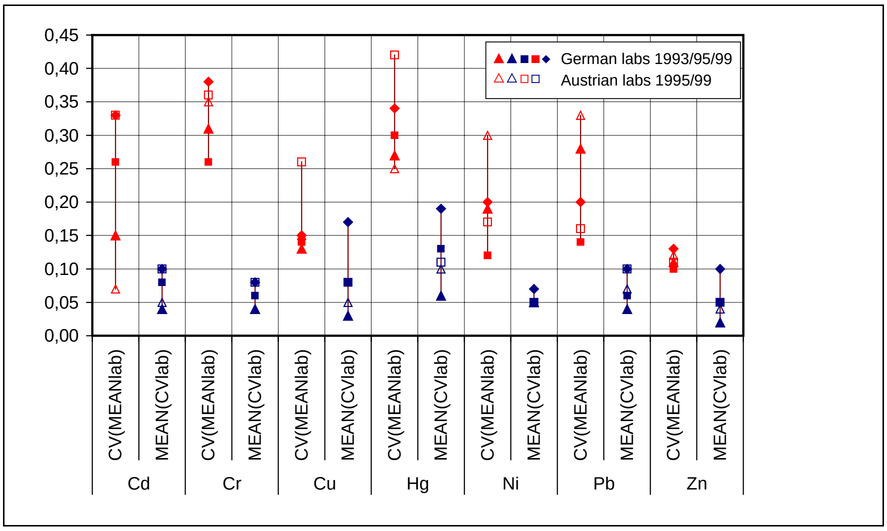


Fig. 7-2: Variance of lab-mean values [CV(MEAN$_{lab}$)] compared to Means of repeated measurements within the labs [MEAN (CV$_{lab}$)]; ILT Germany 1993/95/99 and Austria 1995/99

Table 7-8: Relation between reproducibility [CV(MEAN$_{lab}$)] and repeatability of heavy metal analyses in German ring tests 1995 and 1999 ([CV(MEAN$_{lab}$)] / [MEAN (CV$_{lab}$)])

[f] … fresh samples; [d] … dry samples

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
1995 [f]	3.30	4.30	1.80	2.30	2.40	2.30	2.00
1995 [d]	4.30	3.80	2.00	4.20	3.50	3.30	2.70
1999 [f]	3.47	8.66	3.64	4.47	4.16	6.73	4.59
1999 [d]	3.29	4.10	3.17	5.15	2.67	3.78	3.64

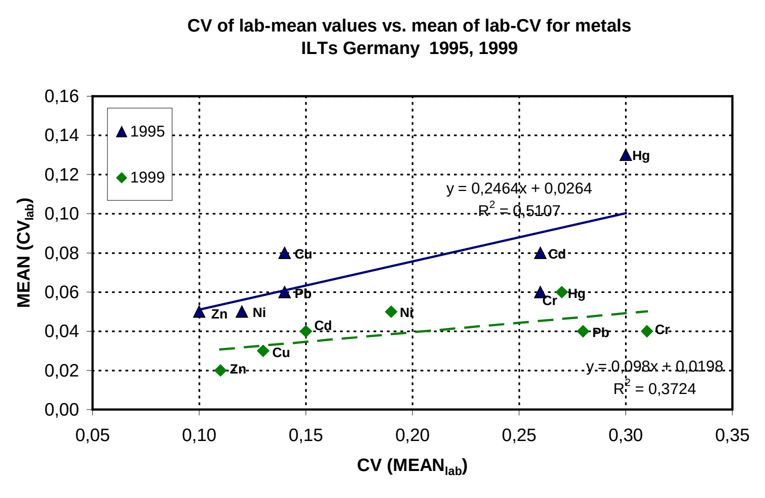


Fig. 7-3: Variance of lab-mean values [CV($MEAN_{lab}$)] of fresh samples vs. mean values of intra lab variance [MEAN (CV_{lab})] for heavy metals; ILT Germany 1995/99

2.) In addition it becomes evident that coefficients for intra-lab repeatability as well as inter-lab reproducibility in general increase at low concentration levels (Fig. 7-4; Fig. 7-5)

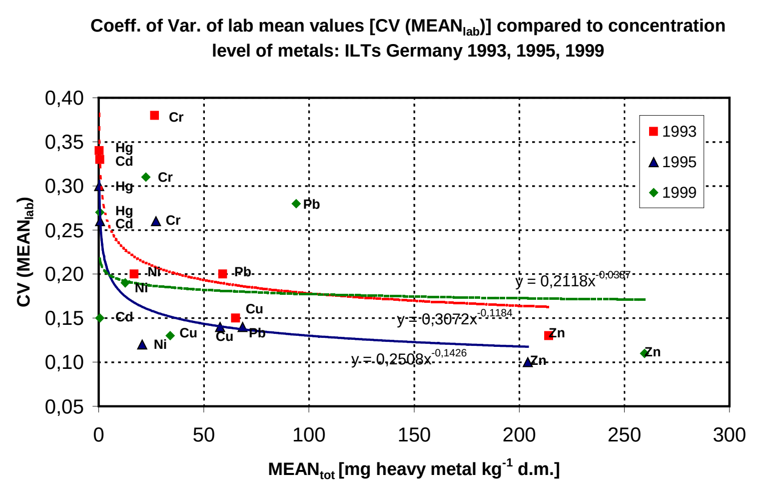


Fig. 7-4: Variance of lab-mean values [CV($MEAN_{lab}$)] of fresh samples vs. concentration levels of heavy metals; ILT Germany 1993/95/99

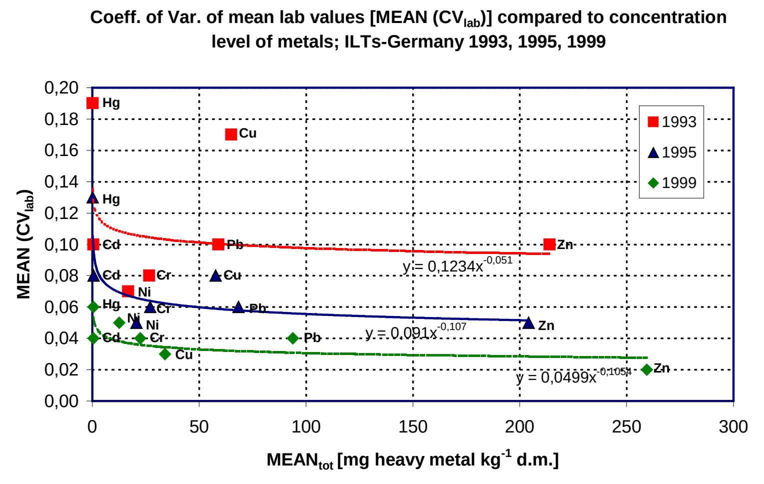


Fig. 7-5: Mean values of lab variance [MEAN(CV$_{lab}$)] of fresh samples vs. concentration levels of heavy metals; ILT Germany 1993/95/99

3.) Most metals variation coefficients are higher in fresh original samples than in dried samples (Fig. 7-6). In the ILTs of 1995 and 1999 the factor between fresh and dry samples ranges between 5.0 (Cr, Austrian labs 1999) and 1,0 (Hg, Austrian labs 1999). Exceptions are Cd, Hg and Ni in 1995, where the relation is reverse. Mean values of this relation for German and Austrian labs in 1995 and 1999 are shown in Table 7-9.

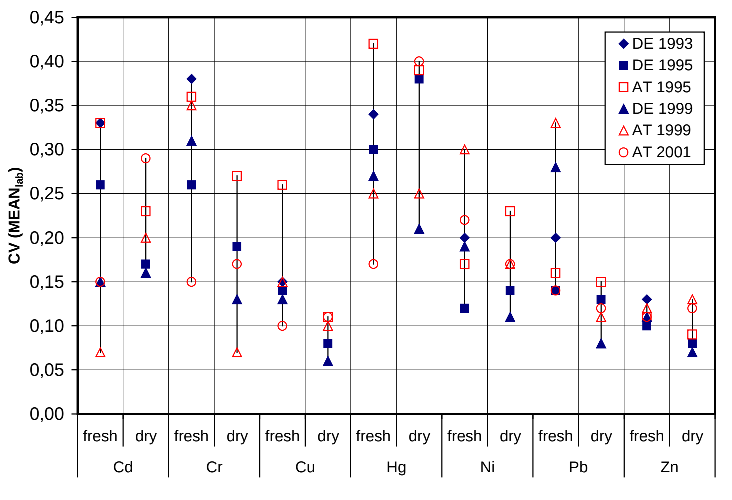


Fig. 7-6: Variance of lab-mean values [CV(MEAN$_{lab}$)] of fresh vs. dry and grinded samples; ILT Germany 1993/95/99 and Austria 1995/99/2001

Table 7-9: Relation between the relative standard deviation in fresh and dried samples; mean value of the results of the ILTs of 1995 and 1999 in German and Austrian labs

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
[CV(MEAN_{lab})] fresh sample/ [CV(MEAN_{lab})] dry sample	1.06	2.52	1.95	1.04	1.27	2.16	1.24

Table 7-10: Comparison of Coefficients of Variability CV(MEAN_{lab}) of lab mean values of the investigated ILTs.

CV(MEAN_{lab}) Results for heavy metals							
	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Germany, BGK 1993 [n=95]	0.33	0.38	0.15	0.34	0.2	0.2	0.13
Germany, BGK 1995 [n=127]	0.26	0.26	0.14	0.30	0.12	0.14	0.10
Austria, BGK 1995 [n=15]	0.33	0.36	0.26	0.42	0.17	0.16	0.11
Germany, BGK 1999 [n=115]	0.15	0.31	0.13	0.27	0.19	0.28	0.11
Austria, BGK 1999 [n=19]	0.07	0.35	0.15	0.25	0.30	0.33	0.12
Austria, KGVÖ 2001 [n=14]	0.15	0.15	0.10	0.17	0.22	0.14	0.11
Austria, ALVA 2001 [n=21]	0.22	0.12	0.09	0.41	0.22	0.27	0.12
Mean of all ILTs	***0.29***	***0.28***	***0.15***	***0.31***	***0.20***	***0.22***	***0.11***
Std Dev % (EN 13650) [n=20]*	1.19	0.73	0.26		0.74	0.42	0.29

* Reproducibility limit relative to conventional mean value after elimination of outliers; calculated at a probability of 95 % R = 2,8 s_R

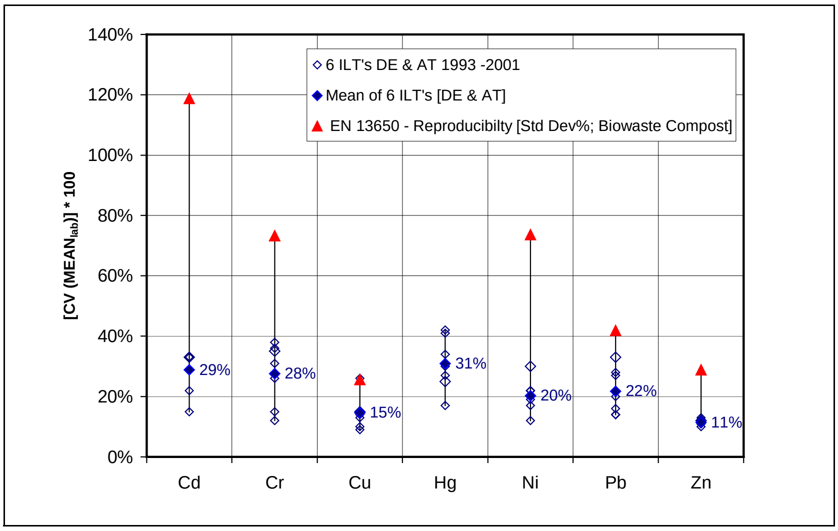


Fig. 7-7: CV of lab-mean values [CV(MEAN_{lab})] of all German and Austrian ILT's- 1993 - 2001 and ILT of CEN TC 223 [EN 13650]

The variability varies with the element concerned. Cu and Zn show a comparatively constant CV and the lowest CV in all ILTs.

Variability of compost analyses is a result of errors stemming from problems with material property (non-homogeneous batches and test material) and inaccuracies or errors made in the course of the entire analytical procedure (from sampling to transcription errors of figures). Bidlingmaier et al. (1996) summarise the errors occurring in the analytical procedure as follows (Table 7-11).

Table 7-11: Possible errors in compost analyses and their causes (Biedlingmaier et al., 1996; modified)

Measure/step (Responsible person/entity)	Influence	Error
Sampling (person taking the sample)	criteria of sample selection sampling technique Nr. of increments mixing; sample reduction procedure	sample not representative for the lot/batch insufficient homogenising systematic sampling effects (when preparing the laboratory sample)
Transport of sample (person taking the sample; shipping company)	duration temperature permeability of packaging material	degradation of organic matter metabolism of organic compounds
Storage of sample (Lab)	duration humidity temperature	degradation of organic matter metabolism of organic compounds
Pre-treatment of sample (Lab)	drying grinding mixing dividing, taking aliquot, representative test samples weighing in the test sample	change of sample (metabolism of organic compounds) insufficient homogenising errors in weighing improper reduction of laboratory sample in order to obtain the representative test sample substance losses by evaporation and adsorption
Analyses of sample (Lab)	dissolution extraction separation enrichment	insufficient homogenising substance losses wrong control test contamination with control test substance losses by evaporation and adsorption
Value determination (Lab)	determination	errors by means of: standard measurement, calibration, measurement reading
Documentation, transcription, and presentation of results (Lab)		Wrong calculation and conversion of data

7.4 Spatial and temporary variation of compost quality –repeated sampling – sampling frequency – forecast of expected quality range

As discussed in section 7.2 and 7.3 environmentally motivated limit values loose their significance, if it is not possible to assess the possible error or difference between the produced analytical result and the *true value*. From the economic point of view as well from the experience of 15 years of organic waste composting and quality control it seems not to make sense to require sampling and analysing of any single compost batch produced.

Effects of sampling and pre-treatment of samples contribute significantly to the statistical uncertainty of analytical results.

This raises the question how the quality control system must be designed so that a satisfactory guarantee can be furnished in the frame of accepted tolerances.

In this context the following questions have to be answered:

A.) What is the necessary sampling frequency for a defined compost type (comparable composition of raw materials) in order to be assured that compost lots produced between two successfully analysed ones meet the quality requirements set and thus regularly analysed ones would represent the entire compost production?

B.) What is the minimum number of analyses within a composting plant in order to satisfactorily predict the quality of individual compost lots or mean value of a continuous produced compost type?

C.) How must the set up of the sampling method be designed so that the analysed laboratory and subsequently the test sample are representative for the entire compost batch concerned?

7.4.1 How many samples are necessary for an accurate prediction of concentration ranges within a defined confidence interval?

In considering this question the *German Bundesgütegemeinschaft Kompost* evaluated the compost analyses conducted between 1991 and 1997 from all members (262 composting plants; 6505 sets of data) of the association (Reinhold, 1998).

The ability to forecast quality parameters, which classify the compost in the frame of legal requirements (limit values), is an essential tool for the set-up of a quality assurance system. These forecasts are needed for analyses of individual lots as well as mean values. The evaluation of mean values yields an assessment of the probability for the fulfilment of set quality requirements. Since single measurements as well as means stem from random samples, only the *expected range* for these random values can be estimated. It is clear that the expected range for single values and mean values respectively within a defined confidence interval can be narrowed down with an increasing number of individual data. The probability limits were fixed at 95 %. The statistical evaluation of the data gave the following minimum numbers of single values for estimating expected value ranges of individual composting plants:

- Assessment of single values -- 6 – 10 individual measurements are needed
- Assessment of mean values -- 10 – 15 measurements individual measurements are needed

The authors conclude that in average 10 individual measurements per composting plant is the minimum number for a verifiable prediction of a quality level that can be guaranteed.

7.4.2 The spatial variability of heavy metal concentrations in compost batches – effects on the sampling procedure

Breuer et al. (1997) analysed composts of 25 different biowaste and green waste composting plants in Baden-Württemberg considering the spatial and temporal variability of compost quality. A repeated sampling (20) of heavy metals and organic compounds in one windrow at a time resulted in standard deviations, which show a wide range (Table 7-12). Heavy metal contaminations on a high level go together with a high variability in the parallel sampling. This indicates that a contamination on a high level mostly has a specific location and is not allocated evenly over the whole windrow. Due to this non-homogeneous structure of composts *a repeated parallel sampling of the windrows with separated single analysis was highly recommended*. In this way one can avoid interpreting outliers caused by non-homogeneous compost heaps as representative of the whole windrow.

Table 7-12: Spatial variability of heavy metal contents of 20 independent samples as compared to one mixed sample (6 windrows in 3 composting plants); Breuer et al. (1997)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
		range of Coefficient of variation [%]						
BWC	4 windrows	11.8 - 93.4	6.5 - 13.4	6.2 - 18.4	17.3 - 20.4	4.8 - 11.8	8.2 - 17.9	3.3 - 7.4
GC	2 windrows	14.0 - 102	6.2 - 10.2	12.9 - 18.7	16.6 - 23.3	3.6 - 8.3	47.7 - 73.3	3.8 - 12.2
Mean value		29.1	11.1	17.5	17.7	9.6	19.8	6.4

In a study on compost quality from different origins and source materials, Zethner et al. (2000) took three independent samples from two biowaste and two green composts in order to evaluate the systematic errors responsible for the variability of the results obtained. The main focus was on the accuracy of the sampling (reduction from a pile with up to 2,000 m^3 to a representative final sample of about 15 litres and further to the laboratory sample). The evaluation of individual differences between the independent samples resulted in an unavoidable deviation from the mean of ± 30% (Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Zn), ± 40% (Pb) and ± 37.5% (AOX). Furthermore Pb exceeded this conventionally set limit in 3, Zn, Cr and Ni in 1 of 4 composts.

Lustenhouwer at al. (1991) concludes from a comprehensive investigation of the determination of heavy metals in compost which included sampling, sample preparation and analyses, that the number of increments taken from a compost lot is the most critical parameter for the verification of a *true value*. This was found to be independent of the size of the concerned lot as well as the compost type and nature of the element. From the viewpoint of precision it was found useless to take more than one test portion from a laboratory (final) sample. These findings led to the NL sampling procedure that requires a minimum of nine independent increments per sampling portion, which are sent to the laboratory. In the laboratory representative sub-samples are then combined for the test portion.

7.5 Analytical / Extraction methods

The most common analytical methods for trace elements are derived from ISO and EN standards. They were summarised in the workshop "Harmonization of sampling and analysis methods for heavy metals, organic pollutants and pathogens in soil and sludge" (Gomez et al., 2001).

Table 7-13 gives a comparative survey on extraction and analytical detection methods for trace elements used for compost matrices on European (CEN) and MSt level.

Analytical figures, which refer to given quality requirements, *must* rely on exactly the same methodology. This covers the type of extractant (strong acid) as well as the pre-treatment of the laboratory sample (drying and grinding with or without or combusting) and sample digestion procedure (time/temperature regime; microwave; material/acid ratio).

From personal communication with experts from the MSts it is reported that in many countries the laboratories still use individual methods. This is the reason why for some MSts no references are given in Table 7-13.

Because differing extractions methods are still used, the need for a consequent implementation of Europe-wide standardised methodology in the frame of a European compost regulation is required. Since the *aqua regia* method was adopted by the Technical Committee on Sewage sludge (CEN TC 308; EN 13346 “Characterisation of sludges – Determination of trace elements and phosphorus – Aqua regia extraction methods”) as well as “Soil improvers and growing media” /CEN/TC 223; EN 13650 “Soil improvers and growing media - Extraction of aqua regia soluble elements”) and the majority of experience with compost is based on this extraction method, it seems to be a reasonable way forward to refer to EN 13650 for further harmonisation.

Table 7-13: Analytical methods for heavy metals in composts used in European countries

Country	**Cd, Cr, Cu, Hg, Ni, Pb, Zn, (As)**	
	Extraction method	**Determination method**
EN 13650	Aqua regia; open digestion; no Hg 1 – 3 g sample (sample: acid = 3.5:1) 21 ml HCI +7 ml $HN0_3$ (=1.66:1) reaction time: 16 hours	ICP-AES [ISO 11885]; FAAS [ISO 11047]
Austria	EN 13650 reaction time: 12 hours	AAS / ICP-AES; DIN38406
Belgium	Heavy metals without Hg: Digestion with 7M Nitric acid total ignition residue of 2 g sample + 30 ml 7 M $HN0_3$ (1 + 1) + ca. 20 ml aqua dest. reaction time: 0.5 hours at 90 ± 10 °C Hg: conc. HNO_3 + conc. H_2SO_4 1 g sample + 4 ml HNO_3 conc. (65 %).+ 3 ml H_2SO_4 conc. (96 %) reaction time: 15 + 15 + 20 min.	AAS / ICP-AES Fluorescence spectrometry; flameless AAS / cold vapour
Denmark	DS 259. in autoclave 1 g sample plus 20 ml 7 M $HN0_3$ (1 + 1) reaction time: 0.5 hours at 120 °C (Compost and sludge on farmland)	AAS/ICP, Hg: cold vapour, As: zAAS, hydrid or ICPMS
Finland	Aqua regia; ISO/DIS 11466	AAS ; As: hydrid; ISO/DIS 11067
Germany	Aqua regia; open digestion; DIN38414 3 – 5 g sample (sample: acid = 3.5:1) 18 ml HCI +6 ml $HN0_3$ (‘pure’ acid ratio =1.66:1) reaction time: 12 hours	AAS / ICP; DIN38406
Italy	In Italy the situation is confused because the legislation does not refer to an analytical method. Consequently laboratories use different extraction methods. - EN 13650 aqua regia (21 ml HCl and 7 ml HNO_3; reaction time 16 hours) is a new official method (2002) and some laboratories a starting to use it. - UNI 10780 – AAS 10 ml HNO_3. Reaction time: 3-4 hours to boiling. Is not widely used - Official methods for fertilisers (law n. 748/84)	ICP-AES [ISO 11885]; FAAS [ISO 11047]

Country	Cd, Cr, Cu, Hg, Ni, Pb, Zn, (As)	
	Extraction method	Determination method
	Only for Cu and Zn; HCl 6M (10 ml for g fertiliser) + 50 ml H_2O; reaction time 30' boiling; Is not widely used - Analytical Methods for sewage sludge (IRSA-CNR, 1985). 10 ml HNO_3 conc. + 2 ml $HClO_4$. Reaction time: boiled until the solution is clear. The most used method.	
Luxembourg	EN 13650	ICP-AES [ISO 11885]; FAAS [ISO 11047]
Netherlands	Aqua regia; microwave; NEN5770 1.5 g sample (sample: acid = 3.1:1) 12 ml HCI +5 ml $HN0_3$ ('pure' acid ratio =1.66:1) reaction time: none	ICP; NEN6426
Switzerland	Heavy metals without Hg: Sample: total ignition residue 10 ml HCl (37 %) + <80 ml H_2O 20 minutes in boiling water quench	AAS
	Hg: 0.1 g sample, dried (40 °C) grinded 3 ml of HNO_3 (65%;) + CrO_3 (1.12 g in 100 ml HNO_3) 30 minutes at 120 °C	AAS / cold vapour
United Kingdom	EN 13650	ICP-AES [ISO 11885]; FAAS [ISO 11047]

7.6 Some conclusions from viewing sampling and analytical methods for compost

Sampling frequency

- Compost types regarding categories of input materials (green waste, biowaste, sewage sludge, other industrial waste) have to be investigated separately
- Random sampling instead of batch sampling is commonly agreed to be a suitable procedure for the quality approval of compost. The number of samples in most cases is raised step by step with increasing treatment capacities.
- One sample per 1,000 to 2,000 tonnes of treated biowaste with a maximum of 12 analyses per year is often considered to be a preferred method.
- In the recognition phase of quality assurance systems (QAS) higher sampling frequencies are required.
- A minimum of ten consecutive analyses at one composting facility has been recognised as a reliable forecaster of the range and mean values of PTE concentrations, which can be estimated with statistical methods. This figure can then also be a reference for a possible reduction of the number of normally required analyses. This case-by-case reduction of standard sampling frequency should only be granted by the competent authority or a recognised quality assurance organisation in the frame of a product certification scheme. By statistical evaluation on facility level expected upper values at a given confidence interval (e.g. 95 %) may give the margin for the identification of facility specific outliers. If an outlier was identified, a repeated sampling and if confirmed specific measures of quality management and a cause study should be applied.

Sample taking

- The *portion sampled has to be in the same condition* as the compost batches marketed under the same specification (e.g. mash size of screen, state of maturation/mineralisation).
- *Comprehensive records* of sample taking is a prerequisite for the reliable approval of the compost.
- Depending on the size of the sampling portion, the *number of increments* (sampling points) range from 4 to 30 from heaps or stocks. Based on a statistical evaluation of standard errors, a minimum number of 9 incremental samples independent of the batch size has been singled out as needed.
- The *sampling points must be distributed evenly* over the volume of the sampled portion (compost lot).
- The *minimum quantity of the incremental sample* depends on the sampling method applied: 1 to 3 litre (drill); up to 20 to l (shovel).
- *Preparing the final (laboratory) sample* by representatively reducing the incremental samples to a quantity of 5 – 20 l .

Repeated sampling and measurements – system of tolerances

In principle we have to distinguish between two types of tolerances for an analytical result:

1.) Tolerance which refers to the standard error of the analytical procedure in order to estimate the *true value*

2.) Tolerance which refers to an analytical result *relative to given quality classes* or limit values

The need and the range of tolerance of the latter is very much dependent on the level of limit values relative to the actually achievable compost quality of the concerned compost specification. If a limit value already takes into account roughly 99 % of the composts produced, almost any site specific and seasonal variability is already included. In this case the need for an individual sampling and analytical tolerance can be less than in the case of tighter thresholds.

Standard deviations or coefficients of variability and according tolerances have to be considered for

[A] Sampling *on behalf of the compost producer*

(i) parallel measurements of *different labs*

(ii) parallel measurements of *one batch (sampled portion)*

- a.) parallel measurements of samples gained from repeated sampling out of one *sampled portion* including the error of sample taking (e.g. preparing cross sections, distribution and number of sample points, mixing and reducing procedure for gaining the representative laboratory samples)
- b.) parallel measurements out of *one combined final (laboratory) sample* including the error of pre-treatment when preparing the representative test sample (mixing and reducing to the amount of necessary sub-sample, screening, grinding)
- c.) parallel measurements out of the readily prepared test sample including the analytical procedure

[B] Sampling *on behalf of the control authority*

tolerance that takes into account the variability of PTE concentrations within one compost type (compost specification and declaration) stemming from one composting facility (seasonal variability including the tolerances covered by [A] (1) & (2))

In case [A] *sampling on behalf of the compost producer* the experience of investigations on parallel sampling of sampling portions (Breuer et al., 1997, Zethner et al., 2000) lead to the conclusion that it may be advisable to carry out parallel, independent sampling from one sampling portion. This would regard the non-homogeneity of compost materials in a better way than a repeated measurement from one combined laboratory sample.

As a consequence, coefficients of variability of parallel sampling have to be considered. A coefficient of variability of ± 30 % would be a reliable maximum value for all elements based on the above-mentioned studies as well as the repeatability factors laid down in analytical standards for composts.

In this case the number of parallel samples has to be limited and the mean value of the repeated measurements has to meet a given threshold value.

Furthermore, a system of tolerance for an individual measurement result in a series of analysed compost batches might be considered. It is referred to the experience of Germany where one sample out of four(25 %) may exceed the limit value by 25 % whereas the sliding mean value of the most recent four samples must be below the limit value.

Tolerances in the case of external control [B]

When it comes to a control measurement on behalf of the competent authority, the entire variability of compost processing and as well as the sampling and analytical procedure has to be considered.

Therefore it is recommended to establish an additional tolerance relative to the set limit values regarding the recognition of a *control sample*. The addition of mean CVs of the discussed ILTs and the batch related CV out of one compost heap (Breuer et al., 1997) cover all relevant standard deviations in the case of a external control (see Table 7-14). It is therefore recommended that a tolerance of 50 % is accepted in the case of an external controlling procedure on the market place.

Table 7-14: Possible approach of a tolerance rule for controlling procedures for composts on behalf of the controlling authority

	Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
	coefficient of variation [CV %]						
Mean CV compost heap	29.1	11.1	17.5	17.7	9.6	19.8	6.4
Mean inter-lab CV	29	28	15	31	20	22	11
Total CV and possible orientation value for an admissible tolerance	**58.1**	**39.1**	**32.5**	**48.7**	**29.6**	**41.8**	**17.4**

8 CONCEPTS OF STATUTORY RULES FOR A SUSTAINABLE USE OF COMPOSTS

8.1 Basic concepts

In line with precautionary soil protection, it is necessary to develop a graduated action concept for the limitation of pollutant discharges that takes into account the type and behaviour of pollutants and provides for some flexibility. The aim of this limitation is to protect soil, particularly with regard to its natural and use specific soil functions.

Diffuse inputs, in contrast to local inputs, especially those in relation to agricultural activities are often a direct consequence of actions necessary to maintain soil functions or plant performance. Fertilisers or soil amendments such as rock phosphate, compost, sludge or animal manure, which are applied to maintain the soil organic matter and nutrient status entail also the addition of contaminants to the soil.

Fig. 8-1 gives a schematic overview of external impacts and those which originate in the agricultural actions and measures itself.

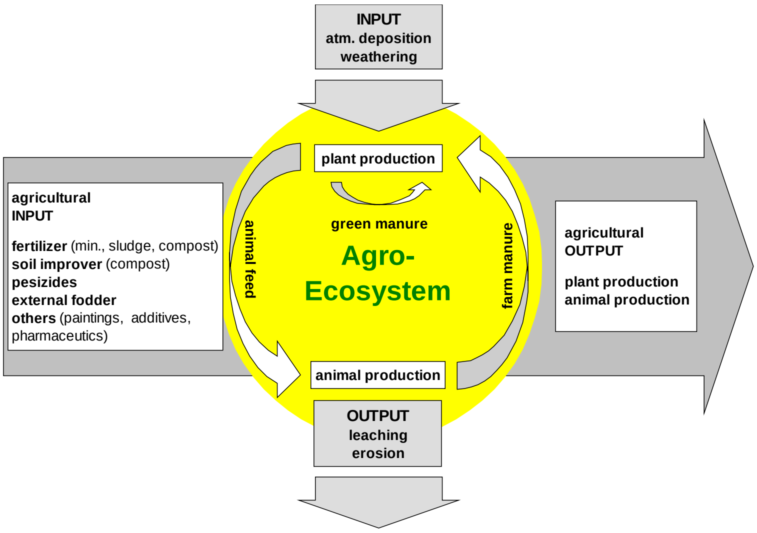


Fig. 8-1: Input and output pathways for contaminants into und out of the Agro-Ecosystem (from Wilcke & Döhler, 1995)

Since recycling strategies always cover both the reuse of resources (i.e. nutrients) as well as the re-import of potentially toxic elements to production processes (i.e. agricultural production), weighing the impact of both in the context of compost and soils is crucial. The beneficial effect of organic matter and nutrient recovery from reuse of organic waste products in agriculture should not counter environmental or specifically soil protection strategies. A reasonable starting point for a number of concepts which have been put forward for the use of secondary raw materials as fertilisers is an equitable estimation of the beneficial effects balanced against the potential risks caused by potential contaminants considering all additional inputs and outputs of pollutants into and from the soil system (Wilcke & Döhler, 1995; Hackenberg & Wegener, 1999; Bannick et al. 2001; Kranert et al., 2001; Bannick et al., 2002; Severin et al., 2002;).

However, the options for minimisation of contaminant inputs into the Agro-Ecosystem are limited and vary with specific sources concerned. The main tools are:

1.) reduction at the source by technological means (principle of *best available technology,* which would also include source separation or the exclusion of certain feedstock materials),

2.) substitution of materials and substances with higher contaminant concentrations than alternative materials with equivalent beneficial effect,

3.) limitation of the use of materials to the minimum needed in order to gain the intended beneficial effect (principle of good [agricultural] practice) and

4.) general load limitations.

A long term sustainable strategy will always seek for a combination of measures which fulfil both:

1.) the maximisation of benefits and

2.) the minimisation of inputs of contaminants that may adversely affect soil functions (and human animal and plant health)

Fig. 8-2 illustrates exemplarily these options.

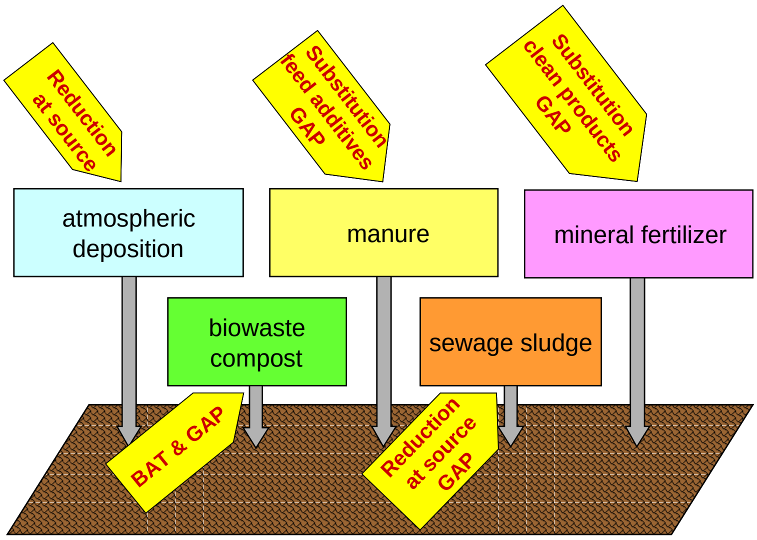


Fig. 8-2: Options to minimise inputs to agricultural land with the examples of atmospheric deposition and fertilisers

Three basic options are available in order to determine "safe" limit values for potential toxic elements (PTEs). They vary according to the viewpoint adopted:

1.) Risk based assessment such as the No Observable Adverse Effect Levels (NOAEL) concept

2.) Mass balance or No Net Accumulation (NNA) in relation to the concentration of contaminants in the soil (precautionary approach)

There are two options for a NNA concept:

i. Limiting **PTE/POP concentrations** in fertilisers and soil amendments to the same level of soil background concentrations ("*same to same*" or "*similar to similar*")

ii. Limiting the **PTE/POP load** so that it matches the amount of tolerable exports from soil via harvested crops, leaching or erosion ("*import = export*")

3.) Between those two polarities manifold hybrid systems and indicators such as the assessment of predicted environmental concentration (PEC) in comparison with the predicted no effect concentration (PNEC) are discussed.

What is commonly agreed is that any concept should provide long-term safe food and feedstuff production, the protection of the water resources and the biodiversity in the soils.

The debate on how far precaution must involve the soil itself and its basic functions (transformation, buffer, filter, biodiversity, genetic heritage and as a result productivity) is still going on and is mirrored by the different approaches.

The main difference may be summarised by quoting the Cornell study (Harrison at al., 1997):

> *"For example, some people place faith in technological solutions and our ability to calculate impacts and risks. Others are more sceptical, believing that history shows that there have been numerous failures of technology resulting in unanticipated environmental and health damage. These are fairly fundamental differences in worldview, leading some to favour precaution while others are willing to proceed until harm is shown to occur.*
>
> *There is no such thing as "safe". Rather, the question is: "What is an acceptable risk, and to whom?"*

A strategy that simply aims for a reduction of inputs following a no net accumulation scenario (input = estimated output) might result in a decline of SOM or nutrient status in the soil below a desirable level according to good agricultural practice (GAP) and sustainable productivity objectives.

On the other hand, a pure risk based approach could result in accumulation (up to a specified level) but this might not be acceptable by neither sustainability nor general political means in order to preserve soil as a multifunctional resource for future generations.

Fig. 8-3 illustrates the results of different approaches in limiting soil concentrations for Cd and Zn.

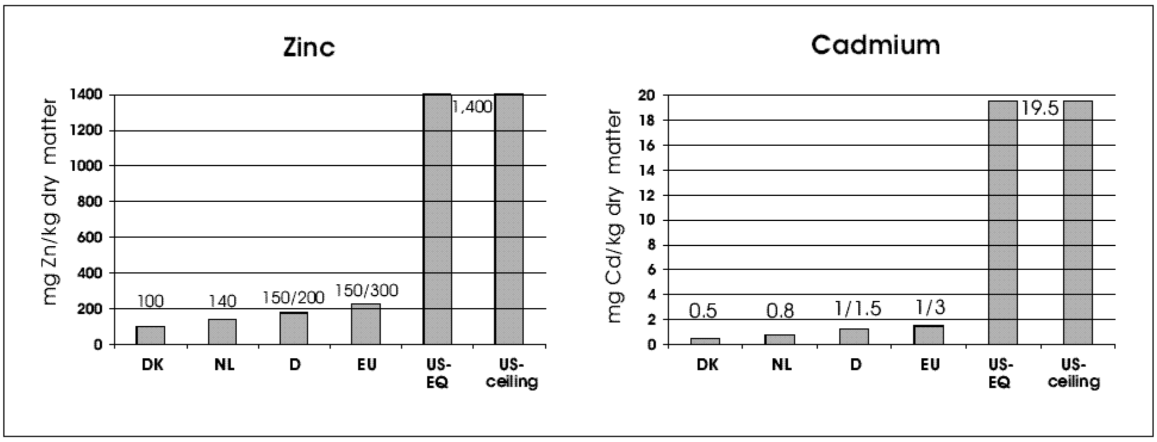


Fig. 8-3: European and US allowable Zn and Cd soil concentrations for sites suitable for sludge application (Harrison at al., 1997)

The following two chapters discuss critically the key elements of *risk based* and *precautionary no net accumulation* concepts. This will lead us to an integrated approach, outlined in chapter 9.

8.2 The US EPA risk assessment NOAEL based strategy (US EPA PART 503 SLUDGE RULES)

The Part 503 risk assessment identifies various potential routes for exposure following the use of sewage sludge in agricultural production. Fourteen different pathways were assessed The risk associated with each of the pathways has been calculated on the basis of available data. The threshold for a particular contaminant is the value generated by the pathway resulting in the lowest concentration that represented an acceptable risk according to the US EPA analysis. For five of the nine regulated contaminants the pathway of direct ingestion of sludge by children was the limiting path, generating the lowest acceptable level.

Many of the assumptions and the underlying approach have been commented on and criticised time and again. To shed more light on this discussion the key arguments of the *Cornell Study* (Cornell Waste Management Institute, Harrison at al., 1997) and Fürhacker et al. (1999) are shown below.

The Cornell Study identifies fourteen different *"Non-Protective Aspects of the US EPA Part 503 Risk Assessment"*. The main scientific as well as strategic criticisms refer to assumptions and decisions of the risk assessment, which do not support conservative or protective approaches. The study of Fürhacker et al. (1999) reviewed the scientifically based methodology and cognition used for Cadmium. The points addressed are:

- Data base for transfer rates shows a variation of 10 – 1,000 fold
- New literature shows higher soil – plant transfer rates
- Assessed diet was very low in vegetables
- Averages are not applicable to particular site or crop
- Use of geometric mean values without considering range of deviations
- Ecological impacts inadequately assessed:
  Other soil microorganisms than those used for the risk assessment are believed to be more sensitive to some heavy metals
- High variety of conditions (type of materials) adopted for the calculations
- Pollution allowed to reach maximum "acceptable" level
- Background soil concentration and soil properties like pH are not considered
- Ground and surface water calculations assume a high dilution/attenuation
- In spite of the uncertain database the concept does not even provide safety or uncertainty factors for the gained figures
- Each exposure pathway was evaluated separately (assessment of any path only for one single PTE) – potential multiple impacts and interactions were not considered
- No obligatory labelling of sludge products was provided

Recently the scientific and technical basis of US EPA Part 503 rules was evaluated by the US National Research Council (NRC, 2002) in order to review (i) the risk assessment methods and data used to establish concentration limits for chemical pollutants, (ii) the current standards for pathogen elimination in biosolids and (iii) to explore whether approaches for conducting pathogen risk assessment can be integrated with those for chemical risk assessment. The task of this evaluation was to focus specifically on human health protection and not ecological or agricultural issues.

The main items of the results of the NRC evaluation were recommended as necessary steps in order to *"reduce persistent uncertainty about the potential of adverse human health effects from exposure to biosolids."* The main findings are summarised as follows:

The committee found the technical basis of the 1993 chemical standards for biosolids to be outdated. EPA has not re-evaluated its chemical standards since promulgation, so the data and methods used for *the original regulations are well over a decade old. There have been substantial advances in risk assessment since then, and there are new concerns about some adverse health outcomes and chemicals not originally considered. Because of the diversity of exposed populations, environmental conditions, and agricultural practices in the United States, it is important that nationwide chemical regulations be based on the full range of exposure conditions that might occur. Furthermore, there is a need to investigate whether the biosolids produced today are similar in composition to those used in the original assessments.*

To illustrate the US EPA Part 503 the effects are shown exemplary:

US EPA approach exceeds the maximum load (1,6 - 2,5 t $ha^{-1}a^{-1}$ d.m.) of existing sludge regulations in European countries (e.g. CH, A, FR) 1.5 to 4 times.

In effect the admissible yearly or cumulative load for Cd of US EPA Part 503 would lead to the following increase of Cd concentration in the soil (20 cm soil depth with 3,000 t ha^{-1}):

BULK SLUDGE:

Cumulative load = 39 kg Cd ha^{-1} ➔ ~ + 13 mg kg^{-1} soil d.m.

BAG SLUDGE:

Yearly load = 1.9 kg Cd ha^{-1} ➔ ~ + 0.63 mg Cd kg^{-1} $year^{-1}$ soil d.m.

Drawing the baseline at typical background concentrations for Cd in European soils (0.20 – 0.70 mg kg^{-1} d.m.) it becomes evident that the NOAEL strategy uses soil as a sink for Cd until an actual adverse effect may be expected for any of the exposures adopted. Protective aspects in the sense of maintaining the soil quality on an existing level are of no relevance.

8.3 Precautionary Concepts

8.3.1 Strict Mass balance approach – no net accumulation

The first subject of protection in the mass balance concept is soil itself.

Precautionary limit values are to safeguard soils against future impacts. Long-term protection is aimed at preserving the possibility of different uses of soils. Focusing on the principles of prevention, precaution and anticipation, the European Commission provided the communication paper "Towards a Thematic Strategy for Soil Protection" (COM (2002) 179 final) emphasising that due to its wide range of vital functions, maintaining soil condition is essential for sustainability. From a soil protective view a zero-balanced input and output of heavy metals and a restriction of organic-compounds according to their degradation-rate (half-life time) in soils mark the ultimate sustainable goal.

In its pure interpretation, the precautionary approach may be regarded as avoiding any increases of soil PTEs concentrations above current values. This means that additions of PTEs from soil improvers, fertilizers and the atmosphere should equal the losses via leaching, plant uptake and erosion.

General options of integrating the needs of sustainability to operational concepts of (agricultural) soil use and pollutant fluxes attached thereto have been formulated by Bachmann et al. (1996):

1.) avoidance and/or decrease of pollutant input to zero; no change of given background concentrations;
2.) limiting input to (provisionally) tolerable output (zero balance and a low level; critical-load-concept)

Whereas option 1) applies to xenobiotics introduced to the environment through human activities (e.g. PCB, DDT), option 2) refers to substances enhancing soil acidification or eutrification, which are naturally present in the environment and should be reduced to a tolerable amount (non-critical load).

Although support exists for this approach, there are no regulations yet that follow this pure interpretation in view of the consequences in agriculture and other sectors of the economy.

A restrictive limitation following a "no net accumulation" policy would lead to maximum compost application (even when high quality composts from source separated garden waste are assumed) of about 3 tons ha^{-1} y^{-1}. would imply the impracticability of beneficial humus management in agriculture.

The concepts discussed in 8.3.3 to 8.3.6 have been brought up in the German discussion aiming at an equal attempt for all types of fertilisers in fulfilment of the requirements of the German Soil Protection Ordinance.

In order to achieve the objective of no net accumulation – at least beneath defined precautionary background values – the following general options are available:

1.) Limit values for PTEs in the fertilisers in combination with maximum application rates
2.) Limiting inputs on the basis of maximum critical PTE loads per ha and year; in principle without specific PTE limits of the fertiliser. The higher the contaminant concentration the more the contaminant limits the maximum application rate,
3.) Limiting inputs on the basis of maximum nutrient supply (as a rule the limiting element in many EOMs is phosphorus, but also nitrogen might be the limiting factor – e.g. in slurry, sewage sludge or poultry manure)
4.) Combination of 1.) to 3.) which can be found in most of the concepts.

8.3.2 Further option seeking a compromise for practicability

In addition to the options described above a third operational concept for integrating the needs of sustainability was formulated by Bachman et al. (1996).

The definition of threshold limits for (provisionally) tolerable enrichment within definite conditions takes into account:

- proportions for different pathways,
- balance of additional inputs (in the range between background and precaution-values) from different sources based on total contents for a time frame of e.g. 200 years and a specific soil use depth,
- mobile fractions and effect concentrations,
- site specific differentiation regarding current pollutant status.

This option has to be applied to the input of substances like heavy metals and organic compounds (e.g. PAH, PCDD/F) where substantial decrease of input is a matter of technical improvement. It has to be based on agreements concerning (i) the considered time frame (exposure time) and (ii) the assumptions of soil properties like bulk density and depth of the considered soil horizon when calculating permissible input loads.

As one example an evaluation scheme for organic and inorganic fertilisers and soil amendments was proposed by Severin et al. (2002) based on a nutrient/pollution ratio which is derived from average nutrient supply in agricultural systems and rated against the background of maximum permissible loads of heavy metals, where the term maximum “permissible load” refers to existing legislative regulations in Germany (Biowaste Directive, Soil Protection Ordinance).

A similar concept was introduced in Switzerland (Eidgenössische Forschungsanstalt für Agrarökologie und Landbau; Herter et al., 2001), where heavy metal concentrations of fertilizers are related to their total content of nutrients (N+P+K). Different fertilisers can be evaluated on this basis.

If a fixed maximum heavy metal load and limited nutrient supply (e.g. phosphorus) according to good agricultural practice are adopted as combined basic conditions, this leads to the following conditions:

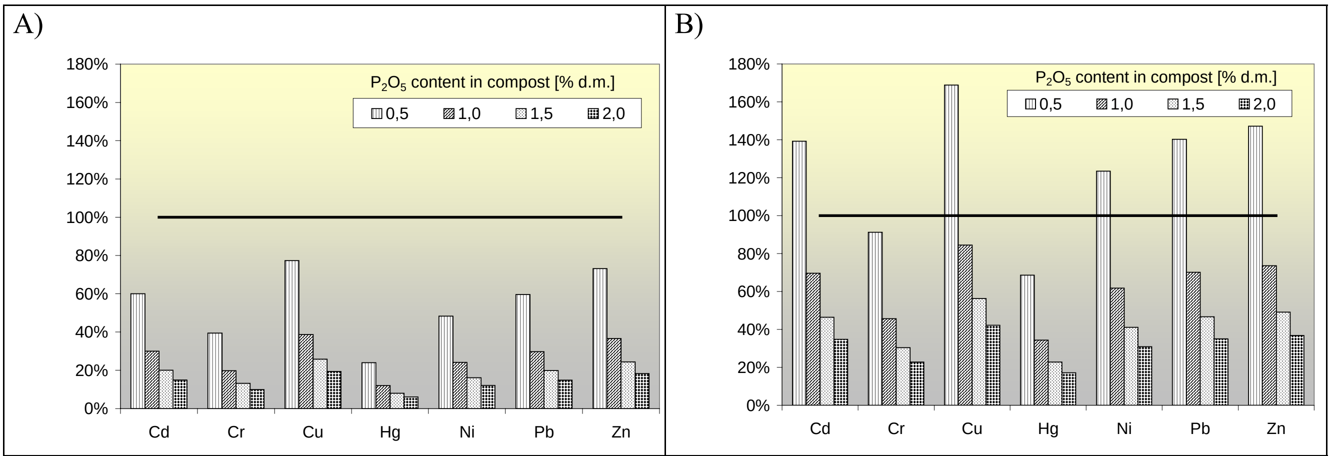
- the lower the nutrient level the higher the required quantity of fertiliser to fulfil the nutrient demand of crops
- when a given metal load must not be exceeded, the heavy metal concentration in the fertiliser has to decrease with declining nutrient contents

The effect for the derivation of heavy metal limits by relating the nutrient and heavy metal concentration is explained with the example of phosphorus and cadmium in Table 8-1

Table 8-1: Example for nutrient related heavy metal limits in waste derived fertilisers (Severin et al., 2002)

Standard fertilisation within crop rotation	60 kg P_2O_5 ha^{-1}y^{-1}
Admissible Cd load	10 g Cd ha^{-1}y^{-1}
Cd limit value at **P_2O_5** content of **0.5 % d.m.**	0.83 mg Cd kg^{-1} fertiliser d.m.
Cd limit value at **P_2O_5** content of **5.0 % d.m.**	8.3 mg Cd kg^{-1} fertiliser d.m.

Fig. 8-4 demonstrates that only composts with low phosphorus (0.5 % d.m.) and high heavy metal concentrations may exceed the maximum heavy metal load, here quoted from the German Biowaste Ordinance (with Cd, Cu, Ni, Pb and Zn).



A)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
HM conc [mg^{-1}kg^{-1}d.m.]	0.50	23.0	45.1	0.14	14.1	49.6	183
Max. load [g ha^{-1}y^{-1}]	10	700	700	7	350	1,000	3,000

Standardised yearly P_2O_5 supply in crop rotation: **60 kg ha^{-1} y^{-1}**

B)

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
HM conc [mg^{-1}kg^{-1}d.m.]	0.87	39.9	73.9	0.30	27.0	87.6	276
Max. load [g ha^{-1}y^{-1}]	10	700	700	7	350	1,000	3,000

Standardised yearly P_2O_5 supply in crop rotation: **80 kg ha^{-1} y^{-1}**

Fig. 8-4: Maximum phosphate supply or max. yearly heavy metal load as limiting factors in dependence of heavy metal concentration and P_2O_5 content in composts.
Assumptions: A) & B) … maximum heavy metal load according to the German Biowaste Ordinance (= 100 %) Standardised yearly P_2O_5 supply in crop rotation: A) … **60 kg ha^{-1} y^{-1}** B) … **80 kg ha^{-1} y^{-1}** heavy metal concentration in compost: weighted MEDIAN or MEAN (A) and 90th percentile (B) value of European surveys (see **Table 9-2**)

Some essential requirements and measures of concepts which lay claim to a comprehensive evaluation system for fertilisers are summarised in Table 8-2

Table 8-2: Main elements of comprehensive fertiliser evaluation systems

Concept element	Remarks
Applicable for all types of fertiliser	This means that risk assessments, limitation of PTE loads and concentration are fixed in such a way as to be applied not only for waste derived products but also for mineral fertiliser and stable manure *Important*: differentiated model calculation is needed for mineral and organic fertilisers depending on the quantity of material that may retain in the soil.
'Equal' treatment of all types of fertiliser	The limitations adopted should not discriminate against any particular fertiliser by, for example, underestimating a specific beneficial value (organic matter supply, creation of a mid term carbon sink, improvement of buffer and sorption capacity)
Maximum PTE concentrations derived from maximum yearly PTE loads	Maximum PTE loads, defined in national regulations (soil protection, sewage sludge regulations, biowaste and compost regulations) constitute the base line for further considerations in order to derive concentration limits *Important*: factual change of soil status (concentration of contaminant) must be taken into account for a proper judgement of the environmental impact of the given load.
Plant nutrient supply following *'Good Agricultural Practice'*	Yearly nutrient supply via fertiliser is limited on the basis of Good Agricultural Practice [GAP] (mostly nitrogen and phosphorus and in addition potassium are the considered nutrients)

8.3.3 The BMU/BMVEL[20] concept with fixed concentration limits

The concept proposed by Bannick et al. (2002) derives permissible concentrations in (organic) fertilisers (sludge, compost, manures and slurries) from the precautionary values of the Soil Protection Ordinance in Germany. The principle adopted here was the "limitation of pollutant loads to a concentration level corresponding to soil background concentration"

The basis: precautionary soil threshold values

Soil precautionary values within the meaning of the Federal Soil Protection Act are understood to mean soil values which, when being exceeded, give rise to assume the (possible) arising of a soil change with possible adverse affects to the environment.

Soils can generally be placed in the three main soil categories clay, loam and sand. They have the largest share in the total area of Germany. The category " Soils with natural or due to urbanisation higher background contents ", can be defined on the basis of (specifically higher) regionally representative background values. This could either be providedby a supplementary list set up on the federal level or by the individual states (Laender) within the frame of soil legislation enforcement. The proportion of these

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[20] German Ministry for Environment and Ministry for Consumer protection, Food and Agriculture

soils, considering the whole territory of Germany is only small. It is significantly increased in certain areas. Soils generally belonging to this category are as follows:

- soils with geogenic increased contents of e.g.magmatites, metamorphites (e.g. granite, gneiss) as well as of sandstone and limestone
- soils with predominately anthropogenic increased contents (due to the settlement structure)
- soils in marshes and flood areas.

Organic soils are considered to be subject to special considerations.

Table 8-3: Precautionary soil values of heavy metals for soils of the German Soil Protection Ordinance (BBodSchV, 1999)

		Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
German Soil Protection Ordinance								
“SAND”		0.4	30	20	0.1	15	40	60
“LOAM/SILT”	mg kg^{-1}d.m.	1.0	60	40	0.5	50	70	150
“CLAY”		1.5	100	60	1.0	70	100	200
soils with naturally increased and large-area settlement-related increased background contents	safe, as far as the release of pollutants or additional inputs pursuant to § 9 paragraphs 2 and 3 of this Ordinance do not give reason to expect any adverse impacts on the soil functions							

Guidance for application of table 1a "Precautionary values for heavy metals":

1. With regard to Cd, Ni and Zn it is evident that the precautionary values of the category "loam" apply to soils of the category "clay" with pH < 6,0 (pH limit value); the precautionary values of the category "sand" apply to soils of the category "loam" with pH < 6.0. This is also true for Pb using, however, a pH limit value of 5.0.
2. Soils consisting of clay stone and marl stone / loess, loess loams and till / sand (pleistocene or alluvial)

For the derivation of precautionary values the following technical requirements apply:

1.) Data on the accumulation and effect of pollutants in soils provide the basis for establishing precautionary values. Reference is made first of all to the variety of data given in literature on ecotoxicological effect thresholds (habitat, buffer, degradation and sink function for substance caused effects). If possible, the effect of soil pollutants on other targets to be protected (e.g. food plants including phytotoxic effects, groundwater and human health) is to be evaluated in addition. The effect thresholds determined shall be checked against soil background values in order to pay regard to the soil as integral part of the ecosystem.

2.) The sensibility of soils shall be taken into consideration by classification of the soil texture (clay, loam, sand) and by using the pH value. Soils with significantly increased background contents shall be evaluated separately. Classification of a given soil into the categories introduced for precautionary values shall be enforceable.

3.) Precautionary values shall be given as total contents for characterization of the long-term ecological relevance of a substance including its potentially mobilisable fraction. In order to particularly consider the soil function as "soil ability to filter, buffer and degrade substances, in particular, for the protection of groundwater" additional data give in mg/l leachate might be used to assess predicted leachate contents (not within the scope of this report).

Table 8-4: Precautionary soil threshold values of organic pollutants for soils (German Soil Protection Ordinance, BBodSchV, 1999)

		PCB (6)	Benzo(a)pyrene	PAH (16)
SOM > 8%	mg kg^{-1}d.m.	0.1	1	10
SOM ≤ 8%		0.05	0.3	3

Steps for the derivation of limit concentrations in fertilisers

The derivation of limit concentrations in fertilisers was done in three steps:

1.) The maximum admissible heavy metal concentration in the organic fertiliser must not exceed the soil threshold values, differentiated for clay, loam/silt and sandy soils respectively. The heavy metal concentration is calculated on the basis of the quantity of organic dry matter stemming from the fertiliser (compost) which can be retained in the long term (71 % of compost dry matter applied, assuming a mineral matter ratio of 63 % and a total of 8 % OM that remains in the soil).

2.) The yearly amount of amended organic fertilisers is derived from average phosphorus supply of 50 kg P2O5 ha-1y-1. The resulting heavy metal load is calculated from the tolerable maximum concentrations of assumption. The export of heavy metals via the harvested crops is added. The resulting yearly amount of applied compost on the basis of 50 kg P2O5 ha-1y-1 was estimated with 6.3 t ha-1.

3.) Finally an empirical sampling and analytical tolerance of 50 % was added to the maximum concentrations.

Table 8-5: Proposed limit values for composts following Bannick et al. (2002) in comparison to existing qualities in Europe.

	Soil type	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Proposed limits	clay	1.63	107.01	70.08	1.10	75.94	107.63	260.57
	Loam/silt	1.10	64.41	48.78	0.56	54.64	75.68	207.32
	sand	0.46	32.46	27.48	0.14	17.36	43.73	111.47
BWC low (1)		0.50	23.0	45.1	0.14	14.1	49.6	183
BWC high (2)		0.87	39.9	73.9	0.30	27.0	87.6	276

(1) average of mean or median values from European compost surveys (biowaste composts;)
(2) average of 90th percentile values from European compost surveys (biowaste composts)

It has been estimated that in Germany on facility level only 10 %, 42 % and 62 % of 376 composting plants may guarantee in average the proposed limit values for sand, loam and clay soils respectively (Reinhold, 2003).

8.3.4 The critical load model of Schleswig-Holstein[21] (Germany)

The critical load model also follows the requirement of the German Soil Protection Ordinance, which limits the yearly PTE load to the soil, when precautionary threshold limits for soils are exceeded. The

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[21] Ministerium für Umwelt, Naturschutz und Landwirtschaft des Landes Schleswig-Holstein (DE): Vorschlag zur Begrenzung des Eintrages von Schadstoffen bei der Düngung landwirtschaftlicher Nutzflächen

model also considers atmospheric deposition which reduces maximum inputs by fertilisers. Secondly this system evaluates the maximum application rate based on an average P_2O_5 supply of 50 kg ha^{-1}y^{-1}. The maximum amount of e.g. compost is then derived from the limiting element, either of one of the PTEs or the P_2O_5 depending on the actual concentration in the fertiliser. It claims to be a more flexible system compared to a concept giving rigid concentration limits and being equally applicable to all types of fertilisers independent of their factual nutrient and PTE concentration.

Table 8-6: Critical load model combined with the limitation of the nutrient supply (slightly adapted from a proposal of Schleswig-Holstein, DE)

Line	Soil type		Cd	Cr	Cu	Hg	Ni	Pb	Zn
1	Atmospheric deposition	g ha^{-1}y^{-1}	2-2.2	5-5.5	30-33	0.3-0.33	8-8.8	30-33	125-137.5
2	Max. **load** (BBodSchV) (1)		6	300	360	1.5	100	400	1,200
3	Precautionary max. **load** through agricultural input		4.0	290	330	1.2	90	370	1100
4	Mean PTE **concentration** in composts	mg kg^{-1}	0.47	25.3	57.7	0.16	16.3	46.4	204.7
5	**PTE derived max application**	**t ha^{-1}y^{-1}**	**8.4**	**11.5**	**5.7**	**7.7**	**5.5**	**8.0**	**5.4**
6	P_2O_5 in compost	kg t^{-1} d.m.	8.3						
7	**P_2O_5 dependant max. appli cation at 50 kg P_2O_5 ha^{-1}y^{-1}**	**t ha^{-1}y^{-1}**	**6.0**						
8	**MAX PTE concentration in composts as additional cover (2)**	**mg kg^{-1}**	**1.00**	**72.5**	**82.5**	**0.30**	**22.5**	**92.5**	**275**

(1) maximum additional load from all types of sources to soils when precautionary soil values are exceeded (BBoSchV, Germany)

(2) additional *'risk cover'* based on the quotient max. PTE load and P_2O_5 based application of 6.0 t ha^{-1}y^{-1} including a 50 % tolerance

In this case no limit values for fertilisers would be necessary. The idea is that at high PTE levels as compared to the nutrient concentration any application would become useless, since the application rate would have to be reduced to an quantity that would exceed technological options. In this way a Zn concentration of 300 mg kg^{-1} would decrease the maximum compost quantity to 3.7 t d.m. ha^{-1}y^{-1}. This would challenge the use of compost since benefits of OM and nutrient supply could not be achieved anymore.

However, the following information would be needed for an implementation of this system into practice:

- Nutrient and PTE concentration of the fertiliser
- Mass balance of applied fertilisers on plot level
- Nutrient and PTE balance on plot level
- Adoption of a calculative time frame for the PTE balance (3 – 5 years or individual crop rotation)
- Adoption of maximum PTE loads in a given time frame

This would require a batch wise PTE and nutrient declaration. One could abandon the knowledge of the PTE soil background concentration since this concept could be applied to all soils and management conditions.

In order to have an additional tool for covering the risk of diluting e.g. low with high quality composts maximum concentration limits are suggested (see Table 8-6, line 8).

Table 8-7 compares those concentration limits with a statistical evaluation of 376 Composting facilities in Germany calculating warranty values for mean values and individual samples on facility scale on a probability level of 95 %. These values incorporate statistical variance for seasonal, sampling and analytical deviations.

It is evident that arbitrary limit setting based on abstract load limits has nothing to do with reality of compost even under best practice of quality management and therefore would undermine the widely agreed efforts for source separation and recycling of organic matter where ever it is needed as an effective tool of soil improvement and protection.

In addition, and most critical, the model assumes the maximum load model which has been created for soils with elevated background concentrations (exceeding the precautionary threshold limits) for all soils.

Table 8-7: Validation of threshold values derived from statistical evaluation of analytical results from 376 composting plants in Germany (Reinhold, 2003[FA545])

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
Warranty values **Means** on plant scale (*)	**mg kg^{-1} d.m.**	1.1	70	110	0,5	60	120	380
Warranty values **single samples** on plant scale		1.8	100	180	1.1	80	190	530
*MAX PTE concentration of the **critical load concept** (Table 8-6)*		***1,00***	***72,5***	***82,5***	***0,30***	***22,5***	***92,5***	***275***

(*) Warranty values calculated on the basis of 376 Composting facilities in the average of 4 subsequent measurements at a precision of $p < 0.05$

8.3.5 The critical load concept of Northrhein-Westfalen (NRW, DE) (König et al., 2003)

This concept seeks to combine the two options of no net accumulation and concludes that the possible loads resulting from

- the limitation of **PTE concentration** in the applied fertilisers fraction, which is retained in the soil after decomposition to the soil background level ("***same to same***") = $\boxed{\text{Load}_{ss}}$
  and
- the limitation of PTE loads at the level of tolerable exports from soil via harvested crops, leaching or erosion ("***import = export***") = $\boxed{\text{Load}_{i=e}}$

may be added in order to maintain the overall contaminant status in soil.

The maximum concentration is calculated as follows:

$$\boxed{\textit{Max. PTE concentration} \;=\; \frac{\textit{Load}_{ss} + \textit{Load}_{i=e}}{\textit{Quantity applied}}}$$

Load_{ss} = long term remaining fraction in soil × tolerable concentration in soil[22]

$\text{Load}_{i=e}$ = tolerable export (harvest + leaching) balanced with other diffuse inputs

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[22] Those are taken from the precautionary soil values for loamy soils of the German Soil Protection Ordinance:

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
mg kg^{-1} d.m.	1.0	60	40	0.5	50	70	150

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Max load [g ha^{-1}y^{-1}]	3	150	180	0.75	50	200	600

The basic data of the calculation for biowaste compost, sewage sludge and pig slurry were:

Table 8-8: Application rate at 50 kg P_2O_5 ha^{-1}y^{-1} and long term remaining fraction in the soil for different fertilisers

		biowaste compost	sewage sludge	pig slurry
mean P_2O_5 conc.	[% d.m.]	0.8 - 0.9	4.9	5.8
application rate at 50 kg P_2O_5 ha^{-1}y^{-1}	[t d.m. ha^{-1}y^{-1}]	6.0	1.0	0.9
Long term remaining fraction in soil	[% d.m.]	50*	40	20
* It has to be stated that the assumption of only 50 % of the total dry matter of added compost would remain in the soil can hardly be accepted since in average the mineral fraction of composts amount at 60 to 75 % !				

Following the calculation model the limit values of Table 8-9 are achieved. The authors claim that this model could be applied also for persistent organic compounds (POPs) analogously.

Table 8-9: Calculated limit concentrations and proposals for limit values after consideration of material specific tolerances and rounding (concept of NRW)

			Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
Biowaste compost [6 t d.m. ha^{-1}y^{-1}]	***Load$_{ss}$***	*g ha^{-1}y^{-1}*	3	180	120	1.5	150	2210	450
	Load$_{i=e}$		3	150	180	0,75	50	200	600
	Total load		6	330	300	2.25	200	410	1050
	Calculated limit concentration	*mg kg^{-1} dm*	1	55	50	0.38	33	68	175
	Proposed limit value 1)		**1**	**60**	**70**	**0.4**	**35**	**70**	**250**
	warranted mean/plant scale 2)		***1.1***	***70***	***110***	***0,5***	***60***	***120***	***380***
Sewage sludge [1.0 t d.m. ha^{-1}y^{-1}]	***Load$_{ss}$***	*g ha^{-1}y^{-1}*	0.4	24	16	0.2	20	28	60
	Load$_{i=e}$		3	150	180	0,75	50	200	600
	Total load		3.4	174	196	0.95	70	228	660
	Calculated limit concentration	*mg kg^{-1} dm*	3.4	174	196	0.95	70	228	660
	Proposed limit value 3)		**1.2** 3)	**170**	**200**	**0.8** 3)	**70**	**60** 3)	**650**
	90th percentile		***1,7***	***110***	***403***	***1,1***	***59***	***110***	***1.201***
Pig slurry [0.9 t d.m. ha^{-1}y^{-1}]	***Load$_{ss}$***	*g ha^{-1}y^{-1}*			7				27
	Load$_{i=e}$				180				600
	Total load				187				627
	Calculated limit concentration	*mg kg^{-1} dm*			208				697
	Proposed limit value 4)				**270**				**900**
	90th percentile		*0,6*	*13*	***402***	*0,03*	*14*	*5*	***1.274***

1) ca. 30 % were added to calculated limit concentrations due to mean background concentrations in German biowaste composts

2) Warranty values calculated on the basis of 376 Composting facilities in the average of 4 subsequent measurements at a precision of $p < 0.05$ (Reinhold, 2003)

3) those figures were adapted to the mean concentrations of Cd, Hg und Pb in German sewage sludge

4) ca. 30 % were added to calculated limit concentrations due to animal nutrition demands

8.3.6 The VDLUFA [23] critical load concept

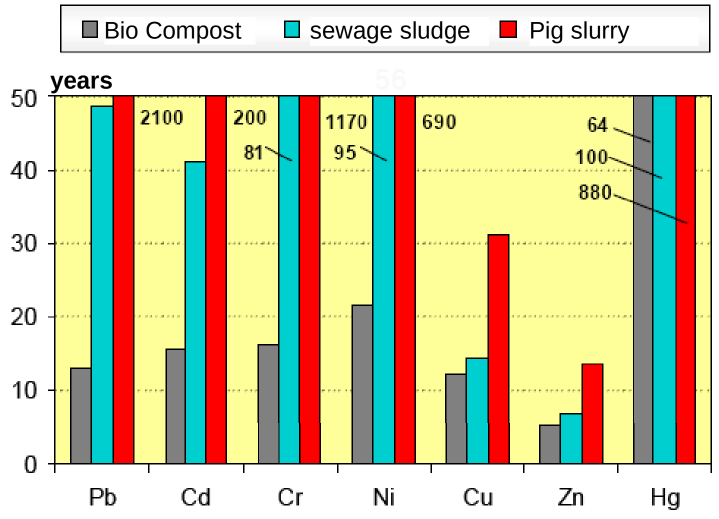
The German VDLUFA presented a concept where heavy metal loads are derived in connection with maximum nutrient supplies following GAP (VDLUFA, 2002). The main criticism against the same-to-same BMU/BMVEL approach (see 8.3.3) referred to the fact that Cu and Zn as essential trace elements in animal and plant nutrition was underestimated and the concept would be far to complex for an effective control.

The concept follows the following premises:

- Setting of critical heavy metal loads which are commonly defined for all types of fertilisers
- Maximum (critical) loads are derived from nutrient supply following GAP

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Max load [g $ha^{-1}y^{-1}$]	6.7	467	467	4.7	233	1000	2000

- 58 kg P_2O_5 $ha^{-1}y^{-1}$ is adopted as reference value for the maximum fertiliser application
- a slow accumulation is adopted for a limited transitional period.
  The accumulation is justified with the fact, that the analytical identification of the PTE increase in the soil would take in minimum 5 years (following application rates at GAP level).



Heavy metal concentration in soil in mg kg^{-}:

mean soil concentration	30	0.2	30	30	20	50	0.2
detectable increase	1.5	0.02	1.5	1.5	2.0	2.5	0.02

Fig. 8-5: Time until soil accumulation can be analysed

Material specific limit values have been calculated by dividing the critical PTE load [g $ha^{-1}y^{-1}$] by the maximum dry matter quantity of fertilizer [t d.m. $ha^{-1}y^{-1}$]. Under consideration of currently achievable PTE concentrations in the different fertilisers and the potential of further measures for the reduction of contaminants a certain tolerance was added. The result as compared to Median and 90th percentile values of biowaste compost, sewage sludge and pig slurry is presented in Table 8-10. Again, even though to a less extent the comparison of the proposed compost limits with mean values which can be warranted on plant scale shows a discrepancy in the case of Cu, Ni and Pb.

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[23] VDLUFA … German Association of Agricultural Research Institutes

Table 8-10: So called *"practicable"* limit values of the VDLUFA proposal (mg kg^{-1} d.m.)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
Biowaste compost	**limit**	**1.0**	**80**	**100**	**0.8**	**30**	**100**	**400**
	Median	*0,4*	*23*	*47*	*0,1*	*14*	*44*	*179*
	90th percentile	*0,8*	*38*	*79*	*0,3*	*25*	*76*	*229*
	warranted mean on plant scale (1)	*1.1*	*70*	*110*	*0,5*	*60*	*120*	*380*
Sewage sludge	**limit**	**2.5**	**200**	**550**	**2.0**	**80**	**200**	**1,400**
	Median	*1,0*	*38*	*217*	*0,4*	*32*	*64*	*753*
	90th percentile	*1,7*	*110*	*403*	*1,1*	*59*	*110*	*1.201*
Pig slurry	**limit** **(orientation value)(2)**	**2.5** **1.5**	**200** **35**	**550** **550**	**2.0** **0.1**	**80** **35**	**200** **15**	**1,400** **1,400**
	Median	*0.3*	*5*	*174*	*0.02*	*8*	*3*	*670*
	90th percentile	*0.6*	*13*	*402*	*0.03*	*14*	*5*	*1,274*

(1) Warranty values calculated on the basis of 376 Composting facilities in the average of 4 subsequent measurements at a precision of $p < 0.05$ (Reinhold, 2003)

(2) In the case of pig slurry with the exception of Cu and Zn for future regulations *orientation values* are suggested. This is based on the fact that metal concentrations are considerably lower than in other media.

8.3.7 The limit values of EU Regulation (EEC) No. 2092/91 – a considerable threat to the continuous production of high quality compost for organic farming

An evaluation of the Austrian data showed that only 38.9 % of 582 BWC (n=501) and GC (n=63) meet the requirements of the EU Regulation (EEC) No. 2092/91 on organic farming. Compost from cities fulfil this requirement only in 20 – 26 % whereas for rural areas the proportion amounts 44.5 %

The 61 % of 501 BWC, which did not fulfil the limit values were examined to determine how far the single metals contribute to exceeeding the given thresholds. The limiting metals are Pb, Zn and Ni, which contribute at a ratio of 50 %, 49 % and 40.5 % of the samples to exceeding the limits (Table 8-11).

Table 8-11: Range of relative contribution to exceeding the limit values for composts which do not fulfil the requirements of Annex II A Reg. (EEC) No. 2092/91 [in %]

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				in %			
range *[in 9 AT provinces]*	0 – 90.9	0 – 6.1	4.1 – 72.7	0 – 13.3	0 – 95.9	8.2 – 81.8	16.3 – 66.7
average *AT total [n=501]*	12.4	1.0	21.2	6.5	40.5	50.0	48.7

The distribution of the elements specifically causing the compost to exceed the limits is shown in Fig. 8-6. ‚n' is the total number of samples, which show concentrations beyond the limit value. The degree by which the limit is exceeded is shown in this figure. E.g. for Cu in 40 % of the samples, the values are less than 10 above the limit value. For seventy percent of the samples the excess is less than 20 % beyond the limit.

Cd, Hg, Cu, Ni and Zn exceed the ranges between 30 and 48 % of the limit values in 75 % of the composts, which do not meet the requirements. Pb shows a much more expanded distribution, 35 % of the analysis exceed the limit of 45 mg kg^{-1} d.m. by more than 50 %.

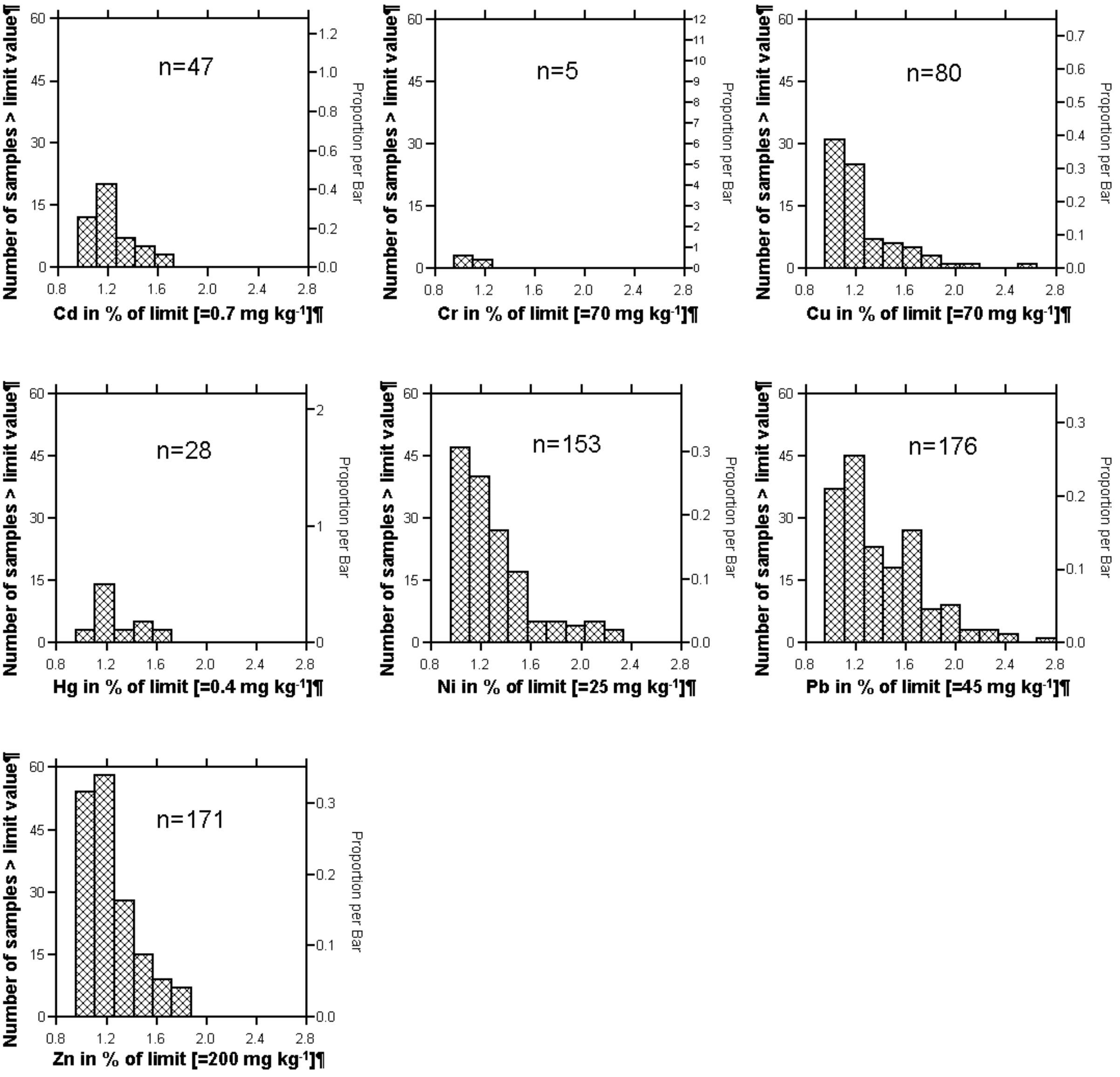

Fig. 8-6: Element specific distribution of exceeding the limit values for 501 BWC according to Annex IIA EC Regulation 2092/91/EC.

Figures give the exceeding as percentage relative to the limit value of all samples, which do not meet the requirement in at least one of the elements ($n_{\text{(all samples)}} = 501$; $n_{\text{(samples > limit values of (EEC) No.2092/91)}} = 306 =$ 61 %).

The statistical evaluation of 376 regulary approved composting plants in Germany (Reinhold, 2003) showed a similar picture. More than 50 % of the composting cannot guarntee to meet the criteria of the organic farming regulation (the mean value of 4 consecutively taken samples is estimeated to exceed the limit value). Only 10 % of the biowaste composting plants may constantly provide composts which fulfil the heavy metal limits of Reg. 2092/91/EC.

Decisive for this are above all the limit values for lead (45 mg kg^{-1} d.m.) and zinc (200 mg kg^{-1} d.m.).

These examples demonstrate that if limit values are set too tight even the best performing source separation schemes would not succeed. In this context tight limit values would fail to support the intended continuous production of high quality compost.

8.3.8 Conclusions as regards the quoted No Net Accumulation concepts

The following disadvantages of the above cited concepts should be considered:

- Maximum PTE loads are non science based arbitrary settings
- The result depends on the assumed phosphorus content in organic fertilisers which shows a considerable variation even for composts (0.4 – 1.2 % P_2O_5 d.m.).
- The organic matter supply especially with composts is underestimated
- The result depends on the assumed plant uptake which again may show enormous variations with crop rotation and sorption properties of the soil
- The rigour of some of the concept may lead to an exclusion of currently achievable compost qualities at proportions of 100 to 50 %
- The concepts are not based on the reality of achievable PTE concentrations considering needed tolerances due to local, seasonal and analytical deviations
- Each of the concepts do not question the factual effect of the application system to the soil at least on a mid term scale. But this would be the key element in order to estimate the environmental impact and the maintenance of soil functions in the agro-eco system
- Thus the concepts tend to be built upon arbitrary abstractions
- The NNA approach typically may represent an "end of pipe" approach, since it hinders beneficial use of source separated materials (since they may be affected by diffuse environmental pollution), whereas farmlands still are subject to other sources of contaminants, primarily atmospheric deposition; the approach should rather be to address the issue for diffuse contamination at the source, improving emissions from industrial processes and transportation, thereby cutting not only direct fall-out onto soils, but also background contamination of organic materials (e.g. food commodities) suitable for application onto soils

9 PROPOSED CONCEPT

The scope of this section is to present a concept for the definition of threshold values of PTEs (*heavy metals*) in compost on a European level. The concept is based on achievable compost qualities (see Table 9-2 and chapter 4) nutrient (P, N) and organic matter supply according to best practice standards and long-term soil protection goals.

Thus it is a hybrid approach following the precautionary principle for the long term protection of soil functions and, at the same time allowing for a controlled and low input of heavy metals in the short and mid term based on the acknowledgment of agronomic needs. By this way it seeks to overcome the not scientifically justified load limitations and disadvantages of the concepts, discussed in chapter 8.

9.1 A step-wise approach towards sustainable fertilising systems

Some of the principles or basic considerations of the aforementioned NNA concepts can be endorsed as reasonable step within a comprehensive perception of fertilising systems. At least one is the definition of beneficial effects and the adoption of GAP in terms of soil improvement and plant nutrition.

In order to recognise the full range of benefits, specifically of organic soil amendments such as compost the potential adverse effects by contaminants inputs have to be assessed on a mid-term basis independently from the application regime which has been derived from GAP requirements.

The latter can differ significantly when considering the site and management specific demands. This can be e.g. (i) a continuous low rate for the maintenance of a given SOM status or (ii) a short-term humus amelioration on degraded sites with higher application rates for a period of e.g. 10 years.

The following general, step-wise approach, would be a feasible procedure which can be applied for all types of fertilisers and contaminants. The variable parameters (background concentrations in soils, leaching rates, applications rates of fertilisers etc.) can be adapted for any individual scenario and fertilisation system.

9.1.1 The four steps

Step I: Identify the benefits of the fertiliser/soil amendment to the agro system (e.g. relevant amounts of OM or plant nutrients)

Agronomic benefit follows good agricultural practice (GAP) (nutrient/OM supply) including eventual necessary limitations related to e.g. water conservation or N_2O emissions.

This in particular, must also include enough flexibility considering the organic sorption/binding dynamic of nitrogen which cannot be neglected in the comparison of e.g. compost, liquid manure or mineral N-fertiliser (Amlinger et al. 2003, Timmermann et al. 2003). Thus short-term and mid-term effects of added organic matter and plant nutrients in terms of plant nutrition, build-up of nutrient and SOM stocks and the likelihood of being leached to the groundwater have to be considered in order to define best practice of beneficial fertiliser management. This gives an optimum range of quantities to be applied under the diverse agronomic/site/cultivation conditions including soil amelioration on degraded sites.

Step II: Identify potential pollutants which may have any adverse effects to the agro-ecosystem, to the environment or the food chain

Identify the potential pollutants (inorganic and organic) and evaluate the potential risks under short/mid and long-term management scenarios defined for good agricultural practice in step I. For PTEs (*heavy metals*) this is apparently an easy exercise though we still have to distinguish between metals which play an essential role as trace elements (Cu, Zn and partly Cr) and others where no beneficial effect is known. But consider, in some cases a "too much" of copper and zinc can be a concern as well as an over supply by plant nutrients. In the case of organic micro pollutants the answer to the question if a certain substance occurring in manure or compost pause a risk is much more complex. There are considerable uncertainties of concentration-dose relation in the considered pathways as well as only sparsely known dynamics of decay, sorption and metabolism of the majority of compounds.

The evaluation and conclusions for organic pollutants are presented in chapter 5.

Step III: Define the limiting factor

This follows GAP with balanced OM and nutrient supply (preventing a surplus over a certain time frame) by observing potential accumulation of PTEs against threshold values for multifunctional use of soils[24]. Subsequently the potential accumulation of pollutants in time (e.g. between 50 and 200 years) must be evaluated and decision must be taken whether the slope of the accumulation or the probable exceeding of precautionary soil thresholds are acceptable or not.

Step IV: Response to the outcome of step III

This can be done by

- further reducing the input by means of technological improvement of the material (source separation, quality management of processing the waste materials)
- limiting the quantity to be applied

or

- accepting the management system (GAP) as a baseline in combination with the strategy that on a long-term scale multifunctional threshold values of soil are not exceeded.
- setting maximum concentrations for fertilisers taking into account tolerances of local, seasonal, sampling and analytical variances

When considering the adoption of scenarios in which we would accept a controlled and very slow input of PTEs to the soil, which does not constitute risk in a given time horizon, this approach

- does take full advantage of beneficial effects of flexible organic matter and nutrients supply especially from compost to the soil eco system
- constitutes a driver for an overall technological improvement of industrial processes over time, in order to have a steady improvement of quality of composts or digestates, its input from feedstock and overall immissions to the environment. This arguably would not happen in the case of a regulatory approach which would imply cutting off the application for much of the composted products even if they originate from source separated organic waste (see concepts discussion in chapter 8.

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[24] The assessment of critical threshold values of soils is not discussed here. Risk based assessments have to consider receptors and path ways of concern as well as dynamic processes in time. The principal approaches see chapter 8. The German *precautionary soil thresholds* are taken as an exemplary reference.

9.2 Concept assumptions

As mentioned before, it is evident that it will be not possible to follow a continuous ecotoxicological derivation of limit values for pollutants in composts. In addition to the scientific evaluation of soil threshold values (whose preservation is one cornerstone in EU environmental policy) pragmatic approaches following the objective of beneficial compost use have to complement the concept.

The assumptions made for a “safe” application scheme and the approach to application limits for compost use are based on existing qualities and intended benefits sought with the application of composted amendments.

9.2.1 Soil threshold values for multifunctional agricultural use

The starting point is the soil and its development in a defined time and management framework. For any concept we have to agree on soil quality parameters and threshold or guide levels of PTEs, which should guarantee agricultural functionality over a long time period.

The following attempt was made:

- The background of existing guide values for soil protection regulations or related proposals have not been questioned in this study. It is assumed that these thresholds are scientifically justified by means of ecotoxicological assessments.
- The “precautionary” values of the German Soil Protection Ordinance for “sand” and “clay” soils (BbodSchV, 1999 [FA549]) were taken as reference and take into account the broad range of relevant soil properties in terms of sorption capacities for PTEs. The guide values were gained by calculating the “safe” aqua regia value from a 2-step correlation. Toxicological impacts on soil functions (microbial activity, adverse effects on the plant, possible accumulation in the food chain, leaching to the groundwater) were correlated with available PTE fraction in the soil and in a 2nd step correlated with aqua regia concentrations. In addition safety factors were introduced. Further explanations are provided in chapter 8.
- The proposal of the Joint Research Centre (JRC) for acidic soils ($6 < pH < 7$) (Langenkamp et al., 2001[FA550]), which were suggested for a revision of the EC Sewage Sludge Directive. These data are based on the actual background concentrations of heavy metals in European agricultural soils (see Table 9-1).
- These data for all PTEs can be found between the two extremes (*sand* and *clay*)of the German Soil Protection Ordinance.
- The proposed concept requires, that under consideration of all other relevant sources and losses in the long run both organic and mineral fertilisation must not exceed these thresholds to a significant extent or if this would occur in a mid term scale further measures should be undertaken to enhance fertiliser quality or reduce the input by other means.

Table 9-1: Soil guide respectively threshold values used for the accumulation scenarios

			Cd	Cr	Cu	Hg	Ni	Pb	Zn
Soil guide value "SAND"	(1)	mg kg^{-1}d.m.	0.4	30	20	0.1	15	40	60
Soil guide value "CLAY"	(1)	mg kg^{-1}d.m.	1.5	100	60	1.0	70	100	200
JRC proposal soil: pH 6-7	(2)	mg kg^{-1}d.m.	1	75	50	0.5	50	70	150

(1) German Soil Protection Ordinance (BbodSchV, 1999)

(2) Proposal for a revision of soil limit values in the frame of an EC Sewage Sludge Directive by the Joint Research Centre (JRC) of the EU Commission, Institute for Environment and Sustainability, Soil and Waste (Langenkamp et al., 2001)

9.2.2 Concentration of PTEs in composts

From the survey in chapter 4 data from those countries which provided detailed information on the distribution of the concentration of PTEs in BWC were taken.

In order to compute representative and realistic scenarios for a *"European compost quality"* the statistically weighted average of each country-median (or mean) and, to provide for an even more precautionary approach, the 90th percentile values were calculated (from 7 country surveys Table 9-2).

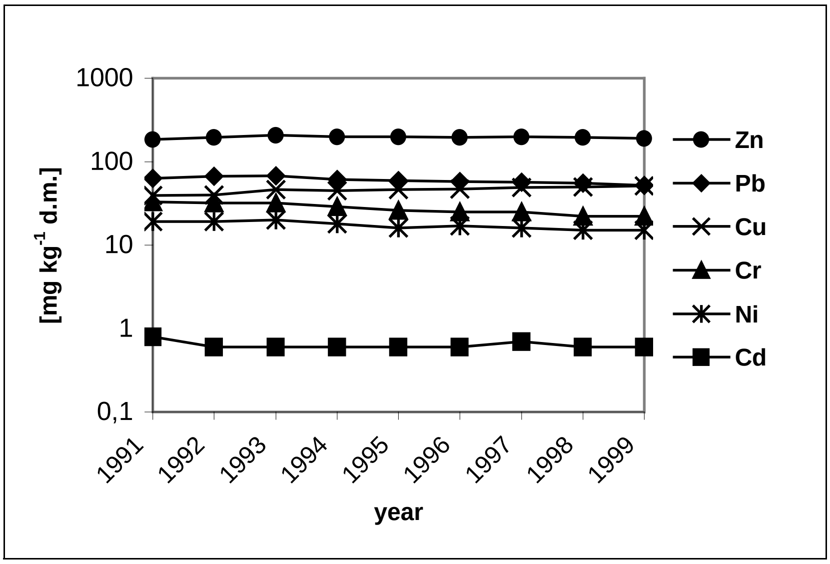
Table 9-2: Median (Med) and 90th percentile (90 %ile) of BWC of national investigations and statistically weighted figures used as representative concentration in the accumulation scenarios.

		AT	DE*	ES	FR	IT	NL	UK	weighted total
	n	*552-582*	*17800*	*48-56*	*25-26*	*22-97*	*624-627*	*60*	*19280 - 19352*
Cd	**Med**	0.38	0.50	1.50	0.86	1.00	0.46	0.51	**0.50**
	90 %ile	0.70	0.87	1.50	1.90	2.00	0.70	1.09	**0.87**
Cr	**Med**	24.1	23.0	27.0	30.2	41.7	19.0	16.0	**23.0**
	90 %ile	40.8	40.0	55.0	69.1	120.6	27.0	31.4	**39.9**
Cu	**Med**	47.4	45.0	88.0	89.0	98.5	32.0	50.0	**45.1**
	90 %ile	74.0	74.0	137.8	162.6	159.0	48.0	90.5	**73.9**
Hg	**Med**	0.16	0.14	0.20	0.50	0.24	0.11	0.20	**0.14**
	90 %ile	0.35	0.30	0.49	1.38	0.97	0.20	0.40	**0.30**
Ni	**Med**	19.0	14.0	23.0	20.2	31.7	9.3	18.0	**14.1**
	90 %ile	33.0	27.0	56.0	54.6	75.9	12.0	29.2	**27.0**
Pb	**Med**	37.0	49.0	55.5	93.0	69.7	67.0	102.0	**49.6**
	90 %ile	69.1	87.0	105.5	187.5	150.9	99.5	159.4	**87.6**
Zn	**Med**	174	183	202	242	256	169	186	**183**
	90 %ile	254	277	306	560	502	201	322	**276**

* mean values; due to the large number of samples these are expected to be very close to the median values

9.2.3 Time frame as a tool of flexibility

The development of compost qualities (see 4.3.3 Temporary and seasonal variations) demonstrates that once a proper separate collection system is implemented, quality variations are comparatively low assuming constant impurity ratios (< 0.5 % m/m). This leads to the conclusion that compost quality mainly depends on back ground concentration in source materials (see Fig. 9-1).



For the accumulation model a time frame of 100 and 200 years was chosen to demonstrate scenarios which may be considered a "ceiling situation" and lead to additional precautionary strategies, so as to stay well within the assumed soil limit values. CSTEE, (2002) took 100 years as reference for the evaluation of the risk assessment from cadmium in fertilisers.

Fig. 9-1: Temporary development of heavy metal concentrations in BWC in Germany [mg kg^{-1} d.m.] (Kranert, 2002)

9.2.4 Beneficial effect of continuous compost application – and loads according to good agricultural practice

P_2O_5 supply

As shown in chapters 6 and 8, phosphorus is identified as a limiting factor for the evaluation of the sustainable use of fertilisers and often serves as key parameter in comparative assessments of different organic and inorganic fertiliser schemes.

The surplus of P supply caused by fertilisation *and* feeding stuff (which affects its concentration in manure) led in Central and Northern European countries to a continuous reduction of the use of P fertiliser. In Germany, for example, the average P fertilisation decreased from 32 kg ha^{-1}y^{-1} (=73.3 kg ha^{-1}y^{-1} P_2O_5) in 1979/80 to 15 kg ha^{-1}y^{-1} (=34.4 kg ha^{-1}y^{-1} P_2O_5) in 1992/93 (Scheffer & Schachtschabel et al., 1998).

Following *good agricultural practices* the *standard* P_2O_5 supply to arable land is estimated 50 to 68 kg ha^{-1} y^{-1} (Bannick et al., 2002; Severin et al., 2002; Schellberg et al., 1999). The CSTEE, (2002) paper evaluating the MSt risk assessment to health and the environment from cadmium in fertilisers presents two scenarios of annual P_2O_5 application, 23 kg ha^{-1}y^{-1} and 69 kg ha^{-1}y^{-1} respectively. The report quotes 30 kg ha^{-1}y^{-1} as a realistic *"worst case"* for continuous annual input. Sweden and Denmark have also introduced similar stringent phosphorus limitations in compost or sewage sludge related standards (SE: 22 – 35 kg ha^{1}y^{-1}; DK: 30 kg ha^{-1}y^{-1}).

This viewpoint is justified for soils with high P status. A phosphorus surplus has not been raised as an issue in Mediterranean countries yet, nor has it led to regulatory provisions to limit its load. Nevertheless, we assume this level as the limiting factor in the lowest base line for an evaluation of long-term effects of compost application in the scenarios.

In order to calculate the quantity of compost for annual application, the P_2O_5 concentration in compost is assumed to be 0.65 % d.m. This equals the median value of 17,500 compost samples in Germany (ZAS, 2002).

These assumptions lead to the following annual application rates:

(1) 30 kg P_2O_5 $ha^{-1}y^{-1}$ ➔ 4.6 t d.m. compost $ha^{-1}y^{-1}$

(2) 60 kg P_2O_5 $ha^{-1}y^{-1}$ ➔ 9.2 t d.m. compost $ha^{-1}y^{-1}$

Organic matter supply

Soil OM substitution by regular compost additions is the main beneficial effect of compost use.

The key role of organic matter, and the importance to increase it in depleted areas, is highlighted throughout the EC Communication on the Soil Strategy (COM, 2002). The EC Communication on the Soil Strategy itself refers to:

- the UNCDD (United Nation Convention to Combat Desertification), which includes both the Northern Mediterranean Countries and many of Candidate Countries in areas at risk of desertification
- the increasing occurrence of floods, landslides, or erosion, whose prevention requires proper attention to be paid to increasing OM

This is particularly true for Southern Europe and, to some extent, for Central Eastern European Countries. The potential role of the soil to act as a "sink" for carbon is now being considered a suitable strategy to tackle climate change after the 6th Conference of the Parties in Bonn and Marrakech.

The IPCC has clearly identified carbon sequestration in soils as one of carbon mitigation measures for agriculture (IPCC, 1996).

The loss of organic carbon in soils has been one of the major environmental consequences of industrial agriculture. Arable top soils in Europe commonly contain 1-3 % of organic carbon. ECAF (not dated) suggests that over about 20 years of tillage, most agricultural soils will have lost about 50% of their organic carbon. The annual net release of carbon from global agricultural activities has been estimated at about 800 Mt/year, or about 14% of current fossil fuel burning (Schlesinger, 1995).

Whereas the importance of organic matter additions is acknowledged for Mediterranean soils, not only for maintaining the yearly OM balance but also for an additional build up, the strategy of creating a C-surplus is sometime critically addressed for temperate zones (Körschens, 2003).

On the other hand long-term experiments situated in Northern Europe have also confirmed a potential decrease of OM under agricultural practices, which do not consider organic fertilisation, either through manure or through other organic fertilisers (Powlson et al., 2002).

The OM concentration in Compost is assumed as 36 % d.m. corresponding to 20.88 % C_{org} (factor: 0,58; median of 17,500 samples in Germany, ZAS, 2002).

At the 2 compost quantity levels this means an carbon input of

(1) 960 kg C_{org} $ha^{-1}y^{-1}$ at 4.6 t d.m. compost $ha^{-1}y^{-1}$

(2) 1,920 kg C_{org} $ha^{-1}y^{-1}$ at 9.2 t d.m. compost $ha^{-1}y^{-1}$

Scheffer & Schachtschabel et al. (1998) report an average C balance resulting from mean annual losses by mineralisation and leaching (ca. –3,400 kg C $ha^{-1}y^{-1}$) and inputs by crop residues (+1,000 kg C $ha^{-1}y^{-1}$) of ca. –2,100 C $ha^{-1}y^{-1}$. The balance with compost fertilisation in the model presented would result in:

balance with compost, quantity (1): 4.6 t d.m. – 1174 kg C $ha^{-1}y^{-1}$

balance with compost, quantity (2): 9.2 t d.m. – 214 kg C $ha^{-1}y^{-1}$

Nitrogen supply

An important consideration is what is the beneficial effect of compost application as far as the N supply and efficiency is concerned. It is well documented that N in compost is organically bound and therefore available to a very low extent (Nortcliff & Amlinger, 2003 Gutser, 1996; Amlinger & Götz, 1999). N-efficiency is reported as 0 – max. 20 % of the initial N input by compost in the year of application (Zwart, 2003; Berner et al., 1995; Amlinger et al., 2003; Timmermann et al., 2003). Immobilisation is frequently occurs especially at initial compost uses.

On the other hand continuous compost application may lead to a build-up of the organic nitrogen pool in the soil, which may increase N-availability over the years, depending on site-specific mineralisation dynamics.

For arable crop rotations recommendations for the total annual N-input via compost range between 80 – 100 kg ha^{-1} (for sites vulnerable to leaching and with additional mineral fertilising; Gutser, 1996) and 250 kg ha^{-1} (for silage maize production in Belgium; Nevens, 2003). The Austrian *rules of good agricultural practice* (BMLFUW, 1999) and the special programme for an *environmentally sound agriculture* (BMLFUW, 2000) acknowledge the organic N binding and set the maximum N-supply from compost at 290 kg for arable land and 350 kg for pasture land respectively.

The *total nitrogen* concentration in compost is assumed at 1.32 % d.m. and 1.9 % N (median and 90th percentile of 17,500 BWC samples in Germany; ZAS, 2002).

The potential N load from compost was estimated at two compost quantity levels – as derived from P loads - assuming the N concentration levels in compost:

Table 9-3: Potential N loads in the used models at two levels of N concentrations in composts

	N concentration in compost *[% d.m.]*	
Compost quantities:	1.32 % d.m. = Median; DE	1.9 % d.m. = 90th percentile; DE
4.6 t d.m. compost ha^{-1}y^{-1}	60.7 kg ha^{-1}	87.4 kg ha^{-1}
9.2 t d.m. compost ha^{-1}y^{-1}	121.4 kg N ha^{-1}	174.8 kg N ha^{-1}

This shows that with the quantities adopted for the calculations the potential nitrogen load will not be a limiting parameter.

Assuming P as a limiting factor especially in the case of the scenario “4.6 t” the need of OM as calculated above is higher than the resulting limit load.

This underlines the importance of a flexible approach whereby various limiting factors, or on the contrary, operational reasons affecting loads of soil improvers, have to be considered. This will be particularly true in the case of Nitrogen whose limiting factor at 170 kg ha^{-1}, taken from the Nitrogen Directive for sensitive sites, might constitute a obstacle to a proper farming pattern in areas where an increase of OM is needed in order to archive a proper performance of soil functions.

Concentration rate of PTEs in soil after compost application due to further mineralisation of the organic compost substance

A further assumption in our calculation model concerns the mineralisation rate of compost once it is applied on soil. The higher the mineralisation is, the less is the remaining proportion of compost incorporated and, at the same proportion, the higher is the accumulation factor or concentration rate PTEs.

Some authors calculated that up to 8 % of the organic carbon applied with compost would remain in the soil on a mid to long-term basis (Bannick et al., 2002; Smith et al. 2001). This of course is essential for the maintenance and build-up of the soil carbon sink and thus a well-designed regular compost management may play an important role in terms of climate change. Respecting a more careful approach towards a reliable accumulation scenario for PTEs we assume 6 % retainment factor for the organic matter (OM) which accumulate together with the mineral substance applied with compost. At an estimated OM level of 36 % d.m. this equals a mineralisation rate of 30 % relative to the total dry matter mass. In other words 70 % of the total dry matter would remain in the soil. This accounts for the resulting accumulation of the PTE concentration in the compost on dry matter basis. Accordingly, the increase of soil mass was set to 70 % of the annually added compost dry matter mass. Therefore the calculation is always related to the soil mass at the assumed soil depth (20 or 30 cm) plus 70 % of the yearly applied compost mass. That is 6.44 t in case of a yearly compost application of 9.2 t ha^{-1}y^{-1}.

Summary of assumptions

Table 5-22 summarises assumptions already specified and further ones made for the accumulation model of PTEs in the soil.

Table 9-4: Assumption for heavy metals used in accumulation scenarios for heavy metals

			Cd	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
Atmospheric deposition	(1)	g ha^{-1} y^{-1}	2.315	15.2	87.45	0	44.9	59	332.6
Export									
Export via leaching	(2)	g ha^{-1} y^{-1}	- 0.28	- 9.2	- 8	- 0.28	- 17.8	- 0.56	- 38
Export via harvest	(3)	g ha^{-1} y^{-1}	- 0.67	- 5.27	- 33.92	---	- 10.29	- 5.92	- 172.9
Total export		g ha^{-1} y^{-1}	- 0.95	- 14.47	- 41.92	- 0.28	- 28.09	- 6.48	- 210.9
Background value soil as base line for the accumulation of PTEs in the soil									
sandy soils	(4)	mg kg^{-1}dm	0.13	7.65	7.27	0.04	6.6	17.57	26.9
clay soils	(4)	mg kg^{-1}dm	0.27	22.45	16.2	0.06	22.53	24.83	61.57
Heavy metal concentration in compost									
Weighted median BWC	(5)	mg kg^{-1}dm	0.50	23.0	45.1	0.14	14.1	49.6	183
Weighted 90%ile BWC	(6)	mg kg^{-1}dm	0.87	39.9	73.9	0.30	27.0	87.6	276
Quantity of compost per ha and year applied									
		P_2O_5 compost: 0.65 % d.m.			max. **30 kg P_2O_5 ha^{-1} y^{-1} → 4.6 t d.m. compost ha^{-1}**				
					max. **60 kg P_2O_5 ha^{-1} y^{-1} → 9.2 t d.m. compost ha^{-1}**				
Soil depths and density		soil density: 1,5 g cm^{-3}							20 cm; → **3,000 t ha^{-1}**
									30 cm; → **4,500 t ha^{-1}**
Time frame for the accumulation model									**200 years**

(1) taken from the latest investigations on the atmospheric deposition of PTEs in the east and south part of Austria from 10 locations (Böhm & Roth, 2000 and 2001)

(2) average total export of heavy metals via leachate water (at a percolation rate of 200 mm) taken from figures in Germany (Bannick et al., 2001):

Concentration in the leachate water *[µg l^{-1}]*	**Cd**	**Cr**	**Cu**	**Hg**	**Ni**	**Pb**	**Zn**
	0.14	4.6	4	0.14	8.9	0.28	19

(3) average total export of heavy metals via harvest (cereals, maize, sugar beet, potatoes) taken from figures in Germany (Bannick et al., 2001)

(4) mean values of sand and clay soils of the country surveys from DK, FR and DE; see also Table 3–3 in chapter 3; latest Version of the report "Trace elements and organic matter content of European soils" are available on the web page http://europa.eu.int/comm/environment/waste/sludge/index.htm

(5) average of median (mean) values from compost surveys (*biowaste composts*) in seven European countries (see Table 9-2).

(6) average of 90th percentile values from compost surveys (*biowaste composts*) in seven European countries (see Table 9-2).

9.3 Results of the accumulation scenarios

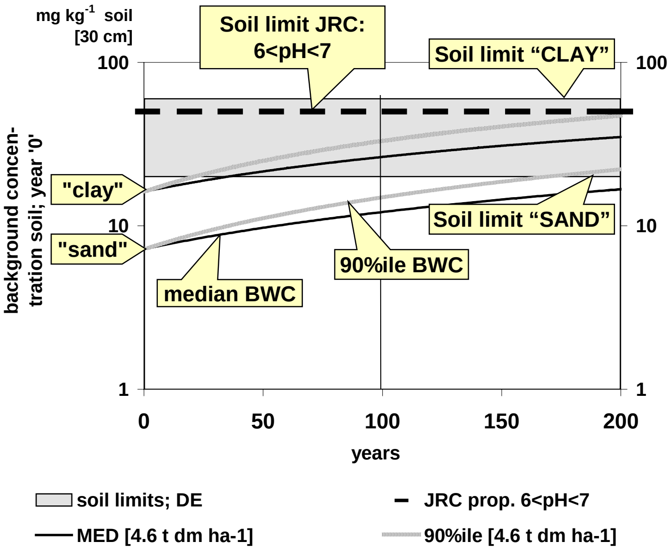
Fig. 9-2 shows the accumulation curve for seven PTEs. The amount of annual compost applications – as calculated by limit P loads – is 4.6 t and 9.2 t d.m. ha^{-1} presented in the left and right column of graphs respectively.

The time frame is 200 years. From the computation of accumulation in a certain soil layer it is evident that the graph is not linear and it would approach an asymptotic curve in relation to the element concentration in compost, taking into account the site and management specific long-term mineralisation rate of the compost.

The lower and the higher starting point (background concentrations) of the accumulation graphs represent sandy and clay soil background values. Accordingly, the lower threshold value (Soil limit "SAND"; see Table 9-1) refers to the graphs **"sand"** and the upper threshold of the shaded range (Soil limit "CLAY"; see Table 9-1) refers to the graphs **"clay"**.

The two PTE concentration levels (weighted median and 90th percentile) were computed for each of the soil types ("sand" and "clay")

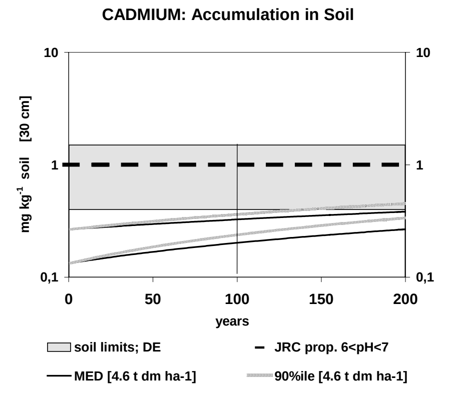
Example graph:

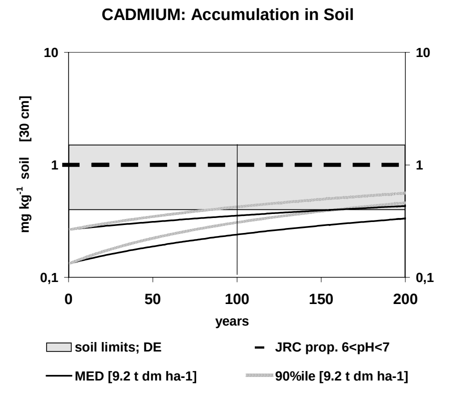


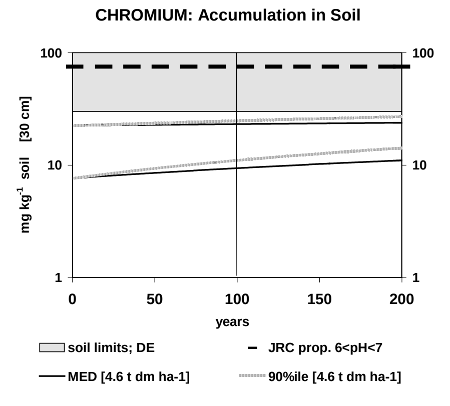
The graphs in Fig. 9-2 are shown for a 30 cm "active" soil layer (the layer into which compost is ploughed).

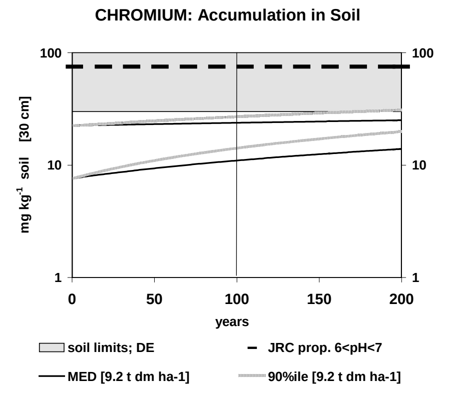
Annual application: 4.6 t d.m. ha^{-1} ▼

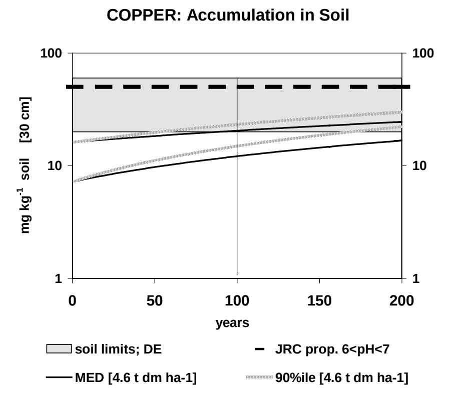
Annual application: 9.2 t d.m. ha^{-1} ▼

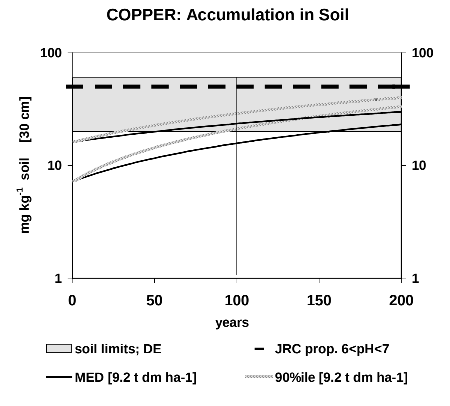


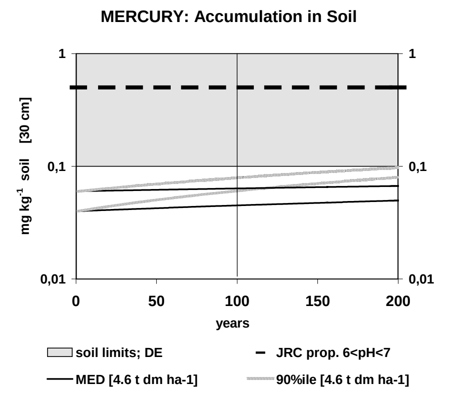


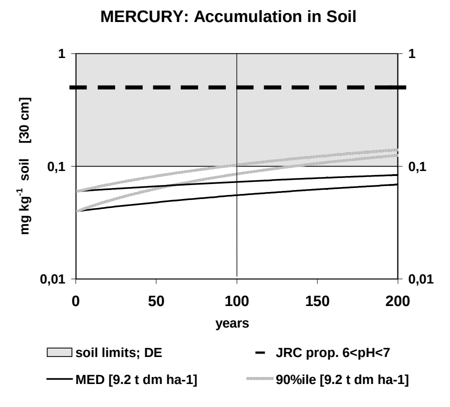












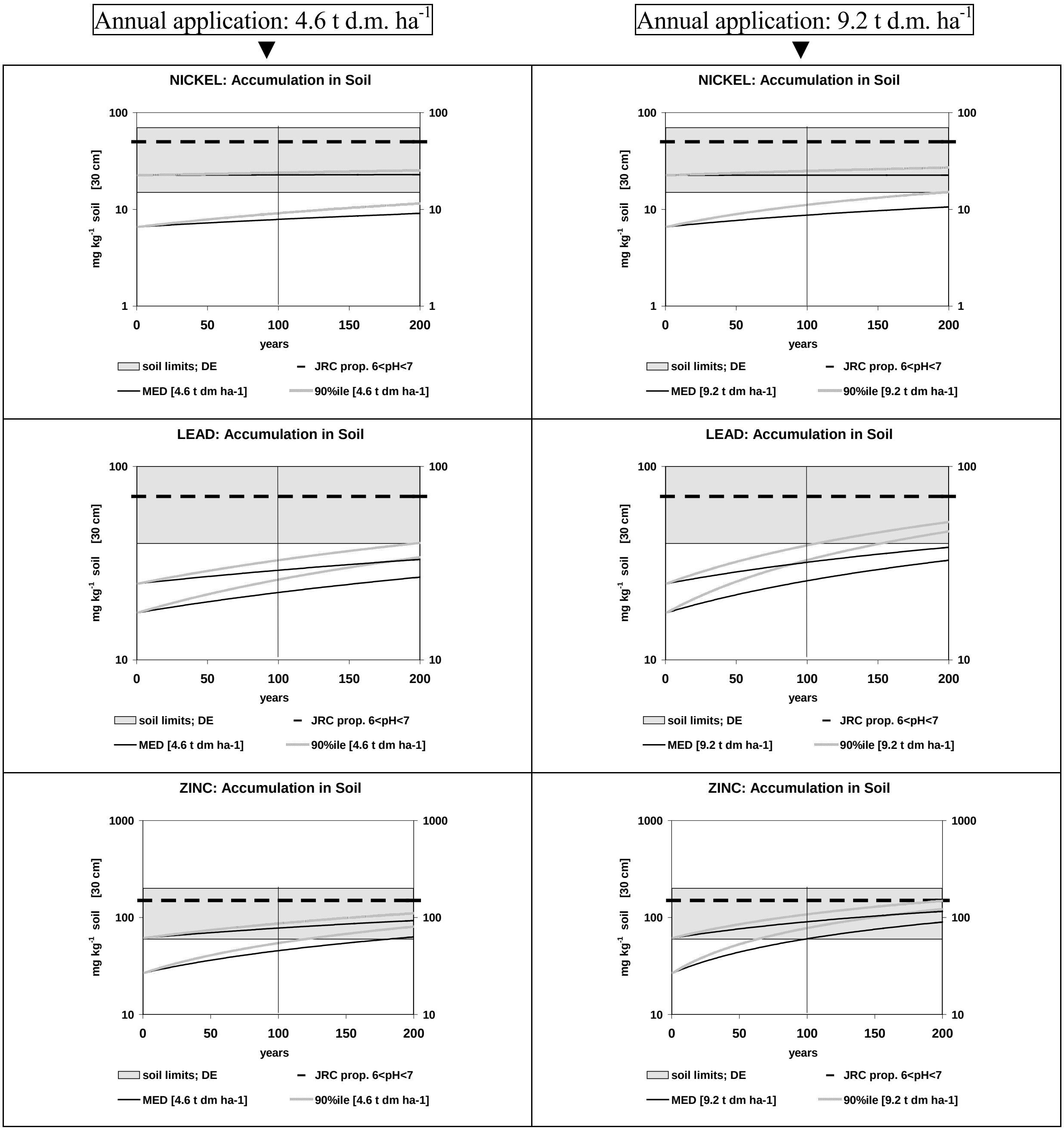


Fig. 9-2: Accumulation scenarios for heavy metals with median and 90th percentile PTE concentrations in European composts

Table 9-5, Table 9-6 and Table 9-7 provide some more detailed information on consequences which the model predicts for the more sensitive assumption made for sandy soils. To further round out Fig. 9-2, numbers for a soil depths of 20 cm are added.

Table 9-5: Soil PTE concentration after 200 years at two rates of annual compost applications, two soil depths and two concentration levels in compost in the scenario on sandy soils *[mg kg^{-1}]*; *bold numbers: > threshold value "SAND"*

	Threshold value soil **"SAND"** *[mg kg^{-1}]*	Concentration in SOIL (sand) in year "0" *[mg kg^{-1}]*	Concentration in COMPOST *[mg kg^{-1}]*	**Concentration in soils after 200 years**			
				4.6 t d.m. compost ha^{-1}y^{-1} = 30 kg P_2O_5		**9.2 t** d.m. compost ha^{-1} y^{-1} = 60 kg P2O5	
				Soil depth:			
				20 cm	30 cm	20 cm	30 cm
Cd	0.40	0.13	(a) 0.50	0.33	0.27	**0.41**	0.33
			(b) 0.87	0.43	0.34	**0.59**	**0.46**
Cr	30	7.65	(a) 23.0	12.54	11.02	16.45	13.92
			(b) 39.9	17.18	14.23	24.83	19.90
Cu	20	7.27	(a) 45.10	**20.98**	16.73	**29.58**	**23.18**
			(b) 73.9	**28.88**	**22.19**	**43.87**	**33.37**
Hg	0.10	0.04	(a) 0.14	0.05	0.05	0.08	0.07
			(b) 0.30	0.10	0.08	**0.16**	**0.13**
Ni	15	6.6	(a) 14.1	10.21	9.09	**12.21**	**10.60**
			(b) 27.0	**13.75**	**11.54**	**18.61**	**15.17**
Pb	40	17.57	(a) 49.5	30.92	26.78	38.87	32.75
			(b) 87.5	**41.36**	33.99	**57.72**	**46.20**
Zn	60	26.9	(a) 183	**79.26**	**63.03**	**114.94**	**89.67**
			(b) 276	**104.80**	**80.66**	**161.09**	**122.58**

Table 9-6: Years until threshold values for SANDY soil of the German Soil Protection Ordinance are met at two rates of annual compost application, two soil depths and two PTE concentration levels in compost (scenario "SAND"); *bold numbers in shaded cells: < 100 years; bold numbers in blank cells: < 200 years;*

				Years when threshold values "SAND" are met			
	Threshold value soil **"SAND"** *[mg kg^{-1}]*	Concentration in SOIL (sand) in year "0" *[mg kg^{-1}]*	Concentration in COMPOST *[mg kg^{-1}]*	**4.6 t** d.m. compost ha^{-1}y^{-1} = 30 kg P_2O_5		**9.2 t** d.m. compost ha^{-1} y^{-1} = 60 kg P_2O_5	
				Soil depth:			
				20 cm	30 cm	20 cm	30 cm
Cd	0.40	0.13	(a) 0.50	283	423	**188**	281
			(b) 0.87	**176**	263	**104**	**156**
Cr	30	7.65	(a) 23.0	>500	>500	>500	>500
			(b) 39.9	>500	>500	282	422
Cu	20	7.27	(a) 45.10	**185**	276	**104**	**156**
			(b) 73.9	**113**	**169**	**62**	**92**
Hg	0.10	0.04	(a) 0.14	>500	>500	300	448
			(b) 0.30	**189**	283	**82**	**123**
Ni	15	6.6	(a) 14.1	>500	>500	343	>500
			(b) 27.0	240	359	**131**	**196**
Pb	40	17.57	(a) 49.5	365	>500	214	320
			(b) 87.5	**188**	281	**102**	**152**
Zn	60	26.9	(a) 183	**122**	**183**	**67**	**99**
			(b) 276	**81**	**121**	**43**	**64**

Table 9-7: Critical limit concentrations in compost when threshold values for SANDY soil of the German Soil Protection Ordinance are met within 100 or 200 years respectively *[mg kg^{-1} d.m.]*;
bold numbers in shaded cells: critical value < median value of European BWC; bold numbers in blank cells: critical value < 90th percentile of European BWC

	Threshold value soil **"SAND"** *[mg kg^{-1}]*	Concentration in SOIL (sand) in year "0" *[mg kg^{-1}]*	Years until soil threshold values are may be met	**Critical concentration in compost *[mg kg^{-1} d.m.]***			
				4.6 t d.m. compost ha^{-1}y^{-1} = 30 kg P_2O_5		**9.2 t** d.m. compost ha^{-1} y^{-1} = 60 kg P_2O_5	
				Soil depth:			
				20 cm	30 cm	20 cm	30 cm
Cd	0.40	0.13	(a) 100	1.6	2.50	0.93	1.35
			(b) 200	**0.75**	1.18	**0.48**	**0.70**
Cr	30	7.65	(a) 100	161	234	87.07	123.80
			(b) 200	86.5	123.1	50.32	68.43
Cu	20	7.27	(a) 100	84	126	**46.73**	**67.65**
			(b) 200	**41.6**	**62.5**	**25.78**	**36.12**
Hg	0.10	0.04	(a) 100	0.47	0.70	**0.28**	0.40
			(b) 200	**0.29**	0.40	**0.18**	**0.22**
Ni	15	6.6	(a) 100	59.3	87	33.5	47.4
			(b) 200	31.5	45.3	**19.7**	**26.6**
Pb	40	17.57	(a) 100	158	231	88.68	125.55
			(b) 200	**83**	119.5	**51.79**	**69.98**
Zn	60	26.9	(a) 100	223	361	**126.7**	**181.1**
			(b) 200	**113**	**167**	**72.28**	**99.13**

The results of Fig. 9-2, Table 9-5, Table 9-6 and Table 9-7 and the underlying calculations may be summarised as follows:

- Chromium will not reach the soil limits under any of the assumed scenarios.
- Starting at mean background concentration for *clay soils* (taken from soil survey from three European countries), the German soil threshold values "CLAY" and the values proposed by the JRC in the range 6<pH<7 will – with the exception of Zn – not be exceeded up to 200 years of compost application. For the most precautionary assumptions (soil depth of 20 cm and 9.2 t d.m. ha^{-1} y^{-1} compost) Zn will reach 150 mg kg^{-1} after 137 years at median compost concentrations.
- After 100 years (mainly with high application rates of 9.2 t ha^{-1}y^{-1} and at the worst compost quality) only Cu, Hg (when calculated to 20 cm soil depth) and Zn exceed the limit values for sandy soils.
- If it is assumed that target values of sandy soils must not be exceeded within 100 years, allowable PTE concentrations could – with the exception of Zn – be even higher than the median values of European BWC. This is valid even if 9.2 t compost per ha and year are applied.
- In contrary, allowable concentrations for Zn and Cu fall below the median values of European BWC when the scope is set to 200 years without exceeding assumed target values for soils. This

occurs for Zn both with the "high application" (9.2 t) and the "low application" (4.6 t compost) rates, whereas for Cu it only occurs at 9.2 t $ha^{-1}y^{-1}$ and in the case of a 20 cm soil horizon.

- In the case of sandy soils, Cu at high application rates and Zn even at the low application rates must be considered the most critical elements compared with achievable BWC quality. However, their function as trace elements essential for plant nutrition has to be taken into account in the overall fertiliser regime.
- Significantly higher concentrations in composts for the elements Cu and Zn (and to a lesser degree for Ni, Pb and Cd) than the 90th percentile assumed in the model calculation might require a site specific evaluation if such composts are to be used continuously in the long run.

9.4 Possible approach towards threshold values for Composts used in Agriculture

Limit values for composts, intended for the use in food and fodder production make sense only if they do not set unreasonably high standards for compost qualities.

On the other hand they have to fulfil the task of minimising risks to the environment while taking into account the agronomic needs for the benefits of soil-plant system.

Regarding temporal and regional variations as well as the variation within an individual composting plant, setting a tolerance factor in addition to the calculated 90th percentile value of European biowaste composts would be a reasonable way feasible.

In Germany, the seasonal coefficient of variation in composting plants averages 28% and ranges up to 50-60% (Kranert, 2002; see also chapter 4). In addition, unavoidable deviations of incremental samples within one compost batch range between ±30 and ±40 % from the mean value. The highest individual variations were found for Cd (100 %) and Lead (73 %) (Breuer et al., 1997; see chapter 7.4.2)

Comparing the statistically weighted 90th percentiles on EU level to two countries in the starting phase of source separation, Spain and the UK, the PTE concentration of the latter is increased by 7 % (Cr) to 56 % (Ni).

Based on these considerations a 50 % tolerance is added to the 90th percentile value of European BWC and the averaged 90th percentile of ES and UK composts. These concentration values are named [level 1] and [level 2] respectively. Table 9-8 gives a comparison of the resulting concentrations with the proposed limit values of the Working Document "Biological treatment of biowaste" (2nd draft) and the averaged limit values for BWC in European countries.

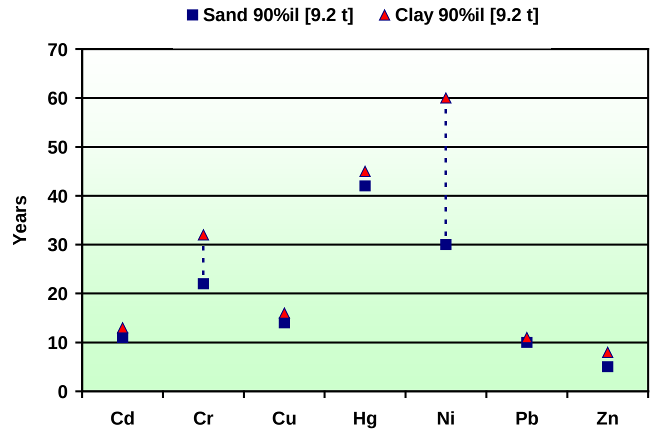
As compared to *class 1 WD 2nd Draft* the calculated [level 1] achieves higher values in the case of Cd, Pb and Zn, lower ones for Cr and Ni and approximately the same level for Cu and Hg.

A similar relation is given for the comparison of [level 2] and *class 2 WD 2nd Draft*. Only for Zn the distance is decreased.

Table 9-8: Possible limit value for compost derived from the accumulation scenarios for sandy soils (mg kg^{-1} d.m.)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
90%ile EU all		0.87	39.9	73.9	0.30	27.0	87.6	276
90%ile UK & ES		1,27	42,8	113,1	0,43	42,1	133,4	314
Difference [(UK&ES)-EU]/EU (%)		+46	+7	+53	+43	+56	+52	+14
90%ile EU + 50 %	**[level1]**	**1.3**	**60**	**110**	**0.45**	**40**	**130**	**400**
90%ile UK & ES + 50 %	**[level2]**	**1,9**	**64**	**170**	**0,65**	**63**	**200**	**470**
Averaged limit values of EU countries		1.4	93	143	1.0	47	121	416
WD 2nd Draft [1]	class 1	0.7	100	100	0.5	50	100	200
	class 2	1.5	150	150	1	75	150	400

[1] EU Commission, DG Environment. Working Document (WD), 2nd draft: "Biological treatment of biowaste"



start soil sand clay	0.13 0.27	7.65 22.45	7.27 16.2	0.04 0.06	6.6 22.53	17.57 24.83	26.9 61.57
Min. measurable increase	0.02	1.5	2.0	0.02	1.5	1.5	2.5
90th percent. compost	0.87	39.9	73.9	0.30	27	87.5	276

Sand...scenario „sand" with 9.2 t compost d.m. ha^{-1}y^{-1}
Clay ... scenario „clay" with 9.2 t compost d.m. ha^{-1}y^{-1}
90th percent. … 90th percentile of PTE concentrations in European BWC

Fig. 9-3: Time in years until a measurable increase of PTEs in the soil concentration can be analysed

Fig. 9-3 demonstrates the period until an analytical detectable increase may be measured when compost of a certain quality is applied. As a precaution, the heavy metal concentrations assumed are the 90th percentile of the European BWC. Depending on the *distance* from background levels of the soil (*"sand"* and *"clay"*), an increase in heavy metal contents in soil to an analytically detectable extent may be expected within a period of 5 (Zn) to 60 (Hg) years.

The accumulation scenarios for [level 1] and [level 2] PTE concentrations under the assumed management and site conditions are presented in Fig. 9-4.

Table 9-9 shows the range of increase of the elements after a period of 10 years differentiated for the two soil types "sand" and "clay".

Table 9-9: Ranges of calculated increase of PTEs in the soil after a period of 10 years of compost application at assumed PTE concentrations of [level 1] and [level 2], differentiated for the soil background concentrations "sand" and "clay"

4.6 and 9.2 t d.m. ha^{-1}; soil depth: 30 cm;	Cd	Cr	Cu	Hg	Ni	Pb	Zn
			------------------ *mg kg^{-1}* ------------------				
Scenario "sand"	0.02-0.04	0.5-1.62	1.05-3.1	0-0.01	0.36-1.1	1.18-3.43	3.74-8.48
Scenario "clay"	0.01-0.03	0.41-0.88	1.18-3.17	0-0.01	0.26-0.9	1.14-3.44	3.51-8.04

These figures show that, within the time frame of 10 years, a measurable increase would hardly be found for both the "sand" and the "clay" scenarios even when assuming those relatively high PTE concentrations as compared to the mean compost quality.

Once we consider concurrently all the assessments in previous sections, we may consider the adoption of scenarios which aim at a very slow input of PTEs to the soil which, at the same time do not constitute risks in the time horizon of technological improvement. As stated before, this is an approach which

- does not hinder beneficial use of organic matter and nutrients in compost and
- constitutes a driver for an overall technological improvement of industrial processes over time, in order to have a steady improvement of quality of composts, feedstock, and overall emissions to the environment.
- The latter is unlikely to happen in the case of a regulatory approach, which would imply cutting off any possible application for much of the composted products.

Thus a more flexible approach posits a certain time horizon during which actions should be taken to reduce environmental pollution from industrial processes.

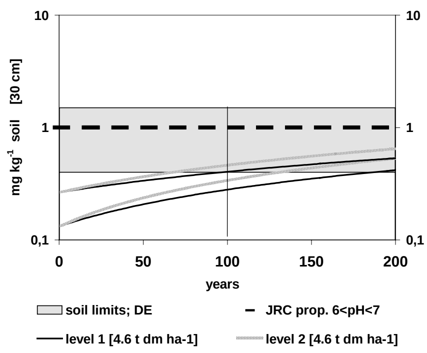
At the same time, the increase of concentration of PTEs due to compost application should be kept by far within safe limits, given:

- Compost loads applied according to good agronomic practice and/or mandated limit loads
- Anticipated concentrations of PTEs in composts (which is the main scope of the investigation in this section)

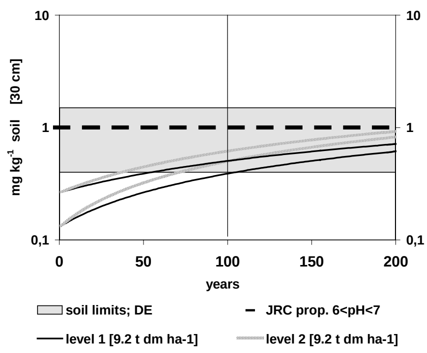
See graphs of Fig. 9-4 giving the accumulation of PTEs in soil for composts at the proposed two limit concentrations.

Annual application: 4.6 t d.m. ha^{-1} ▼

Annual application: 9.2 t d.m. ha^{-1} ▼



CADMIUM: Accumulation in Soil



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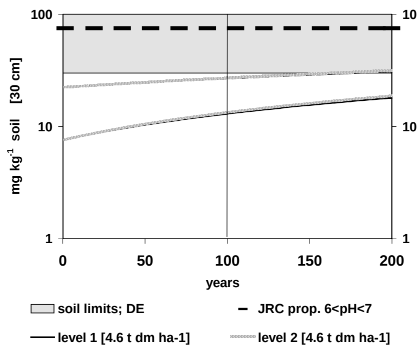
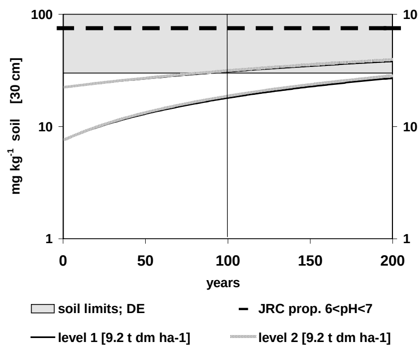


CHROMIUM: Accumulation in Soil



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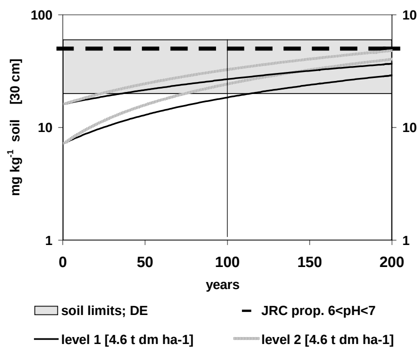
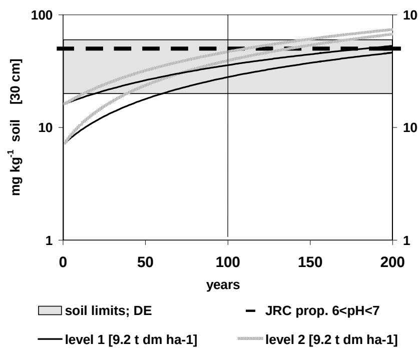


COPPER: Accumulation in Soil



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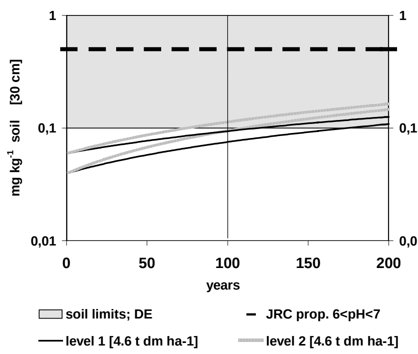
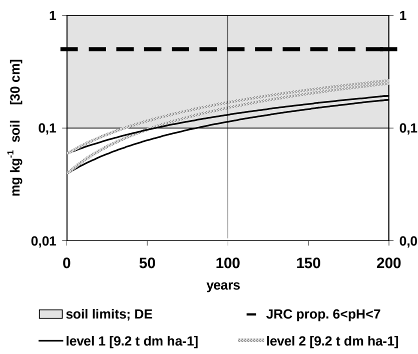


MERCURY: Accumulation in Soil



MERCURY: Accumulation in Soil

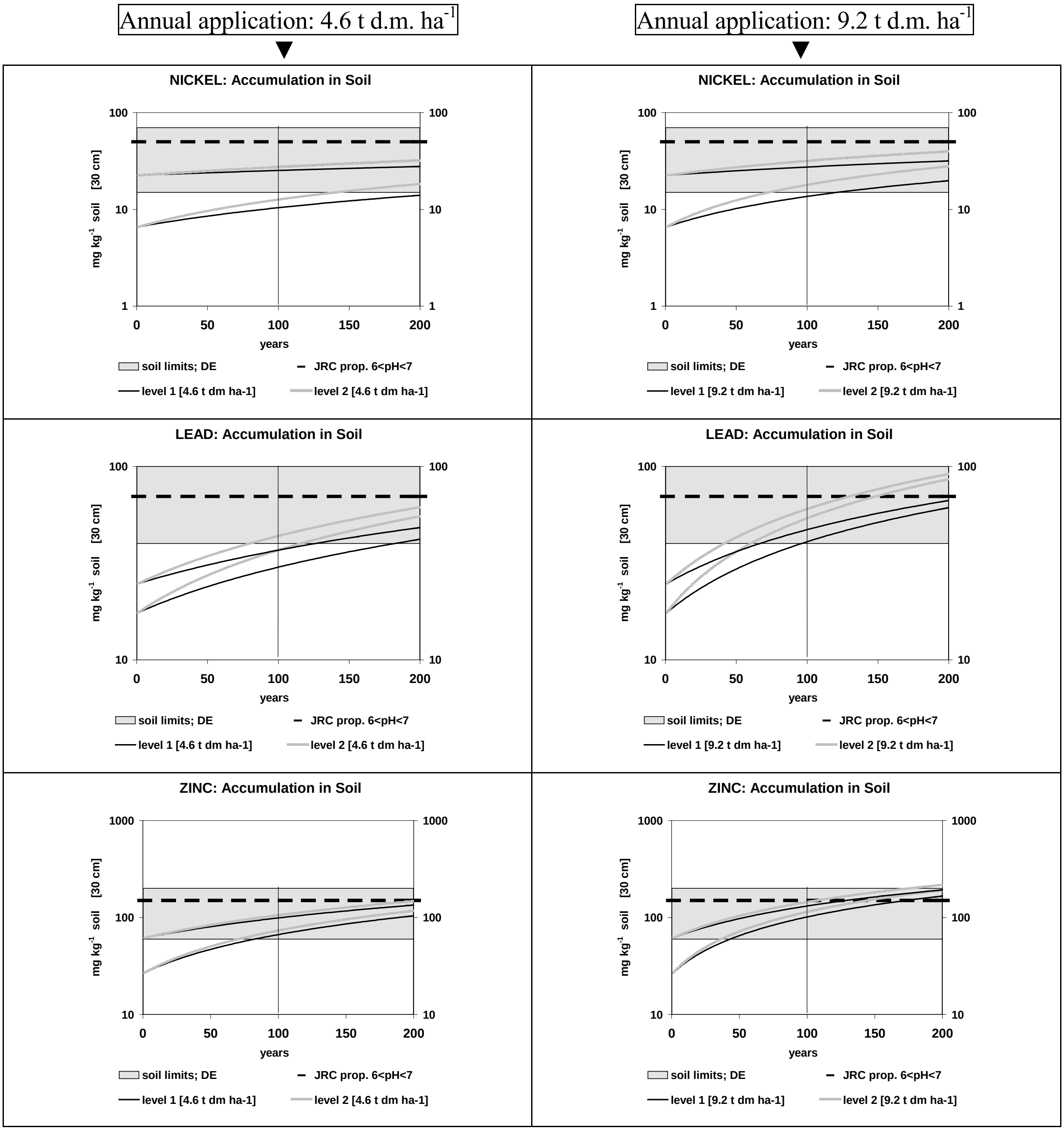


Fig. 9-4: Accumulation scenarios for PTEs at [level 1] and [level 2] PTE concentrations in composts following the assumptions given in Table 5-22 and Table 9-8.

<u>From the above it can be concluded:</u>

[B] Scenario findings and justification of the proposed approach towards <u>threshold values for composts used in agriculture</u>:

- The regional and seasonal variations of Biowaste and Green Composts are based on unavoidable background concentrations which fluctuate even in similar collection schemes and purity levels.
- It has to be recognised that due to specific regional background concentrations, the 90th percentile of country/Europe-wide produced composts may actually represent the average quality of composts achieved at a single composting plant in a certain region

- Countries in the starting phase of separate collection systems show 10 to 50 % higher PTE concentrations in Biowaste Compost and Green Compost. This has to be considered in the light of (i) the still limited data set and the comparatively low reliability of sampling/testing methods (ii) the inability to distinguish between facilities producing compost from biowaste only and those also including sludge (frequently considered jointly in available statistics as "source selected materials").
- Based on the evaluation of material specific, regional and temporal variability a 50 % tolerance was added to the 90th percentile value of European BWC [level 1] and the averaged 90th percentile of ES and UK composts as representatives for *starting phase countries* mentioned above [level 2]. Table S1 gives a comparison of the resulting concentrations with the proposed limit values of the Working Document "Biological treatment of biowaste" (2nd draft) and the averaged limit values for BWC in European countries.

Table S1. Possible approach for compost limit values derived from the accumulation scenarios for sandy soils (mg kg^{-1} d.m.)

		Cd	Cr	Cu	Hg	Ni	Pb	Zn
90-percentile EU all		0.89	37.4	79.5	0.35	29.7	105.2	284.2
90-percentile UK & ES		1.27	42.8	113.1	0.43	42.1	133.4	314.0
Difference [(UK&ES)-EU]/EU (%)		+47	+7	+53	+43	+56	+52	+14
90-percentile EU + 50 %	**[level1]**	**1.3**	**60**	**110**	**0.45**	**40**	**130**	**400**
90-percentile UK & ES + 50 %	**[level2**	**1.9**	**64**	**170**	**0.65**	**63**	**200**	**470**
Averaged limit values of EU countries		1.4	93	143	1.0	47	121	416
WD 2nd Draft 1)	class 1	0.7	100	100	0.5	50	100	200
	class 2	1.5	150	150	1	75	150	400

1) EU Commission, DG Environment. Working Document (WD), 2nd draft: "Biological treatment of biowaste"

- [level 1] can be recommended as general threshold for sustainable, regular use of compost in food and feeding stuff production as well as hobby gardening.
- In order to guarantee comparability between well "stabilised" or mineralised composts and "young" ones (in which concentration of PTEs due to the loss of *organic matter* has not fully occurred, yet) measured PTE concentrations should be standardised to a fixed organic matter content (e.g. 35 or 40 % d.m.). This would be a fair proof of compliance with set limit values and balances the sometimes highly varying degree of mineralisation in composted materials.
- In any case, the calculated increase of PTE concentrations in the soil due to annual compost applications is hardly detectable by analytical means prior to 10 years of application.
- The allowance of a transition period of about 10 to 15 years for the use of [level 2] composts (see Table S1) in food and feeding stuff production may be acceptable from the viewpoint of environmental and soil protection. This would be a viable way in order to
    - allow countries in the starting phase of separate collection systems to improve collection schemes and quality management during the implementation phase;
    - give enough time to improve – consistently with the "Programmes for the prevention of pollution" envisaged by last working documents for the revision of the Sludge Directive – sludge qualities from industrial and urban waste water treatment for the use as feedstock of composting.
- However, when sewage sludge or animal manure (specifically pig manure) is used as source material, PTE concentrations of [level 2] could in some cases not be met in the case of Cu and Zn. This requires further efforts for improvement of quality at the source.
- In terms of practicability and a flexible application system, applied quantities may be seen as average yearly dosage within a time span of 20 years. In other words for land reclamation projects for the restoration of degraded soils a maximum incorporation of approximately 200 tonnes per hectare should be granted under consideration of all relevant environmental aspects (i.e. water conservation etc.)
- The graphs presented in chapter 9 of the study show that even at high application rates (9.2 t d.m. ha^{-1}) [level 1; see Table S1] composts would not lead to an accumulation beyond soil reference concentrations for clayey soils within 200 years. The JRC proposal would only be exceeded within 130 years in the case of Zn.

- Thus, for clayey soils, there is still a comparatively wide scope and flexibility for the use of compost. This might be of special importance where a distinct need for the amelioration of degraded soils is recognised (for example, in Mediterranean soils).

- The time span after which reference values for sandy soils would be exceeded when using composts of both proposed qualities [level 1 and 2] is shown in Table S2.

Table S2. Years until precautionary threshold values for sandy soils* are exceeded**

PTE concentration according to:	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				years			
[Level 1]	103	237	60	74	122	97	44
[Level 2]	70	218	39	53	73	61	37

* taken from the German Soil Protection Ordinance

** at 9.2 t compost $ha^{-1}y^{-1}$

- It is worth noting that at the proposed levels of threshold concentrations, the limiting factor for compost application is still the phosphorus supply rather than the PTE loading. This is particularly true in Central and Northern European Countries, where specific regulatory provisions are being implemented on P supply.
- Some open issues for research and monitoring may be defined, along the following lines:

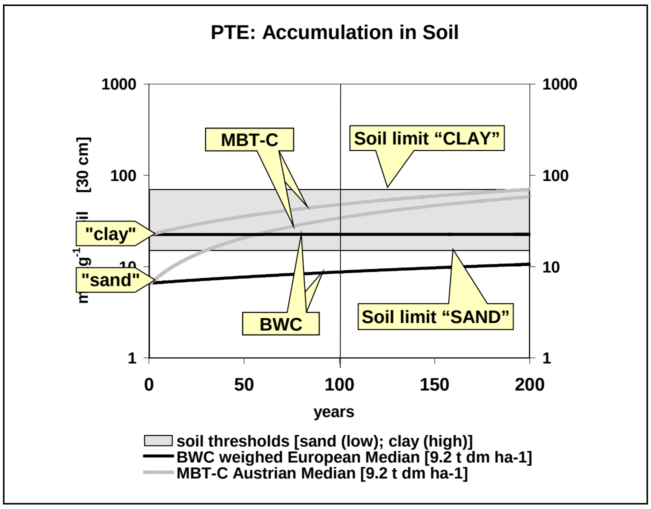
iii. Soil concentrations of Cu, Hg and Zn should be regularly monitored on sandy soils which show *high background concentrations* every 100 to 150 tons of compost application. The definition of high soil background concentration should be made by local authorities based on national soil surveys.

iv. Monitoring and research of PTE availability/solubility/mobilisation within pilot schemes with plots fertilised with compost is an important tool for further evaluation of potential impacts due to the input of contaminants by fertilisation systems. It would be of great advantage to establish a cooperative research and monitoring network which integrates existing as well as new field trials covering the most important climate, soil and management conditions for a better understanding of the dynamics of the elements and substances concerned in the soil-plant-groundwater system. In any case, the slow accumulation profile caused by the (comparatively) low thresholds made possible by source separation, allow for the possibility of even long-term research programmes, before any risk can be envisaged.

9.5 A comparative view on accumulations scenarios with sewage sludge and municipal solid waste compost

9.5.1 Accumulation of heavy metals after regular application of stabilised residual waste (MSW compost)

Fig. 9-5 shows the result of an input/output balance comparing the regular application of biowaste composts (BWC) against residual solid waste compost (MSW-C) form mechanical biological treatment. The assumptions for soil back ground concentrations, precautionary soil threshold values, heavy metal concentration in Biowaste, export by harvest and leaching and input by deposition are the same as considered above:



MSW-C: MEDIAN of Austrian residual waste compost stemming from mechanical biological treatment plants:

Cd	Cr	Cu	Hg	Ni	Pb	Zn
mg kg^{-1} d.m.						
2.7	209	247	1.3	149	224	769

Application rate: 9.2 t d.m. ha^{-1}y^{-1} (according to approx. 60 kg P2O5 ha^{-1}y^{-1})
Soil depth: 30 cm = 4500 t ha^{-1}

The outcome for the more critical scenario on sandy soils is shown in Table 9-10.

Table 9-10: Years until precautionary threshold values for sandy soils are exceeded. Comparison between Biowaste compost and compost derived from biological treatment of residual mixed waste *

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
				years			
Biowaste composts	281	>500	156	448	>500	319	100
MSW compost	50	57	27	25	30	54	23

* However, time span may vary when the assumptions of soil background concentrations are changed

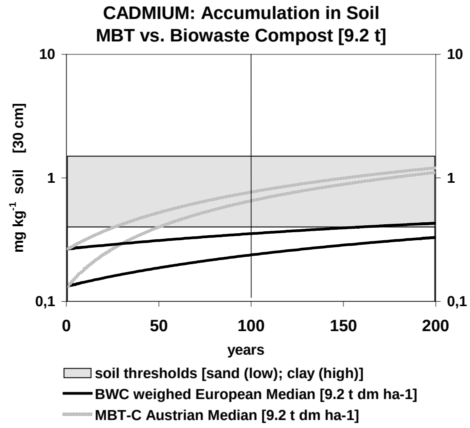
Taking the German soil thresholds as reference it is evident that with actual biowaste qualities no problem occur even in the long run. The situation is completely different for MSW or MBT-Compost.

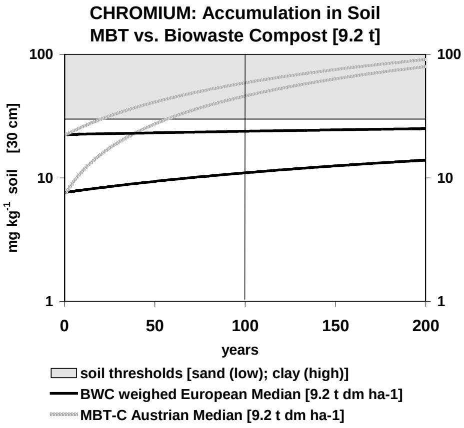
The main pressure is given for Cu and Zn on clay and sandy soils, for sandy soils also for Hg and Ni because of the exceeding of soil thresholds in less than 30 years.

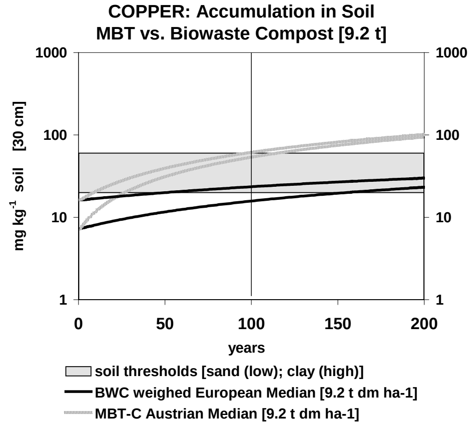
It can be clearly seen that, even considering modern splitting and separation technologies via MBT the accumulation rates caused by stabilised residual waste would not satisfy the needs of precautionary and thus sustainable soil management.

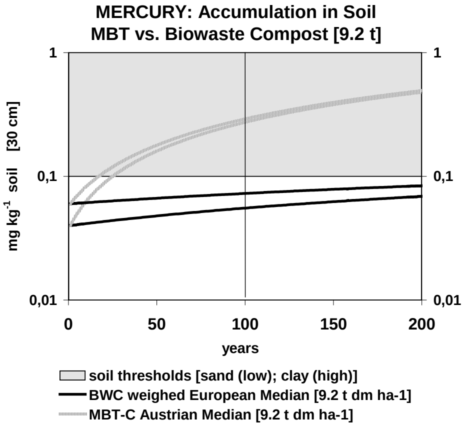
Thus creating drivers for the separate collection of organic waste from industries and households can be addressed not only as a matter of waste management but is definitely affecting the demands of soil protection.

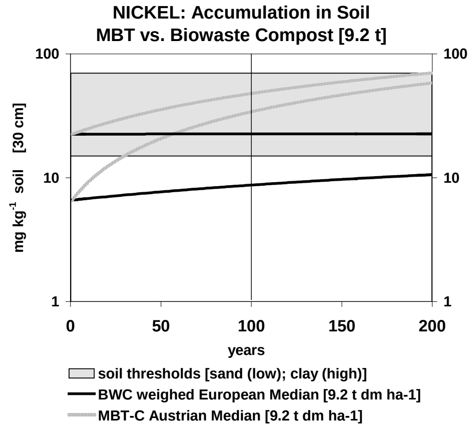
This is the only guarantee to maintain a consequent high level of quality of composts used as organic soil improver under the manifold geographical and infrastructure conditions.

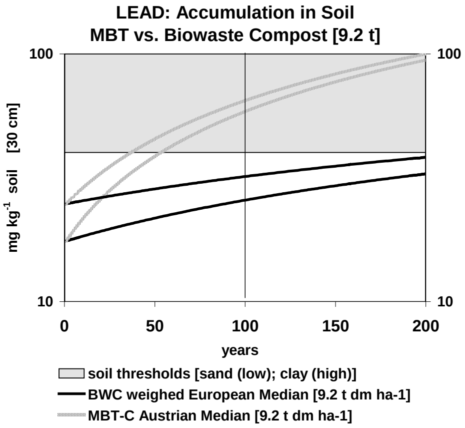












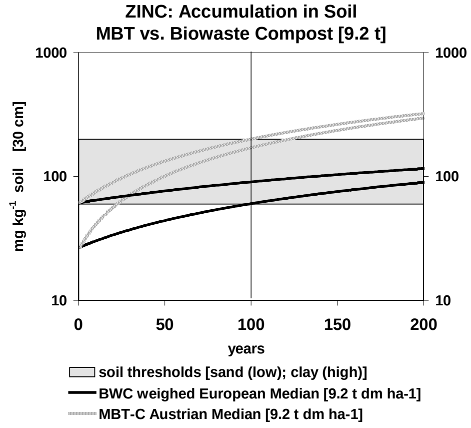


Fig. 9-5. Comparative accumulation of heavy metals due to the regular use of MBT Compost compared to Biowaste Compost (BWC)

- 2 scenarios for sandy and clay soils respectively
- PTE concentration MBT-C = MEDIAN [AT]
- PTE concentration in BWC = MEDIAN [EU]
- Further underlying assumptions see above.

9.5.2 Accumulation of heavy metals after regular sewage sludge application

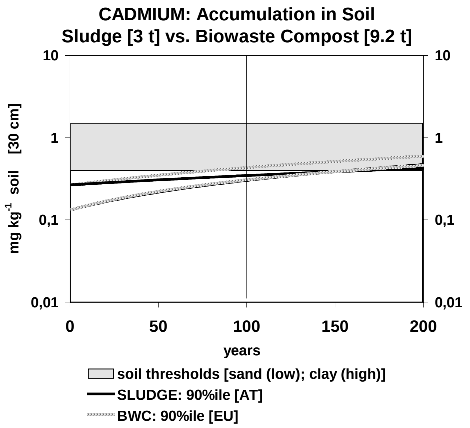
For comparison an additional plot scenario was computed for sewage sludge (see Fig. 9-6)

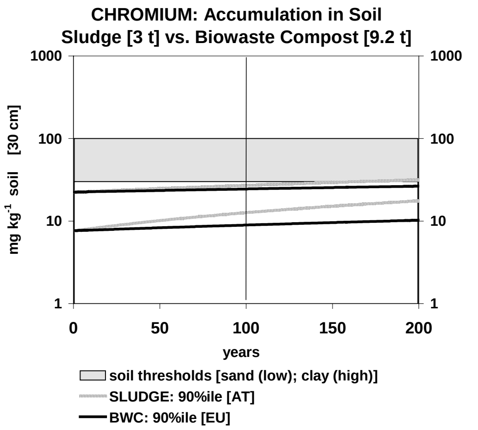
The assumptions for soil back ground concentrations, precautionary soil threshold values, heavy metal concentration in Biowaste, export by harvest and leaching, input by deposition and soil layer are the same as considered in the scenarios for biowaste compost (chapter 9.2)

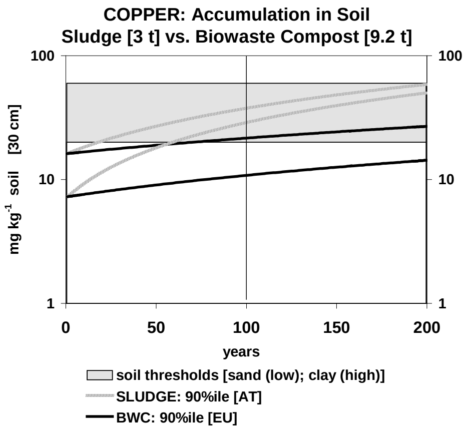
- heavy metal concentration in sludge:

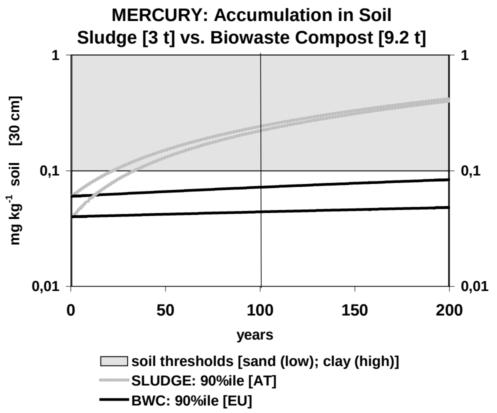
Cd	Cr	Cu	Hg	Ni	Pb	Zn
90th percentile of Austrian sludge *g ha^{-1} y^{-1}*						
2.2	80	320	2.9	45	110	1,400
Limits WD on sludge, 3rd draft *g ha^{-1} y^{-1}*						
10	1,000	1,000	10	300	750	2,500

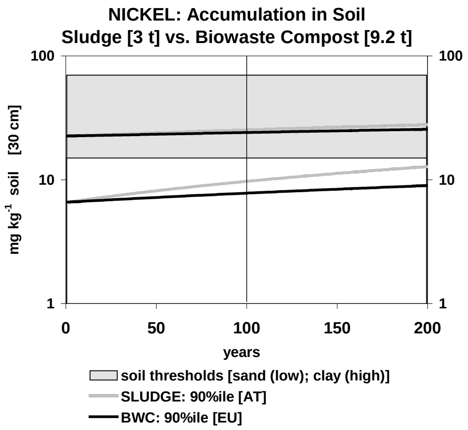
- Application rate: 3 t d.m. ha-1y-1 (according to 75 –150 kg P2O5 ha-1 y-1)
- Organic matter content in sludge: 60 % d.m.; complete mid term mineralisation (40 % of the applied dry matter remains in the soil)

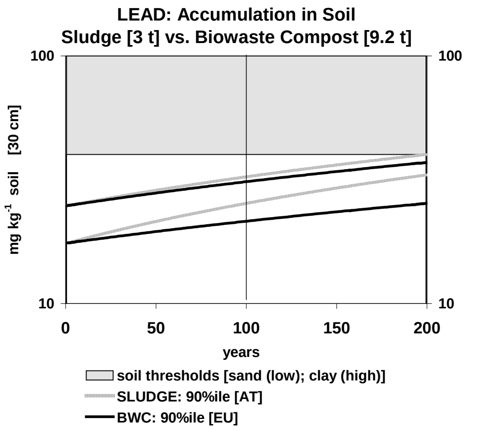












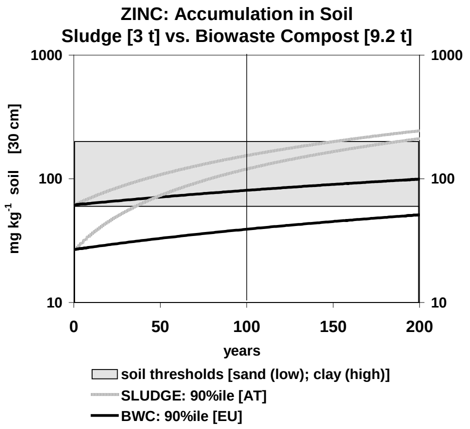


Fig. 9-6. Accumulation of heavy metals due to the regular use of Sewage Sludge compared to Biowaste compost.

- scenarios for sandy and clay soils respectively
- PTE concentration in sludge = 90th percentile [AT]
- PTE concentration in BWC = 90th percentile [EU]

Observations from the accumulation models for sludge application under the assumptions described here:

- Actual sludge qualities would not lead to considerable threats to soil functions also in the long term perspective with the exception of Copper, Zinc and Mercury, where German threshold values for sandy soils would be exceeded after 30 to 60 years.
- This would lead to the necessity for further efforts to reduce the Zn, Cu and Hg inputs. Site specific evaluations are recommended in order to assess the need of Cu and Zn as micro nutrients.
- This leads to the recommendation that limit values for municipal sewage sludge must be adapted to the factual concentration which can be achieved in practice. Otherwise precaution principles would be neglected.

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