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SCIENTIFIC COMMITTEE ON TOXICITY, ECOTOXICITY AND THE ENVIRONMENT (CSTEE)

Opinion on the Report on "Risks to Health and the Environment Related to the Use of lead in products" (TNO report, STB-01-39 final)

Adopted by the CSTEE during the 37th plenary meeting of 1st April 2003

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INTRODUCTION

A report on the risks to health and the environment by the use of lead has been prepared for the European Commission by TNO. Based on the conclusions made in this report, a number of questions were put forward to the CSTEE. These specific questions are addressed in this opinion. The CSTEE had recently issued (May 5, 2000) an opinion on a similar topic (Lead - Danish notification 98/595/DK) which also deals with health and environmental risk caused by the use of lead for a variety of purposes. This extensive CSTEE-opinion provides detailed background information on the problem.

Terms of Reference

The following questions were put forward by the European Commission to the CSTEE:

1. Accumulation of lead in the environment (in particular soil)

One of the main concerns raised by the study is that the continuing high level of the consumption of lead would lead to an increase of the substance, particularly in the top layer of soils. Depending on the soil characteristics a doubling of concentrations could occur over a timescale varying between 40 years and several centuries. The study stresses, however, that all the values in these enrichment studies are "highly tentative, particularly with regard to the food circle".

Are the arguments presented for such an enrichment of lead in soil scientifically convincing and in line with current research?

2. Methodological validity of the substance flow analysis (SFA) approach

In estimating the long-term trends the study applies the SFA approach in order to identify, and to the extent possible quantify, the main sources of emissions.

Does the SCTEE consider this approach as appropriate to assess the long-term trends and can the ranking of emissions to the various environmental compartments be used for a priority setting when deciding about the urgency of further work on risk reduction measures?

3. Potential need for additional marketing and use restrictions

According to the results of the study, it seems to be appropriate to consider marketing and use restrictions in two applications, which cause lion share of the emissions:

Lead shot

According to the calculations of the consultant, lead shot is the most important contributor of lead emissions to soil. Due to the fact that virtually no recovery and recycling takes place, the quantity used is almost the quantity emitted. The quantities emitted are higher than all other emissions to soil together. Direct effects are, however, limited, although acute poisoning of birds (which had swallowed lead bullets) was reason enough to ban it in parts of the US. National use restrictions in

Europe exist (at least) in the NL, DK and S (notified). In its comments to the Danish Lead ban the CSTEE stated in May 2000: "The problem of lead poisoning due to the ingestion of lead has been generally acknowledged and has been the reason for a ban on the use of lead shot in several countries including Denmark".

Is the CSTEE therefore of the opinion that the eco-toxicological evidence would call for a restriction of lead shot (on the Community level)?

Lead sheet

Lead sheet is used in a number of member States (in particular in the UK) in large quantities for roofing. The run-off of lead leads to high local concentrations in the vicinity of buildings. Corrosion from lead sheet is suspected to be the main source of lead in the inflow to waste water treatment plants and subsequently to sewage sludge. The study envisages a long-term problem due to the high accumulation of lead in the technosphere by this application with a life-time of several decades.

Does the CSTEE consider the argumentation on lead sheet and in particular the applied corrosion factor of 0.008%/year as appropriate?

4. Lead intake of young children from dust

The study identifies the intake of lead from dust as the potential predominant source of lead for young children, which may result in a very low margin of safety for this age group. The CSTEE expressed some skepticism on a similar assessment in its opinion on lead of May 2000 and a recent publication of the OECD seems to confirm this view.

Is the CSTEE aware of any new findings, which would bring it to a different opinion on this issue today?

OPINION

The CSTEE in general agrees with the approach, the procedures and most of conclusions in the TNO report. It also endorses the importance of the statements of uncertainties resulting from the many assumptions regarding exposure scenarios made in the report. The responses to the questions posted are outlined in order of the numbering of questions made above.

1. Accumulation of lead in the environment (in particular soil)

The concern on the potential accumulation of lead in soil expressed is – as clearly indicated in the report - based on a (large) number of assumptions and estimated releases taken from the SFA or literature. Using calculation reported in different studies (Annema, 1995; Janus et al., 1999; Molenaar, 1998, Danish EPA, 1998) and their own calculations, the report concludes that the lead emissions will result in an annual enrichment of 0.2 to 0.06% or 0.5 to 0.2%, respectively (compared to natural background of 10 to 30 mg Pb/kg). These enrichment figures would result in an average doubling of lead concentration in European soil in 200 to 500 years. However, based on Janus et al. (1999) the report also states that for grasslands the accumulation might increase the lead soil lead content by 10 mg/kg every 40 years, implying a doubling of the soil concentration (natural background 10 mg/kg) every 40 years.
It is the opinion of the CSTEE that these predictions do not include all lead emission sources and are based on too many assumptions to allow science-based conclusions to be made. It is suggested that the uncertainties and assumptions are identified and analysed so that a focused

literature review and possible dedicated research can be performed to reduce uncertainties associated with the present calculations. Possibly, this can be suggested as an action item in the context of the presently ongoing Voluntary Risk Assessment.

The CSTEE has commented on the potential accumulation of lead in soil in a previous opinion (CSTEE, 2000) regarding the prediction of a very small, but steady increase of lead concentrations in soil predicted by the Danish EPA. A potential accumulation of lead in soil by atmospheric deposition and a number of intentional and unintentional uses by app. 0.1 % was predicted implying a doubling of background soil concentrations within more than 1 000 years. The CSTEE concluded that even when the basic assumptions for emission rates used by the Danish authorities were correct, this is not expected to cause a significant impact on environmental species for many hundreds of years and that bioavailability of lead from this deposition is also unclear. In this opinion, the CSTEE encouraged environmental monitoring of lead to identify if changes in lead levels will occur as a possible basis for remedial actions.

2. Methodological validity of the substance flow analysis (SFA) approach

Lead is emitted during production, use, recycling and from the incineration or deposition of the products containing this element. Lead is used in many different applications and the emissions have to be estimated separately for these applications. The main difficulties are to find good measures of the emissions from the many different products containing lead. The method used in this report is called substance flow analysis (SFA), often also called metal flow analysis (MFA) when applied to metals. This method is especially useful for the elements, such as lead, as these are not formed or destroyed in the environment. The limitation in this approach is often the availability of good emission factors for the different materials containing the specific metal. The emissions are slow processes and predictions of future releases can be correlated to predicted use volumes for the different applications. The CSTEE considers the SFA an appropriate and valid method to estimate release of lead from applications.

The separation between functional and non-intentional use of lead made in the report is relevant. The functions identified in the report are covering the major volumes of the produced lead. In a recently reported Swedish research program the flows of heavy metals in Stockholm were studied. This program highlighted a few additional product types, such as balance weights for wheels, asphalt wear, brake linings and “Falu-Red”. The last one may be a typical Swedish product used to paint a lot of local houses red, but the others may also be significant in other parts of Europe. Several of these are giving considerable emissions, and the Dutch report may thus have underestimated some of the lead flows to the environment. The Swedish project identified ammunition and sinkers used for sport fishing as the major emission routes of lead in Stockholm (Sörme et al., Water, Air, and Soil Pollution: Focus 1, 213-227, 2001).

The diffusive emission of lead from products where it has been intentionally added is difficult to estimate. As an example of these difficulties, the corrosion from lead sheets used on buildings, which is one of the major routes, can be mentioned. Release of lead has been reported to be very high initially, but already after a couple of months the values decreased by an order of magnitude (Persson and Kucera, Water, Air and Soil Pollution: Focus 1, 133-150, 2001). The explanation given for this is a possible formation of protective corrosion products as a top layer preventing further release of lead. It is not clear how the factor used in the report was derived, but it seems to be close to the value observed for the fresh material, and thus these emissions may have been overestimated.

The emission factors used for ammunition, other products and landfill are rather speculative and should be discussed. In landfills for example, 0.23% is used for “free lead”, a figure that seems rather high. On the other hand, no emission of lead in PVC is expected, which may not be true when the PVC is degraded in the site.

The main non-intentional uses of lead include the use of fertilisers and fossil fuels, mainly coal. The emissions from agriculture are based on the situation in the Netherlands, which is extrapolated to the EU. The emission factors used for the emissions to air from combustions are those developed within EMEP, which probably are the best available for the EU.

3. Potential need for additional marketing and use restrictions

Lead shot

Lead emissions into the environment are dominated by the release of lead from ammunition, which is calculated with a high corrosion factor (1 % per year). In its previous opinion (May, 2000), the CSTEE has recognised the problem of acute lead poisoning due to ingestion of lead shot by waterfowl.
In wetlands, which are shot over, lead poisoning of waterfowl has almost invariably been found to occur. As a result of this evidence, not a formal risk assessment, a four-year voluntary phase out of lead shot began in the UK in 1995 and culminated with the UK Government introducing legislation in September 1999 to restrict the use of lead shot. The UK Environmental Protection prohibits the use of lead shot over the wetland sites of special scientific interest (SSSI) and over areas below the high water mark; for shooting the specific species of waterfowl. The objective of these regulations was not to unnecessarily restrict shooting but to alter shooting practices and to ensure that spent lead shot does not cause unnecessary deaths in waterfowl. Although the CSTEE supports the approach taken by the UK to reduce the use of lead shot in (local) environmentally sensitive areas to decrease the incidence of lead poisoning in waterfowl, the CSTEE does wish to express concerns on the reliability and accuracy regarding the relative importance (magnitude) of lead emission estimates of ‘ammunition’ to soil. According to Table 3.31 of the report, lead shot/ammunition is responsible for 80 % of total lead emission estimates to soil in 2030 (15 EU countries). The assumptions on the loss of lead shot in the environment and on the corrosion rate are based on limited (if any) data. As such the CSTEE would like to recommend that these uncertainties and assumptions are further examined and analysed through a focused literature review and possible dedicated research in order to reduce uncertainties associated with the present calculations. This may be suggested as an action item in the context of the presently ongoing Voluntary Risk Assessment.

Lead sheet

The factor of 5 g/m2.yr for the corrosion of lead and the use of an emission factor of 0.008% per year from lead in buildings chosen in the report are deemed acceptable by the CSTEE. Even if one can accept the assumption that the progressive formation of patina will slowly reduced the emission factor with time, this remains to be demonstrated and convincing arguments (data) must be presented.
In addition, lead sheet is frequently used in balconies, flat roofs and terraces where exposure to rainwater is direct on an horizontal surface. This seems not to have been taken in consideration in the TNO assessment and counterbalances the possible overestimation by taking 25 % of the lead sheet as being directly exposed to the environment and subject to run-off.

4. Lead intake of young children from dust

The CSTEE has commented on the issue of soil ingestion and lead exposure in children (opinion of May 5, 2000) and is not aware of new scientific findings to support a revision of this opinion. The CSTEE has also recently commented on other sources of potential high lead exposures (release of lead from candle wicks during burning of candles) in the indoor environment.

The TNO -report uses TDI-values as a basis for health risks assessment for humans and derives exposures from levels of lead in air, the food basket and in soil and dust. For adults, the calculated total exposure based on a average concentration of lead in air of 0.01 µg/m^3, a realistic drinking water concentration of 2.6 µg Pb/L and a daily intake of 27 µg lead for adults reaches 13 % of the TDI. Using air, drinking water and food basket concentrations for lead exposure of children, 30.3 % of the TDI was calculated to be reached, on average. The approach used in the report for estimating soil and dust uptake by children suggests that this pathway is a significant contributor to total lead exposure of children, resulting in estimated exposure about half the TDI (55.6 %).

The authors discuss the problems and difficulties in the exposure assessment for children using assumed intakes of soil and dust. The TNO report uses an average intake of 0.2 g of soil and/or dust in children and an average concentration of 100 mg lead/kg soil/dust, noting that worst case assumptions of 3 g soil/dust intake per day and lead concentrations in soil of more than 1000 mg/kg have been reported in the literature. The average value for a lead concentration of 100 mg/kg dust is based on data measured as an average lead concentration in indoor dust in the 1990 and the consultant notes that maximum values measured were much higher.
Using the traditional risk assessment process based on these assumptions, the consultants come to the result that the TDI for children is almost reached (30 % from food and 56 % from soil/dust) and that dust intake contributes to a significant part of the lead exposure in children.

The CSTEE considers the estimates used for lead concentrations in indoor dust and dust intake reasonable. Mean concentrations of lead in household dust were determined in a number of studies in addition to those used by the consultants. For example, a mean of 57 mg Pb/kg of dust with a 95th-percentile of 178 mg Pb/kg of dust was found in a large survey of samples collected in Germany in the 1990 (Seifert *et al.* 2000), median dust concentrations of lead ranged from 100 to 135 mg Pb/kg of dust in urban and mining towns in Sweden (Berglund *et al.* 2000), and median dust concentrations were 125 mg Pb/kg of dust in a smelter town in Germany in 1993 (Meyer *et al.* 1999).

A number of recent studies have also addressed the dust/soil intake in young children (Calabrese *et al.* 1997; Stanek and Calabrese 1995, 2000) suggesting that the soil/dust intake used by the consultants (0.2 g/day) represents about the upper 95th percentile which is reasonable to use. A mean soil intake by children 1-4 years old, residing on a superfund site, was estimated to about 30 mg/day (median less than 1 mg/day) and 95th percentile 160 mg/day (Calabrese *et al.* 1997). In a later study, Stanek and Calabrese (Stanek and Calabrese 2000) report an average intake of 31 mg/day (median 17 mg/day) with 95th percentiles of 106-133 mg/day, depending on observation time, for children at a contaminated site.

However, the results on dust and soil intake in the case of environmental lead exposure need to be balanced with data on blood levels of lead in the children, because the bioavailability of soil/dust lead may vary considerably. A number of studies have assessed blood levels of lead in children and some of these studies have also determined lead concentrations in dust and/or soil.

The CSTEE opinion of May 2000 was based on the results of a Swedish study (Berglund *et al.* 2000) whose results suggested that lead in soil from mining activities does not make a major contribution to lead body burden in children, most likely due to poor bioavailability and low soil/dust intakes due to the climate. There were indications of some influence of soil/dust lead to blood lead in the urban area. The blood lead levels peaked at about 2 years of age, peaked in season with more outdoor activities, and were slightly higher in the urban (median 28 µg/L) than in the rural town (median 20 µg/L). However, it was concluded that, at present in Sweden, food is the main source of lead exposure in children even in areas with high soil lead concentrations and that lead in soil and dust contributes little to total lead intake. As the lead concentrations in urban soil, which still has some remaining lead from previous automobile exhaust (when leaded gasoline was use), will decrease with time, it is likely that the exposure contribution from soil/dust also will decrease further.

Further support for a small role of household dust and the inadequacy of the traditional risk assessment approach for this problem can be derived from these observations:

- In the Berglund (Berglund *et al.* 2000) study, median dust concentrations in the houses and day care centers were between 100 and 135 mg Pb/kg of dust and thus similar to the value used in the TNO-report, but blood levels of lead in the children using these buildings were low (median values 19-28µg lead/L)

- In the German environmental (Seifert *et al.* 2000) survey with a mean of 57 mg Pb/kg of dust with a 95th-percentile of 178 mg Pb/kg of dust, geometric means of blood levels were also low (32 µg lead/L with a 95-percentile of 62 µg lead/L)

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