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

"The environmental impact (reduction in eutrophication) that would result from banning sodium tripolyphosphate (STPP) in household detergents"

Adopted by the CSTEE during the 40th plenary meeting of 12-13 November 2003

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#### **Terms of reference**

DG Enterprise has recently requested the opinion from the SCTEE on the WRc study ("Phosphates and alternative detergent builders"), commissioned by both DG ENTR and DG ENV, and dealing with the impact of phosphate-loaded detergent discharges on eutrophication.

The SCTEE has adopted by written procedure on 10 March 2003 a final opinion on the WRc report on the impact on the environment (reduction in eutrophication) that would result from substituting phosphates in household detergents. Given several weaknesses of the report, the SCTEE was of the opinion that the conclusions of the WRc report are not adequately substantiated.

DG Enterprise would therefore very much appreciate a further opinion of Scientific Committee on Toxicity, Ecotoxicity and the Environment (CSTEE) based on a more complete analysis of the literature dealing with the eutrophication issue, covering in particular the items not adequately treated in the WRc report, as pointed out in the above mentioned opinion. This examination of the available data should enable the SCTEE to deliver a new opinion on the following issues:

**1a**/ Is phosphate-based detergent a substantial contributor to eutrophication in the enlarged EU (including at least Poland, Hungary and Czech Republic) and does this contribution lead to a significant risk to the environment?

**1b**/ To what extent would detergent limited to a phosphate content of 1% reduce the occurrence of eutrophication?

2/ Would the substitution of STPP by Zeolite A in detergents substantially reduce eutrophication, and would such a substitution by Zeolite A impose any additional risks or adverse effects on the environment (including the aspect of accumulation of sludge)?

**3**/ Would the accumulation of sludge (resulting to the addition of Zeolite) be greater than the accumulation of sludge resulting from the Phosphate removal process in the waste water treatment plant?

#### **Overall considerations**

The risk of eutrophication related to anthropogenic phosphate loads plays a role when the following key factors appear simultaneously in the spatial and temporal scales:

- The ecosystem can respond to the additional nutrient load with an increase in algal productivity resulting in structural and functional changes
- Phosphate is the limiting nutrient

Increase in phosphate loads will results in eutrophication problems only in those locations and points in time when these conditions are fulfilled.

The role of STPP becomes critical when the contribution of the additional load related to STPP plus the phosphate loads from other sources exceed the threshold for euthrophication of the exposed system. It must be stressed that this threshold will be different for different water bodies.

The information submitted to the CSTEE indicates that in some locations STPP represent a contribution to the euthrophication problem, but it was not possible to address in a quantitative way the extent (e.g. percentage, location, significance) of the water bodies, within the EU and the enlarged EU, where this situation occurs.

The CSTEE is aware of the large amount of published information on the relevance of phosphate on eutrophication, covering exposure and effect aspects, not considered in the submitted documents.

A quantitative assessment of the extent of eutrophication in EU water bodies in relation to phosphorus load from different sources, and in particular in relation to STPP contribution, could be performed on the basis of a literature review on existing experimental and modelling information, produced on the evolution of the eutrophication problem and on the recovery of eutrophic water bodies.

# Question 1a

As remembered in the previous opinion of the CSTEE on phosphorus and eutrophication, it is currently recognised that excess phosphorus is responsible for eutrophication in most inland water bodies, as well as in some coastal marine areas.

Phosphorus loads originate from point and non-point sources, in particular:

- urban sewage, in which two different components are present: phosphorus from human metabolism and phosphorus from synthetic detergents;
- animal farms;
- discharges of some specific industrial typologies;
- runoff and drainage from agricultural soils;
- runoff and drainage from natural soils.

The contribution to eutrophication of phosphate-based detergents is extremely variable country by country and, even in the same country, it can be different in different hydrographic basins as a function of human activities and land use.

In the TNO (2003) report on "Agricultural phosphate emissions to surface waters", the amount of phosphorus in wastewater that can be attributed to detergents, in countries where no actions for reducing STPP in phosphate containing detergents were undertaken, is as high as 40%.

This figure is reliable enough, at least as an average, and is in substantial agreement with comparable values reported by other literature sources. Nevertheless, substantial differences may occur in detergent consumption in European countries as a function of water characteristics. In the hard waters common in Mediterranean countries, detergent use is higher than in the soft water common in northern Europe (see table 1), and this led to higher phosphorus contribution.

Table 1 -	Detergent	consumption	(kg/year per	capita)	in some	European	countries	(From	Fox e	t al.,
2002)										

Country	Kg/y	Country	Kg/y
Finland	3.8	Greece	10.2
Sweden	4.5	France	11.8
Norway	4.9	Portugal	12.2
Denmark	6.5	Spain	12.4
The Netherlands	7.5	Italy	12.9
European Ave	erage	10	

In Italy, before restrictions on STPP in detergents, the contribution to phosphorus in wastewater was evaluated as higher than 50% (Chiaudani et al., 1978).

More difficult is the assessment of the percentage of the <u>total</u> phosphorus load represented by phosphate-based detergents. The same TNO report (TNO 2003) indicates the following relative contribution from different sources:

- Point sources: 50-75%
- Agriculture: 20-40%
- Natural loading : 5-15%

These figures, referred to an European average basis, are more variable and controversial and need to be clarified.

- *Point sources.* These include urban sewage and wastes from some specific types of industrial sources, as well as direct discharges to surface water from intensive animal farms. Even if the maximum permitted amount of animal manure is used in agriculture as natural fertilizer, in areas with high livestock density emission due to direct discharge is not negligible.
- *Agriculture*. This includes contributions, through runoff and drainage, from synthetic fertilisers as well as from animal manure and sludge from urban treatment plants. It must be highlighted that, unlike synthetic fertilizers, manure and sludge can often be improperly used, by being applied to agricultural soil as a "waste to be disposed". This may led to an excess of losses from soil and to surface and groundwater pollution.
- *Natural loading*. This represents the contribution from natural soil (forests, woods, etc.). In hydrographic basins with low human impact, this may be responsible for the major percentage of the phosphorus load. In these cases, eutrophication phenomena are possible but unlikely to occur.

The relative amounts of the different sources can be extremely variable, depending upon land use. A significant example of the variability that may occur, even in the same country, in water bodies subject to eutrophication, can be the assessment of the phosphorus load to sub-alpine lakes and to Northern Adriatic coastal waters in Italy, before the measures for phosphorus control were undertaken.

In the major sub-alpine lakes phosphorous resulting from agriculture is low or negligible. Major phosphorus contributions come from urban emissions, and natural loads can represent a relatively high percentage of the total phosphorous. The contribution of different phosphorus sources was calculated for six large European alpine lakes before measures for reducing STPP in detergents were undertaken and the results are reported in table 2 (Vighi and Chiaudani, 1987). It can be seen that point sources are the main contributor.

Table 2 – Contribution of point and non-point sources to phosphorus load in some alpine lakes.

Lakes	Contribution to phosphorus load (%)				
	Point sources	Agriculture	Natural		
			background		
Zurich	86	9	5		
Geneva	88	5	7		
Constance	79	11	10		
Iseo	68	15	17		
Lucerna	61	27	12		
Maggiore	69	11	20		

The River Po valley is the major contributor to Northern Adriatic eutrophication and is the area with the highest human impact (urban, agriculture, livestock, industry) in Italy. It was calculated that about 50% of the total phosphorus load produced in Italy was discharged into the Adriatic through the River Po (Chiaudani et al., 1978). In this case, agricultural contributions play a more significant role.

Moreover, it must be taken into account that, at present, many interventions have been undertaken in Europe to reduce phosphorus in urban emissions, either by reducing STPP or by urban sewage treatment (biological and chemical). It follows that, in the present European situation, the relative contribution of non-point sources has been increased, due to the reduction of the point emissions.

Some examples of the present contribution of non-point sources in different European river basins is given by Macleod and Haygarth (2003). The authors indicate that the phosphorus contribution from non-point sources in various river basins in Europe in the 1990s ranges from 2% to 60%. The lowest figure refers to basins without point sources control, while highest values occur when point sources, and thus total phosphorus inputs, are substantially reduced. In the river Thame, non-point sources contribute 15% and 36-53% before and after phosphorus treatment in sewage respectively; in the river Kennet the same figures are 2% and 29-45%.

In Italy, before STPP ban, the total phosphorus load to surface waters was about 59000 tons/year and agriculture and livestock represented about 35% of the load (Chiaudani et al., 1978). After the ban the total load decreased to 45000 tons/year and the percentage due to agriculture and livestock rose to 47% (Pagnotta and Ghergo, 1996, Capri and Pagnotta, 2003). A trend of phosphorus loads and their sources in Italy is shown in figure 1.



Figure 1 – Trend of phosphorus load in surface water in Italy in the last five decades (data from Chiaudani et al., 1978, Capri and Pagnotta, 2003).

In Switzerland there are also many experimental examples of the reduction in phosphorus load and concentration in running and lake water, as a consequence of the detergent phosphorus ban in 1986 (Wehrli et al., 1997; Muller, 1997).

The present situation of STPP use in detergents is described by the WRc report. In some EU countries regulations were implemented for an almost total (e.g. Italy, Belgium) or partial (e.g. Austria, Germany) ban of STPP. In other countries (e.g. Scandinavian countries, The Netherlands), even in the absence of a precise regulation, voluntary agreements were obtained which led to a substantial reduction of phosphorus quantities.

At present, only in Greece, Luxembourg, Portugal and Spain have no actions (legislative or voluntary) for phosphorus reduction in detergents been taken. It has been estimated that, as a consequence of these actions, phosphorus emissions from detergents in surface water have been reduced by about 77% from 1985 up to 2000.

Less information is available about the situation in new candidate member states of the European Union, but it seems that, in some countries, some reduction measures have been undertaken.

Additional information is reported in a document of ZEODET, showing the picture of the market shares of phosphate-free detergents in European countries in 1998 reported in table 3.

To better evaluate the present contribution of STPP to the total load of phosphorus in surface waters of European countries, data from table 3 should be combined with data on the consumption of detergents (table 1) and with data on the percentage of population connected to treatment plants.

Table 3 – Market share of phosphate free detergents in European countries in 1998.

Netherlands	100 %	Croatia	60 %
Norway	100 %	Great Britain	55 %
Italy	100 %	France	50 %
Germany	100 %	Greece	50 %
Switzerland	100 %	Spain	40 %
Austria	100 %	Czech/Slovakia	35 %
Slovenia	95 %	Hungary	30 %
Finland	90 %	Portugal	30 %
Belgium	90 %	Poland	15 %
Sweden	85 %	Romania	5 %
Denmark	80 %	Bulgaria	5 %

Low levels of phosphate free detergents are typical of some EU candidate countries, where the percentage of population connected to treatment plants is also relatively low (see comments to Question 1b).

In Eastern European countries, severe eutrophication problems are documented. For example, there is a problem of eutrophication in Hungary's major Lake, Lake Balaton. Moreover, the WRc report mentions the eutrophication problem in the Black Sea, to which Hungary and other countries discharging to the Danube contribute. The WRc report suggests that approximately half of the P load to the Danube Basin is from agriculture (but not from the Hungarian part of the basin where the domestic load represents 67% and agriculture 13% of the total), which may be decreasing slightly but is difficult to control. A substantial reduction in P from other sources could be very effective in controlling eutrophication of the NW shelf of the Black Sea.

It should be stresses, that the HERA (2003a) on STPP performed by the industry has not addressed the eutrophication hazard associated to the use of polyphosphates in detergents. The argument mentioned in the HERA report "nutrient enrichment is not addressed in this document because a PNEC cannot be defined for such as effects" is not acceptable. Obviously, a PNEC for eutrophication cannot be defined as a single number applicable to all ecosystems, but as a set of values or even a function or set of functions addressing the diversity and influence of related factors on the eutrophication process. The basic rules for environmental risk assessment are however applicable, although a higher tier assessment should be required, e.g. a landscape evaluation with probabilistic outcomes for each landscape scenario.

In conclusion, in order to answer to Question 1a, it is opinion of the CSTEE that:

- in the absence of measures for reducing the STPP content in detergents, the contribution of this phosphorus source to the total phosphorus load in surface water can be very variable (roughly speaking from 10 to 40%) as a function of different human activities and land use; even at the lower end of the interval, this contribution is not negligible especially in areas that can be subject to eutrophication processes; therefore, STPP in detergents would produce a significant increase of phosphorus load in surface water and a significant risk for eutrophication in some areas of the enlarged EU;
- the present situation in Europe has substantially changed in comparison with the 1980s, because many European countries have undertaken measures to reduce STPP; in these countries, detergent phosphorus is no longer a substantial contributor and other sources contribute a higher percentage to the overall phosphorous load.

#### **Question 1b**

The analysis of the extensive amount of information available on phosphate loads and eutrophication indicates that a quantitative estimation, at the river basin levels, of the areas of Europe where phosphate-based detergent could be a substantial contributor to eutrophication could be estimated from available river-basin phosphate load models and indicators on detergent and non-detergent based loads (demography, livestock densities, land uses, etc.). However, this estimation has not been included in the documents submitted to the CSTEE. Therefore the CSTEE cannot provide a quantitative answer to this question, and only qualitative assessment can be provided.

As a consequence of the answers to Question 1a, it follows that contributions to the total phosphorus load to surface waters derive from different point and non-point sources, whose relative magnitudes are very variable. Thus a complete solution to the eutrophication problem will require, in most cases, interventions on all the major phosphorus sources, given that enough time is then allowed for P removal from overloaded sediments, and perhaps from overloaded soil which may enter the watercourse via erosion.

The control of phosphorus emission from urban sewage, either by reducing STPP in detergents and by Waste Water Treatment Plants, cannot be sufficient, in some cases, to completely cure the problem of eutrophication in surface water. On the other hand, the contribution of urban sewage is, in most cases, so relevant that it would be very difficult, if not impossible, to get a solution without controlling this source of phosphorus.

About the relevance of different options for controlling phosphorus emissions from urban waste water (STPP ban and/or waste treatment) interesting comments can be derived from the document on the current Swedish position on STPP in detergents (Wallgren, 2003).

In Sweden no official measures were undertaken to reduce STPP. Some producers reduced STPP on a voluntary basis, to get a "quality" label.

STPP reduction is not considered as a priority intervention because 95% of urban sewage is collected in treatment plants and over 90% is treated with chemical phosphorus reduction. Phosphorus concentration in treated water is 0.5 mg/L and there is a trend to reduce it to 0.3 mg/L. Notwithstanding this position, according to ZEODET data, STPP free detergents cover 85% of the Swedish market.

The situation in many European countries is very variable. First of all, in many countries, the percentage of treated urban sewage is much lower than in Sweden.

Moreover, tertiary sewage treatment for phosphorus removal is not a general rule. In many countries, phosphorus treatment is required only in "sensitive" areas. For example, in Italian law, a general limit for phosphorus in effluents is set at 10 mg/L and in sensitive areas the limit is lowered to 0.5 mg/L. The selection of sensitive areas is made by local administrations and, in general, is restricted to effluents discharging in lakes or in some very vulnerable coastal marine sites (e.g. northern Adriatic areas with high tourist relevance). This means that in all rivers of the Po valley, carrying phosphorus in the eutrophic Adriatic Sea, no phosphorus treatment is required.

According to Eurostat data (Vall, 2001), in some EU countries the percentage of population connected to treatment plants is close to or higher than 90%, and inn some cases there will be a high percentage of advanced treatment (e.g. Scandinavian countries, The Netherlands, Germany). In other countries the percentage connection is lower than 50%, with a very low incidence of advanced

treatments (e.g. Belgium, Spain, Portugal). Relatively low treatment levels are also typical of some candidate countries: in Poland, the population served by treatment plants is 52%, with 15% having advanced treatment, in Hungary the figures are 26% and 3% respectively.

An additional reason for supporting STPP restrictions is its character of preventive intervention, to be preferred to treatment for the sake of sustainability. Moreover, the reduction of phosphorus content in sewage will reduce the amount of phosphorus in sludge.

The ban of STPP in detergents in Switzerland, in 1986, led to a substantial reduction of phosphorus concentration in all major Swiss lakes as well as in the River Rhine at Basel. This intervention strongly increased the effect of the chemical removal of phosphorus in treatment plants (Müller, 1997). As a result of the combination of phosphorus ban with improved sewage treatment efficiency, the load of phosphorus has reduced by about 60%. Moreover, the lower phosphorus input in sewage treatment plants increased the efficiency and allowed using half the amount of phosphorus precipitation agents (Siegrist and Boller, 1997)

The most relevant eutrophication problem of the whole Mediterranean area is probably represented by Italian Northern Adriatic coastal waters. It was recognised as a problem of high concern, for environmental and economic reasons, in the early 1970s.

In 1982, the progressive reduction of STPP began in Italy, concluding in 1989 with total STPP elimination. This produced a reduction in the total phosphorus load to the Adriatic sea of about 30%.

All other phosphorus sources were not significantly reduced. The effect of the increased number of urban sewage treatment plants was negligible, because, as previously mentioned, chemical phosphorus reduction was applied only in a small percentage of plants. The reduction of phosphorus load produced a substantial improvement in Adriatic quality (Pagnotta and Ghergo, 1996; Regione Emilia Romagna, 2002). The problem is not yet solved, but severe eutrophication episodes are strongly reduced. A complete solution would be achieved with additional interventions on sewage treatment, livestock and agriculture.

As previously mentioned, in some circumstances non-point phosphorus loads may represent a relevant percentage of the total load. Particularly in areas where measures for reducing point sources are undertaken, agricultural loads may represent the major contribution and a complete solution of eutrophication cannot be achieved without some control measures on these emissions.

Nevertheless one must be aware that these measures are usually more difficult and can not give a complete solution of the problem. Reducing phosphorus releases from runoff and drainage of agricultural soil without affecting crop productivity needs a complex strategy based on:

- using more environmental friendly agricultural practices;
- optimising and rationalising the application of animal manure and sludge;
- developing a general strategy of land use management.

Therefore there is the need for changing the behaviour of farmers and this result cannot be achieved in the short time. Moreover, even if such a strategy could substantially reduce phosphorus losses from agricultural soil, loads can be only partially reduced. It is commonly accepted, among agronomists, that a reduction of more than 50% of agricultural loads can hardly be achieved, even if it is difficult to find sound literature references to support this statement. A review paper evaluating the maximum reduction of agricultural phosphorus load that could be achieved by applying best agricultural practices without reduction of crop productivity, would be very useful. The production of such a document is strongly encouraged by the CSTEE.

It follows that controlling non-point sources can be a relevant contribution but not the complete solution for the problem of eutrophication.

In conclusion, a quantitative assessment of the extent of eutrophication reduction achievable by reducing STPP content up to 1% is not possible with the information provided, nevertheless, it is opinion of the CSTEE that:

- the control of point sources through a combined approach of significant to complete reduction of STPP in household detergents and completing the implementation of the Urban Waste Water Treatment Directive is a necessary in some areas, even if in some cases not sufficient, measure to cure the problem of eutrophication in the EU;
- there are significant examples of a substantial, even if not complete, reduction of eutrophication obtained only with the elimination of STPP;
- in particular STPP restrictions are the only possibility to reduce phosphorus loads from urban emissions in all situations where connection to sewage treatment plants is difficult (small communities, rural areas) or if phosphorus removal by advanced sewage treatment is not planned;
- point source control is the most effective measure to reduce phosphorus load; the control of non-point sources needs more complex strategy, in particular with respect to changes in agricultural practices, , and, in any case, can produce only a partial reduction of agricultural emissions; this may be necessary in specific cases, however.

# **Question 2**

An extensive review of the literature on the environmental effects of Zeolite A has been performed in a report on Human & Environmental Risk Assessment produced by HERA 2003.

From the experimental evidence available it can be concluded that, on a toxicological and ecotoxicological point of view, there is no risk for man and the environment. Moreover there is no evidence for the mobilisation of heavy metals in the aquatic environment due to Zeolite.

After discharge to surface water Zeolite A hydrolyses to amorphous minerals, or in the presence of environmental calcium and phosphate to poorly soluble calcium aluminium silicate phosphates. The half-life for these processes is less than 2 months, with 12 to 30 day half-lives, depending upon pH, seen in the presence of calcium and phosphate concentrations that are typical of natural waters. These amorphous materials have no ion exchange capacity, and will be unable to bind metals in the environment. Thus, after hydrolysis, Zeolite A should be environmentally inert.

Nevertheless, there is limited information on additional possible effects on the aquatic environment, in particular due to the increase of suspended solids and of sedimentation in natural aquatic ecosystems.

On the other hand it must be taken into account that the emission level of suspended solids is regulated by and subject to a legal level given in the discharge permit. Therefore, for treated effluents, the use of Zeolite would not produce an increase in suspended solids load to natural surface water bodies. Assuming that suspended solids sedimentation does not require advanced treatment, it can be concluded that, according to the previously quoted Eurostat data on population connected to sewerage systems, in most European countries, including some Candidate countries, the large majority of domestic effluents are subject to at least mechanical treatment.

Notwithstanding this general situation, there are some relevant exceptions in some EU countries (e.g. Belgium, Portugal) as well in Candidate countries (e.g. Hungary), where significant amounts of domestic effluents are discharged without treatment.

Precise figures allowing evaluation of the increase of suspended solids in untreated effluents due to Zeolite use are not provided, nevertheless, from some rough evaluations, it should be around 3-6 %, in any case not higher than 10%. In terms of total increase of suspended solid load in surface water, including the contribution from runoff and soil erosion, Zeolite contribution would be even lower.

Untreated effluents discharged in surface water produce serious pollution problems, due mainly to organic matter, including organic suspended solids which, besides the effects on oxygen balance, would also increase sedimentation.

It is opinion of the CSTEE that, among the consequences on the overall quality of surface water bodies produced by the discharge of untreated effluents, a moderate increase in suspended solids due to Zeolite can be assumed as negligible. The need for increasing the level of treatment must be supported for reasons other than Zeolite.

As a final comment on the effects of Zeolites in surface water bodies, it can be highlighted that no environmental problems are documented in those countries (e.g. Switzerland, Italy) where Zeolites have been used for more than 15 years.

In conclusion, in order to answer to Question 2, it is opinion of the CSTEE that:

- there are no toxicological or ecotoxicological problems related with the use of Zeolites in detergents, and no environmental problems have been documented in those areas where the use of Zeolites is already common, although monitoring programmes downstream of wastewater discharges are relatively common;
- in treated wastewater effluents the amount of suspended solids is regulated, therefore the use of Zeolites will not increase the emissions of suspended solids in surface waters over those values accepted for the receiving waters;
- non treated effluents produce severe damages to the surface water environment due to many other pollution factors (e.g. organic matter and oxygen depletion); in these conditions it is the opinion of the CSTEE that the contribution of Zeolites would be minor, even in relation with the potential increase in sedimentation due to organic sediments;

# **Question 3**

As most of the Zeolite used in washing products leaves the sewage treatment plant as part of the sewage sludge, it is necessary to consider whether the increase in sludge volume due to Zeolite will be significant, and whether problems may arise from the disposal of any excess sludge. Similar considerations apply for any additional sludge produced by the two main phosphate removal routes, namely biological phosphate removal and phosphate removal via various precipitation processes.

Approximately 90% of the Zeolite which enters the sewage treatment facility is incorporated into the sewage sludge (HERA, 2003b). Although this typically causes an increase of approximately 10% in the dry weight of the sludge produced the sludge volume has been shown not to increase, as Zeolites aid sludge settling (Zeodet, 2000). Thus sludge transport costs will not be increased by the use of Zeolite in washing products.

Sludge will also be produced during the removal of phosphate incorporated in washing powder. Biological phosphate removal is potentially more sustainable then phosphate removal by chemical precipitation, but has specific requirements for the level of phosphate in the waste water (more than 5 - 10 mg/l phosphate is preferred) and the BOD/P ratio (at least 35), which often requires the addition of short chain fatty acids or other suitable low molecular weight organic substrates (SCOPE, 1999). Also, biological phosphate removal typically removes only 40 to 70% (exceptionally, up to 85%) of the phosphate present. Thus it may not, on its own, be able to meet some of the strictest phosphate consent levels found in the EU (SCOPE, 1999). For these and other considerations (financially, biological P removal has higher capital costs, but lower running costs than chemical phosphate removal), chemical phosphate removal is currently more prevalent in EU countries. Sludge production in a biological P-removal WWTP, can be slightly less than in an activated sludge treatment plant, with overall average reductions in biosolids production (dry solids) estimated at 2–8%. (SCOPE, 2001).

Phosphorous removal by chemical precipitation, often with iron chloride as the precipitating agent, produces excess sludge, with contributions both from the suspended solids removed and from the chemicals added to induce precipitation.

For example, iron P stripping using a 5/1 Fe/P ratio to achieve outflow levels from a primary treatment tank of 0.2 mg/l ortho phosphate increased total suspended solids (TSS) removal from 50% to 75% during primary treatment, and increased primary sludge solids by a total of 94%: 50% due to increased TSS removal plus 43% due to precipitation chemicals. In another example, a 54% increase in sludge due to P precipitation with iron chloride was observed in a plant in Saint Paul, Minnesota, with a 5/1 Fe/P ratio used to obtain 2mg/l P in the outflow stream (SCOPE, 1998a, 1998b).

The OIE-CEEP report (2001) assumes an increase of only 15% of total sludge production due to chemical precipitation with iron chloride. However, the WRc report uses both 15% and 35% as estimates of additional sludge produced by chemical precipitation of phosphate. Thus it seems that there will be an increase in sludge if chemical precipitation is used to remove P during wastewater treatment. The estimated sludge increase ranges from 15% of total sludge to 94% of primary sludge, depending upon the specific operation considered and the specific operating conditions. In addition, chemical treatment systems increase the amounts of chloride or sulphate discharged to receiving waters. However, the chemical phosphate removal process, which can operate at any phosphate concentration and temperature, can remove 95% or more of the phosphate present in sewage, and can remove phosphate down to very low concentrations, sufficient to meet the most stringent regulatory discharge limits (SCOPE, 1999).

The above considerations show that there will be no significant increase in sludge volume for an activated sludge process if Zeolites are used to replace phosphate in washing products. Biological phosphorous removal will also cause no increase, and perhaps a small decrease, in sludge volume. However, chemical phosphorous removal will cause an increase in sludge, due to both additional solids removal in primary treatment and to the additional mass of chemical added to induce phosphate precipitation. Estimates of the increase in sludge production range from about 15% to greater than 50%.

Concerning the quality of the sludges produced by each process, the use of Zeolite, which decomposes to natural aluminosilicates, will cause no problems if the sludge is deposited on agricultural soil as a fertiliser. Sludges from biological P removal processes contain additional P, with the highest percentage (85–95% P removal) processes producing sludges with a P content of 2–7%, as compared with a waste activated sludge P content of 1.5-2.5% from an activated sludge process alone (SCOPE, 1998a, 1998b). In many cases this will increase their suitability for use in agriculture, provided that the soils to which they are applied are not already saturated with respect to phosphorous. However, sludges resulting from chemical precipitation of phosphate may not be suitable for agricultural use (SCOPE, 1998c). Thus either the use of Zeolites, or biological rather than chemical phosphorous removal processes, should provide sludges of acceptable quality.

In conclusion, in order to answer to question 3, it is opinion of the CSTEE that:

- the use of Zeolites in detergent products should not increase the amount (volume) of sewage sludge produced, or lead to a sewage sludge of unacceptable quality for agricultural use;
- use of phosphates in detergent products will not increase sludge volume, and may increase the suitability of sludge for use as a fertilizer, if biological phosphate removal processes can be used in sewage treatment;
- in contrast, chemical phosphate removal (the most effective and extensively used procedure for phosphate removal in Europe) will lead to an increased amount of sludge, at a lower sludge quality.

#### Additional phosphate substitutes in detergents

The function of phosphate in detergents cannot be replaced by a single substance; a combination of various compounds is needed. In addition to the most important builder in detergents Zeolite A, other phosphate substitutes are used such as polycarboxylates, phosphonates, NTA, citrate and soda. Therefore, even if not explicitly mentioned in the Terms of Reference, the CSTEE recognise the suitability of adding some comments on possible additional phosphate substitutes.

Polycarboxylates are water soluble, linear polymers characterized by numerous carboxylate groups. The main function of polycarboxylates in detergents is their "threshold effect"; that is, they act as dispersing agents to prevent the deposition of salts on the fabric. Polycarboxylates are used in substoichiometric amounts, so that a fraction of 2% to 6% in detergents in combination with other builders is sufficient. Softening the water, by the addition of Zeolites is required for the carboxylates to work properly. The polycarboxylates are not completely degraded during sewage treatment, and can be accumulated in the sewage sludge, so they will generally end up in the terrestrial environment, after sewage treatment and incorporation into sludge.

Phosphonates, the salts of the organic phosphonic acids, are characterized by the presence of one or more  $C-PO(OH)_2$  groups in each molecule. Four different phosphonates are commonly used in detergents. Phosphonates in detergents both prevent the deposition of incrustations on fabric as well as stabilize the bleaching agent during washing and storage. Because of the phosphonates' threshold effect (optimal effectiveness at very low concentrations), they comprise only 0.2 to 0.5 weight % of detergent components in combination with other builders. Phosphonates have properties that differentiate them from other chelating agents that greatly affect their environmental behaviour. Although phosphonates are very water soluble, they have a very strong interaction with surfaces, which results in a significant removal in technical and natural systems. No biodegradation of

phosphonates during water treatment is observed but photodegradation of the Fe(III)-complexes is rapid (Nowack, 2003 and therein cited literature). The lack of information about phosphonates in the environment is linked to analytical problems of their determination at trace concentrations in natural waters.

The softening effect of NTA and citrate on water occurs via the complexing of calcium and magnesium ions. The good biodegradability has been extensively documented.

The CSTEE considers that a risk assessment of all substitutes should be conducted.

#### References

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