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

"RISKS TO HEALTH AND THE ENVIRONMENT RELATED TO THE USE OF MERCURY PRODUCTS"

(RPA FINAL REPORT: J372/MERCURY)

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

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INTRODUCTION

The consultant was required to estimate releases of mercury to the environment and associated risks to the environment and human health from the use of mercury in a number of specific applications, i.e. dental amalgam, batteries, measuring instruments (such as thermometers and manometers), lighting.

The report assesses the use of mercury in these specific applications within the European Union, performs a life-cycle assessment and estimates releases from these products. Environmental concentrations and human exposures to mercury and its compounds from the use of these products are then predicted using EUSES. Based on the results of the predicted exposures and human and environmental effects of mercury, it is concluded that significant risks to the environment and human health are unlikely to be associated with the mercury containing products under study.

Since care needs to be taken when applying EUSES to metals and their derivatives, the CSTEE was asked to examine the conclusions of the report and provide specific answers to the following questions:

1. Do the results achieved with the refined EUSES model reliably predict the risks to the environment and health associated with the use and disposal of mercury containing products? If not, does the model lead to an over- or underestimation of the risk?

2. Does the CSTEE agree to main conclusion that potential risks to the environment related to mercury in products should be considered as local risks close to manufacturing sites (see page 104 of the report)? Would they agree to the conclusion that the risks to human health are low for the general population? Are the margins of safety estimated sufficient given the specific properties of mercury?

3. If the EUSES model, even if modified, is not considered to be a useful tool to predict the risks to the environment associated with mercury in products under consideration, is the CSTEE aware of any other simple and validated model that could be used for a risk assessment on mercury.

The CSTEE has examined the document and has a number of questions and comments on the document and the approaches and assumptions used. These comments will be detailed below after responses to the specific questions.

Question 1

As also stated by the authors, the results of this study should be treated with caution. EUSES is not directly suitable for the modelling of distributions of metals in the environment. Adaptations can be made to circumvent some of the problems when modelling distributions of metals with EUSES, but the problem of speciation of mercury and the complex environmental chemistry and biochemistry of mercury cannot be solved in a satisfactory way in EUSES. Due to the high uncertainty of the outcome of the

model calculations and thus high uncertainties regarding predicted environmental and human exposures to mercury, no science-based conclusions regarding risk assessment are possible.

Question 2

The analysis below shows that both over-and underestimation of exposures to different mercury-species can result from the approach followed by the consultants. This results in a high uncertainty in the estimated values. Therefore, the CSTEE cannot support the statement that the potential risks related to mercury in the products studied are limited only to local risks close to production sites. Due to the problems with the application of EUSES to metals and the many assumptions on mercury emissions from the products under study, no scientifically justified conclusions on potential risks are possible based on the present report.

Question 3

Speciation models are available to predict the behaviour of metals in a particular compartment, but an integrated modelling of the behaviour of metals in all environmental compartments simultaneously is not possible with these systems. A major problem in the applications of models to predict the fate of mercury and thus environmental concentrations and human exposures to the different toxicologically relevant species of mercury (Hg^0, Hg^{++}, CH_3Hg^+) is the limited knowledge of the transformation reactions between the different mercury species in environmental compartments.

The evaluation results in the following overall conclusions and recommendations by the CSTEE:

- The EUSES model is not adequate for the prediction of environmental and human exposures to mercury and its compounds from the products under study; therefore, no conclusions are possible based on the report.
- The incomplete comparison of predicted mercury concentrations with measured data on mercury concentrations further reduces the confidence in the conclusions made in the report.
- On the regional and continental scale, an assessment of the contribution of the products under study in the report may be made based on a comparison of emissions from the products under study with total mercury emissions from all sources.
- Regarding local emissions, the risk assessment should rely on measured data.

GENERAL COMMENTS

The report reflects the application of EUSES to estimate risks from mercury at production and in specific products. Aside from the question whether this is a good idea, the authors have done a reasonable job. Provided that the model is suitable, it is legitimate to calculate the additional risk of mercury compounds in specified products in relation to natural and anthropogenic background levels of mercury from all sources.

The EUSES distribution models at the local and regional scales are linear, e.g. a doubling of an emission to a compartment will also lead to a doubling of the risk characterisation ratio.

This also means that the question of the overall contribution of the products considered to the total risk of mercury could have been answered on the basis of emissions, i.e. the contribution of the emissions from the products under discussion to the total emission at the regional and continental scale. If the aim of the study also is to characterise the absolute risks of mercury in each of the product categories, then

this approach would not be sufficient and risk assessments are needed on the basis of measured data and/or modelling.

As the authors point out themselves, modelling metals in general, and mercury in particular, is a very difficult task in view of the speciation, which for mercury can change over time and depends on the environmental compartment in which mercury is present. This speciation will result in different fates and environmental behaviour and in changing bioavailability to organisms. The speciation will also result in a different toxicological assessment since the toxicities of elemental mercury, mercury ions and methyl mercury are different. The specific comments below will address some of these difficulties.

The CSTEE agrees with the warning in the report "that the results generated must be treated with a degree of caution" (paragraph 5.5.1).

However, the uncertainties in the risk assessment for mercury-containing products using EUSES should have been addressed more thoroughly in the report, among others by a comparison between measured and modelled concentrations. Given the difficulties of modelling, the exposure estimation for mercury should preferably be based on local and regional measured data. The most severe error is the underestimation of the emissions at primary production.

The exact calculations performed with EUSES are not transparent. Among others, the following data are not reported:

- The fraction of the main source (local assessment).
- The number of emission days (local assessment).
- The regional tonnage.
- $PEC_{oral,fish}$ and $PEC_{oral,worm}$.
- PECs for all life cycle stages: although not clearly stated it seems that for the local environment only the highest PECs are reported.
- Assessment factor used for the derivation of the PNEC for predators.
- The derivation of the disposal emission factor for wastewater of dental amalgam and measuring and control equipment: it cannot be derived from Table 3.8.

The EUSES export files, submitted by RPA to the CSTEE, show the data with regard to bullet 1-6. However, the rationale for the choice for the fractions of the main source, the number of emission days and the regional tonnage is not clear. For instance: for the use pattern "batteries", the regional tonnage is chosen to be 1 tonne/year (EU consumption stated to be 8 tonnes/year) and the fraction of the main source 0.3 for formulation and 0.15 for processing.

From the export files it is clear that indeed the highest local PECs per use pattern are reported and that the correct assessment factor is used for the derivation of the PNEC for predators.

SPECIFIC COMMENTS

Emissions

The application of EUSES is necessary especially for estimating PECs at the local level. Comparison of the emissions would probably give a picture of relative risks away from local sources, and perhaps should have been done and compared to the corresponding output of EUSES for the regional and continental levels.

Emissions to the atmosphere after disposal are accounted for, but not emissions to soil and ground/surface water. Depending on the disposal conditions, this may lead to an underestimation of the PECs for these compartments.

Emission factors for different sites in some cases seem to have been pooled for the local assessment, e.g. in the case of emissions to the atmosphere from incineration, scrap steel and deposition of lamps in landfills. This may result in an overestimation of the PECair for disposal at different sites.

Emission factors at production are applied to the total consumption of mercury in 2000 in Europe (page 74, last paragraph). However, this consumption (120 t/year and 33% of this value, i.e. 40 t/year, assumed to be related to primary production) is only approximately 25% of the total production in Spain. This results in a severe underestimation of the risks from primary mercury production. If the emission factors were applied to the total production by mining in Spain (500 t/year), the PEC/PNECs for primary production will all be approximately 10 times higher. The relative importance of the risks of mercury release from non-ferrous metals processing and secondary production will be far less.

In paragraph 5.2 (p. 74) it is stated that an emission factor to air for primary production of 0.01 has been suggested in recent EU guidance (EU, 2001), but "a much lower figure may be appropriate given the IPPC requirements", and that MAYASA claim a value of <0.001. As input for EUSES a value of 0.001 was adopted, based on non-referenced "report for MAYASA". Given that, as the report acknowledges, the impact of the primary production emission factor to air on the EUSES output could be significant, the selection of the low value should be justified.

The estimates on which life-cycle assessments are based carry high uncertainties and seem to be based only on very limited life-cycle measurements.

Behaviour in air

The assumption is that all mercury emitted to the atmosphere is either elemental mercury or mercuric chloride. In the first case, this would mean an overestimation of concentrations in air (slow deposition), in the second case an overestimation of the PEC_{soil} (fast deposition).

The fraction bound to aerosol is assumed to apply only to inorganic mercury. It is highly variable: the higher this fraction, the lower the PEC of inorganic mercury in air and the higher the PEC_{soil}. A default of 0.09 is chosen. Although not reported, the report seems to assume that the fraction of elemental mercury bound to aerosol is 0. Other reports, however, assume a fraction of 1-5% (e.g. Slooff *et al.,* 1995).

Behaviour in water

Chemical and biochemical transformations of elemental mercury and inorganic mercury in water cannot be modelled with EUSES. The kinetics of these transformations are strongly dependent on pH and redox potential and the presence of ligands (chloride, sulphate, humic acids). The report assumes, as a worst case, mercury to be present as inorganic mercury and takes an additional 10% of the calculated PEC for methyl mercury. This is a valid approach. With regard to sorption to suspended sediment, the value used for the K_p is acceptable.

The use of the value of the Henry's law constant for elemental mercury is, however, not correct. This parameter is used to describe the flux from water in case of emissions via wastewater occurring at manufacture and disposal. Using the high value for elemental mercury will lead to a strong overestimation of PEC_{air} (from the STP) and underestimation of PEC_{water} considering that inorganic mercury is the major species present in water. From the data presented it seems as if the PEC_{air} is only based on the direct emission to air from the point source or landfill and not on the emission to air from the STP. The error only will influence the outcome of the study in the latter case.

Behaviour in soil/sediment

The value used for the K_p for sediment is acceptable. The value used for soil, however, seems very high. A value of 158 (log K = 2.2) can be derived from a US study with 11 soils (Bockting et al., 1992; Slooff *et al.*, 1995). High values up to a $\log K_p$ of six have been found for adsorption to iron oxides and bentonite clays. High chloride and humic acid concentrations may strongly reduce sorption. The approach taken will lead to an overestimation of the PEC_{soil} and an underestimation of the concentration in porewater and leaching to groundwater.

Methylation of inorganic mercury and washout of organic mercury may occur in soil and particularly in sediment through chemical and microbiological action. The assumption of 100% presence of inorganic mercury in soil is not worst case, but seems acceptable. The assumption of 10% organic mercury in sediment is worst case and acceptable for this analysis.

Behaviour in biota – environment

The BCF-values used apparently are based on field studies and as such include bioconcentration and bioaccumulation. The values chosen are worst case and acceptable for this analysis.

In section 7.2.3., PNECs are alleged to be compared to "the water and soil PECs". However, the comparison of the PNECs should be with the estimated or measured PECs in fish and worm. However, the EUSES export files show that the calculations have been performed according to the TGD.

The assessment factors used in the derivation of the PNEC for predators are correctly chosen. It is noted, that according to the updated TGD higher assessment factors should be used.

Human and environmental exposure assessment

Concentrations of mercury and its compounds in environmental media and human exposures attributed to the specific uses of concern are estimated by EUSES. This is what the mandate asked. However, the presentation of these results alone, without being placed into the perspective of the total exposure, constitutes a serious deficiency which does not permit correct conclusions at the risk management level.

Local and regional PECs are derived from the model output and compared to PNEC, and, for most

endpoints considered, PEC/PNEC ratios are significantly smaller than 1 with regard to the production and use of the products under consideration. However, PEC/PNEC ratios close to 1 are calculated for CH_3Hg^+ due to the high potential for bioaccumulation. The authors consider that some risk reduction measures may be needed. If the background PEC/PNEC is already above 1, the contribution of the sources under consideration in the report should be estimated and clearly pointed out for risk management. In fact, the information needed to put the specific use-associated vs. total exposure-associated risks into perspective is present in the report, but the comparison is not completed.

The first line of each of Tables 5.8-5.11 (p.81-84) give the values of measured environmental concentrations, which represent the contribution of all sources, while below in the same Tables the corresponding concentrations estimated by EUSES for the specific uses as a group (*they should also have been given for each use individually)* are shown. The latter are used to calculate the corresponding MOS values (shown in the other columns of these Tables) by dividing by the relevant RfD's. If the total exposures were also used for similar MOS calculations, the results would have placed the use-specific estimated risks into the right perspective. For example, inspection of the figures given in Table 5.8 (p.81) suggests inhalation exposure (and hence the associated risks) are substantially increased in the vicinity of primary production plants of mercury, and even for some of its products, even if the estimated MOS values (Table 7.5, p.105) are higher than 1.

The estimated human exposure levels for the various scenarios are not presented in the report, but only the resulting MOS values are given (Table 7.5 p.105 and p. 109).

Regarding the general human exposure to mercury and its compounds from all sources, 10 µg/day are used and it is assumed that 50 % of this dose is in the form of methyl mercury. The other 50% are assumed to result from amalgam fillings.

Based on a recent surveys and the IPCS-document on methyl mercury, total human mercury exposure from environmental sources is varies from 3 to 13 µg/day (depending on fish consumption) for the general population with more than 90 % of methyl mercury from seafood. Exposure is highly dependent on fish consumption and consumption of a single portion freshwater fish from contaminated lakes in Scandinavia may give a dose of up to 200 µg methyl mercury/person.

The report should separately assess risks from methyl mercury in sea food and from elemental mercury in dental amalgams. The inclusion of direct exposures (to the individual with the fillings) to dental amalgams in the exposure assessment is questioned since the report focuses on environmental exposures to mercury and its compounds. The individual exposures due to the release of mercury from dental amalgams may be highly variable and, for example, are dependent on the use patterns for chewing gum. The integration of indirect exposures to Hg from the preparation and application of amalgams is correct. Moreover, Hg^0 exposure is different regarding health risks from methyl mercury.

Toxicology of mercury and methyl mercury

The report does not attempt to describe the toxicity of mercury in any detail, but aims to come to a selection of limit values based on a number of recent authoritative reviews (WHO 2000, ATSDR 1999, DEFRA 2002; UNEP 2002). The key effects upon which the limit values are based are correctly identified as effects on the nervous system for inhaled mercury vapour, the kidney for ingested inorganic mercury compounds and the central nervous system of children for ingested organic mercury compounds. The CSTEE notes that no significant information modifying these conclusions has appeared more recently.

The limit values adopted by the report (NOAELs for oral, dermal and inhalation exposures to inorganic

or organic mercury) are also taken from recent authoritative assessments (ATSDR 1999, USEPA 2001) and, where more than one such value has been derived by different organisations, the adopted values represent the most conservative ones. One limit value which is not mentioned in the report is the latest (2003) PTWI for methyl mercury developed by JECFA, which revises its previous value from 3.3 µg/kg per week to 1.6 µg/kg per week, corresponding to 0.23 µg/kg per day. However, this does not affect the report's conclusions since the corresponding limit value adopted is even lower, 0.1 µg/kg per day, derived by USEPA (2001) as the corresponding RfD. So the overall conclusion is that the report does not have any errors in its handling of the human toxicity aspects, including the limit values. However, the CSTEE notes the following problems regarding the handling of some limit values:

a) Those for dermal exposure are given as "2 µg/kg/day" for inorganic mercury and "0.1 µg/kg/day" for methyl mercury, which causes confusion since they have the wrong units (they would be expected to be in µg/cm2/day). If, as seems likely, the above figures are meant to indicate the corresponding absorbed doses, the implied assumption of 100% dermal absorption is neither explicitly stated nor justified.

b) Similarly, the limit value for inhalation of methyl mercury is assumed to be 10% of the corresponding value for inorganic mercury, without adequate justification and despite the fact that it is stated that no relevant data have been found (Tables 6.6 and 6.7, and p. 96, last paragraph).

The concept of MOS values is used in a different way from the standard way described in the TGD. Normally, the exposure values (measured or predicted) would be divided by the NOAEL or LOAEL values directly observed in humans or animals to derive the MOS. These would then be compared with a minimum MOS value which would incorporate safety factors taking into account interspecies and interindividual variation and other considerations. Unacceptable risk would be indicated if the calculated MOS is smaller than the minimum MOS.

In the report, the predicted exposures of mercury and its compounds are divided by Reference Dose Values (RfD) derived by the US EPA (erroneously referred to as NOAEL, Table 6.7, p. 95), which represent the maximum daily dose which can be expected to be without adverse effects, and which already contain various safety factors. The derived MOS values are then considered as reflecting no unacceptable risk if they exceed a value of 1. While this is not in accordance with the methodology of the TGD and causes confusion, the final conclusions regarding the risk assessment are not affected.

Ecotoxicology of mercury and methyl mercury

The assessment factor used to derive the PNEC for aquatic ecosystems is stated to be 5. However, according to both the new and the old TGD this factor should be 10, since NOECs for 3 different taxonomic groups are available. Use of a lower assessment factor should be fully justified.

REFERENCES

Bockting, GJM, Van de Plassche, EJ, Struijss, J and Canton, JH (1992) Soil-water partition coefficients for some trace metals. Bilthoven, RIVM, Report No. 679101003

Barregard, L, Sallsten, G, and Jarvholm, B (1995) People with high mercury uptake from their own dental amalgam fillings. Occup. Environ. Med., 52 (2): 124-128.

Slooff, W, van Beelen, P, Annema, JA and Janus, JA (eds.) (1995) Integrated Criteria Documents Mercury. Bilthoven, RIVM, Report No. 601014008

Gebel, T, and Dunkelberg, T, (1996) Influence of chewing gum consumption and dental contact of amalgam fillings to different metal restorations on urine mercury content. Zentralbl Hyg Umweltmed, 199 (1): 69-75.

Sallsten, G, Thoren, J, Barregard, L, Schultz, A, and Skarping, G (1996) Long term use of nicotine chewing gum and mercury exposure from dental amalgam fillings. J. Dental Res. 75 (1), 594-598.