Agricultural use of sewage sludge - Is there a need to revise the Swedish regulations pertaining to heavy metals?

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Background

This document is a complement to and update of the unpublished report "Land application of sewage sludge - review of national and international reports on allowable heavy metals loads" by Lage Bringmark (2002-04-19, Naturvårdsverket). It should therefore be read in conjunction with that report. The aim of the document is to specifically answer the question if there is new evidence necessitating a revision of the existing regulations on the agricultural use of sewage sludge that pertain to heavy metals in sludge and soil (SNFS 1994/2). No attempt is made to carry out a comprehensive, in-depth review of the scientific literature, but instead the Swedish limit values for heavy metals in soil are compared with those in other countries (mainly EU member states). A comparison is also made with recent soil quality standards for heavy metals in soils that are used in conjunction with risk assessment of contaminated land. Where it is deemed necessary to clarify the scientific basis for the limit values reference to the scientific literature is made but this does not represent an exhaustive review of the literature. No assessment is made of the need to revise limit values for cadmium as there is a broad consensus in Sweden for a policy to reduce cadmium loadings to a level corresponding to zero accumulation (see Bringmark 2002 for further details on the risk assessment of cadmium). Cadmium (and mercury) appear moreover on the EU list of priority substances with the aim that emissions, discharges and losses into water of these substances should cease (EC, 2001). For the elements that are currently not regulated but that have been deemed to potentially pose a risk, a cursory search of the scientific literature was carried out as basis for a tentative, qualitative risk assessment. In addition the KemI-Riskline Database[2], and information available through the websites of the EFSA (European Food Safety Authority[3]) and the US ATSDR (Agency for Toxic Substances and Disease Registry[4]) were consulted.

The document restricts itself to discussing the scientific evidence for regulations on heavy metals and does not comment on judgements of what constitutes acceptable levels of risk which are a matter for policy. Limit values for heavy metal contents in sewage sludge are hence only discussed in relation to whether there is a scientific basis for a precautionary approach to metal loading rates.

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2 http://apps.kemi.se/riskline/index.htm
3 http://efsa.europa.eu/EFSA/efsa_locale-1178620753812_home.htm
4 http://www.atsdr.cdc.gov/

Conclusions and recommendations

- The Swedish maximum permissible concentrations of potentially toxic elements in soil to which sewage sludge is applied (SNFS 1994/2) are comparable to the lowest found in other countries. They are also comparable to the target values for soil quality that are believed to reflect the maximum soil metal concentrations that give full protection to soil organisms, processes and functions, as well protection of humans directly or indirectly in contact with the soil, and of surrounding ecosystems, including groundwater. These target values include those recently proposed for Sweden (Naturvårdsverket, 2009a). It is therefore concluded that there is no need to revise current limit values for the concentration of the regulated heavy metals in soil.
- In some countries limit values for soil are, as are the target values for soil quality, differentiated on basis of soil properties that affect metal bioavailability and toxicity. Swedish limit values only differentiate the limit value for Zn on basis of background concentrations in soil. It is recommended that an assessment is made whether there is a need to further differentiate limit values on basis of soil properties.
- The existing Swedish limit values are based on a limited amount of data and not a complete pathway analysis approach. It is recommended that in future use is made of the comprehensive pathway analysis approach that is used to derive target values for soil quality as a complementary tool in any further risk assessment of sewage sludge application to agricultural soil.
- At current concentrations in soil and sewage sludge (Eriksson, 2001), application of sewage sludge according to current regulations will increase soil concentrations of Au (gold), Ag (silver), B (boron), Sn (tin), Sb (antimony), W (tungsten) and Mo (molybdenum) at a nominal annual rate of >0.2%. These elements have therefore been deemed to constitute the largest potential risk. A cursory search of scientific literature, as well as comparison with target values that are available for some of these elements, suggest that the risk for phytotoxicity, ecotoxicity and for negative effects on human health due to food intake for most of these elements is low. The exceptions are for silver for which the risk for phytotoxicity is potentially moderate and the risk for ecotoxicity potentially moderate to high, and for Mo for which the potential risk to human health is moderate.
  On basis of the tentative risk assessment there appears to be no need to introduce regulations for B, Sn, Sb, W and Mo. No conclusions could be made concerning Au.
- The amount of evidence available for the risk assessment of the above, unregulated metals is extremely sparse and insufficient to establish any limit values for soil. Given the potential toxicity of silver it is recommended that effort is made, possibly through regulation, to further reduce the silver content of sewage sludge.
- Given the uncertainties in risk assessment of both the currently regulated and the unregulated elements, and the evidence which suggests that there may not be thresholds for toxicity effects on soil microorganisms warrant that the precautionary approach inherent in the current regulation on metal loading rates is maintained.
- The content of potentially toxic elements and other pollutants in sewage sludge changes over the years as it reflects current use of these substances in society.

There is therefore a need to periodically re-evaluate the presence of such substances in sewage sludge. One type of substance that has not been evaluated in this report are synthetic nanoparticles which in some products are combined with elements such as titanium, zinc, silver or gold. Currently, little is known about the toxicity of these substances or how nanoparticles may affect the toxicity and environmental behaviour of the elements with which they are combined. There is, however, awareness of this lack of knowledge and new evidence is likely to emerge in the coming years that should be assessed for its implications for the agricultural use of sewage sludge.

- The models used to derive target values for soil quality build often on a number of assumptions while the amount of data available is often considered to be very limited. There continues therefore to be a need to verify the derived target values through environmental monitoring of long-term field experiments and contaminated sites. Indeed, much of the initial evidence for toxicity of heavy metals to plants, invertebrates and microorganisms came from field studies of contaminated sites, e.g. around smelters or mining sites. Similarly, the potential toxicity of heavy metals in sewage sludge to soil microorganisms was not taken into account until evidence emerged from studies on long-term field experiments.

Maximum permissible concentrations of potentially toxic elements in soil

Bringmark (2002) compiled a table (Table 3 in his paper) comparing maximum permissible concentrations of potentially toxic elements in soil (limit values) in the sludge regulations of several countries. The table has been updated (UK, Denmark) and the German and the new Swedish generic guideline values (soil quality standards) indicating concentrations where the risk for negative effects on human health, the environment or natural resources is acceptable, have been added (Table 1).

A comparison of the numeric values in Table 1 suggest that the Swedish limit values for soil metal concentrations are very similar to the lowest found among other EU member states as well as to those of Norway and Switzerland. They are also similar to the target values for soil metal concentrations in Germany, The Netherlands and Sweden. These target values are believed to reflect the maximum soil metal concentrations that give full protection to soil organisms, processes and functions, as well as protection of humans directly or indirectly in contact with the soil, and of surrounding ecosystems, including groundwater. On the face of such a comparison there would not appear to be any urgent need to revise the Swedish limit values.

It may, however, be argued that, as is evident in the UK limit values as well as the German soil quality standards, there may be a need to differentiate limit values on basis of soil properties. Differentiation of the limit values was already introduced in the Swedish regulations in 1998 when a higher limit value for zinc (150 instead of 100 mg/kg) was introduced for soils with high natural Zn background concentrations. It is more common, however, to differentiate on basis of soil properties such as soil pH, or clay and organic matter content that are known to affect mobility and bioavailability of many of the regulated metals. A recent study by Smolders et al. (2009) showed how derived PNECs (Predicted No Effect Concentration) for several of these metals were highly dependent on soil properties, including soil background concentrations. The PNEC for zinc, for example, ranged from 28 mg/kg in a highly sensitive soil to 286 mg/kg in a weakly sensitive soil. Compared to the latter, the highly sensitive soil was characterized by a low pH (4.5), low organic carbon (1.0%), a low clay (5%), a low eCEC (4 $cmol_c$/kg) and a low zinc background concentration (8 compared to 155 mg/kg). The PNEC in these soils for Cu and Ni were also affected by soil properties, though not background concentrations, and between the most and least sensitive soil varied between 30 and 162 mg/kg for Cu and 8 and 93 for Ni (values are for amounts added, not concentrations). The Swedish limit values were set so as to protect even sensitive soils (Witter, 1992), but the results from the study by Smolders at al. (2009) suggest that there may be a need to review if the existing Swedish limit values for soil need to be further differentiated on basis of soil properties. In its simplest form such a differentiating could specify that soils receiving sewage sludge must have a minimum pH value (e.g. 5.5), clay (e.g. 10%) and organic carbon (e.g. 1-2%) content, and/or eCEC value (e.g. 10-20 $cmol_c$/kg). A similar recommendation was already made in 1992 (Witter, 1992). It is unlikely that such a specification would greatly affect opportunities for agricultural use of sewage sludge as most agricultural soils will have values above those indicated by way of example.

Table 1. Maximum permissible concentrations of potentially toxic elements in soil in the EU sludge directive, in the sludge regulations of some EU member states and those of Norway, Switzerland and the United States, as well as soil quality standards in Germany, The Netherlands and Sweden. The soil quality standards indicate maximum soil metal concentrations that give full protection to soil organisms, processes and functions, as well as protection of humans directly or indirectly in contact with the soil, and protection of surrounding ecosystems.

Country	**Year**	**Cd**	**Cu**	**Cr**	**Ni**	**Pb**	**Zn**	**Hg**
Council Directive 86/278/EEC	1986	1-3	50-140		30-75	50-300	150-300	1-1.5
France	1988	2	100	150	50	100	300	1
Germany[5]	1992	1.5	60	100	50	100	200	1
Italy		3	100	150	50	100	300	
Spain	1990	1	50	100	30	50	150	1
United Kingdom pH 5.0 < 5.5	1996	3	80	400	50	300	200	1
United Kingdom pH > 7.0	1996	3	200	400	110	300	300	1
Denmark	2006	0.5	40	30	15	40	100	0.5
Finland	1995	0.5	100	200	60	60	150	0.2
Sweden	1994 (1998)	0.4	40	60	30	40	100 (150)	0.3
Norway		1	50	100	30	50	150	1
Switzerland	1992	0.8	50	75	50	50	200	0.8
United States[6)]	1993	20	750	1500	210	150	1400	8
Soil quality standards:								
Denmark[7]	2008	0.5 (0.3)	500 (30)	500 (50)	30 (10)	40 (50)	500 (100)	1 (0.1)
Germany[8], sand	1999	0.4	20	30	15	40	60	0.1
Germany, loam	1999	1	40	60	50	70	150	0.5
Germany, clay	1999	1.5	60	100	70	100	200	1
Netherlands[9]	2000	0.8	36	100	35	85	140	0.3
Sweden[10]	2009	0.5	80	80	40	50	250	0.25

5 New limit values have been proposed that conform to the soil quality standards (BMU, 2007).

6 Calculated as the maximum cumulative pollutant loading mixed into ploughed soil layer,

The Swedish limit values were derived in 1992 on basis of a limited analysis of available data and a lack of data for many of the regulated metals (Witter, 1992). Later studies, such as those forming the basis for the limit values in Denmark, as well as the very comprehensive analysis underpinning the German, Dutch and Swedish target values for soil quality, nevertheless arrived at values not dissimilar from the Swedish limit values. This may simply be a fortuitous outcome of the precautionary approach used when the Swedish limit values were derived in 1992. There are some important differences in how the Swedish limit values were derived and how, for example, the Swedish soil quality standards were derived. The target values for soil quality standards were derived from risk analysis using a multiple pathway analysis approach and considers targets such as groundwater which were not considered in the approach used by Witter (1992). The latter, however, focused more on evidence from long-term field experiments with sewage sludge as these provide a realistic scenario for assessing the risks associated with agricultural use of sewage sludge. Witter (1992) considered mainly soil (micro-)organisms, plants and human health as targets, and, except for Cd, limit values were determined by toxicity to soil (micro-)organisms. The two different target values for soil quality established in Denmark confirm that ecotoxicity is an extremely sensitive target (Table 1). Similarly, the Swedish target values for the regulated metals Cu, Cr and Zn were determined by their ecotoxicity, while the target values for Hg, Pb and Cd were determined by human health, and that for Ni by protection of groundwater (Naturvårdsverket, 2009b). Not all of the pathways in the latter study may, however, be relevant for when sewage sludge is applied in agriculture.

It would be wrong to assume that the apparent consensus between the Danish, Dutch, Swedish, Norwegian and Swiss limit and target values is due to a lack of ambiguity in the available toxicity data or its interpretation. To some extent the apparent consensus is due to similarities in approach and selection of targets. For example, the Swedish guideline values make use of the data used in the derivation of the Dutch standards (RIVM, 2001).

Even when targets and approach are similar, there are considerable uncertainties in the validity of the approach. For example, the approach used in the derivation of the guideline and intervention values for ecotoxicity is based on the so-called ‘‘species sensitivity distribution’’ or SSD approach (EC, 2003). In this approach the available data on toxicity to soil organisms and processes is combined and from these a value to indicate protection of all or a certain percentage of all soil organisms and processes is derived statistically. The approach builds on that organisms and processes differ in their sensitivity to metal toxicity and that this, after correction of the data for differences in bioavailability, is reflected in the differences in the toxicity data. As discussed by Giller et al. (1998) several other factors, of both a physico-chemical, a methodological and a biological nature, may however contribute to the differences in the toxicity data while there is uncertainty in how far the correction for differences in bioavailability are adequate. Smolders et al. (2009) attempted to circumvent some of these short-comings and found that differences in metal bioavailability was an important contributing factor to differences between soils in the observed toxicity of many metals to soil plants, soil invertebrates and soil

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without regard to soil background concentrations.

7 Soil quality criteria and eco-toxicological soil quality criteria in brackets (DEPA, 2002). The values conform to those in Miljøstyrelsen, miljøministeriet. Liste over kvalitetskriterier i relation til forurenet jord og kvalitetskriterier for drikkevand, opdateret december 2008.

8 Precaution values for soils pursuant to § 8 parargraph 2 No. 1 Federal Soil Protection Law. Federal Soil Protection and Contaminated Sites Ordinance (BBodSchV) dated 12 July 1999.

9 Generic target value (VROM, 2000).

10 Generic guideline values for “sensitive” soil use (Naturvårdsverket, 2009a).

microbial processes. The PNEC values derived from this study indicate that in sensitive soils where bioavailability is high metal loadings as low as 24 mg Zn/kg soil, 30 mg Cu/kg soil, and 8 mg Ni/kg soil will lead to soil concentrations corresponding to the PNEC values. The results from the latter study have so far not been taken into account in the derivation of any of the target values summarised in Table 1.

The SSD approach is also problematic when applied to soil microorganisms, an important target group for the ecotoxicity of metals. A major assumption in the SSD approach is that the ‘‘species’’ tested are fully representative of, and a random sample from all populations in the ecosystem to be protected, which in practice is rarely the case (Giller et al., 2009). There are two other reasons why this approach may not be directly applicable to soil microorganisms. First, the species concept is not really applicable to microorganisms, especially bacteria. Second, the majority of effects on microorganisms reported in the literature focus not on species, but on microbially-mediated processes, which may be mediated by consortia of microorganisms, with consortia composition differing at different sites and at different times, but performing the same processes. What the SSD actually represents is the likelihood of finding effects in other studies based on the same assays as were used to construct the original frequency distribution. It also remains to be determined whether organisms with particularly important functions are more sensitive than the estimated PNEC (Giller et al., 2009).

The SSD approach is therefore problematic and has many uncertainties, but is currently perhaps the best pragmatic approach we have to derive PNEC values from the available toxicity data in order to arrive at limit values that meet the aim of protecting all or some of the ecosystem species and functions.

As Giller et al. (1989) stated, continued research is likely to find negative effects on soil microorganisms at smaller and smaller metal loading rates. This prediction has largely been borne out and, moreover, it appears that for some effects a threshold for toxicity effects may not even exist (Bünemann, et al., 2006). Effects are therefore sometimes seen at even very low metal loading rates (Dahlin et al., 1997; Witter et al., 2000; Chaudri et al., 2008). Some of the effects at low metal loading rates are changes in the structure of the microbial community and the development of metal tolerance among the soil microorganisms. It is still unclear what the implications of such changes are for the maintenance of soil ecosystem functions.

In conclusion it can therefore be said that the Swedish limit values for soil metal concentrations are consistent with the lowest limit values among EU member states, and with the German, Dutch and Swedish soil quality standards[11] that are believed to give full protection to soil organisms, processes and functions, as well protection of humans directly or indirectly in contact with the soil, and of surrounding ecosystems. There seems therefore to be no urgent need to revise existing limit values. New evidence on the importance of soil properties in determining bioavailability and thereby toxicity of several heavy metals, however, suggests that it may be necessary to differentiate metal limits according to soil properties, or to alternatively, prohibit the application of sewage sludge on the most sensitive soils.

Meanwhile, uncertainties in the SSD approach and in the interpretation of the available toxicity data, especially that pertaining to soil microorganisms, means that the derived PNEC, limit values, or soil target values should not be interpreted to guarantee full protection of all soil organisms and ecosystem functions.

11 Reasons why the USEPA limit values markedly deviate from these is discussed by Bringmark (2002)

Is there a need to regulate other potentially toxic elements?

Historically, only some of the potentially toxic elements are regulated for the agricultural use of sewage sludge. This in part reflects the potentially toxic elements in sewage sludge that several decennia ago were considered to be most problematic, and in part earlier technical limitations to analyse some elements at low concentrations. The EU Council Directive on the agricultural use of sewage sludge (86/278/EEC) regulates 7 elements and in their adoption of the directive EU member states as well as Norway have, as far as I am aware, restricted their regulations to these elements. The USEPA Part 503 Biosolids rule initially also included molybdenum, but this was subsequently dropped. In a currently ongoing re-evaluation of the agricultural use of sewage sludge in Norway, the Norwegian Scientific Committee for Food Safety (Vitenskapskomiteen for mattrygghet) has asked that the following additional elements should also be evaluated: arsenic (As), silver (Ag), tin (Sn), selenium (Se), bismuth (Bi), vanadium (V), antimony (Sb), tungsten (wolfram, W), beryllium (Be) and molybdenum (Mo). This evaluation includes an assessment of whether the contents of these elements in sewage sludge in Norway constitute a potential risk. That an element appears on this list does therefore not imply that the Norwegian Scientific Committee for Food Safety has deemed that the content in sewage sludge poses a potential risk. The Danish EPA published a study 6 years ago in which the potential hazard of Sb, Be, Bi, B, Ga, In, Li, Mo, Pd, Pt and V was evaluated. The elements were chosen on basis of their potential for significant adverse health and environmental effects, including long-term effects and that they are used in significant or increasing amounts (Kjølholt et al., 2003). The choice was, however, not related to their content in sewage sludge.

There is broad agreement among interested parties in Sweden (Naturvårdsverket, LRF and Svenskt Vatten) that there is a need to assess whether there is a need to regulate elements for which their soil concentration in soil would double within a time-span of 500 years (nominal annual accumulation rate >0.2%) at current loading rates when sewage sludge is used in agriculture need to be regulated. Somewhat depending on what data is used for current concentrations in sewage sludge there appears to be agreement amongst these parties that the elements that need to be assessed include the following (in order potential of rate of accumulation in soil): Au (gold), Ag, B (boron), Sn, Sb, W and Mo.

Toxicity data for these elements is very limited. Table 2 shows the soil quality standards that are available. For most of the elements no data (nd) is available. The derived values for soil quality (corresponding to no effect) or for effects (generic soil clean-up trigger values) indicate that these are at least 5 to 10-fold over background concentrations in Swedish soils for the elements (Ag, Sn, Sb, Mo) for which standards have been derived. The derived values are, however, based on few data and are therefore highly uncertain.

A preliminary risk assessment of these elements is also given in Table 2 and indicated as low, moderate or high. With this is meant the likelihood that a doubling of concentrations in soil will lead to undesirable toxicity effects. The assessment is based on a mere cursory search of the scientific literature and the assessment is therefore highly tentative, but is intended to give an indication of the risks associated with their accumulation in agricultural soils.
No assessment was made of the risk of these elements to livestock, groundwater or surrounding ecosystems. The assessment of risk to human health is cursory and only based on exposure through intake of foodstuffs and is made on the assumption that a doubling of soil concentrations for an element will also double the concentration in the crop.

The toxicity of some of these elements is highly dependent on their speciation. This has not been taken into account in the tentative risk assessment. Tin is highly toxic as a metal-organo compound, the most commonly used being tributyl tin. In the tentative risk assessment such metal-organo compounds have not been considered.

Table 2. Soil content (25 percentile, median and 75 percentile; Eriksson, 2001) and soil quality standard for the metals with an estimated nominal accumulation rate >0.2% per year when sewage sludge is applied to land. nd = no data. The tentative risk assessment assesses the risk if doubling of soil concentrations will lead to adverse effects.

	Soil content	Soil quality standard				Tentative risk assessment		
	mg/kg 25%-median–75%	Den-mark	Ger-many	Nether-lands[12]	Sweden	Phyto-toxicity	Eco-toxicity	Human toxicity
Au	< 0.005	nd	nd	nd	nd	nd	nd	nd
Ag	0.06 – 0.09 – 0.15	1	nd	nd (15)	nd	Moderate (?)	Moderate - High (?)	Low
B	3.2 – 4.8 – 6.6	nd	nd	nd	nd	Low	Low (?)	Low
Sn	1.0 – 1.4 – 2.3	20	nd	19 (900)	nd	Low (?)	Low (?)	Low
Sb	0.20 – 0.25 – 0.30	nd	nd	3 (15)	12	Low	Low	Low
W	0.98 – 1.3 – 1.7	nd	nd	nd	nd	Low (?)	Low (?)	Low (?)
Mo	0.20 – 0.36 – 0.67	2	nd	3 (200)	40	Low	Low (?)	Moderate (?)

Gold

There is virtually no information available on either the phyto- or ecotoxicity of gold in the soil environment. Plants are able to take up gold from soil but the bioavailability of gold in soil is very low and uptake is enhanced in the presence of chelating agents (Bapula et al., 2008). These authors also reviewed evidence that gold is taken up in ionic form but is then reduced in the roots to Au(0), presumably reducing its toxicity. No data on its phytotoxicity appears to have been reported. Similarly, I have not been able to find any reports on the ecotoxicity, nor human toxicity of gold. The US Agency for Toxic Substances and Disease Registry does not have a toxicological profile for gold and no information is available from EFSA (European Food Safety Authority).
Bringmark (2002) arrived at a similar conclusion.

Silver

Silver ions are highly toxic to microorganisms in an aqueous environment. However, silver readily binds to soil mineral particles and to organic matter which greatly reduces its bioavailability (Throback et al., 2007). For example, despite its aqueous toxicity no strong inhibitory effect on microbial activity in sewage treatment plants

12 Generic target value and intervention value in brackets. For Sn there is no target value and the value for natural background concentrations is given. For Ag there are no data on target nor background values. For Sb the target value is the same as the background value. There are no intervention values for Sn and Mo and instead the value indicating serious soil pollution is given.

are generally observed because of its reduced bioavailability due to rapid complexation and adsorption (Anonymous, 2002).

Bringmark (2002) concluded that silver in the soil environment is highly toxic, especially to microorganisms rather than plants. He arrived at this conclusion mainly on basis of the results of Johansson et al. (1998) who showed effects of addition of silver salts to soil at 5 times background concentrations. The sensitivity of denitrifying bacteria to silver was confirmed in a follow-up study (Throback et al., 2007), but the derived EC50 values were twice as high as in the earlier study which the authors ascribed to higher background concentrations of silver in the later study. Murata et al. (2005) found that even though silver ions present in an aqueous growth medium was highly toxic to microbial growth, when added to soil inhibition of dehydrogenase activity in two soils was not inhibited until soil silver concentrations were 100 times background concentrations. In comparison, in the same study copper inhibited dehydrogenase activity at 50 times background concentrations.

In general, accumulation of silver by terrestrial plants from soils is low, even if the soil is amended with silver-containing sewage sludge or the plants are grown on tailings from silver mines, where silver accumulates mainly in the root systems (Anonymous, 2002). Addition of silver ions has been shown to reduce growth of lettuce in soil (Brandt et al., 2005), but studies in aqueous nutrient solutions suggest that silver is less toxic to root growth than copper, nickel, lead, chromium, mercury and zinc (Samantaray et al., 1996).

The European Food Safety Authority uses a migration limit value for the content of silver in food of 0.05 mg Ag/kg (wet weight) food in its assessment of food additives and materials in contact with food (EFSA, 2005). Eriksson (2001) found that the median content of silver in wheat grain was < 0.0005 mg Ag/kg (dry weight) with a highest value of 0.0014 mg/kg. Given the enormous disparity of at least 2 orders of magnitude between the limit value and current wheat grain concentrations the risk associated with doubling soil Ag concentration for human toxicity via dietary would seem to be negligible.

It is clear from the above studies that silver is toxic, especially to microorganisms in aqueous media, but far less so when added to soil. Added to soil, it appears to be not more toxic than other metals such as copper or zinc and uptake by plants is very low. The available data is very scant, but suggests that it is unlikely that even a doubling of soil background concentrations would result in phytotoxic effects or unacceptable human exposure through dietary uptake. It can, however, not be excluded that soil microorganisms can be negatively affected.

Boron

Boron is an essential plant nutrient with crop requirements ranging between 0.1 to 1.0 kg B per year. Soils are believed to be deficient if the boron content of the soil falls below 0.5 – 1.0 mg/kg and recommended rates of fertilization with boron to deficient soils are between 1 and 2 kg/ha and year (Swedish Board of Agriculture, 2008). This can be compared with an annual load of on 45 g B/ha when sewage sludge with a weighted average B content is applied at a rate of 22 kg P/ha, while application of farmyard manure at the same P rate would supply 55 g B/ha (Eriksson, 2001). Given that the load of B with sewage sludge is similar to that of farmyard manure and 20-40 times less than when B is applied as fertilizer it seems highly unlikely that the load of B with sewage sludge will give rise to any negative effects. The highest concentration of B found in sewage sludge by Eriksson (2001) was about 6 times the weighted average and therefore still below amounts applied as fertilizer but similar to crop requirements. At high concentrations boron can be phytotoxic but plants appear to have a considerable tolerance. The boron content of wheat grains, for example, can be raised more than 20-fold without negative effects on seed germination and seedling growth (Marschner, 1995).

In its Directive relating to plastic materials and articles intended to come into contact with foodstuffs the EC uses a migration limit of 6 mg B/kg (fw) food (EC, 2002). This restriction can be compared to a median and maximum content in wheat grain in Sweden of 0.74 and 0.83 mg/kg (dw), respectively (Eriksson 2001). It would therefore seem highly unlikely that even a doubling of B concentrations in soil and grain would lead to that the restriction value would be exceeded.

From the above, and on the assumption that normal fertilizer practice for boron does not negatively affect soil organisms or crop quality, the risk for phytotoxic, ecotoxic or human toxic effects through the food chain due to B in sewage sludge would seem to be negligible.

Tin

Bioavailability and toxicity of inorganic tin compounds is thought to be low (Rüdel, 2003). It has not been possible to find any studies on the phytotoxicity of tin, although Bringmark (2002) remarked that tin is strongly toxic to crops under acidic conditions, but that uptake (and hence presumably toxicity) is low at neutral soil pH values. Little is known about the ecotoxicity of inorganic tin, but Rüdel (2003) quoted a study showing that its toxicity to daphnids may be 200 times less than that of copper.

Highest concentrations of tin in foodstuffs are found in canned foods. The EC has a maximum allowable tin concentration of 200 mg Sn/kg food (fw) (EC, 2006). This can be compared with a median and maximum concentration of Sn in wheat grain of 0.098 mg/kg (dw) and 0.12 mg/kg (dw), respectively (Eriksson, 2001). Doubling of the concentration of Sn in crops will therefore still leave Sn concentrations well below the EC limit value.

On basis of the above, and the values for tin in the Dutch and Danish soil quality standards (Table 2) which are more than 10 times current background concentrations in soil suggests that despite the sparsity of information there is little reason to believe that a doubling of tin concentrations in Swedish soils is likely to result in any negative effects on plants or soil microorganisms, or to lead to any adverse health affects due to increased concentrations in foodstuffs.

Antimony

At the time Bringmark (2002) wrote his report little was published on the toxicity of antimony. Since then one review has been published on the phytotoxicity (Tschan et al., 2009) and one study on the ecotoxicity (Oorts and Smolders, 2008) of antimony.

Antimony is used in relatively large amounts in flame retardants, electronics and cosmetics (Kjølholt et al., 2003). Toxicity of antimony is not well known, but Sb(III) species are usually more toxic than Sb(V) species and is comparable in its biochemical behaviour with arsenic and bismuth (Bapula et al., 2008). It can be toxic to both humans, animals and aquatic organisms (Kjølholt et al., 2003). Bapula et al. (2008), however, reported that bioavailability of Sb in soil is very low and that it appears that after uptake, plants convert the more toxic Sb(III) to the less toxic Sb(V) form, thus allowing plant tissue concentrations to increase without causing phytotoxicity. This transformation of Sb(III) to SB(V) appears also to take place in the soil solution. Oorts and Smolders (2008) found that within 2 days more than 70% of the added Sb(III) in the soil solution had been converted to Sb(V). Phytotoxic effects have been observed at soil Sb concentrations equivalent to 510 mg Sb/kg if all the Sb added solubilises in the soil solution, but no phytotoxicity was observed if the added Sb was allowed to equilibrate in the soil for one week (Oorts and Smolders 2008). It would therefore seem unlikely that phytotoxicity will occur under field conditions.

Inhibition of microbial processes (nitrification) was observed at the same concentrations as the phytotoxic effects, and were also absent if the added Sb was allowed to equilibrate for 1 week in the soil (Oorts et al., 2008). Also Murata et al. (2005) found no toxic effects to soil microorganisms (dehydrogenase activity and PFLA profiles) at Sb application rates up to 100-fold of background concentrations, even though Sb did inhibit microbial growth in aqueous solution.

The main effects of antimony on human health relate to the inhalation of dust particles (ATSDR, 1992). In experimental animals, the critical effect of antimony exposure is its effect on the respiratory passages. Lung tumors have been observed in female rats exposed to sparingly soluble antimony compounds (antimony trioxide, antimony trisulfide). Antimony compounds have been shown to be genotoxic in vitro, but there is no conclusive in vivo evidence of genotoxicity (Criteria group for occupational standards, 2000). Uptake of antimony by plants appears to be in proportion to soluble Sb concentrations in the soil, but it is only in plants grown on “heavily Sb-contaminated sites” that risks of toxicities to humans or animals is likely to occur (Tschan et al., 2009).

Antimony trioxide is used as a catalyst in the manufacture of PET and the EFSA has set a migration limit for Sb in food of 0.04 mg/kg (EC, 2007). This can be compared with a median and maximum concentration of Sb in wheat grain of 0.0004 mg/kg (dw) and 0.0007 mg/kg (dw), respectively (Eriksson, 2001). Doubling of the concentration of Sb in crops will therefore still leave Sb concentrations well below the EC limit value.

From the above, and comparison of the soil quality standard for Sb in The Netherlands which is 10 times higher than background concentrations in Swedish soils (Table 2), it appears that with a reasonable amount of confidence it can be stated that Sb has relatively low toxicity in the soil environment and that the likelihood of toxic effects to soil microorganisms, plants and consumers of plants is negligible at current application rates and Sb contents in sewage sludge.

Tungsten

Although tungsten is mainly associated with its use by the military (in ammunition) and as a filament in incandescent light bulbs, its presence in tyre studs is the main contributor to W in run-off water from roads in Sweden (Wik et al., 2008) from where it may enter sewage treatment plants.

Babula et al. (2008) cited studies showing that tungsten can both enter the food chain via the soil-plant system, and that it can cause phytotoxic effects. The phytotoxic effects are related to nitrate reductase activity in the root where W interferes with molybdenum in enzyme complexes. Similarly, tungsten can replace molybdenum in the enzyme nitrogenase in N_2-fixing microorganisms thus causing disruption of N_2 fixation and toxicity effects to the microorganisms (Wichard et al., 2008).

In a comprehensive study of the ecotoxicity of tungsten Strigul et al. (2005) found that W was highly toxic in aqueous solutions, but that toxic effects on plants, invertebrates and microorganisms in soil were only found at concentrations at or exceeding 1000 mg W/kg. These results are not dissimilar to those of Inouye et al. (2006) who found that W was toxic to earthworms, but at concentrations in soil above 700 mg/kg. These toxicity thresholds correspond to more than 500 times background concentrations in Swedish soils.

Tungsten is known to be toxic to humans and animals (Wilbur et al., 2007). Recently released evidence from the US military, however, appears to suggest that tungsten exhibits relatively little human-health toxicity (Pardus et al., 2009). The US Agency for Toxic Substances and Disease Registry concluded that members of the general public were unlikely to experience any health effects that would be related to exposure to tungsten or tungsten compounds (ATSDR, 2005). EFSA appears not to have any standards for tungsten in foodstuffs.

The evidence therefore suggests that tungsten has low toxicity in the soil-plant system, although there are numerous uncertainties and the available data is very scarce.

Molybdenum

Molybdenum is present in alloys such as stainless steel from where it can be leached. It can also be found in flame retardants, pigments, plastics and as a dietary supplement so it has several routes by which it can enter sewage treatments plants (Kjølholt et al., 2003). As pointed out by Bringmark (2002) molybdenum is an essential micro-nutrient for plants, microorganisms and ruminants. Molybdenum deficiency in plants is most likely to occur on acidic soils, and legumes are more sensitive to Mo-deficiency than other plants. Liming is often recommended to correct Mo-deficiency in crops. The Swedish Board of Agriculture has no recommendations on application rates for Mo in the case of deficiency. Plants have a high tolerance for Mo: the difference between threshold concentrations in plant tissues between deficiency and toxicity can be as high as a factor of 10^4 (0.1 – 1000 µg Mo/g dw), whereas that for B is a factor of 10 (Marschner, 1995). The likelihood for phytotoxicity is therefore very low. Somewhat elevated concentrations of Mo (5 – 10 µg Mo/g dw) in forage plants may, however, be sufficient to induce an imbalance between Mo and Cu in the diet of animals, especially ruminants, leading to a condition known as molybdenosis. There appears to be no information on the extent of molybdenosis in livestock in Sweden. The so-called “mysterious wasting-disease” observed in elk during the late 1990's is, however, thought to have been partially due to molybdenosis (Frank, 2004) but is entirely unrelated to Mo in sewage sludge.

Per amount of P the concentration of Mo in sewage sludge is lower than that in animal manures (Eriksson, 2001). It is therefore unlikely that the application of Mo-containing sewage sludge will contribute to occurrence of molybdenosis in livestock, were it to be used on livestock farms which is in any case unlikely.

There are few studies on the ecotoxicity of Mo in agricultural soils, but Åkerblom et al. (2007) found that in the mor-layer of a forest soil Mo was less toxic than either Zn, Cr and Pb. In an assessment of toxicity of metals to the benthnic worm *Tubifix tubifix* Mo was found to be less toxic than Cu, V, Hg, Mn, Ni, Cd and Cr, but more toxic than Pb, Sn and As (Fargasova, 1999). Kjølholt et al. (2003) concluded on the basis of toxicity tests on aquatic organisms that Mo has a low to moderate ecotoxicity.

Although the available data was scarce, the EC Scientific Committee on food established a Tolerable Upper Intake Level of Molybdenum which is six times the mean estimated intake of 100 µg Mo/day for adults in 11 different countries and exceeds the upper range of intakes for The Netherlands (96 µg/day), Sweden (260 µg/day), the UK (400 µg/day), Germany (500 µg/day), and Finland (150 µg/day) (EC, 2000). Doubling intake of Mo in Sweden would therefore bring it close to this upper limit, and similar to current intake in Germany.

It can be concluded that the risk for phytotoxicity due to the application of sewage sludge under the current Swedish guidelines is minimal. The risk for ecotoxicological effects appears to be small on basis of the studies cited above, and the soil quality standards shown in Table 2. The uncertainty in the data is, however, very high. The risk to human toxicity is deemed to be moderate as doubling intake of Mo would bring it close to the tolerable upper intake level.

Is there a need for a precautionary approach to metal loading rates?

In the regulation for the agricultural use of sewage sludge in Sweden limit values for metal concentrations in soil represent the maximum concentration below which significant toxic effects on crops, livestock, soil organisms and humans consuming crops grown on such land is minimal. Despite a fair degree of consensus between the Danish, Dutch, Swedish, Norwegian and Swiss limit and target values, there remains a high degree of uncertainty in the derived limit values, especially so for the elements currently not regulated in SNFS 1994/2. Moreover, as for cadmium toxicity to humans, there may not be a threshold value for metal toxicity to soil microorganisms. There are therefore valid scientific reasons to maintain a precautionary approach to metal accumulation in soil.

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