

# Proposal for recommended limits for impurities in zinc-based fertilisers

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CSIRO Land and Water Science Report xx/11 January 2011

Report for The International Zinc Association



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# **EXECUTIVE SUMMARY**

- The International Zinc Association requested CSIRO Land and Water to develop a set of proposed guideline values for selected contaminants in zinc (Zn) fertilisers – arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb) and mercury (Hg).
- A static 100-year simulation of Zn fertiliser use, combined with a range of environmental limit values (for harvested produce, soil, surface and groundwaters), and using a probabilistic approach, produced fertiliser contaminant limit values for As, Cd, Cr, Hg and Pb in Zn-based fertilisers.
- This approach was part of a larger risk assessment of contaminants in N, P, K and trace element fertilisers, as well as industrial wastes, conducted by CSIRO Land and Water for the Commonwealth of Australia Fertiliser Working Group, managed under the Department of Agriculture, Food and Fisheries in Australia.
- Fertiliser Contaminant Trigger Values (FCTVs) produced are summarised below.

| Element | FCTV values<br>(mg/kg Zn) | FCTV values<br>(mg/kg ZnO) | FCTV values<br>(mg/kg<br>ZnSO₄.7H₂O) |
|---------|---------------------------|----------------------------|--------------------------------------|
| As      | 6000                      | 4820                       | 1365                                 |
| Cd      | 680                       | 546                        | 155                                  |
| Cr      | 8100                      | 6508                       | 1843                                 |
| Hg      | 130                       | 104                        | 30                                   |
| Pb      | 530                       | 426                        | 121                                  |

• These values have a high likelihood of protecting intensively-fertilised agricultural systems from accumulating impurities (applied in the Zn fertiliser) to levels likely to impact on food quality, animal health, soil health or contamination of surface and groundwaters.

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# 2. BACKGROUND

Zinc (Zn) fertilizers play an essential role in maintaining and/or increasing world food production and improving human health (Cakmak 2008). Many soils of the world are Zn deficient (Alloway 2004), and crops respond well in these regions to small applications of Zn fertilisers, either alone, or in combination with nitrogen, phosphorus and potassium fertilisers. The main soil types which are Zn deficient and on which the best response to Zn fertilisers have been observed are alkaline soils (soils with pH values above 7.0), and in particular calcareous alkaline soils (soils with high pH and containing limestone).

Some inorganic fertilizers are known to contain contaminant metal, metalloids and radionuclides. Concentrations of contaminants in fertilisers are either derived from the source materials from which fertilisers are manufactured or are introduced into the fertiliser during the manufacturing process. The latter is very uncommon and the dominant source of contaminants in manufactured fertilisers is the raw material used for manufacturing (Stacey et al. 2010).

The International Zinc Association requested CSIRO Land and Water to develop a set of proposed guideline values for selected contaminants in zinc fertilisers – arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb) and mercury (Hg).

The report sets out the methodology used to determine permissible concentrations of As, Cd, Cr, Hg and Pb contaminants in Zn-based fertilizers – the methodology is more fully described in Sorvari et al. (2010).

# 3. SCOPE OF THE PROJECT

## 3.1. Fertilisers and applications

The project considered two different agricultural sectors that represent high-fertilizer use scenarios, so that if protective limits are developed for these, all other agricultural sectors should be protected:

- Horticulture includes vegetables and flowers, and "recreational horticulture" (e.g. turf farms, golf courses and sports grounds); and
- Dairy production.

## 3.2. Hazards to be considered

The specific hazards that were considered are:

- Increased concentrations of contaminants leading to negative effects on soil ecosystems, aquatic ecosystems or plants, in the short or long term;
- Accumulation or concentration of contaminants in the food chain producing unacceptable concentrations from a trade or human health perspective;
- Potential for contaminant mobilisation and off-site migration at concentrations sufficient to cause negative impacts on off-site ecosystems (e.g. surface water, ground water); and
- Toxicity to domestic animals, livestock and wildlife.

The introduction of fertiliser contaminants to agricultural soil can lead to contamination of other environmental compartments (e.g. plants, animals, surface water and groundwater) due to increased release or bioavailability, accumulation and/or on- and off-site transport of contaminants. The primary transport pathways from soil include leaching to groundwater, run-off to surface water, absorption to plant roots and particle transport via air. After distribution from soil to the different environmental compartments, further contaminant transport can occur via secondary pathways i.e. from groundwater and air to a water body, from surface water to sediment, from sediment to surface water, from soil water to plant roots and from irrigation water to plants. After the distribution of contaminants into different environmental compartments has occurred, a number of potential uptake and exposure routes of contaminants to humans, livestock and biota exist. These exposure and uptake routes together with the contaminant transport pathways that are relevant for human and livestock are presented in Figure 1 while those relevant for ecosystems are presented in Figure 2.



**FIGURE 1.** The transport pathways and exposure routes relevant to human and livestock health considered in this project. The potential transport pathways of contaminants from fertiliser amended agricultural soil are indicated with dashed arrows and the resulting exposure and uptake routes with solid arrows.



**FIGURE 2.** The transport pathways and uptake routes relevant to ecosystem health considered in this project. The potential transport pathways of contaminants from fertiliser amended agricultural soil are indicated with dashed arrows and the resulting uptake routes with solid arrows.

# 4. PROJECT METHODOLOGY

## 4.1. Overview

The methodology used to calculate Fertiliser Contaminant Trigger Values (FCTVs) is fully described in Sorvari et al. (2009). FCTVs were derived so that after 100 years of application to agricultural soil the contaminant concentration would not exceed any environmental benchmarks (e.g., guidelines, limits, standards) established for the following environmental compartments - soil, surface water, groundwater, plants, sediments, fish, seafood and livestock. Conservative scenarios were adopted throughout in relation to contaminant accumulation and behaviour in each compartment (Table 1). Within each compartment all relevant benchmarks were collated and the lowest benchmark for each compartment was used to calculate the trigger values.

The recommended FCTVs were calculated using a Probabilistic Hazard Assessment and in order to understand the level of protection they provide it is necessary to understand how they were calculated. The FCTVs were calculated in the following manner. 2500 simulations of the FCTV calculations were conducted for each contaminant. For each contaminant the lowest FCTV calculated in each simulation was determined. A cumulative distribution of the 2500 lowest FCTVs for each contaminant was then generated. The 5%ile of this distribution became the recommended FCTV for a particular contaminant in a Zn fertiliser. Hence, if the FCTVs are adopted then there is at least a 95% probability that none of the environmental compartments that were considered (i.e., soil, livestock, groundwater, surface water, sediment, plants, fish and seafood) and for which FCTVs are available will exceed the corresponding benchmarks. **TABLE 1.** Details of the 'conservative' scenarios adopted to calculate the FCTVs for different environmental compartments.

| Environmental<br>Compartments | "Conservative' scenario  | Notes  |
|-------------------------------|--|--|
| All compartments              | 100 years application to soil at the current maximum recommended application rate.   |  |
| Soil                          | 1) Part of the contaminants <b>are removed</b> by:   | Leaching can be important, depending on the infiltration, soil texture, land cover, and contaminant.   |
|                               | - leaching<br>- surface runoff   | Surface run-off can be important in some areas, but insignificant in flat crop lands due to the minimal slope  |
|                               | <ul> <li>2) There is <u>no</u> removal of contaminants through <ul> <li>volatilisation</li> <li>dusting</li> <li>uptake by plants, humans, animals, human activities</li> <li>further dilution of contaminants</li> <li>transformation and ageing</li> </ul> </li> <li>3) Contaminant distribution between soil solid phase and pore water is governed solely by chemical partitioning which can be determined using the partition coefficient K<sub>d</sub>.</li> </ul> | <ul> <li>Volatilisation is insignificant in case of metals.</li> <li>Dusting - its importance varies depending on land cover (type, fraction), soil disturbance, rainfall, and surface soil texture.</li> <li>Uptake - part of the fertiliser contaminants will be removed from soil particularly by plant uptake. Grazing animals may also ingest some soil containing contaminants. However, part of such contaminants is also expected to return to soil with non-harvested, decaying plant parts and manure.</li> <li>Dilution may take place after fertiliser application due to addition of clean soil.</li> <li>Ageing is important for some metals but was not modelled here.</li> </ul> |
| Ground water                  | All contaminants dissolved in soil<br>pore water enter the groundwater<br>(maximum leaching). There is<br>- no evapotranspiration<br>- no resorption to soil particles or<br>precipitation<br>- dilution between pore water and<br>ground water (assessed using<br>the lowest calculated dilution<br>factor).<br>It is also assumed that<br>contaminants are evenly mixed in<br>the aquifer.   | See leaching above. Adsorption and precipitation of<br>dissolved contaminants to soil particles decreases the<br>amount of contaminants in aqueous phase. Since the<br>processes are seldom fully irreversible (i.e. leaching may<br>take place again if environmental conditions change), the<br>prediction of the importance of re-sorption in the long-term<br>is difficult.  |
| Surface water                 | All contaminants dissolved in soil<br>pore water enter the water phase<br>in surface water body (maximum<br>surface runoff)<br>- see assumptions for ground<br>water   | Water-induced surface runoff can be significant particularly<br>in some dairy agricultural areas (higher slope factor<br>compared with crop cultivation areas)   |

#### Table 1 continued.

| Environmental<br>Compartments | "Conservative' scenario  | Notes  |
|-------------------------------|--|--|
| Sediment                      | All contaminants in soil solid<br>phase enter the sediment (max.<br>erosion). There is no dilution or<br>losses due to e.g. resuspension,<br>volatilisation or dissolving. | In practice, resuspending and dissolving can remove<br>contaminants from sediment. However, in case of stagnant<br>water bodies and constant fertiliser application to soil,<br>saturated concentrations of contaminants in sediment are<br>possible.  |
| Air                           | Volatiles: not considered<br>Dust: A conservative estimate of<br>soil deposited on plant surfaces<br>is considered.  | Only benchmarks for human and livestock consumption<br>consider air as a transport route for contaminants.<br>Therefore, only terrestrial plant PECs will consider air<br>transport of contaminants. No air PECs will be derived.                      |
| Plants                        | Uptake arises from rainsplash,<br>dust deposition and direct<br>contact with soil. No losses from<br>soil (see list above in medium<br>'soil') considered.                 | Uptake of contaminants into above and below ground plant<br>parts varies considerably depending on the plant, climatic<br>conditions and soil properties. Here, conservative models<br>will be used to assess contaminant concentrations in<br>plants. |

#### $K_d$ = the soil-water partition coefficient

Selecting the 5% ile as the 'protection level' is in line with the approach adopted in most developed countries to protect soil and water ecosystems from adverse effects (e.g. Denmark (Petersen & Pedersen 1995), Australia and New Zealand (ANZECC and ARMCANZ, 2000; Heemsbergen et al., 2008, 2009a, 2009b); Hong Kong (Chapman and Warne, 2000), South Africa (Roux et al., 1996), USEPA (1986, 1994), the Netherlands (e.g. Van der Plassche et al., 1993) and organisations such as the OECD (OECD, 1995) and the European Union (Carlon, 2007; EU, 2000).

The values that define the distribution used for the Zn fertiliser application rate are defined in the following. The minimum value was equal to the minimum calculated median of the typical application rates for the two agricultural sectors. The maximum value was equal to the maximum calculated median of the typical application rates while the likeliest value was the median of the medians of the typical application rates (Table 2).

**TABLE 2**. The numerical values that define the triangular distribution used for the application rate of different fertiliser types.

| Fertiliser type | Values that define the triangular distributi<br>(kg Zn/ha/year) |           | ular distribution |
|-----------------|---|-----------|-------------------|
|                 | Minimum   | Likeliest | Maximum           |
| Zn fertilizers  | 0   | 2.5       | 5.0               |

The FCTV values were calculated using the calculations described in Sorvari et al. (2009) and a Monte Carlo re-sampling technique on Crystal Ball<sup>™</sup> software (Oracle, 2008. Version 11.1.1.0.0). FCTV values were derived for each contaminant in Zn

fertilisers 2500 times (i.e. 2500 simulations)<sup>1</sup>. For each simulation a single value from the set of values for each parameter was randomly selected and the calculations conducted. Thus, at the end there were 2500 FCTVs for each contaminant. From this a cumulative distribution of FCTV values was generated for each contaminant.

A similar process to the above is used to derive environmental quality guidelines (EQGs) for water and soil (e.g. the Australian and New Zealand water quality guidelines (ANZECC and ARMCANZ, 2000), European soil guidelines (Carlon et al., 2007) and the proposed methodology for deriving Australian soil quality guidelines (Heemsbergen et al., 2008) and for deriving Australian biosolids guidelines (Heemsbergen et al., 2009)). These guidelines are generated using a species sensitivity distribution (SSD) in which a cumulative distribution of the sensitivity of species to a toxicant is plotted against the toxic concentration. From the cumulative distribution the concentration that should theoretically protect any selected percentage of species can be determined. The usual level of protection that is provided by these guidelines is 95% of species (e.g. ANZECC and ARMCANZ, 2000; Carlon et al., 2007; USEPA, 1986). However, there is no scientific basis for this particular level of protection. Rather it is an arbitrary value which it is felt will protect the vast majority of species and will be sufficient to preserve the form and function of ecosystems. Details of how the SSD methods work and the validity of the assumptions that they make are presented in Warne (1998) and papers cited therein. Given, the predominance of protecting 95% of species in EQGs throughout the world this level of conservatism was also adopted for the FCTVs (i.e., the adopted FCTV was the 5% ile of the distribution of the FCTV values).

## 4.2. Benchmarks to be applied

Various benchmarks have been derived for the protection of humans, livestock and ecosystems from the harmful effects of contaminants in different environmental compartments (e.g. for soil, drinking water, food items, sediment). Examples of benchmarks include the Australian Ecological Investigation Levels for soils (NEPC, 1999), the Australian and New Zealand Water Quality Guidelines (ANZECC and ARMCANZ, 2000) and the Australian maximum levels for contaminants in food (FSANZ, 2008). The benchmarks that were used in this study for the calculation of FCTV values are given in Table 3 and are more fully explained in Sorvari et al. (2009). If there were multiple benchmarks for a contaminant in an environmental compartment, then the lowest benchmark for that contaminant was used to derive the FCTVs.

In the Netherlands, soil benchmarks based on ecological risks and different protection levels have been developed (Verbruggen et al., 2001). In this project three types of soil benchmarks were used: the Dutch soil benchmarks (Verbruggen et al., 2001), the USEPA Ecological Soil Screening Levels (ECO-SSLs) (USEPA, 2008a); and unofficial benchmarks for cadmium derived as part of the Australian National Biosolids Research Program in Australia (Heemsbergen et al., 2009a). For all other metals/metalloids, the lower of the Dutch or USEPA values were adopted, as both methods have some limitations. These particular sets of benchmarks were selected because:

<sup>&</sup>lt;sup>1</sup> In Monte Carlo sampling, the number of simulations needs to be defined. The higher the number of samplings the more accurate are the individual values of the FCTVs in the statistical distribution curve i.e. the percentiles of FCTVs. On the other hand, the higher the number of samplings the longer time it will take to run the simulation. 2500 simulations were used as this typically leads to a 2 % confidence limit in the median value of the parameter being estimated (in this case FCTVs).

- they are available for a large number of contaminants; and
- they consider secondary poisoning i.e. toxic effects in higher trophic levels owing to predation.

**TABLE 3.** Benchmarks (BMs) to be used in the calculation of FCTVs for the protection of (A) human health, (B) ecosystems and (C) livestock.

| Environmental<br>compartment or<br>exposure medium         | Benchmarks for direct human exposure  | Benchmarksforsecondaryhumanexposure   |
|--|---|---|
| Ground water   | Australian Drinking Water Standards   | Australian Water Quality<br>Guidelines for irrigation and<br>general use                          |
| Surface water  |   |   |
|  | none applicable   | Australian Water Quality<br>Guidelines for aquaculture<br>and human consumers of<br>aquatic foods |
| Food items: crops,<br>fruits, vegetables,<br>meat, seafood | Australian: 1) Maximum Limits (MLs)<br>for agricultural and veterinary<br>chemicals; 2) Maximum Residue<br>Limits (MRLs) in food and animal<br>feedstuff. | none applicable   |
|  | Non-Australian: Codex Maximum<br>Levels (MLs) and Guideline Levels<br>(GLs) for contaminants and toxins in<br>food.                                       |   |

(A) Benchmarks used for the protection of human health.

(B) Benchmarks used for the protection of ecosystems.

| Environmental compartment or exposure medium | Benchmarks   |
|--|--|
| Soil   | The lower of the Dutch soil quality benchmarks and USEPA ECOSSLs   |
|  | Unofficial Australian values for Cd that are based on crop uptake under field conditions (McLaughlin et al. 2006).                 |
| Surface water                                | Australian and New Zealand Water Quality Guidelines<br>for marine and fresh water to protect aquatic<br>ecosystems and aquaculture |
| Sediment                                     | Australian Interim Sediment Quality Guidelines to protect aquatic ecosystems   |

**TABLE 3 (CONT).** Benchmarks (BMs) to be used in the calculation of FCTVs for the protection of (A) human health, (B) ecosystems and (C) livestock.

Environmental compartment or<br/>exposure mediumBenchmarksGround waterAustralian and New Zealand Water Quality Guidelines<br/>for livestock drinkingSurface waterAustralian Maximum Residue Limits (MRLs) in animal<br/>feed commodities (only for organic contaminants)

(C) Benchmarks used for the protection of livestock.

Additionally, the selected Dutch and Australian benchmarks for soil have been derived using a method (i.e. a species sensitivity distribution method) that is very similar to that used to derive the Australian and New Zealand water quality guidelines (ANZECC and ARMCANZ, 2000). This method has also been proposed to derive new Ecological Investigation Levels for Australian contaminated sites and soils (Heemsbergen et al., 2009b) and new Australian guidelines for contaminants in biosolids (Heemsbergen et al., 2009a).

### 4.3. Contaminant trigger values for zinc fertilizers

Using the methodology summarised above and detailed in Sorvari et al. (2009), FCTVs for Zn fertilisers were developed (Table 4). Equivalent contaminant concentrations in zinc oxide (ZnO) and zinc sulfate heptahydrate (ZnSO<sub>4</sub>.7H<sub>2</sub>O) are given for comparison.

**TABLE 4.** The recommended Fertiliser Contaminant Trigger Values (FCTVs) for inorganic contaminants in Zn fertilisers.

| Element | FCTV values<br>(mg/kg Zn) | FCTV values<br>(mg/kg ZnO) | FCTV values<br>(mg/kg ZnSO <sub>4</sub> .7H <sub>2</sub> O) |
|---------|---------------------------|----------------------------|---|
| As      | 6000                      | 4820                       | 1365  |
| Cd      | 680                       | 546                        | 155   |
| Cr      | 8100                      | 6508                       | 1843  |
| Hg      | 130                       | 104                        | 30  |
| Pb      | 530                       | 426                        | 121   |

These can be compared to limits for the same contaminants suggested by US EPA (Table 5).

| Element | Maximum<br>allowed<br>concentration<br>per 1% Zn<br>(mg/kg) | Maximum<br>allowed<br>concentration<br>per kg Zn<br>(mg element/kg<br>Zn) | Maximum<br>allowed<br>concentration in<br>ZnO<br>(mg element/kg<br>pure ZnO) | Maximum allowed<br>concentration in<br>ZnSO <sub>4</sub><br>(heptahydrate)<br>(mg element/kg<br>pure ZnSO <sub>4</sub> .7H <sub>2</sub> O) |
|---------|---|---|--|--|
| As      | 0.3   | 30  | 24.1   | 6.8  |
| Cd      | 1.4   | 140   | 112.5  | 31.9   |
| Cr      | 0.6   | 60  | 48.2   | 13.7   |
| Hg      | 0.3   | 30  | 24.1   | 6.8  |
| Pb      | 2.8   | 280   | 225.0  | 63.7   |

**TABLE 5.** Limits on contaminants in Zn fertilizers in USA (US EPA, 2002) and equivalent contaminant concentration per kg pure ZnO and  $ZnSO_4.7H_2O$ .

It can be seen that the proposed contaminant limits are less restrictive than the US EPA limit values. However, the US EPA limit values were not based on risk assessment, but on technological considerations.

# 5. SUMMARY/CONCLUSIONS

A static 100-year simulation of Zn fertiliser use, combined with a range of environmental limit values (for harvested produce, soil, surface and groundwaters), and using a probabilistic approach, produced fertiliser contaminant limit values for As, Cd, Cr, Hg and Pb in Zn-based fertilisers.

This approach was part of a larger risk assessment of contaminants in N, P, K and trace element fertilisers, as well as industrial wastes, conducted by CSIRO Land and Water for the Commonwealth of Australia Fertiliser Working Group, managed under the Department of Agriculture, Food and Fisheries in Australia (Sorvari et al. 2009).

Fertiliser Contaminant Trigger Values (FCTVs) produced are summarised below.

| Element | FCTV values<br>(mg/kg Zn) | FCTV values<br>(mg/kg ZnO) | FCTV values<br>(mg/kg ZnSO <sub>4</sub> .7H <sub>2</sub> O) |
|---------|---------------------------|----------------------------|---|
| As      | 6000                      | 4820                       | 1365  |
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These values have a high likelihood of protecting most intensively-fertilised agricultural systems from accumulating impurities (applied in the products) to leves likely to impact on food quality, animal health, soil health or contamination of surface and groundwaters.

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