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SOIL GUIDELINE VALUE FOR THALLIUM

Moanataiari Subdivision, Thames

REPORT

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1.0 SCOPE

Thallium is a chemical element, symbol Tl, sitting in the fifth row of the Periodic Table between mercury and lead. Highly toxic to humans, it was once used as a rat poison, insecticide, and ringworm treatment, but today it has only a few specialist uses. Tl can be emitted by metal smelters and coal-fired power plants, and is among the suite of elements associated with sulfide minerals.

Thallium has been detected in soils of the Moanataiari subdivision, Thames, Waikato, at levels of up to 29 mg/kg, associated with elevated arsenic and/or lead. This is well above Tl's typical natural abundance of approximately 1 mg/kg (Kabata-Pendias and Pendias 2001). The health risk posed by such concentrations is not clear, as Tl is not on the Ministry for the Environment's (MfE) list of priority contaminants, so there is no New Zealand Soil Contaminant Standard (SCS) for it.

At the Moanataiari Project Management Group (PMG) meeting on 18 July 2012, Thames-Coromandel District Council (TCDC) instructed Golder Associates NZ Ltd. (Golder) to calculate a soil guideline value (SGV[1]) for thallium (meeting minutes refer). This report presents that calculation. It has been carried out in a manner consistent with the existing SCS (refer 'the Methodology', MfE 2011a) incorporated by reference in the relevant National Environmental Standard (NES: 2011).

This report is provided subject to the limitations in Appendix A.

2.0 REFERENCE HEALTH STANDARD

The first requirement of this calculation is to obtain a suitable reference health standard (RHS) protective of human health (refer MfE 2011b).

No RHSs have been set for thallium in New Zealand; there is no drinking water standard and no maximum residue limit for Tl in food.

International jurisdictions for which there are SGVs for thallium include Canada, the Netherlands and the United States of America. Table 1 summarises the RHS underlying these SGVs and the bases of the assessments. These authorities do not consider that there is evidence that Tl is a carcinogen or mutagen, so they take it to be a 'threshold' substance – all RHS 'prescribe a daily level of exposure over a lifetime where there is no appreciable risk to human health' (refer MfE 2011b). This level is often referred to as a tolerable daily intake (TDI) or a reference dose (RfD).

Table 1: Reference health standards for thallium in other jurisdictions.

Country	Reference	RHS type	RHS
Canada	CCME 1999	Provisional TDI	0.07 µg/kg/day
Netherlands	RIVM 1998	Provisional TDI	0.2 µg/kg/day
USA	USEPA 2007	RfD	0.08 µg/kg/day
	USEPA 2009	Candidate TDI	0.01 µg/kg/day
	USEPA 2009	Candidate TDI	0.003 µg/kg/day

These derivations are discussed further below.

[1] There are at least as many terms for the concept 'soil guideline value' as there are jurisdictions. For clarity, this memorandum will keep to the single acronym SGV, regardless of the actual term used by a given source.

There is extensive literature describing thallium poisoning incidents and serious health endpoints. However, there is little information about the long-term effects of exposure to Tl in the environment.

All the authorities cited in Table 1 rely on a single study (variously Stoltz et al. 1986 and Stoltz et al. 1988[2]). Stoltz et al. described a single 90-day gavage study using rats, which were administered 'high', 'medium' or 'low' doses of Tl as the sulphate. They concluded that there were no significant clinical effects on the rats at the high dose level, 0.2 mg/kg/day (expressed as Tl). This dose was therefore considered to be a 'no observable adverse effect level', or NOAEL.

However, a NOAEL for rats may not protect people. The three authorities listed in Table 1 applied an uncertainty factor of 10 for interspecies effects – i.e. people may be more susceptible to Tl poisoning than rats. Another factor of 10 was applied for intraspecies effects – some people may be more susceptible to Tl poisoning than others. And a third factor of 10 was applied because the Stoltz et al. study lasted only 90 days, which may not have been long enough for all clinical effects to manifest themselves. This is a standard approach.

Canada and the USA applied a further factor of 3. They cited a lack of information on Tl's carcinogenicity, reproductive effects, and ability to cross the placental barrier. This caution appears excessive: studies have found that Tl can easily cross the placental barrier, and does have reproductive effects, but these are not the first clinical indications of Tl toxicity (IPCS 1996, USEPA 2009). An uncertainty factor of 1,000 already accounts for these factors, therefore **the Dutch RHS of 0.2 µg/kg/day appears most appropriate for New Zealand purposes.**

A recent review cast doubt on this whole approach (USEPA 2009). The female high-dose rats in Stolz et al. 1986 had exhibited some hair loss, one of the hallmarks of Tl poisoning. Although the control group had also suffered some hair loss – not unusual in rats – this caused the USEPA to abandon the high dose as a NOAEL for rats, and consider instead either the medium dose level of 0.04 mg/kg/day, or a value of 0.01 mg/kg/day estimated from the clinical observations using modern modelling methods. Ultimately, however, the USEPA was not confident in any of these NOAEL, and therefore they withdrew the RHS altogether (USEPA, 2009).

However, a second line of evidence supports the Dutch RHS. A World Health Organisation report (IPCS 1996) considered, on the basis of 1980s German epidemiological studies of people living around the Lengerich cement plant, that Tl exposures leading to urinary concentrations of up to 5 µg/L were unlikely to cause adverse human health effects. The authors acknowledged that there was considerable uncertainty and cited a counter opinion that 'Strong individual variation in sensitivity prevents an estimation of the thallium concentration in the urine at which no effects occur' (Dolgner and Wiegand 1982, cited in IPCS 1996). Nonetheless, the 5 µg/L threshold is supported by recent Chinese observations (Xiao et al 2012).

Converting a urinary threshold to a RHS is straightforward. For example, for bisphenol-A, daily exposure has been estimated based on urinary concentrations by assuming a typical urine volume of 1.6 L/day for US men (Lakind and Naiman 2011). Converted using the same assumption, the WHO indicator level translates to 8 µg/day excreted via the kidneys. This is the principal route for Tl (IPCS 1996, USEPA 2009); the WHO report cites an unnamed study using radioactive Tl as finding that more than 70 % was excreted in this way. Multiplying out by this figure, the daily intake unlikely to cause adverse health effects would be approximately 11 µg/day.

Finally, by taking 70 kg as the standard adult weight (MfE 2011b), a RHS of approximately 0.16 µg/kg/day is obtained. This value is within 20 % of the 0.2 µg/kg/day used in the Netherlands' SGV.

[2] These are two versions of the same internal USEPA report, apparently without any substantive differences (USEPA 2009).

3.0 CONTAMINANT UPTAKE BY VEGETABLES

Many soil contaminants can be taken up by garden vegetables, potentially posing a health risk to people who grow their own vegetables. The ratio of the concentration of Tl in vegetables to the concentration of Tl in soil is called a bioconcentration factor (BCF). BCFs are key parameters in SGV derivation, and the Methodology calls for separate BCFs for root vegetables, tubers and leafy vegetables.

Thallium is easily available to plants. This appears to be because, biogeochemically, Tl can behave very similarly to potassium, a major plant nutrient. Tl uptake depends on Tl concentration, soil pH, the origin of the Tl and the type of plant. Tl may not be evenly distributed around a plant, concentrations may be quite different in roots from shoots, flowers, seeds, bark etc. (IPCS 1996, Kabata-Pendias and Pendias 2001).

Brassica species have an exceptional ability to accumulate thallium, often in greater concentrations than in the surrounding soil (IPCS 1996, Kabata-Pendias and Pendias 2001).[3] Common food brassica include broccoli, Brussels sprouts, cabbage, cauliflower, cress, horseradish, mustard, radish, turnip and watercress. Rapeseed (canola) is also a brassica.

There are no widely recognised BCFs for thallium. However there are studies that present sufficient information to suggest BCF values.

- At Lengerich in Germany, soils were contaminated with Tl due to air discharges from a cement kiln. Tl concentrations in rapeseed plants were sometimes found to be greater than 100 mg/kg (IPCS 1996). In one study, vegetables grown in soil with 4.5 mg/kg Tl were analysed and classified into five groups based on Tl uptake (Scholl and Metzger 1982, cited in IPCS 1996):
  - green cabbage, at 22.6 mg/kg, and Savoy cabbage, at 8.5 mg/kg;
  - turnip, broccoli, kohlrabi and white cabbage at 3.1 mg/kg;
  - stock beet and others (unspecified), 1.4 mg/kg;
  - red beet, rhubarb and spinach, 0.7 mg/kg; and
  - red cabbage, Brussels sprouts, onion, salad, carrot, bean, tomato, cucumber and potato at 0.5 mg/kg.
- At Lanmuchang in China, horticultural soils are contaminated with 40-120 mg/kg Tl from acid mine drainage used for irrigation water. Green cabbages accumulated Tl at concentrations averaging two to eight times higher than in the surrounding soil. Chinese cabbage, chilli, rice and corn contained much less Tl, up to approximately 5 mg/kg. Villagers exhibited symptoms of chronic Tl poisoning and their urine contained high concentrations of Tl (Xiao et al. 2012).

Taking these two studies together, there seems reasonable evidence that green cabbage has a BCF of approximately 5 for thallium, and other brassica also take up considerable amounts of Tl. On this basis, and considering that brassica are common vegetables that evidently tolerate being grown in contaminated soil, this study will use a BCF of 5 for green leafy vegetables in general. For roots and tubers, a BCF of 0.1 will be used. Nonetheless, these values are based on limited studies and considerable uncertainty.

[3] It could follow that these plants would be good choices for phytoremediation of soils (Kabata-Pendias and Pendias 2001).

4.0 DERMAL ABSORPTION

Soluble forms of thallium are readily absorbed through the skin (IPCS 1996). However, Tl in soil is a different matter, because it will largely be bound in or on soil particles. No published estimates of the dermal absorption factor for Tl from soil are available. There also appear to be no published dermal absorption factors for metals such as silver or rubidium that have similar geochemistry. Typically, uptake of inorganic contaminants through skin is considered to be negligible, even for contaminants such as arsenic, chromium and nickel that are known to have effects on skin. For cadmium, MfE (2011b) uses a dermal absorption factor of 0.001, based on USEPA practice. For this report, the same factor will be used for Tl.

5.0 BACKGROUND EXPOSURE

Little is known about background exposure to thallium in New Zealand. It does not appear to have been included in the Total Diet Survey. Thames' drinking water supply has recently been tested for Tl and none was detected (WDHB 2012).

During recent years, Waikato Regional Council has surveyed elemental composition of soils across the Waikato region (Taylor et al 2011). Seventeen samples of unmodified natural soils had a mean of 0.21 mg/kg Tl and a 95th percentile of 0.56 mg/kg. Sixty-seven samples of urban soils had a mean of 0.25 mg/kg and a 95th percentile of 0.61 mg/kg. Forty-six samples of horticultural and arable soils had a mean of 0.52 mg/kg and a 95th percentile of 1.16 mg/kg.

Background exposure information for thallium in other jurisdictions appears limited (see Table 2).

Table 2: Background exposure to thallium.

Country	Reference	Estimated typical thallium intake
[Europe]	RIVM 1998	2 µg/day dietary
United Kingdom	Sherlock and Smart 1986 (cited in ATSDR 1992 and CCME 1999)	5 µg/day dietary – principally in brassica
USA	ATSDR 1992	2 µg/day in drinking water (Dietary intake assumed equal to United Kingdom)

Additionally, IPCS (1996) cite a "carefully controlled" Italian study (Minoia et al. 1990) as determining a background urinary concentration of 0.42 µg/L. Using the same conversion factors as above, 1,600 mL/day urine and 70 % excretion through the kidneys, this would translate to an intake of approximately 1 µg/day.

Worldwide, natural Tl concentrations in soils are generally less than 1 mg/kg (Kabata-Pendias and Pendias 2001).

Based on the absence of Tl from the Thames water supply, background intake in drinking water is assumed to be negligible for the purposes of this report. Background dietary intake is assumed to be 5 µg/day, as in the UK.

6.0 CALCULATING A SOIL GUIDELINE VALUE

6.1 Soil Ingestion

The Methodology (MfE 2011a) uses this equation to estimate pathway-specific SGVs for ingesting soil containing threshold-type substances:

$$SGV_{ing} = \frac{(RHS - BI) \times BW \times 365{,}000{,}000}{IR \times EF} \text{mg/kg}$$

Three of these are generic parameters:

BW = body weight = 13 kg for a child

IR = ingestion rate = 50 mg/day for a child in a standard residential setting

EF = exposure frequency = 350 days/year

The other two are contaminant-specific. For Tl,

RHS = reference health standard = 0.16 µg/kg/day

BI = background intake = 0.07 µg/kg/day

Accordingly the pathway-specific SGV for Tl in a standard residential setting is estimated to be

SGV_{ing} = 35 mg/kg

6.2 Produce Consumption

The Methodology uses this equation to estimate pathway-specific SGVs for home-grown vegetable consumption from soil containing threshold-type substances:

$$SGV_p = \frac{(RHS - BI) \times BW \times 365}{IP \times EF \times P_g \times \sum_{veg}(BCF + SL) \times p} \text{mg/kg}$$

Fruit are assumed to make a negligible concentration, and other produce such as eggs must be considered on a site-specific basis.

RHS, BI, BW and *EF* are the same as in the soil ingestion equation. There are several more generic parameters:

IP = daily vegetable consumption = 10.5 g/day for a child

P_g = proportion of vegetables home-grown = 10 % for a standard residential setting

SL = soil loading on vegetables = 0.001 for roots and tubers, 0.0002 for leafy vegetables

p = proportion of vegetables of each type = 60 % tubers, 10 % roots, and 30 % leafy vegetables

The bioconcentration factors *BCF* are contaminant-specific: for Tl, 0.1 for roots and tubers, 5 for leafy vegetables.

Accordingly the pathway-specific SGV for Tl in a standard residential setting is estimated to be

SGV_p = 1.1 mg/kg

SGV_p is driven almost solely by consumption of Tl-accumulating brassica. If they are not grown, so that BCF_{leafy} becomes 0.1, SGV_p increases to 16 mg/kg.

6.3 Dermal Contact

The Methodology uses this equation to estimate pathway-specific SGVs for dermal contact with soil containing threshold-type substances:

$$SGV_d = \frac{(RHS - BI) \times BW \times 365{,}000{,}000}{AR \times AH \times AF \times EF} \text{ mg/kg}$$

RHS, BI, BW and *EF* are the same as in the soil ingestion equation. There are two more generic parameters:

AR = area of exposed skin = 1900 cm^2 for a child in a standard residential setting

AH = soil adherence factor = 0.04 mg/cm^2/exposure for a child in a standard residential setting; it is assumed that there is a single exposure event per day

The final parameter *AF* is the contaminant-specific dermal absorption factor, for Tl taken to be 0.001

Accordingly the pathway-specific SGV for Tl in a standard residential setting is estimated to be

SGV_d >> 10,000 mg/kg

Dermal exposure is negligible and will not be considered further.

6.4 Dust Inhalation

The Methodology does not calculate pathway-specific SGVs for inhaling soil dust containing threshold-type substances. This pathway is believed to make a negligible contribution to standard residential exposure and will not be considered further.

6.5 Combining the Pathway-Specific Values

The Methodology combines pathway-specific SGVs using this equation:

$$SGV_{all} = \frac{1}{SGV_{ing} + SGV_p + SGV_d}$$

Using the values derived above, the SGV for Tl is 1.0 mg/kg. This value is the same as the SGV derived by Canada based on background concentration in soil and is lower than other regulatory values identified for Tl (Table 3).

7.0 SOIL GUIDELINE VALUES IN OTHER JURISDICTIONS

Three other jurisdictions have developed soil guideline values for thallium. These are shown in Table 3.

Table 3: Soil guideline values for thallium in other jurisdictions

Country	Reference	SGV	For protection of	Derivation of SGV
Canada	CCME 2007	1 mg/kg	All end uses	Background concentration in soil
Netherlands	VROM 2000	1 mg/kg	Target value	Background concentration in soil
		15 mg/kg	Human health and the environment	Integrated value
USA	USEPA 2007	6 mg/kg	Residential	Ingestion + dermal exposure
		91 mg/kg	Outdoor worker	Ingestion + dermal exposure
		160 mg/kg	Indoor worker	Ingestion + dermal exposure
USA – Region 9	USEPA 2004	5.2 mg/kg	Residential	Ingestion + dermal exposure
		67 mg/kg	Outdoor worker	Ingestion + dermal exposure

This SGV of 1 mg/kg is more than the 95th percentile for Waikato urban soils, estimated at 0.6 mg/kg. More relevantly, since consumption of brassica appears to be a critical risk factor and residents' main source of vegetables is likely to be in the wider Waikato region, it is below the 95th percentile for Waikato horticultural and arable soils, estimated at 1.2 mg/kg.

If thallium-accumulating brassica were not grown, the site-specific SGV would increase to 11 mg/kg. And if home-grown produce consumption was eliminated altogether, the soil ingestion pathway would dominate – the SGV would be 35 mg/kg.

8.0 CONCLUSIONS

A soil guideline value of 1.0 mg/kg has been calculated for thallium in the standard residential use scenario based on the generic criteria presented in the Methodology (MfE 2011a).

The risk driver is consumption of home-grown vegetables, particularly cabbage and other brassica species. Brassica are known to be thallium accumulators; the thallium concentration in the plant can be several times that in the surrounding soil. For the wider Waikato region, background thallium concentrations in horticultural soils are typically less than 1.2 mg/kg. These horticultural soils grow the green vegetables that the Moanataiari residents would normally eat. Therefore it would be unreasonable to impose any soil guideline value lower than 1.2 mg/kg on sites in this region, such as Moanataiari. **A site-specific SGV of 1.2 mg/kg is recommended.**

If residential land could be managed to ensure that brassica were not grown in Moanataiari soils, a site-specific soil guideline value of 11 mg/kg would be acceptable. And in circumstances where there was no home-grown vegetable consumption at all (in Moanataiari soils), a value of 35 mg/kg, based on incidental soil ingestion, could be used.

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APPENDIX A

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