

'Organic Materials Guidelines - Contaminant Review'

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Peer reviewed by Jacqui Horswell (and a further review to be decided)



REPORT INFORMATION SHEET

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August 2014

Table of Contents

1.	Executive summary	1
2.	Introduction and context	2
	2.1. Purpose of the revised report2.2. Existing knowledge on trace element contaminants in organic wastes	2 3
3.	Aims and approach	4
	3.1. Justification of N loading rates in different application scenarios3.2. Justification for including TE limits	4 5
4.	Recommendations and conclusions	9
	 4.1. Revised guideline values for TE contaminants in biowaste for soil application 4.2. Recommendations for a revised biowaste application guideline 4.3. Possible wastes that may be included in a new guideline 	9 10 11
5.	Acknowledgements	12
6.	References	12
7.	Appendix	19



1. EXECUTIVE SUMMARY

- This draft report comprises the major findings of a literature review on guideline limits for the concentrations of the chemical elements in biowastes. This preliminary report is for use by Water NZ and not intended to be used as the final document upon which the guidelines are eventually based.
- The land application of biowastes improves physical, chemical and biological soil conditions, but can also result in excessive nitrate leaching and / or TE accumulation in soil and plants. In this respect, the addition of biowastes to soil is similar to the application of fertiliser.
- Currently, biowaste application to land is mainly limited by its N content, to avoid excessive N leaching. However, while N is regulated on the basis of total N, only mineralised N can be taken up by plants. Therefore, the N application via biosolids is often insufficient for adequate plant growth; hence current guidelines are too restrictive for certain application scenarios.
- For the continual application of biosolids, nitrogen concentrations should not exceed current regulatory guidelines of 200 kg N / ha / year, whereas biosolids application to rebuild degraded soils should be limited based on a concentration of 150 kg / ha of mineral N.
- Currently, the trace element concentrations in New Zealand biosolids can be used to categorize the biosolids as grade "A" or "B". The threshold values for trace element (TE) concentrations in grade "A" biosolids currently prevent the beneficial reuse of these materials. In most situations, using biosolids that do not meet grade "A" TE concentrations would not cause soil TE concentrations to exceed internationally-recognised guideline values. Moreover, the threshold concentrations for TEs in NZ's grade "A" biosolids are significantly lower than international values.
- In many cases, the addition of Zn and Cu to soil via biosolids could provide agronomic benefits to many NZ soils that are deficient in these essential micronutrients.
- Biosolids and other biowastes could be safely and beneficially reused if their application does not cause soil TE concentrations to exceed internationally-recognised guideline values. Therefore, the current TE concentration thresholds for biosolids to be categorized as grade "A" could be abandoned, while leaving in place current grade "B" thresholds to prevent dumping of overly-toxic materials.
- Current NZ soil guideline values for biosolids disposal are higher than those used internationally. We suggest that soil guideline values for biowastes are consistent with the NZ national standards currently under development. Assuming these are similar to internationally-recognised guideline values, the eventual NZ standards will not be overly restrictive for the beneficial reuse of biowastes. In contrast, the proposed system will enable the beneficial reuse of more biowastes.
- Example scenarios have been calculated using European soil TE guideline values. Calculations reveal that most NZ soils could be amended with at least 125 tonnes (total over several years) of biowastes at the current grade "B" threshold without causing soils to exceed these guidelines. If NZ adopts higher soil guideline values, then greater amounts of biosolids could safely be added.
- This approach for the beneficial reuse of biowastes is consistent with the approaches used by fertiliser companies, for example the Tiered Fertiliser Management Strategy for cadmium, which is used by the NZ fertiliser industry.

2. INTRODUCTION AND CONTEXT

2.1. Purpose of the revised report

Organic wastes are derived from animal manures, crop residues, food processing wastes, municipal biosolids and wastes from some industries (Westerman & Bicudo, 2005). Increases in population and wealth have resulted in an increased production of biowastes that require sustainable management strategies for their disposal and recycling (Panagos *et al.*, 2013; Río *et al.*, 2011). The application of organic wastes to land can improve the physical, chemical and biological fertility of soils. However it is well known that these wastes can also carry unwanted TEs that may impact soil health and function when present in high concentrations (Bolan *et al.*, 2014).

The current *Guidelines for the Safe Application of Biosolids to Land in New Zealand (2003)* (NZWWA, 2003), state that risk management considerations, permissible loading rates and public health risks apply depending on its land use. A new *Organic Material Guideline* should consider that biowastes can be used either as a continual soil conditioner, or an amendment that is used in a single application, for example to rebuild a degraded soil. The environmental outcomes of each practice are distinct and consequently different limits and guideline values will be given for each practice. The purpose of this report is to:

- a. Summarise existing knowledge on trace element contaminants in organic wastes relative to the current *Guidelines for the Safe and Application of Biosolids to Land in New Zealand* (2003)
- *b.* Review justification for the nitrogen (N) loading rate of no more than 200 kg N per hectare per year (averaged over three years)
- *c.* Review the justification for the inclusion of limits for the following TEs: arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), and zinc (Zn)
- *d.* Determine if other organic wastes contain additional TEs of concern that should be included in a new guideline
- e. Review the allowable concentrations of trace elements in biosolids (Table 4.2 in current *Guidelines for the Safe Application of Biosolids to Land in New Zealand* (2003), and provide recommendations for new limits for waste that will not cause waste-amended soil to approach the soil contaminant limits (using the current soil limits in Table 4.2 noting that these might change under the new Soil Health Indicators Envirolink Project).
- *f.* Provide recommendation for the above with supporting logic
- *g.* Determine possible wastes that may be included in a new guideline include but are not limited to:
 - Household organic (Food waste green waste)
 - Paper and cardboard
 - Primary sector related organic waste (e.g. Agricultural wastes (big in volume but small in practice in some cases), Meat works waste, Manure, chicken manure)
 - Sewage sludge
 - Pulp and paper waste
 - Nappies and sanitary
 - Medical Waste

2.2. Existing knowledge on trace element contaminants in organic wastes

Major sources for most Trace Elements (TEs) to enter the soil are fertiliser, manure and organic amendments. Due to their long residence time in soil, and long-term application the accumulation of TEs in soil are potential risks for quality assurance in the human food chain but may also impact soil health and function and plants growth. High concentrations of TEs can result in reduced plant growth (Zn, Cu, Ni), endanger the human food chain via crop uptake (Cd) and direct ingestion (Hg), or negatively influence animal health (Cu, Pb) and soil microbial processes (Zn) (Smith, 2009). However, various parameters have to be taken into account when assessing TE application via organic waste amendments, since TE bioavailability in soil depends on the chemical association within the soil matrix, soil pH, the concentration of the element in soil and amendment as well as plant uptake (Smith, 2009). Treatment technology and processing of organic wastes have increased during recent years, and land application as a waste management strategy has become increasingly popular (Park *et al.*, 2011). As a result, there is a strong need to monitor the quality and hence the concentrations of different TEs in organic wastes. Examples of TE concentrations in animal manures, biosolids and municipal organic wastes, representing the major sources of TE inputs into soil, are given in Table A1 (Appendix).

2.2.1. Animal manures

The application of animal manures has become a major source of TE inputs into soil, and repeated applications can result in an accumulation, especially of Zn, Cu and Ni (Park *et al.*, 2011). The use of TEs such as Fe, Mn, As, Se and Co as animal dietary supplement has also resulted in an enrichment of these TEs in organic waste streams. However, the ultimate concentration of TEs in manures depend on several chemical, biological and physical production factors, e.g. age and size of animals, manure collection or storage and handling of the manures (Park *et al.*, 2011). In this context, a repeated application of pig slurry has resulted in soil Cu and Zn values of 183 kg and 266 kg per ha, respectively (Martinez & Peu, 2000). Poultry manure amendments in addition may contain high levels of Cd and As, but also B (Table A1, Appendix), which can cause accumulation in the long term (Wuana & Okieimen, 2011) or enter the food chain, when taken up by edible plants (Jinadasa *et al.*, 1997). In New Zealand, Cd accumulation in manure from sheep and cattle has led to restrictions and export barriers for meat to overseas markets (Loganathan *et al.*, 2008).

2.2.2. Biosolids

Urban wastewater treatment plants create large quantities of biosolids. Depending on its origin, biosolids can carry high loads of TEs, including As, Cd, Cr, Co, Hg, Mn, Ni, Pb, Se and Zn (Park et al., 2011). Because of physical-chemical processes involved in wastewater treatment, TEs that are existent in the wastewater accumulate in the generated biosolids (Bai et al., 2012). To reduce the amounts of fertiliser for agriculture, but also to rebuild degraded land, land application of biosolids has become attractive, since biosolids contain high concentrations of nutrients, especially N and P (Bai et al., 2012). The long-term disposal of biosolids to landfill will become prohibitively expensive. In the United States, some 5.6 million dry tonnes of biosolids is produced with around 50% reused for land application and agricultural purposes (Wuana & Okieimen, 2011). Australia produces more than 175 000 dry tonnes of biosolids per year, most are applied to agricultural land and incorporated into the soil (McLaughlin et al., 2000). In China, more than 5 million dry tonnes of biosolids are produced annually, with land application as the most cost-effective option of disposal (Tang & Zhao, 2005). New Zealand produces a yearly amount of around 1.5-2 million wet metric tonnes of biosolids with varying moisture contents, whereas 90% is disposed into landfill, however striving towards a higher percentage of biosolids reuse (Goven & Langer, 2009). Recent studies in New Zealand have shown increased concentrations of Cd, Ni and Zn in drainage water and soils after biosolids application to soil-grassland systems (Keller et al., 2002; Speir et al., 2007) and undisturbed soil (McLaren et al., 2004), illustrating a strong need to monitor TE concentrations in biosolids subject to soil application.

2.2.3. Municipal solid wastes and composts

TEs are present in many household products, including batteries, body-care products, pesticides and medicines, and are present in many plastics, ink cartridges and paints (Bardos, 2004). Consequently, when transferred and composted, municipal solid waste represent a major source for TEs. Zinc, Cu,

Ni, Cd, Pb, Cr and Hg are of major concern (Smith, 2009). However there are differences in TE concentrations between different types of municipal solid wastes, depending on their origin, separation processes and treatments (Amlinger *et al.*, 2004; Bardos, 2004). Mechanically segregated composts in general tend to contain higher concentrations of TEs compared to source segregated ones, nevertheless due to advancing technology, both types can comply with e.g. UK limits (Smith, 2009). Compost from municipal solid wastes has similar properties to biosolids, and is treated and utilised similarly when in terms of land application. With the exception of Pb, in mechanically segregated composts, TE concentrations are likely to be 30 – 50% lower than in biosolids (Smith, 2009), whereas the Pb content can be similar or even up to 80% higher. Recent research however has shown that municipal solid wastes have a high potential for TE sorption, hence limiting their solubility and bioavailability in soil. Lead in this context has been shown being the most strongly bound element whereas Ni the weakest; intermediate sorption characteristics have been shown for Zn, Cu and Cd (Smith, 2009).

3. AIMS AND APPROACH

3.1. Justification for nitrogen (N) loading rates in different application scenarios

Current guidelines recommend that biosolids are applied in accordance with the agronomic N needs of crops and assumes all N will eventually become mineralised and hence have potential for leaching as nitrate (Barbarick *et al.*, 2010; Lagae *et al.*, 2009; Sullivan *et al.*, 2009). Currently there is no regulation of biosolids P application, but because of concerns of effects of repeated applications on soil P and P loss to surface waters, a similar approach has been considered as for fertiliser P and manure P application (Lu *et al.*, 2012).

When using biosolids to rebuild degraded land or low-fertility soil, biosolids are often applied at greater than agronomic rates to build up soil organic matter and improve soil fertility. The required high application rates can exceed guideline values for nutrients, which were set to avoid excessive leaching or run off into lakes and streams causing eutrophication (Tian *et al.*, 2006). The *Organic Materials Guidelines* should take into account these application scenarios with recommendations for the safe application of biosolids. However, the impact and accumulation of TEs after application to either agricultural or degraded land has to be assessed.

3.1.1. Biosolids application in a continual application scenario

Biosolids when applied at agronomic rates can increase grain yields compared to conventional N fertilization (Koenig *et al.*, 2011; Sullivan *et al.*, 2009), but high rates can also lead to yield loss through waterlogging (Mantovi *et al.*, 2005) or moisture stress (Cogger *et al.*, 1998). Within the European Union, specific directives have been established to ensure a correct biowaste application management to avoid excessive run-off and leaching of nutrients into surface water and groundwater (Park *et al.*, 2011). A continual application of biosolids can increase the soil N supply, which requires an adaption of application rates to avoid N loss in the long term (Cogger *et al.*, 2013; Hernández *et al.*, 1991; Moss *et al.*, 2002; Uggetti *et al.*, 2012; Walter *et al.*, 2000). Active soil testing would provide valuable information for a sustainable management of biosolids at sites with repeated applications, whereas determining residual soil nitrate in the fall will help guide N application rates at long-term application sites

In New Zealand, an application rate of 200 kg N per ha per year is recommended for pasture. However, this rate is based on plant available N, and as already stated in the current *Guidelines for the Safe Application of Biosolids to Land in New Zealand* (2003) (NZWWA, 2003), application rates should be based on site-specific assessments including climate, soil characteristics, mineralisation rates and agronomic N requirements of the crops. A recent study suggested that high agronomic biosolids application rates of between 450 and 600 kg N per ha every two years have resulted in more vigorous vegetative plant growth, which probably led to more rapid soil moisture depletion hence depressing grain yields (Cogger *et al.*, 2013). Effects like this should be taken into account when averaging application rates over several years.

However, for a continuous biosolids application to agricultural productive land, a rate of 200 kg N per ha per year still represents an adequate guideline value. According to decreasing mineralization rates after the first year of application, it is unlikely that the total amount of available (mineralized) N each year, including plant available N already present in soil, will add up to cause excessive N leaching.

3.1.2. Biosolids applications to rebuild a degraded soil

Depending on its composition and treatment, biosolids can contain high concentrations of nutrients. A soil application of biosolids can restore organic matter and provide nitrogen, phosphorus, and sulphur (Uggetti *et al.*, 2012). Several studies have demonstrated that biosolids can reclaim degraded lands (Dere *et al.*, 2012; Mbakwe *et al.*, 2013; Meyer *et al.*, 2001; Oladeji *et al.*, 2013; Speir *et al.*, 2003; Stehouwer *et al.*, 2006). Large amounts of organic material added via biosolids have been found to promote topsoil development, increase water holding capacity and stimulate microbial activity and nutrient cycling (Stehouwer *et al.*, 2006). Most N that exists in biosolids is present in organic form, hence unavailable for plant uptake and not subject to leaching (Pu *et al.*, 2008). To minimise N leaching and runoff, N mineralisation rates need to be estimated.

Biosolids application for reclamation purposes often only requires one single application. After biosolids application to soil, between 15 and 55% of the organic N can be mineralised during the first year after application (Adegbidi & Briggs, 2003; Binder *et al.*, 2002; Eldridge *et al.*, 2008; Mendoza *et al.*, 2006; Robinson *et al.*, 2002; Vieira *et al.*, 2005). Degraded lands are characterised by a loss of soil organic matter, soil structure and nutrient availability, hence a biosolids application rate based on 200 kg total N per ha won't provide enough N_{min} to ensure proper plant growth. Conversely, high application rates of between 50 – 100 tons of biosolids (2500 - 5000 kg total N assuming a biosolids N content of 5%) per ha have been proven suitable for re-vegetation and long-term plant growth (Hearing *et al.*, 2000), but excessive N can be lost from these systems via leaching (Dere *et al.*, 2012; Stehouwer *et al.*, 2006)

However, mineralisation rates are highly dependent on the composition of biosolids on the one hand, and the soil properties on the other hand (Pu *et al.*, 2012), making it difficult to determine the appropriate amount of biosolids to be applied unless they are assessed on a case-by-case basis. Generally, in the first year after application mineralization rates are 30% for aerobically digested biosolids, 20% for anaerobically digested biosolids and 10% for composted biosolids, these figures may provide a good starting point for further calculations (Hearing *et al.*, 2000). Since the biosolids total N content varies from 1% to 6% depending on its origin and treatment (NZWWA, 2003), an application of 50 tons of biosolids per ha could result in either 125 kg N_{min} per ha or 750 kg N_{min} per ha, assuming a mineralisation rate of 25% within 12 months after application. Runoff and loss of N via leaching is likely in the latter, but negligible in the first case.

For a more appropriate and safe application of biosolids for reclamation purposes the revised guidelines could allow one-off applications based on a biosolids N_{min} value of 150 kg N_{min} per ha. Further applications can be made with a time-lag of three years and a biosolids application rate of 150 kg N_{min} content measured in soil prior to application. Guidelines should be flexible enough to allow assessments on a case-by-case basis.

3.2. Trace elements in biowastes

The TE concentrations in biowastes have been limited to guarantee its safe application to land. A policy framework for the beneficial reuse of biowastes in New Zealand has been released with the *Guidelines for the Safe Application of Biosolids to Land in New Zealand (2003)* (NZWWA, 2003). However, since there is growing interest in increasing the recycling of organic wastes on the one hand while simultaneously preventing TE accumulation in soil on the other hand, recent literature and revised international standards have to be taken into account to review *Guidelines for the Safe Application of Biosolids to Land in New Zealand* (2003). The actual level of contamination and its risk to the food chain is strongly related to the bioavailability of TEs in soils and plants, which depends on factors like soil pH, plant species and their cultivars, growth stage, biosolids source, soil condition and the chemistry of the element (Warman & Termeer, 2005).

TEs in biosolids of most concern are Cr, Cu and Zn, Pb, Ni, Cd. Boron, Mn, Se, As and Hg can also be present in high concentrations, depending on the origin and treatment of the biosolids (Park *et al.*, 2011; Silveira *et al.*, 2003). TEs can be grouped into essential and nonessential for humans/plants/animals, whereas Cr, Cu, Zn, B, Se and Mn are assigned to the former, and Pb, Ni, Cd, As and Hg to the latter group. Nonessential TEs can cause major health problems, but essential TEs can also be potentially toxic at high concentrations (Nicholson *et al.*, 2003), hence limits in biowastes for soil application purposes are necessary to prevent high amounts of TEs accumulating and entering the food chain.

3.2.1. Organic wastes as a source for essential human trace elements

Zinc (Zn)

Zinc can be present in high concentrations in biosolids and composts (Table A1, Appendix). It is relatively labile and is readily transferred to plant tissues (Smith, 2009). Recent research has shown the use of biosolids application to increase Zn concentration in plants to reduce animal deficiencies (Anderson *et al.*, 2012). At high concentrations however, Zn can also influence soil activity by negatively influencing the activity of microorganisms and earthworms hence slowing down organic matter degradation (Wuana & Okieimen, 2011), and an accumulation in plants has been reported to cause rolling of young leaves, death of leaf tips and chlorosis (Rout & Das, 2009). Generally Zn is important in many biological functions, but recent studies increasingly show free ionic Zn (Zn²⁺) as more biologically toxic than traditionally presumed (Plum *et al.*, 2010). Due to its high mobility, but also its potential to accumulate in soil in the long term, a threshold should be included for a safe biosolids application to land to prevent accumulation in soil and plant damage.

Chromium (Cr)

Chromium was proposed as an essential element in mammals and humans around fifty years ago, with a role in maintaining proper carbohydrate and lipid metabolism (Pechova & Pavlata, 2007). In high concentrations it is associated with allergic dermatitis in humans (Wuana & Okieimen, 2011). In soil and soil amendments, Cr is characterised as an element with extremely low bioavailability (Smith, 2009), hence does not accumulate in above ground plant parts or the food chain. However, it can accumulate in soil where it can be toxic to plants as chromate (Cr⁶⁺), or can be transported into surface waters by runoff, or leach into groundwater (Epstein, 2002); (Wuana & Okieimen, 2011). Therefore, a guideline value is necessary to prevent phytotoxicity and contamination of waterways.

Copper (Cu)

Copper is an essential micronutrient required for growth-related processes in plants, animals and humans. Its availability is limited in biosolids and biosolids amended soil due to the extent of its complexation, particularly with organic matter (Smith, 2009). Most Cu that is introduced is quickly stabilized and does not pose any risk to the environment. It is therefore of less concern regarding land application of organic wastes (Wuana & Okieimen, 2011). Plant uptake is well regulated, hence concentrations in plant tissues and the risk of bioaccumulation in the food chain is usually low (Smith, 2009). Copper in high doses can cause anaemia, liver and kidney damage, and stomach and intestinal irritation (Wuana & Okieimen, 2011). A Cu limit in biosolids for land application is necessary to protect the ecosystem health.

Manganese (Mn)

High amounts of Manganese (Mn) may be added to soil with organic wastes, especially with the application of manure and sewage sludge (Table A1, Appendix). Mn is known as an essential TE, but can become toxic when present in excess (Millaleo *et al.*, 2010). In plants, Mn toxicity may lead to biomass reduction or chlorotic leaves, but due to different tolerance mechanisms, individual plant species may respond with toxicity symptoms earlier than others (Millaleo *et al.*, 2010). In general, Mn toxicity mainly occurs in acid soils and under water logging. Recent studies have shown Mn toxicity to be strongly dependent on soil pH, hence proposed soil guideline limits between 500 and 2500 mg per kg according to the pH (Hernandez-Soriano *et al.*, 2012). The concentration of Mn can be monitored so as not to exceed soil guideline values, rather than implementing Mn thresholds for biowastes.

Boron (B)

Boron is an essential TE required in small concentrations, usually detected in soil from 2 to 200 mg B per kg (Diana, 2006). However, the range between B deficiency and toxicity is comparatively small, and plant toxicity has been shown when soil concentrations increase above 2.5 mg per kg (Goldberg, 1997; Robinson *et al.*, 2007). B is mobile and does not accumulate in soil to any great extent; however, depending on its origin, certain biowastes may contain high B concentrations (Table A1, Appendix; (Park *et al.*, 2011), potentially toxic to plants when applied at high rates. Generally in NZ soil, boron concentrations are known to be more deficient rather than toxic (Sherrell, 1983), this is because boron readily leaches in our high-rainfall climate. We do not recommend that boron concentrations be regulated in biowastes or soil where biowastes may be applied.

Selenium (Se)

Selenium is increasingly becoming an environmental concern, and the gap between deficiency and toxicity levels in animals and humans is narrow. Selenate-Se is soluble and highly mobile, hence liable to leaching and plant uptake (Dhillon & Dhillon, 2003). Organic wastes, especially biosolids can carry higher concentrations of Se (Table A1, Appendix), but absorption and accumulation of biosolids-applied Se by plants strongly depends on soil characteristics as well as the plant species itself (Dhillon & Dhillon, 2003). However, Se in New Zealand should not pose problems because of generally low concentrations in soil (Sheck *et al.*, 2010), hence guideline concentrations do not necessarily have to be implemented in a new organic materials guideline.

3.2.2. Nonessential trace elements applied to soil with biosolids

Lead (Pb)

Lead is s non-essential element for plants and humans, and can be toxic to people if ingested in large amounts. Usually concentrations in biosolids and biosolids amended soil are low. Due to a limited plant uptake the soil-plant barrier means that generally Pb concentrations in plant tissues are low and human food chain impacts are not a concern (Smith, 2009). Lead is mostly applied with biosolids and composts, where it is bound in stable forms that have low availability (Amir *et al.*, 2005; Zheng *et al.*, 2004). Although it is possible that some Pb is taken up by plants if soil concentrations are high, studies have shown Pb does not readily accumulate in the fruiting parts of vegetable and crops (Wuana & Okieimen, 2011). Even long term application with high loads of municipal solid waste – composts have shown only limited mobility of Pb into crops (Gigliotti *et al.*, 1996). In South Australia, lead is proposed to be no longer regulated in biosolids applications because concentrations in biosolids do not represent a risk to human health (ANZBP, 2009). However, the most serious source of exposure to soil Pb is through direct ingestion, hence limits are required to prevent toxic concentrations endangering human health via the consumption of root crops or leafy vegetables, where eating of soil is a concern (Wuana & Okieimen, 2011).

Mercury (Hg)

Soil contamination with Hg has led to environmental concerns. Mercury is a highly toxic and nonessential element that is regarded as an environmental pollutant because of its toxicity, mobility, and long residence time in the atmosphere (Pedron *et al.*, 2013; Wang *et al.*, 2005). It can be readily taken up by plants and accumulate in the food chain, but can also directly affect plant growth, yield production and nutrient uptake if present in toxic concentrations (Patra & Sharma, 2000). Including ceiling concentrations in biosolids for soil application is important to prevent Hg accumulation in soil and hence to prevent increasing concentrations in plant products and potential human health impacts.

Cadmium (Cd)

Together with Hg and Pb, Cd is among the big three toxic elements causing environmental concern, with no known essential biological function (Wuana & Okieimen, 2011). Cadmium has similar chemical properties to Zn, hence able to substitute Cd in certain metabolic processes. Zinc is an

essential micronutrient for plants and animals, therefore its substitution by Cd may cause malfunctioning of metabolic processes (Campbell, 2006). High concentrations in soil and plants pose risks to human health through direct consumption of Cd contaminated food products, but also indirect via e.g. soil ingestion by grazing animals (Park *et al.*, 2011). Agricultural inputs such as fertilisers, pesticides or biosolids can increase the total Cd concentration in soils, hence guideline values in soil as well as limits for Cd in soil amendment are required to prevent its accumulation in the environment (Wuana & Okieimen, 2011).

Arsenic (As)

Arsenic is a potentially toxic element, widely distributed in the environment. Arsenic containing compounds have been extensively used in agriculture and forestry, e.g. to control cattle ticks or pests in banana, or to preserve timbers (copper chrome arsenic, CCA) (Wuana & Okieimen, 2011). In soil, As strongly binds to the clay fraction in the form of arsenate, reducing the mobility through soils as well as plant uptake. In its form of arsenite (formed under anaerobic conditions) it is less adsorbed on soil particles, hence As in the soil solution can be phytotoxic to plants (Epstein, 2002). As phytotoxicity protects As entering into the food chain, but guidelines for soil and soil amendments should prevent an accumulation and hence contamination of soils.

Nickel (Ni)

Nickel is a known potentially toxic in soil and routinely monitored in biosolids subject to land application. However, Ni toxicity is rarely reported in practice, and its hazardous role in the food chain or in terrestrial ecosystems is negligible compared to other TEs (Smith, 2009). Nickel concentrations in biosolids and other biowastes have not been shown to be of environmental significance (Speir *et al.*, 2007) hence guideline values in soil amendments are not necessarily required for a safe application of biowaste to land (Smith, 2009).

3.2.3. Necessity of trace element guideline concentrations in biowastes

A review by Smith (2009) regarding the bioavailability and impact of TEs in biowastes showed various types of biowastes, irrespective of their source, contain higher concentrations of TEs compared to the soil background values. Therefore, long-term application will cause TEs to slowly accumulate in soil until soil limits are approached. Several studies have shown an increase of certain TEs after biosolids application to soil. In general little effect has been shown regarding Ni and Cr; but mainly due to their high availability in biosolids, considerable increases in concentrations of Cd, Cu, Pb and Zn has been detected (Gartler *et al.*, 2013; Illera *et al.*, 2000; Simmler *et al.*, 2013). In a long-term field experiment García-Gil et al. (2000) showed significant accumulation of Zn and Cu in soils following the application of 80 t per ha per year of municipal solid waste compost for five years.

Copper levels have been increased in agricultural soils due to repeated fertilisers use or the application of fungicides to protect vines, citrus trees, and other fruit crops against fungus diseases (He *et al.*, 2005; Schuler *et al.*, 2008). Certain types of biowastes, especially manures, may contain high concentrations of Cu (Table A1, Appendix), hence when applied to soil, may cause exceedance of soil guideline values.

Cadmium is naturally found in phosphate rock (Mar & Okazaki, 2012), hence to date, it has been continuously applied to agricultural soil with phosphate fertilisers. The New Zealand fertiliser industry has implemented a self-governed strategy to address Cd management in soils, progressively decreased to a current maximum of 280 mg Cd per kg P (MAF, 2009). For a continuous and sustainable application of biowastes in replacement of conventional fertiliser, Cd could be regulated in a similar way, which would not result in higher Cd concentrations in soils compared to the continuous use of phosphate fertiliser.

A similar system to the "Tired Fertiliser Management Strategy" (TFMS (MAF, 2009) could be used for biowaste applications, where soil concentrations are continuously monitored and further applications become more restricted with higher soil TE concentrations. Using mass balances and simple models, the impact of certain TE concentrations in biowastes on the total soil concentration can be assessed to avoid soil guideline exceedences with biowaste application (Table 3). The total TE concentration in soil (TE_{soil}) could be calculated by adding the concentration applied with the

biowaste (TE_{biow}) to the concentration already present in soil (TE_{i soil}), but reduced by the amount taken up by plants (TE_{plant}) and lost via leaching (TE_{leach}).

 $TE_{soil} = TE_{i soil} + TE_{biow} - (TE_{plant} + TE_{leach})$

4. RECOMMENDATIONS AND CONCLUSIONS

4.1. Revised guideline values for TE contaminants in biowaste for soil application

Currently, biosolids are used in agriculture, forestry, public recreation and rehabilitation of degraded land. In New Zealand, the *Guidelines for the Safe Application of Biosolids to Land in New Zealand* (2003) (NZWWA, 2003) were established to prevent accumulation of TEs and excess N input into ground and surface waters. However, the availability of TEs in soil is highly variable and strongly depends on soil characteristics. Therefore soil concentrations have to be monitored in combination with concentrations in biowastes to prevent soil contaminant concentration limits being reached. Current international limits of TEs in biowastes and soil are summarized in Table 1 and 2, respectively.

values in mg/kg DV	classification	Cadmium	Copper	Nickel	Lead	Zinc	Mercury	Chromium	Arsenic
		Cd	Cu	Ni	Pb	Zn	Hg	Cr	As
Biosolids (New Zealand)	grade a	1	300	60	300	300	1.0	600	20
	grade b	10	1250	135	300	1500	7.5	1500	30
Biosolids (India)	pH < 7	20	1000	300	750	2500		1000	
	pH > 7	40	1750	400	1200	4000		15000	
Sewage sludges (China)	pH < 6.5	5	800	100	300	2000	5.0	600	75
	pH≥6.5	20	1500	200	1000	3000	15.0	1000	75
Compost (Canada)	class A	3	100		150	500			
	class B	20	757		500	1850			
Biosolids (South Australia)	grade C	20	2500			2500		1 (Cr 6+)	
Biosolids (USA)	grade A	39	1500	420	300	2800	17.0	1200	41
Biosolids (EU)	grade 1	20	1000	300	750	2500	16.0		

Table 1: International guideline concentrations of TEs in biowastes for land application.

Table 2: International soil guidelines for TE contaminants of environmental concern.

values in mg/kg DW	soil properties	Cadmium	Copper	Nickel	Lead	Zinc	Mercury	Chromium	Arsenic
		Cd	Cu	Ni	Pb	Zn	Hg	Cr	As
New Zealand		1.0	100	60	300	300	1.0	600	20
Canada		1.4	63	50	70	200	6.6	64	12
	clay	1.5	60	70	100	200	1.0	100	
Germany	silt	1.0	40	50	70	150	0.5	60	
	sand	0.4	20	15	40	60	0.1	30	
Austria		1.0	100	60	100	300	1.0	100	20
France		2.0	100	50	100	300	1.0	150	
Switzerland		0.8	40	50	50	150	0.5	50	
India			100	<mark>60</mark>	100	300		50	

The revised guidelines have to take into account the use of biowastes for either a continual soil conditioner, or an amendment that is used in a single application, for example to rebuild a degraded soil. The environmental outcomes of each practice are distinct, and consequently different limits and guideline values are recommended for the most beneficial and safe use of biowastes.

Table 1 shows that NZ's current guideline values for biosolids of grade "B" are similar, or even more conservative, to overseas guidelines. Table 1 also shows that NZ's current guideline values for biosolids of grade "A" are, in many cases, an order of magnitude lower than overseas guidelines. The current guidelines for grade "A" biosolids were developed to allow the continual application of

these materials to land without causing excessive accumulation in soils, grain and livestock products (ANZBP, 2009). However, many, if not most, biosolids in NZ do not meet grade "A" criteria with respect to TEs (MfE, 2005). Moreover, many of these biosolids exceed guideline values for Cu and Zn. These elements can have agronomic benefits when added to NZ soil (Gartler *et al.*, 2013; McLaren *et al.*, 1990).

Rather than protecting soils by setting low guidelines for biosolids TE concentrations guidelines, we propound that the amount of biosolids added to soil should be determined on the soil's TE concentration. We suggest that the NZ soil guidelines that are currently under development, be used as threshold values that should not be exceeded by biosolids addition.

Comparing the current soil guideline values for biosolids with internationally-recognised guideline values (Table 2) reveals that for some TEs, the New Zealand values are considerably higher. These international-values have been based on decades of research into the effect of TEs on soil function. Therefore, the new soil TE guidelines that NZ eventually adopts may well be lower than the current biosolids guidelines. Here, we aim to show that even if these soil guidelines are lowered, this will not prohibit biosolids addition to soil with respect to their TE concentration. Table 2 shows that, even using conservative guideline values, consistent with internationally-recognised guideline values, at least 125 tonnes of biosolids at the upper limit for grade "B" could be applied before the guideline values are met for Cr.

Table 3: The minimum amount of biosolids that could be applied, over several years (i.e. the biosolids/ soil ratio is small), without breaching even "precautionary" soil guideline values. These calculations assume that biosolids are mixed into the top 30 cm of soil (over several years), a soil density of 1.3 g/cm³, and negligible leaching or removal in crops.

			Mass of biosolids (to applied before limit r	nnes / ha) that can be eached
Element	Nominal agricultural soil	Precautionary soil guidelines (following EU example)	Biosolids at grade	Biosolids at grade "B" limit
Cd**	0.43	1	2223	222
Cu*	15	60	585	140
Ni*	12	50	2470	1098
Pb*	12	50	494	494
Zn*	68	150	1066	213
Cr*	27	75	312	125
As*	4.1	20	3101	2067
Hg***	0.1	0.5	1560	208

* Reiser et al. (2014). Journal of Environmental Quality

** Taylor M.D., Ltd M.W.-L.R.N.Z. (2007) Soil maps of cadmium in New Zealand Ministry of Agriculture and Forestry. *** Estimated

4.2. Recommendations for a revised biowaste application guideline

This report considers that biowastes can be used either as a continual soil conditioner, or an amendment that is used in a single application, for example to rebuild a degraded soil. The environmental outcomes of each practice are distinct and consequently different values will be given for each practice.

Nitrogen:

a. For the continual application of biosolids, nitrogen concentrations should not exceed current regulatory guidelines of 200 kg / ha / yr or 400 kg / ha every two years, based on evidence that the organic nitrogen present in biosolids is eventually mineralised.

b. When using biosolids to rebuild a degraded soil, the nitrogen limit should be based on mineral nitrogen (e.g. 150 kg / ha), because the organic N given in a single application will not mineralise at a rate sufficient to cause excess nitrate leaching.

Trace Elements:

We propose the abandonment of metal concentrations to categorize biowastes as either "A" and "B". The guidelines for all biowastes will be based around the old "B" limits. This enables the reuse of a much greater number of biowastes in NZ, while preventing the soil disposal of toxic waste. These former "B" guidelines are consistent with overseas guidelines for biowastes. We propose that biosolids can be added *ad lib* with respect to TEs as long as the soil TE concentration stay's within NZ's soil guideline values, which are currently under development. We do not assume to make a suggestion on what these are.

4.3. Possible wastes that may be included in a new guideline

To reduce and avoid prohibitive costs for landfill, an increasing effort is made with recycling waste material. However, treatment technology and procession of organic wastes have increased during recent years, and land application as waste management strategy has become increasingly popular (Park *et al.*, 2011).

The application of animal manures has become a major source of TE input to soil, and repeated applications easily result in an accumulation, especially of Zn, Cu and Ni (Park *et al.*, 2011). The uses of TEs like Fe, Mn, As, Se and Co as animal dietary supplement additionally has resulted in an enrichment of these TEs in organic waste streams (Park *et al.*, 2011).

TEs are present in many household products, including batteries, bodycare products, pesticides and medicines, and are present in many plastics, ink cartridges and paints (Bardos, 2004). Consequently, when transferred and composted, municipal solid waste represents a major source of TEs, Zn, Cu, Ni, Cd, Pb, Cr and Hg are of major concern (Smith, 2009). However there is a huge difference in TE concentrations between different types of municipal solid wastes, depending on its origin, separation processes and treatments (Amlinger *et al.*, 2004; Bardos, 2004).

The majority of medical waste worldwide is incinerated in a high temperature oxidation process, resulting in a significant reduction of waste volume and weight, but producing incinerator ash (Shams *et al.*, 2012). Different types of ash (bottom ash, fly ash) can contain high concentrations of TEs as well as organic compounds. Although bottom ash is generally considered to be safer than fly ash, a recent study showed high concentrations of TEs in bottom ash (Shams *et al.*, 2012). Proper management is required to avoid serious environmental problems (Shams *et al.*, 2012). According to our definition, biowaste should have an organic carbon content of >30%, thus incinerated wastes or other ash would fall outside these guidelines containing <30% organic carbon.

We suggest that pulp and paper be considered under the nitrogen and TE guidelines for other biosolids. We suggest that, given the high C:N ratio, pulp and paper waste may be beneficially blended with other wastes such as biosolids to further improve soil fertility whilst mitigating nitrate leaching.

5. ACKNOWLEDGEMENTS

We acknowledge the financial support provided by the Ministry of Business, Innovation and Employment (MBIE C03X0902), WaterNZ and WasteMINZ.

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valu+D6:R58es in mg/kg DW	Arsenic As	Boron B	Cadmium Cd	Cobalt Co	Chromium Cr	Copper Cu	Mercury Hg	Manganese Mn	Molybdenium Mo	Nickel Ni	Lead Pb	Selenium Se	Zinc Zn
Cattle manure													
Dairy manure					15.0	7		372		0.6	6		67
Dairy liquid and solid manure	1.3	8.1	0.2			139			2.5	0.8	2	3.0	191
Cow dung						200		700					800
Caw manure	6.8		0.7	2.2		18	<0.4	172		9.6	8		
Cattle manure (composted)	5.2		0.4	3.6	14.4		<0.4	357		8.7	5	0.5	164
Feed lot manure						17		149					6480
Dairy cattle FYM	1.6		0.4		5.3	38				3.7	4		153
Dairy cattle slurry	1.4		0.3		5.6	62				5.4	9		209
Beef cattle FYM	0.8		0.1		1.4	16				2.0	2		81
Beef cattle slurry	2.6		0.3		4.7	33				6.4	7		133
Poultry manure													
Poultry dropping						400		1800					2300
Broiler litter	34.6		4.9		6.9	9		501		2.5		1.2	743
Broiler / turkey litter	9.0		0.4		17.7	97				5.4	4		378
Layer manure	0.5		1.1		4.6	65				7.1	8		45
Poultry litter	43.0	51.0	3.0	6.0		748		956	6.0	15.0	11		718
Deep-pit poultry litter		19.0	2.0	8.0	6.0	19		271		14.0	13		252
Poultry manure		390.0	0.5		7.3	54		465	7.7	7.0	2		22
Swine manure													
Swine manure			0.3		33.0	1338		869		12.4	14		<u>14</u>
Swine dung						1000		2100					2900
Cu-enriched swine manure		17.8				1279		197					231
Swine FYM	0.9		0.4		2.0	374				7.5	œ		431
Swine slurry	1.7		0.3		2.8	351				10.4	2		575
Sewara cludra					-								
Sewage sludge (Athens. Greece)	5.0		11.2		75.1	55		1248		53.4	2	1.2	294
Urban compost (unspec.)			0.5		71.0	119		214		15.0			
Sewage sludge (unspec.)			11.4		645.0	870		497		479.0	226		178
Sewage sludge (Denver, USA)	8.1		26.0	7.1	280.0	816	7.8	220	84.9		950	4.6	1672
City sewage sludge (unspec.)	14.3		104.0	9.6	1441.0	1346	8.6	194	14.3		1832	3.1	2132
Sewage sludge (Austin, USA)	9.4		3.3	4.1	106.0	300	1.5	430		36.7	87	2.6	563
Sewage sludge (Milwauke, USA)			7.2	4.1	2940.0		1.1	142		31.2	130	1.0	450
Anaerobic sewage sludge (unspec.)			4.4		5.1	709		129		5.3	67		407
Municipal solid waste													
Fresh municipal solid waste			6.0		16.0	139		816		25.0	216		2677
Degraded waste			3.0		53.0	173		643		21.0	420		1658
Municipal solid waste			0.6		55.0		1.2				330		
Green waste			0.1		20.0	37					87		214
Mixed refuse compost					5.5	274					513		1510

7. APPENDIX