# Quantifying heavy metal inputs from organic and inorganic material additions to agricultural soils in England and Wales

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#### Abstract

Heavy metal inputs to agricultural soils in England and Wales were estimated from all major sources, including atmospheric deposition, biosolids, livestock manures and footbaths, composts, anaerobic digestates, industrial 'wastes' (including paper crumble, food 'wastes', water treatment sludges), ash (from poultry manure incineration) and canal/river dredgings. Across the whole *agricultural land area*, atmospheric deposition was a major source of metals ranging from 8 to 85% of total inputs, with livestock manures and biosolids also important sources as a result of the large quantities of these materials applied. Metal addition rates at the *individual field* level from pig and poultry manures and composts were similar to (and sometimes greater) than those from biosolids. Moreover, metal inputs from 'new' materials which are increasingly being applied to agricultural land (e.g. food wastes) also sometimes exceeded those from biosolids. The study has provided baseline information upon which to develop and focus future policies limiting heavy metal inputs to and accumulation in topsoils.

# **Key Words**

Heavy metals; organic materials; agricultural soils; inventory, soil quality

## Introduction

The soil is a long-term sink for the group of potentially toxic elements often referred to as heavy metals. Leaching losses and plant offtakes are usually relatively small compared with the total quantities entering the soil from different diffuse and agricultural sources. As a consequence, they slowly accumulate in the soil over long periods of time, which can have implications for the quality of agricultural soils. Therefore, reducing heavy metal inputs to soils is a strategic aim of soil protection policies in England and the EU (Defra, 2009; EC, 2002). However, information on the significance and extent of soil contamination from heavy metals in different material additions is required so that appropriate actions can be effectively targeted to reduce inputs to soil.

A previous version of the "Agricultural Soil Heavy Metal Inventory" (Nicholson *et al.*, 2003) identified atmospheric deposition as the principal source of most metals entering soils at a national level, with biosolids (treated sewage sludge) and livestock manures also significant sources, albeit to limited land areas. Subsequently, a number of important changes have taken place which are likely to have affected heavy metal inputs to agricultural soils in England and Wales. These include reductions in the maximum permitted levels of trace elements in livestock feeds, decreasing metal atmospheric deposition rates, the increased recycling of organic materials such as compost to agricultural land and the application of 'new' materials (e.g. anaerobic digestate, paper crumble etc.) which are likely to be of increasing importance in the future. In this paper, we discuss the effects of these changes on the relative importance of the different sources of metals at a *national* and *individual field level*, and the implications for agricultural soil quality.

## Methods

This paper presents a quantitative inventory of heavy metal inputs (zinc - Zn, copper - Cu, nickel -Ni, lead - Pb, cadmium - Cd, chromium - Cr, arsenic - As and mercury - Hg) to agricultural soils in England and Wales. The major sources included were atmospheric deposition, biosolids (i.e. treated sewage sludge), livestock manures and footbaths, composts (green and green/food), anaerobic digestates, industrial 'wastes' (including paper crumble, food 'wastes', water treatment sludges), ash (from poultry manure incineration), canal/river dredgings, inorganic fertilisers and lime, agrochemicals, metal corrosion, lead shot (from recreational shooting) and irrigation water.

The inventory was based on recently published data on the heavy metal contents of the above materials and estimated quantities applied to agricultural land, including data from a survey of metal concentrations in c.190 livestock manures sampled between 2007 and 2009. The relative importance of the different sources of metals at a *national* level was estimated from the total quantities of materials applied to agricultural land and their typical metal concentrations. Relative importance at the *individual field* was based on a typical rate of material addition per hectare of farmland, which for most organic materials was equivalent to c.250 kg N/ha

(Defra/EA 2008). We also assessed the implications of these input rates in terms of the time required to reach soil metal limit concentrations where biosolids are recycled to agricultural land (DoE 1996).

#### Results

#### Inventory of heavy metal inputs

Estimated total annual heavy metal inputs to agricultural land in England and Wales (Table 1) were lower than those previously reported (Nicholson *et al.*, 2003) mainly due to lower estimated inputs from atmospheric deposition. Inputs from atmospheric deposition, which were previously based solely on a national monitoring network (Alloway *et al.*, 2000), were adjusted to correct for a potential over-estimate of the contribution of dry deposition, as well as allowing for reductions in deposition since the original measurements were taken (1995-98). Nevertheless, atmospheric deposition still accounted for *c*.20-30% of the total annual inputs to agricultural land for Zn, Cu, Ni, Cd, Pb and As, and *c*.80% for Hg. Livestock manures were estimated to account for *c*.30% of Zn, Cu and As inputs, but were a less important source of the other metals. Biosolids contributed between 10% (Cd) and 40% (Cr) of total metal inputs, and composts less than 5% of total inputs (except for Pb). Industrial 'wastes' (including paper crumble, food 'wastes', water treatment sludges and canal/river dredgings) were an important source of Ni (22%), Pb and Cd (*c*.10%), whilst inorganic fertilisers (mainly phosphate fertilisers) and lime contributed *c*.30% of Cd and Cr inputs.

Table 1. Estimates of total annual metal inputs (t/yr) to agricultural soils in England and Wales from diffe	erent
sources.	

Source	Zn	Cu	Ni	Pb	Cd	Cr	As	Hg
Atmospheric deposition	1010	330	70	100	6	20	8	9
Livestock manures	1070	380	20	40	2	30	10	<1
Biosolids	700	360	40	170	2	100	7	2
Industrial wastes <sup>1</sup>	180	80	50	50	2	20	2	<1
Compost <sup>2</sup>	120	60	10	60	<1	10	<1	<1
Anaerobic digestate	<10	<10	<1	<1	<1	<1	nd	nd
Footbaths <sup>3</sup>	30	10	nd	nd	nd	nd	nd	nd
Fertilisers and lime	150	60	30	10	7	70	5	<1
$Ash^4$	150	40	nd	nd	nd	nd	nd	nd
Plant protection products	20	<10	nd	nd	nd	nd	nd	nd
Irrigation water	<10	<1	<1	<1	<1	<1	<1	<1
Corrosion	30	nd	nd	nd	nd	nd	nd	nd
Lead shot				$[3250]^5$				
Total	3450	1330	220	430	19	255	31	11

nd: no data

<sup>1</sup>Including paper crumble, food 'wastes', water treatment sludges, canal/river dredgings

<sup>2</sup>Including green compost and green/food compost

<sup>3</sup>Only includes the proportion of footbaths disposed directly to land. Metals in footbaths disposed to slurry/manure stores are assumed to be included in the contribution from livestock manures

<sup>4</sup>Ash from the incineration of poultry litter

<sup>5</sup>Not included in total due to uncertainty of estimate

Although atmospheric deposition was an important source of heavy metal inputs to agricultural land on a national scale, metal addition rates at an *individual field* level were small compared with other sources. For some metals (Zn, Cu, Cr and As), the highest metal addition rates at a field level were from biosolids (applied at 6.5 t dry solids/ha/yr, *c*.250 kg total N/ha/yr), although Zn and Cu addition rates from some pig and poultry manures (applied at 250 kg total N/ha/yr) were of a similar magnitude (Figure 1). Metal addition rates from composts (applied at 20-30 t fresh weight/ha, *c*.250 kg total N/ha/yr) were similar to or greater than those from biosolids. 'Wastes' from food production (e.g. dairies, soft drinks manufacture etc.) are generally low dry matter liquids and as a results are commonly applied at high rates to agricultural land (*c*.180 m<sup>3</sup>/ha). Consequently, Ni and Cd addition rates with these materials were estimated to be higher than from biosolids (Figure 1). Similarly, water treatment sludges were also estimated to add high rates of metals, in particular Ni, to agricultural soils. Anaerobic digestate (applied at 30 t fresh weight/ha or *c*.250 kg total N/ha/yr) did not add significant quantities of metals at the field level compared to many of the other sources considered. Notably, metal addition rates from all the sources discussed here were below the current maxima for biosolids application to agricultural soils (DoE, 1996).

#### Implications for soil quality

The above field level heavy metal addition rates were used to estimate the time (number of years) required to raise topsoil metal concentrations from background values (i.e. mean concentrations in England and Wales; McGrath and Loveland, 1992) to the maximum permissible concentrations for heavy metals stipulated in the Code of Practice for Agricultural Use of Sewage Sludge(DoE, 1996), assuming that all fields received inputs from atmospheric deposition and had the same rate of metal loss via crop offtake and leaching. The limiting metal for most organic and inorganic material additions was Zn, with the limit value of 200 mg/kg soil reached after approximately 100 years of biosolids, pig slurry and compost additions. Clearly, these times would decrease if soil Zn concentrations were already above background values, if more than one material was applied in a year, or if application rates or input material Zn concentrations were higher than those assumed here. In comparison, it was estimated to take >1,000 years for cattle farmyard manure (FYM) applications to raise topsoil Zn concentrations to the limit value. For food 'wastes', the limiting metal was generally Cd, where the maximum permitted concentration of 3 mg/kg soil was estimated to be reached after *c*.250 years.

#### Conclusion

In response to concerns over the impact of heavy metal inputs on long-term soil fertility and the potential transfer of certain metals to human diets, an updated inventory of heavy metal inputs to agricultural soils was compiled for England and Wales. Whilst atmospheric deposition was a major source of total heavy metals inputs to agricultural land at the national level, livestock manures and biosolids were also important sources because of the large quantities of materials applied. The results also showed that metal addition rates at the field level from some pig and poultry manures and composts were similar to (and sometimes greater than) those from biosolids. Moreover, metal inputs from 'new' materials which are increasingly being applied to agricultural land (e.g. food 'wastes') also sometimes exceeded those from biosolids. This study has provided baseline information for the development of strategies to reduce heavy metal inputs to agricultural land and to effectively target policies for minimising long-term accumulation in soils.

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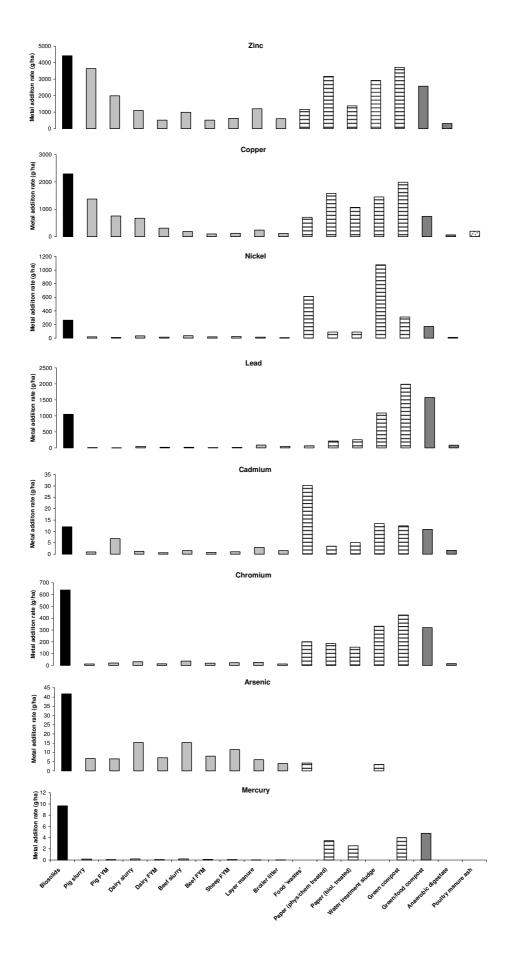


Figure 1. Estimates of metal addition rates (g/ha/yr) from selected organic and inorganic materials applied to agricultural land in England and Wales at typical rates.