A CONTEMPORARY AND HISTORICAL ANALYSIS OF THE TRACE ELEMENT COMPOSITION OF SEWAGE SLUDGE IN THE UK

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Abstract

Sewage sludge is the essential by-product of wastewater treatment and approaching 100% of the sewage sludge generated in the UK is recycled to agricultural land. In collaboration with three major Water Utility companies in the UK, a critical statistical analysis of historical and contemporary sludge quality data was completed to demonstrate the long-term patterns and improvements in the trace element (TE) content of sludge recycled to agriculture since 1989, and to indicate the current status of sludge quality in the UK. Comparisons with pollutant emission inventory data showed most of the TE concentrations in sludge were strongly linked to declining environmental emissions. A soil accumulation model showed that zinc and copper would be the first to approach their statutory soil limits in the long-term and that nickel, cadmium, lead and mercury are no longer significant and, from a practical perspective, could be removed from the regulatory controls on agricultural use.

Keywords

Agriculture biosolids; land spreading; sewage sludge; sludge quality; soil; trace elements

Introduction

Municipal and industrial waste materials and sludges have significant agronomic and fertiliser replacement value and recycling to land enables value recovery, contributing to a sustainable circular economy for nutrients, and reducing disposal by other routes with high environmental impact, such as landfill. However, metallic trace elements (TEs) slowly accumulate in the soil following the long-term application of sewage sludge to agricultural land (Fijalkowski *et al.,* 2017). Most of the metals present in sewage sludge are considered to be harmful if accumulative concentrations in soil exceed safe thresholds, including the microelements essential to living organisms, such as copper (Cu) and zinc (Zn), as well as those elements that represent a potential risk to the human food chain, including cadmium (Cd), lead (Pb), arsenic (As) and mercury (Hg) (Milik *et al.,* 2017). Therefore, it is important to ensure the chemical and microbiological quality of recycled materials to protect human health and the environment. Recycling of municipal/industrial materials in agriculture is therefore controlled and regulated to ensure the practice is consistent with good agricultural and environmental stewardship.

The wastewater sector in the UK has a long history of recycling treated sewage sludge (biosolids) to agriculture, successfully increasing the proportion recycled to land from 44% in 1992 to 80% in 2010 (Ofwat, 2016), and it is currently close to 100% as sludge incineration has almost been phased out. Recycling to agricultural land is recognized as the Best Practicable Environmental Option for sludge management (Kroiss and Zessner, 2007). In Europe, recycling sludge to agriculture is a highly controlled activity regulated by EU Directive 86/278/EEC (CEC, 1986) and, in the UK, the Directive was implemented in 1989 through The Sludge (Use in Agriculture) Regulations (SI, 1989 amended in 1990). Sludge management is supported by a Code of Practice for the Agricultural Use of Sewage Sludge (DEFRA, 2018), which provides further control measures to protect the environment and human health. The Safe Sludge Matrix (ADAS, 2001) also prescribes operational and numerical microbiological parameters, as well as land-use restrictions, depending on the treatment to conventional or advanced status. The application of sewage sludge in agriculture is beneficial to soil and plants, it supplies nitrogen (N), phosphorus (P) and other nutrients for plant growth, and organic matter to improve soil physical conditions.

The chemical composition of, and presence of microbial indicator organisms in, sewage sludge is routinely measured by the UK Water Utilities and the UK Government is required to report the content of specific trace elements specified in the Sludge Directive (CEC, 1986) to the European Commission. Previous surveys of UK sewage sludge quality were reported by Sleeman (1984), CES (1993) and Gendebien *et al*. (1999). These demonstrated significant

reductions in TE concentrations in sludge due to industrial discharge controls and improved industrial practices (Gendebien *et al*., 1999). However, no information on the TE content in UK sludge has been released into the public domain since 1999. Therefore, changes and improvements in the quality of sludge which have taken place in recent years, that underpin the practice of beneficial land spreading, are not generally known.

The aim of this research, therefore, is to provide information on the chemical quality of sewage sludge used for agricultural application in the UK, to examine the relationships between industrial emissions of TEs on sludge composition, and to assess how this may affect the long-term patterns in TE concentrations in sludge-amended agricultural soil in future.

Methodology

Data information

Sludge quality data were provided by three major Water Utility companies from 75 wastewater treatment plants (WWTPs) in the UK, for the period 1989 to 2017. The data collected from Company 1 included results for 5658 individual sludge samples from 50 WWTPs for the period 2000 to 2017. Company 1 also collected composition data in relation to sludge type (raw (untreated) cake, digested cake and limed-treated cake). There were 4416 recorded samples in the dataset from Company 2, representing 13 WWTPs for the period 1989 to 2017. Records for Company 3, from 1998 to 2017, included 1486 samples from 12 WWTPs. Trace element concentration data were available for regulated elements (SI, 1989): Zn, Cu, nickel (Ni), Pb, Cd and Hg, and advisory elements (DEFRA 2018): chromium (Cr), molybdenum (Mo), As, selenium (Se) and fluorine (F). All concentrations are total values and were reported as mg/kg dry solid (DS).

Statistical procedures and data analysis

The IBM SPSS Statistics 25 and Excel computer programmes were used for statistical analysis calculations. Sludge quality data were initially combined and consolidated into a consistent format. A screening process was applied to the combined dataset to remove extreme outliers larger than 3 times the interquartile range, using the boxplot method (Frigge *et al.,* 1989). The numerical mean, annual concentration for each TE was calculated, and analysis of variance (ANOVA) and post hoc tests (Pallant, 2010) were applied to determine the statistical significance of differences in TE concentrations between years and sludge treatment type (for this subset of sludge data from Company 1). Percentile diagrams were constructed with a 5-year time step to illustrate the changes and distributions of TE concentrations since the previous survey of sludge quality by Gendebien *et al*. (1999).

The UK National Atmospheric Emissions Inventory (NAEI) provides comprehensive reporting on the emission of pollutants to the atmosphere from different sources (https://naei.beis.gov.uk/). We used this information to examine the statistical relationships between the emissions and concentrations of the reported elements measured in sludge for the period 1989 – 2017, following standard, linear regression analysis procedures. Whilst this data does not account for direct discharges to wastewater and, therefore, to sludge, it will indicate the extent of diffuse inputs from pollutant deposition onto paved surfaces and run-off into the combined sewer system. Thus, it can provide a representative assessment of the overall extent of environmental emissions from different sources potentially influencing the TE concentrations in sludge. Total emission data was not available for Mo.

A soil accumulation model was developed to calculate the most limiting elements to sludge recycling on agricultural land and the time period (years) required by annual applications to raise the soil concentration from the median background to the soil limit value for each of the regulated elements (Zn, Cu, Ni, Pb, Cd, Hg) (SI, 1989 amended in 1990) and Cr (DEFRA, 2018). The quantity of sludge applied to land was calculated based on a maximum recommended N application rate of 250 kg/ha/year (DEFRA, 2009) and the mean total N concentration measured in 2017 (45700 mg/kg DS) was used as the representative content in sludge applied to land. As TEs are retained in the cultivated layers, sludge was assumed to be applied onto a hectare of agricultural land with 23cm cultivation depth, at a frequency of once per year. It was further assumed that the soil density was equivalent to 1.3g/cm^3 and there was no sludge degradation factor. The calculations are listed as follows:

Annual quantity of sludge applied = 250/0.0457 = 5470 kg/ha/year (1)

Quantity of cultivated soil = 10000*0.23*1300=2990000 kg/ha/year (2)

The soil median total TE concentration data were from Rawlins *et al.* (2012) for all the elements, except Hg, which was from McGrath and Loveland (1992), and mean and 95th percentile TE concentrations in sludge were used in the soil accumulation calculations from the combined dataset in 2017 (Supporting Information). Mean concentrations indicated representative TE values and 95th percentile values were also used to determine the effects of upper range concentrations in sludge on the changes in soil conditions. The blended TE concentration of sludge with soil after each annual application was calculated as:

$$\text{Blending concentration} = \frac{(\text{mass of sludge} \times \text{sludge concentration}) + (\text{mass of soil} \times \text{soil concentration})}{(\text{mass of sludge} + \text{mass of soil})} \quad (3)$$

The same procedure was applied in each subsequent year to calculate the soil concentration after the annual addition of sludge and to estimate the number of years necessary to reach the soil limit values.

Results and discussion

Patterns in TE concentrations in sludge with time (year)

The annual mean concentrations of TEs measured in sludge for 1989 to 2017 are presented in Figure 1. Percentile values (Figure 2) were generated for the combined dataset for the elements: Zn, Cu, Ni, Pb, Cd, Cr and Hg for the years: 2000, 2005, 2010, 2014 and 2017 to illustrate the changes and patterns in TE concentrations since the previous survey of sludge quality (Gendebien *et al*., 1999). However, it is important to draw attention to the very large reductions in concentrations of all the elements that occurred in the 10 year period prior to 1999, as noted by Gendebien *et al*. (1999). The numerical mean, median, 5th and 95th percentile TE concentration data for each year are summarised in the Supporting Information.

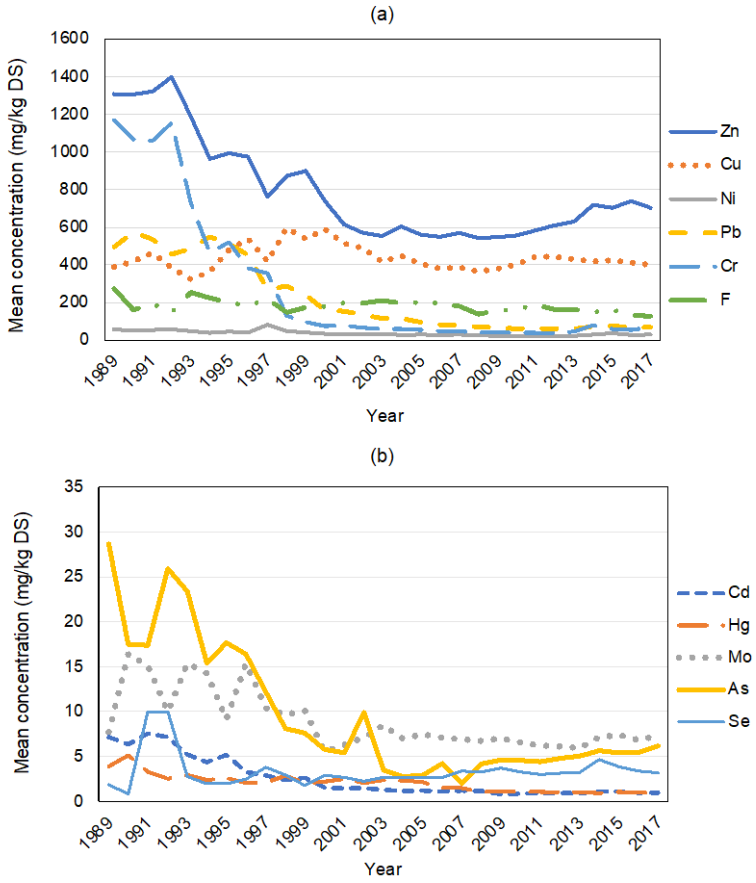

Figure 1 Mean annual total concentrations of (a) Zn, Cu, Ni, Pb, Cr and F, and (b) Cd, Hg, Mo, As and Se in sewage sludge for the period 1989 - 2017

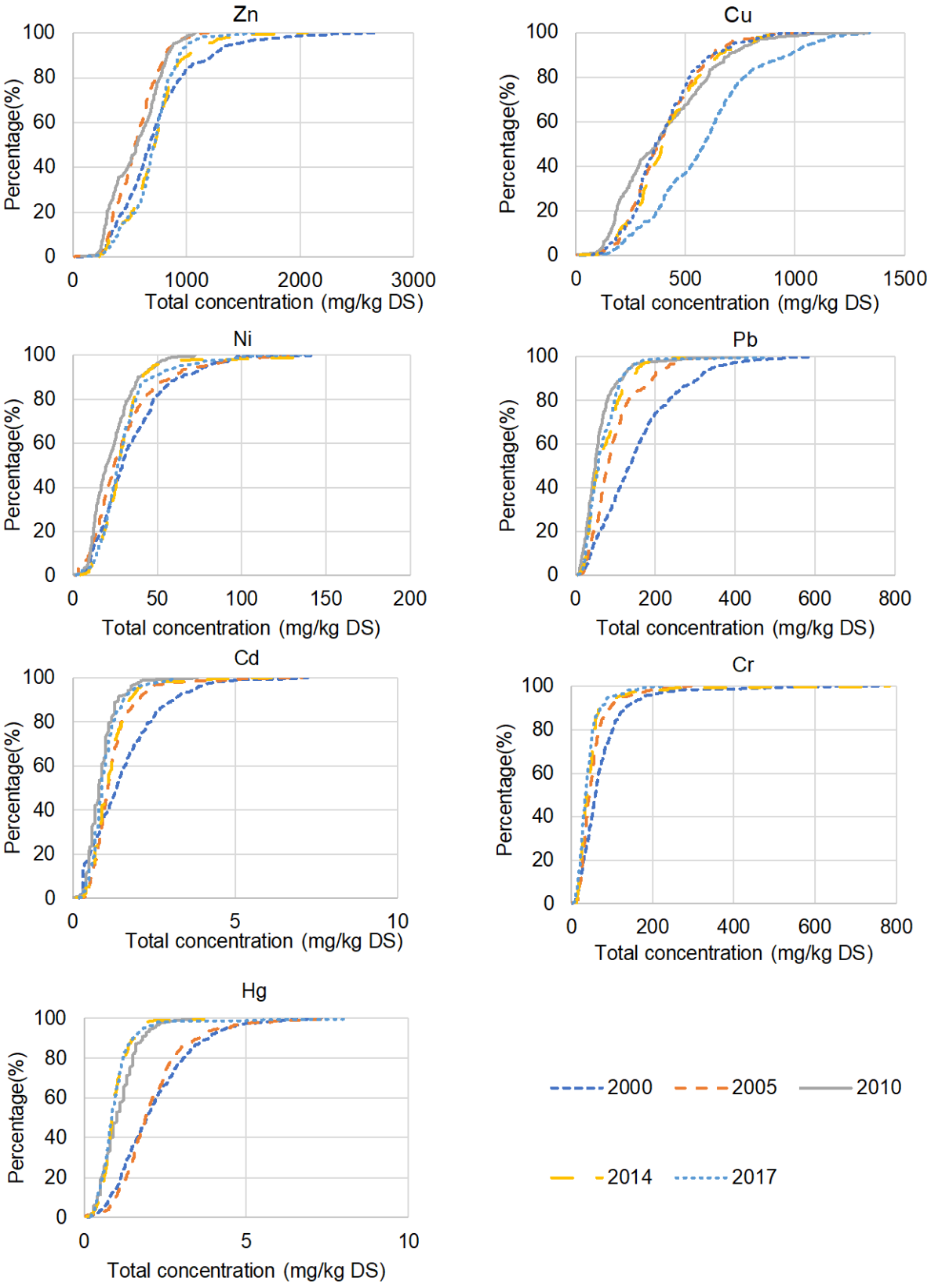

Figure 2 Percentile diagrams of the total concentrations of Zn, Cu, Ni, Pb, Cd, Cr and Hg in sludge in 2000, 2005, 2010, 2014 and 2017

For example, in the period 1990-2000, the average Pb content in sludge declined by over 70% from approximately 500 to 150 mg/kg DS (Figure 1). Since 2000, the mean Pb content in sludge has continued to decrease further, by more than 50%, and the mean value in 2017 was 68 mg Pb/kg DS (Supporting Information). The largest overall reduction in concentration was observed for Cr, which fell from a maximum mean value of over 1000 mg/kg DS in the early 1990s to a minimum mean concentration equivalent to 38 mg/kg DS in 2011, representing a reduction of more than 96%. Significant reductions in annual mean concentrations of Cd, Hg and F were also observed for the period 2000 to 2017, with reductions of 35, 55 and 30 % from 1.6 to 1.0, 2.2 to 1.0 and 180 to 130 mg/kg DS, respectively.

Interestingly, the mean Cu concentration has remained relatively static and contemporary concentrations are similar to those observed in the early 1990s, equivalent to approximately 400 mg/kg DS (Figure 2). An apparent rise in sludge Cu from 1995 to 2001 (Figure 1), was not observed for any of the other elements and is difficult to reconcile. However, the concentration patterns of specific elements reflect the opposing effects of the downward movement due to reduced emissions, and an upward trend due to increased solids destruction, for instance, by more intensive sludge treatment, such as anaerobic digestion. Thus, a stable concentration observed for several elements reflects a balance of these mechanisms and that, overall, emissions are probably continuing to decline. Zinc, on the other hand, showed a significant drop in mean concentration from approximately 1300 mg/kg DS in 1989 to 540 mg/kg DS in 2008, equivalent to a decrease of 60%, clearly indicating a major reduction in emissions of this element. Subsequently, however, concentrations showed a small rise to approximately 700 mg/kg DS from 2014. A similar pattern was also shown in As concentrations during this period, although the amount of As in sludge is small and typically approximately 5 mg/kg DS on average, representing 10% of the soil limit value (for pH >5.0, 50 mg/kg dry soil (ds)). Indeed, the 10th percentile total As concentration in soil (Rawlins *et al*., 2012) and the 95th percentile DS value in sludge (see Supporting Information) are almost identical at 8.6 mg/kg ds and 8.7 mg/kg DS, respectively, thus sludge is not a significant contributor to the As status of soil. By contrast, Ni, Mo and Se concentrations in sludge have remained relatively consistent over the past 20 years with mean values typically in the ranges: 25 – 30, 6.0 – 7.0 and 3.0 – 4.0 mg/kg DS, respectively. Within this group, the mean values of Mo and Se measured in sludge were moderately or slightly higher than their soil limits (for pH >5.0, Mo: 4.0 mg/kg ds and Se: 3.0 mg/kg ds).

It is interesting to note that the 95th percentile concentrations of Ni, Pb, Cd, Cr, As and F (63, 139, 1.9, 129, 8.7 and 274 mg/kg DS, respectively, see Supporting Information) in contemporary sludge samples are all significantly below their respective soil limits (in mg/kg ds; Ni at pH 6.0 - 7.0, 75; for pH >5.0: Pb, 300; Cd, 3.0; Cr, 400; As, 50 and F, 500).

The TE distribution patterns for the past approximately 20 years (Figure 2) show a declining trend in both the upper range and median concentrations measured in sludge. Trace element concentrations in the higher ranges have decreased to the greatest extent, whereas those in the low range have shown relatively little movement, presumably because these concentrations already reflect background values. However, as may be expected, the rate of reduction has diminished as concentrations approach the nominal background status of wastewater catchments. Thus, ANOVA and post-hoc tests for each element, comparing annual mean concentrations with the data for 2017, showed there was a statistically significant reduction initially, but there was no significant difference ($P > 0.05$) detected between the mean concentrations measured after 2006 – 2008 compared to 2017.

Relation between environmental emissions and TE concentrations in sludge

Industrial emissions of potentially toxic elements (PTEs), have been significantly reduced by the Integrated Pollution Prevention and Control (IPPC) Directive (CEU,1996), and other source control measures, including the Dangerous Substances Directives (CEC, 1976), and its many amendments (which can be viewed at ECOLEX (2020) and OOPEC (2004)), which was repealed by the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) Regulations (EPCEU, 2006), and also the Water Framework Directive (EPCEU, 2000; Ostoich, *et al.,* 2009). All these measures have contributed to a reduction in environmental emissions of pollutants and, consequently, have had a profound impact on reducing TE concentrations in sludge.

Highly statistically significant correlations ($P < 0.05$) were detected between annual mean TE concentrations in sludge and the total emissions reported in the NAEI (2019) database (Figure 3). Very strong relationships were found for Cd, Pb and Cr, with R^2 values in each case ≥95%. Arsenic, Zn and Hg in sludge were also strongly related to the emissions, recording R^2 values >70%, and Ni and F were also statistically significant.

A good example of the direct effects and benefits of pollution control measures on the TE content in sludge is shown by the withdrawal of Pb based anti-knocking additives to petrol, which were a major source of Pb emissions to the environment and contributed 74% of UK Pb emissions in 1990. Leaded petrol was withdrawn from normal sale in the UK fuel market by the end of 1999 and this action reduced the total amount of Pb emitted from petrol to just 1% of UK emissions from 2000 onwards (NAEI, 2019). Overall atmospheric emissions have declined by 97% since 1990 and these factors are a major reason why Pb concentrations in sludge have fallen to such an extent (Figure 1), following a logarithmic pattern (Figure 3). Thus, the reduction in Pb concentrations in sludge has increased as emissions to the environment have declined.

The relationship between total Cr concentrations in sludge and UK emissions was also described by an exponential function (R^2 = 0.95, P < 0.001), however, in contrast to Pb, the largest reductions in sludge Cr occurred as maximum emissions of this element declined (Figure 3). This may be explained because Cr originated from multiple sources that were gradually phased out over time, compared to Pb, which was controlled to a large extent over a relatively short period following the ban on unleaded petrol. The main sources of Cr before 2000 were the chemical industry (eg, for use in pigments, wood preservatives, leather tanning) and metal production industry, but these industries and the use of Cr in manufacturing processes and marketing of Cr containing products have declined gradually over time with the introduction of effective source controls.

Zinc represents the largest elemental concentration in sludge, and mainly enters the wastewater system in run-off from gutters, roofing and galvanised surfaces, and from the use of detergents and washing powders, and body care products containing Zn (Dokulilová *et al.,* 2018). The decline in non-ferrous metal manufacture, and particularly Zn smelting, which ended in the UK in 2003, and improved emission standards for municipal solid waste incineration, have also contributed to a large reduction in Cd emissions (NAEI, 2019) and, therefore, the decline in concentrations of this element in sludge.

Copper is the second most significant element in sludge (Figure 1). However, in contrast to Zn, and the other elements examined, except for Se, no statistically significant relation between the concentration in sludge and the emissions of this element was detected (P = 0.74, data not shown). The Cu concentration in sludge has also remained relatively consistent over the past 30 year recording period. The unique behaviour of this element may be explained because, in contrast to the other TEs, the results show that this element mainly enters the wastewater system directly and atmospheric transfer is less important, and also that it originates largely from diffuse sources. Thus, Cu in sludge mainly comes from the dissolution of Cu pipes that are ubiquitous in plumbing systems in the UK built environment (Comber and Gunn, 1996), and this is particularly evident in catchments with hard mains water, where more aggressive dissolution takes place (Edwards *et al.*, 1994).

Similar to Cu, Se in sludge was not statistically correlated (P = 0.24) with atmospheric emissions, suggesting this was a minor route of entry to wastewater and sludge, and concentrations have also remained relatively consistent in sludge. Industrial sources of Se include the electrical and electronics industry, the manufacture of semiconductors, rectifiers, ceramics, glass, pigments, alloys and catalysts (Tjandraatmadja, *et al*., 2010). However, the consistent concentration profile suggests that Se originates mainly from diffuse domestic

sources, as it can be found in food products and supplements, shampoos and other cosmetics (Thornton *et al*., 2001).

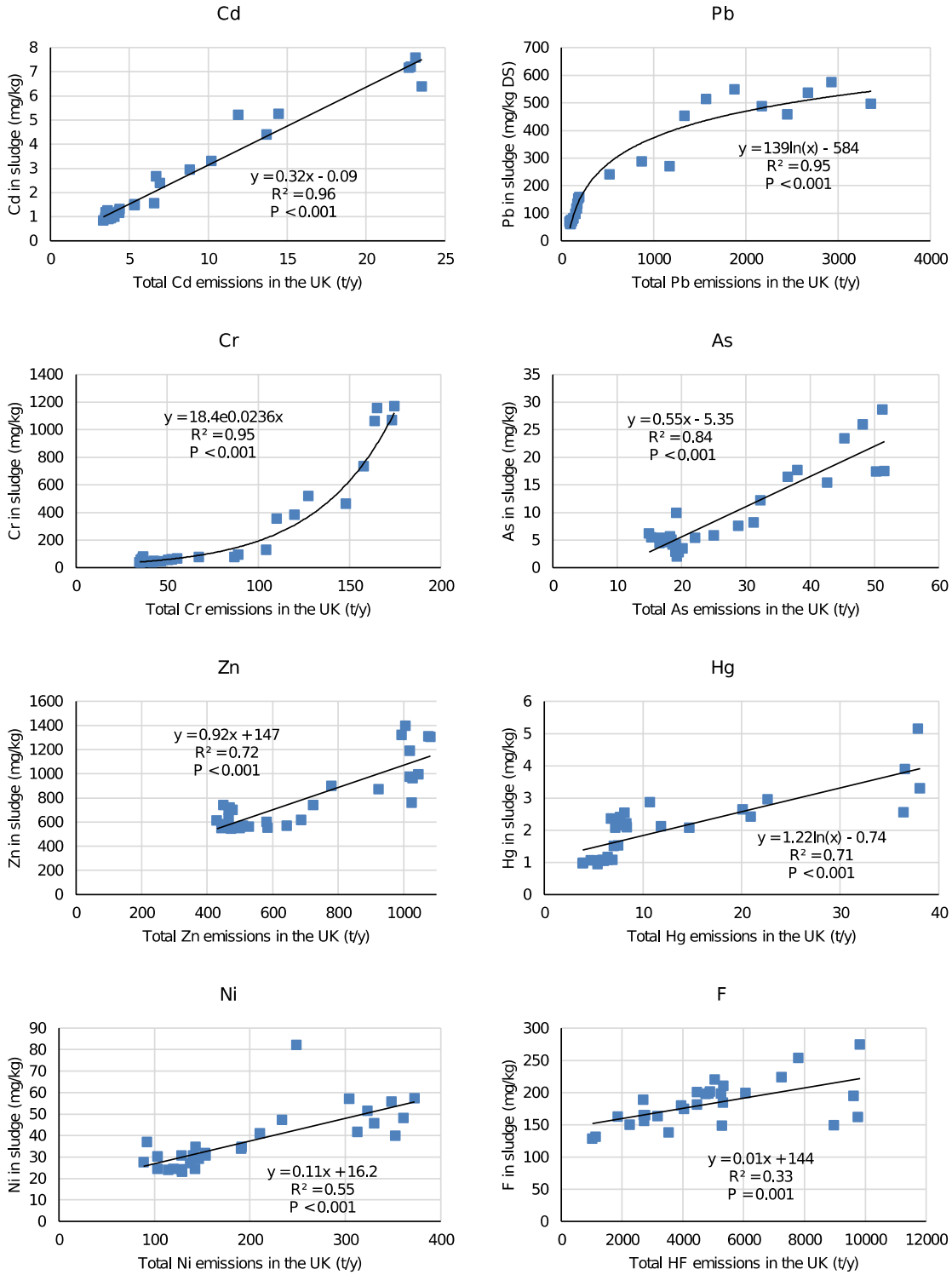

Figure 3 Relationships between mean annual total concentrations in sewage sludge and total annual atmospheric emissions of Cd, Pb, Cr, As, Zn, Hg, Ni and F between 1989 and 2017 (TE emission data from NAEI, 2019)

Sludge type comparison

ANOVA and post-hoc tests were applied to examine the effect of sludge type on TE content, based on the data provided by Company 1 for the period of 2000 to 2017. A statistically significant ($P < 0.05$) effect of sludge type was detected, and mean TE concentrations for the different types of sludge examined are presented in Table 1. As may be expected, TE concentrations in lime-treated sludge were reduced compared to raw sludge, due to the large rates of lime addition required by this form of sludge treatment. This is pertinent when considering the declining patterns in TE concentrations towards the end of the 1990s and in the early part of the next decade as this coincided with a short-term expansion in lime-treatment following the ban on the use of untreated sludge in agriculture from 31 December 1999 (ADAS, 2001). Digested sludge, on the other hand, contained significantly larger mean concentrations of most elements compared to raw and lime-treated sludge. This is explained because TEs are conservative elements that sorb strongly to the sludge solids and, as volatile solids are destroyed during the anaerobic digestion (AD) process, so the concentrations increase. More than 60% of sewage sludge was treated by AD in the UK in 2010 - 2011, and this increased to 80% in 2014 – 2015 (Ofwat, 2016). Thus, a decline in lime-treatment and corresponding expansion of AD (Ofwat, 2016) may explain the small increase (albeit not statistically significant) observed in concentrations of some TEs after 2010, for example, Zn (Figure 1 and 2).

Table 1 Mean total trace element concentrations (mg/kg DS) according to sludge type (Company 1)

Sludge type	Zn	Cu	Ni	Pb	Cd	Cr	Hg	Mo	As	Se	F
Raw (untreated)	466	402	20.8	68.8	0.9	32.8	2.2	4.5	8.5	8.5	115
Lime-treated	337	234	13.2	47.5	0.7	21.3	0.9	3.7	2.9	1.9	97
Digested	682	583	33.0	97.7	1.3	52.8	2.0	7.3	4.1	4.1	186

Trace element accumulation in agricultural soil

Official statistics report that 80% of the sewage sludge produced in the UK is recycled to agricultural land (Ofwat, 2016). However, this is currently approaching 100% as all but one of the sludge incinerators in the UK have been decommissioned due to reaching their design life

in favour of advanced digestion and recycling to agricultural land (pers comm Tom Taylor, Yorkshire Water). The concentrations of TEs in sludge have remained relatively consistent (for example, Cu), associated with diffuse inputs , or have continued to fall in the 20 years since the previous sludge quality statistics were reported (for example Pb, Cd and Hg) in response to source and emission controls and changing industrial practices (Figure 1). Nevertheless, they are larger than the background concentrations present in soil, therefore TEs will accumulate to varying degrees in soil following the long-term application of sewage sludge. A soil accumulation model was therefore developed to assess the effects of repeated applications of sludge to agricultural land on the soil-sludge-limit TE balance. The effects of applying sewage sludge with mean and 95th percentile TE concentrations on the total soil content were calculated and are shown in Figure 4.

The model showed that annual applications of sewage sludge increased TE concentrations in soil following a diminishing pattern with time, as total soil concentrations approached those in the sludge. As may expected from the TE profiles observed in sludge (Figure 1), Zn and Cu were the principal elements limiting sewage sludge recycling to agricultural land in the long term (Figure 4a). Thus, the maximum soil limits for Zn and Cu will be approached in 121 and 198 years, respectively, at the mean concentrations of these elements measured in sludge (Table 2). Under acid soil conditions (pH<6), the soil limits for Cu and Ni are reduced (DEFRA, 2018), however, this would only affect the accumulation of Cu, which would reduce the number of annual applications to approximately 96 years in this case.

Zinc and Cu were also the most limiting at the 95th percentile sludge concentration and, as would be expected, annual applications of sludge in this representative upper TE range would increase the soil concentration to the maximum permissible values of these elements within a shorter time frame, equivalent to 78 and 99 years, respectively (Figure 4b). After Zn and Cu, Hg is the next element most likely to limit the recycling of sewage sludge to agricultural land, however, the soil accumulation model showed that the soil limit value for Hg would be approached in approximately 408 years (at the 95th percentile sludge concentration), a condition, which, given the other more restrictive elements, could never be achieved by land application of sewage sludge.

The analysis of soil TE accumulation showed that Zn and Cu were the most important elements for the agricultural use of sludge under the maximum soil limit regimes applicable in both neutral and acid soil conditions, and also when upper, representative, 95th percentile TE values in sludge were assumed. Given the wide differences in the accumulation of Zn and Cu, and especially Zn, in soil, compared to the other TEs and their limit values, Zn and Cu represent the only elements that are relevant to recycling of contemporary sewage sludge to

agricultural soil in the long term. The concentrations of the other regulated elements: Ni, Cd, Pb and Hg, and also Cr (which is not regulated, but is considered to be one of the major TE contaminants in sludge), have declined to such an extent, relative to Zn and Cu, and to their soil limits, that they will never approach their maximum permissible limits in soil (Table 2 and Figure 4). Therefore, Ni, Cd, Pb and Hg are no longer significant parameters dictating the environmental quality of sewage sludge and could be removed from the regulatory regime for agricultural application.

Table 2 Total trace element concentrations in sludge and the maximum permissible limit and background concentrations in soil and following annual application of sludge for 10, 100 or 1000 years, or until the UK maximum permissible soil limit is reached

	Zn	Cu	Ni	Pb	Cd	Cr	Hg
Median soil concentration (mg/kg ds)[1, 2]	76.0	19.0	21.0	49.0	0.33	68.0	0.1
Mean sludge concentration (mg/kg DS)[3]	702.9	401.6	30.2	68.3	1.0	68.9	1.0
95th Percentile sludge concentration (mg/kg DS)[4]	1013.2	718.7	62.9	138.5	1.9	129.0	1.8
Soil limit (mg/kg ds)[5]	200	135	75	300	3.0	400	1.0
Soil concentration after 10 years (mg/kg ds)[6]	87.4	25.9	21.2	49.4	0.3	68.0	0.1
Soil concentration after 50 years (mg/kg ds)[6]	130.8	52.4	21.8	50.7	0.4	68.1	0.2
Soil concentration after 100 years (mg/kg ds)[6]	180.7	82.9	22.5	52.2	0.4	68.2	0.3
Soil concentration after 1000 years (mg/kg ds)[6]	121 years to reach the limit	198 years to reach the limit	28.7	65.2	0.9	68.8	0.9

(1) McGrath and Loveland (1992): Hg data; (2) Rawlins *et al*. (2012): Zn, Cu, Ni, Pb, Cd, Cr; (3) Mean total trace element concentrations in 2017; (4) 95th percentile trace element concentrations in 2017; (5) DEFRA (2018) for pH between 5 – 7 for Zn, pH 6 – 7 for Cu and Ni and pH ≥5 for other elements; (6) Soil concentration after annual application of sludge with mean total trace element concentrations in 2017 and N limit of 250 kg/ha; ds, dry soil; DS, dry solids

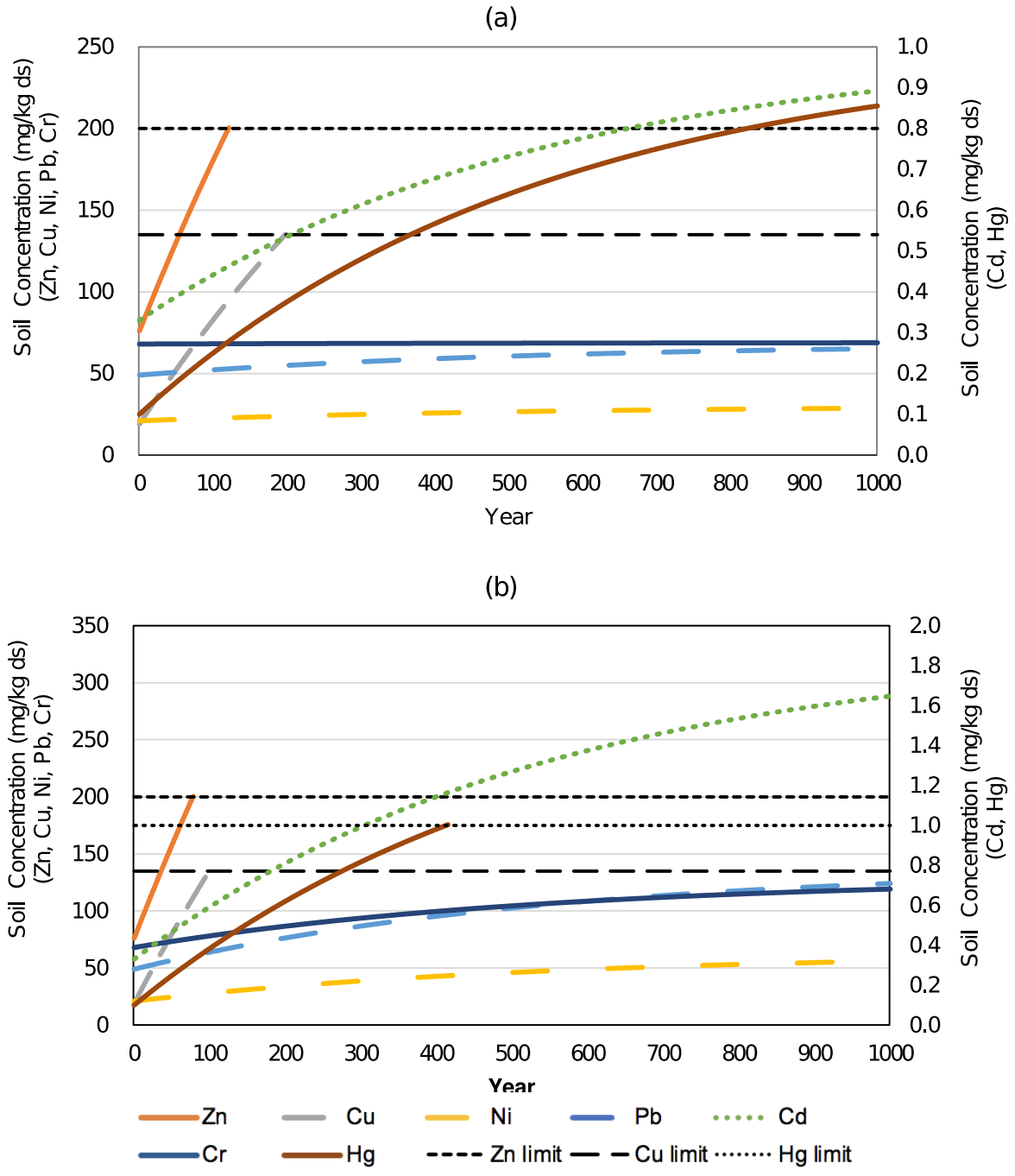

Figure 4 Accumulative total concentrations of trace elements in soil (mg/kg dry soil (ds)) following annual application of sewage sludge at a rate of 250 kg N/ha and containing (a) mean, and (b) 95th percentile total concentrations of trace elements; maximum permissible soil concentrations are shown for the most limiting elements, Zn (pH 5-7), Cu (pH 6-7), and also Hg (pH ≥5), although Hg is not limiting to long-term agricultural application; other elements will not reach their respective soil limit values

Conclusions

Trace element concentrations in sewage sludge have decreased significantly in response to declining pollutant emissions, demonstrating the environmental benefits of effective source control and cleaner technologies. This trend has continued in the past 20 years for several TEs in sludge including: Pb, Cr, Cd, Hg and F. In contrast, the concentrations of Cu, Ni, Mo and Se have remained relatively consistent over this period, whereas the monitoring data suggest Zn and As may have slightly increased. Indeed, contemporary concentrations of TEs in sludge reflect the balance between declining emissions, albeit at a slower rate compared to earlier when concentrations were often greater, and the effect of more intensive sludge treatments, and particularly AD, that increase volatile solids destruction and hence the TE content. Although this balance has resulted in a small apparent rise in mean sludge As, a comparison of conservative total concentrations shows that sludge is not a significant contributor to the soil As status.

The analysis of TE accumulation in sludge-amended agricultural soil, based on N restricted annual applications, showed that Zn and Cu were the most limiting to agricultural recycling, reaching their maximum soil limit concentration values in approximately 120 and 200 years, respectively. In contrast, Ni, Cd, Cr, Pb and Hg are unlikely to approach their maximum permissible limits, relative to Zn and Cu. The results emphasise that the TE quality of sludge in the UK has increased to the extent that the statutory soil limits for Ni, Cd, Pb and Hg have effectively become obsolete and could therefore be removed from the regulatory controls on the agricultural use of sewage sludge.

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