Chemosphere 111 (2014) 478–484

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Concentrations of trace substances in sewage sludge from 28 wastewater treatment works in the UK

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HIGHLIGHTS

- Sludge samples from 28 UK WwTWs were analysed for >40 trace substances.
- Concentrations were broadly similar across all sludge samples.
- Concentrations were generally below regulatory standards for sludge.
- Predicted concentrations in soil indicated negligible environmental risk.

ARTICLE INFO

Article history:
Received 23 December 2013
Received in revised form 8 April 2014
Accepted 9 April 2014

Handling Editor: I. Cousins

Keywords:
Sewage Sludge Directive
Sludge
Priority substances
Trace substances
Chemical Investigations Programme

ABSTRACT

Concentrations of trace substances in sewage sludge have been measured in a survey of 28 wastewater treatment works (WwTWs) in the UK carried out over a period of 12 months. Approximately 250 samples were analysed for more than 40 trace contaminants, including trace metals, pharmaceuticals, polycyclic aromatic hydrocarbons (PAHs), 'emerging' and regulated organic pollutants. All substances investigated were found to be present in at least some of the sludges sampled. Concentrations were relatively homogenous across all the WwTWs, irrespective of the treatment process, influent and effluent concentrations, and the location of the sludge sampling point within each works. Analysis of the results against existing regulatory and proposed thresholds suggested that levels are mostly below the limits set in the Sewage Sludge Directive, and proposed new limits for sludge used in agriculture. Predicted soil concentrations after application of sewage sludge to land were below the predicted no effect concentrations (PNEC) for all determinands. Predicted concentrations of pharmaceuticals in soil were also below thresholds deemed to indicate negligible environmental risk.

1. Introduction

Sewage sludge is increasingly applied to agricultural land, a practice that is deemed both economically and environmentally advantageous (e.g. Eriksson et al., 2011). Production of sludge is expected to increase from 11.5 M tonnes of dried sludge (2010) to 13 M tonnes of dried sludge by 2020 (Palfrey, 2010). In parallel, raised public awareness of environmental pollution and new environmental regulation (e.g. the 'Water Framework Directive' (Official Journal of the European Commission, 2000); the 'Priority Substances Directive' (Official Journal of the European Commission, 2008)) have led to an increased interest in the presence of trace substances in wastewaters and effluents (WwTWs; e.g. Rule et al., 2006; Gasperi et al., 2008; Clara et al., 2012). Greater awareness of the presence of contaminants in wastewater raises the issue of the extent to which various chemicals are present in sewage sludge and consequently might be transferred to land, impacting soil organisms or being transferred up the food chain. As a result, the presence of trace chemicals in sewage sludge has been investigated in several studies over the last decades (see for example, reviews by Eriksson et al., 2008; Fytili and Zabaniotou, 2008; Eriksen et al., 2009; Clarke and Smith, 2011). The current paper is an account of the concentrations of trace substances in sludge, determined as part of the Chemical Investigations Programme (CIP) in the UK.

The CIP was a £25 M nationally-coordinated investigation of the risks posed by trace contaminants in the wastewater treatment process in the UK, examining effluent quality, the effectiveness of different treatment processes and sources of substances in the sewer catchment (Gardner et al., 2012; 2013). Determinations of sludge quality were made, as part of the wider CIP process investigations, at 28 WwTW sites.

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http://dx.doi.org/10.1016/j.chemosphere.2014.04.025
0045-6535/

2. Materials and methods

WwTWs for process investigation were selected to be representative of different process types applied across UK sites. 14 biological-filter (BF) WwTWs, 12 activated sludge process (ASP) WwTWs, one biological nutrient removal (BNR) WwTW and one membrane bio-reactor (MBR) WwTW were included in the study. Sludge samples were collected at the participating works over a period of approximately 12 months, with 7–15 sampling occasions at each site. Sludge samples were collected at one selected point per WwTW. Samples consisted of primary sludge (collected from the primary settlement tank), secondary/biological sludge (e.g. humus sludge) or mixed sludge (mixture of primary and secondary/biological sludge) and were analysed for suites of substances including nutrients, metals, emerging and regulated organics, polybrominated diphenyl ethers (BDEs) and pharmaceuticals. The frequency and extent of the sampling meant that results presented here for each parameter are based on approximately 250 samples, sometimes with additional replicates taken on the same date. A smaller number of data points (derived from 6 or 7 WwTWs) were available for a limited number of determinands; these were diclofenac, nonylphenol and its mono, di- and tri- ethoxylates. Where WwTWs collected replicate sludge samples on the same sampling occasion, concentrations across replicates have been averaged and the average value has been used in all subsequent statistical calculations. All laboratories taking part in this study were accredited to ISO17025 standard for their quality systems. An Aqua Regia digest was applied for the analysis of metals, and organic substances were analysed by LC/MS or GS/MS. All concentrations are reported as mg kg^{-1} dry weight, with the exception of dry solids concentrations which are reported as a percentage. Where results were reported as less than the limit of detection (LOD), the approach was taken of substituting half the reported value (as specified at EU level; Official Journal of the European Commission, 2009).

3. Results

3.1. Trace substances concentrators in sludge

Key statistics for all parameters considered are presented in Table 1. All substances included in the analysis suite for this study were detected in at least some of the sludge samples. In the metals group, zinc exhibited the highest concentrations, with a median

Table 1
Summary of sludge data. sd: standard deviations, 25%ile: 25th percentile, 75%ile: 75th percentile CoV: coefficient of variation; WwTW: wastewater treatment works.

	Mean	Median	sd	25% ile	75% ile	CoV	Number of WwTWs sampled
Dry solids %	2.6	1.3	2.3	0.9	4.3	0.88	25
Nitrogen	41733	38409	12418	32941	53281	0.30	28
Phosphorus	19898	17742	10836	10491	27397	0.55	28
Potassium	3313	2171	2795	1574	4195	0.84	27
Metals							
Nickel	29.9	25.1	19.9	19.3	32.8	0.67	28
Lead	68.9	48.3	52.4	38.1	82.6	0.76	28
Copper	344	269	228	172	414	0.66	28
Zinc	607	505	309	454	642	0.51	28
Cadmium	0.8	0.7	0.5	0.6	0.9	0.61	28
Mercury	0.7	0.7	0.2	0.6	0.8	0.34	28
Silver	2.7	1.7	3.8	0.5	2.7	1.44	28
BDEs							
2,2′,4,4′-tetrabromodiphenyl ether (PBDE47)	0.023	0.021	0.014	0.015	0.031	0.59	28
2,2′,4,4′,5-pentabromodiphenyl ether (PBDE99)	0.032	0.033	0.019	0.022	0.043	0.60	28
2,2′,4,4′,6-pentabromodiphenyl ether (PBDE100)	0.007	0.006	0.004	0.005	0.009	0.56	28
2,2′,4,4′,5,5′-hexabromodiphenyl ether (PBDE153)	0.006	0.005	0.003	0.004	0.008	0.58	28
2,2′,4,4′,5,6′-hexabromodiphenyl ether (PBDE154)	0.005	0.005	0.002	0.003	0.005	0.53	28
'Emerging' and regulated organic substances							
Diethylhexylphthalate (DEHP)	19.0	11.0	20.6	3.0	30.8	1.08	28
Nonylphenol 4-nonylphenol	4.4	3.8	2.9	2.3	5.8	0.67	28
Tributyltin compounds (Tributyltin-cation; TBT)	0.02	0.02	0.01	0.01	0.02	0.70	28
Triclosan	4.9	4.7	3.1	2.1	7.0	0.64	28
Bentazone	0.07	0.02	0.06	0.02	0.11	0.95	28
Bisphenol-A	0.34	0.21	0.35	0.12	0.56	1.05	28
Nonylphenol Monoethoxylate	5.0	6.1	2.9	2.7	7.1	0.59	6
Nonylphenol Diethoxylate	1.1	1.1	0.6	1.0	1.3	0.51	6
Nonylphenol Triethoxylate	176.0	0.6	1.7	0.3	0.9	1.41	6
PAHs							
Anthracene	0.11	0.10	0.07	0.07	0.13	0.66	28
Fluoranthene	0.70	0.52	0.51	0.41	0.77	0.73	28
Naphthalene	0.43	0.21	0.73	0.10	0.40	1.71	28
Benzo(a)pyrene	0.38	0.32	0.20	0.19	0.50	0.52	28
Benzo(b)fluoranthene	0.38	0.31	0.21	0.25	0.46	0.54	28
Benzo(k)fluoranthene	0.29	0.25	0.21	0.13	0.38	0.74	28
Benzo(g,h,i)perylene	0.33	0.27	0.22	0.16	0.44	0.66	28
Indeno(1,2,3-cd)pyrene	0.32	0.28	0.18	0.18	0.42	0.58	28
Pharmaceuticals							
Diclofenac	0.06	0.07	0.03	0.05	0.07	0.41	7
Ibuprofen	0.27	0.22	0.19	0.12	0.39	0.69	28
Propranolol	0.14	0.12	0.08	0.10	0.18	0.54	28
Erythromycin	0.06	0.05	0.04	0.03	0.06	0.63	28
Ofloxacin	0.22	0.20	0.12	0.14	0.27	0.56	28
Oxytetracycline	7.63	4.00	9.25	2.65	8.66	1.21	28
Fluoxetine	0.13	0.12	0.05	0.09	0.18	0.42	28