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Time-trends of metals and organic contaminants in sewage sludge

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ABSTRACT

The occurrence of chemicals in sludge from sewage treatment plants (STPs) is of concern for human health and the environment. Legislations and regulations are put in place to minimize the release of harmful chemicals into the environment and arable land, e.g. via application of sewage sludge. Temporal trends analysis of sludge contaminants can be used to assess the effectiveness of such actions. Such analyses can be performed retrospectively, using sludge stored in environmental specimen banks, to investigate new or emerging environmental contaminants. The present study provides data from time-trend analyses of metals, persistent organic pollutants, pharmaceuticals, personal care products, and other organic compounds in sludge from Swedish. The analysis showed that sludge is a suitable matrix for time-trend studies and it can take on average 12 years (range, 5–26 years) to track an annual change of $\pm 10\%$ (with a power of 80%). Statistically significant trends were found for 18 out of the 77 compounds subjected to analysis, of which 75% showed decreasing trends. Triclosan and the antibiotic norfloxacin followed the same trend as the national recorded usage and decreased annually by 65% and 60%, respectively. The opposite was true for the methylsiloxanes, which showed an annual average increase of about 30%. A downward trend (about 20% year⁻¹) was observed for the polybrominated diphenylethers (PBDEs 154 and 183), while PBDE 209 increased by 16%. Further measurements are required to determine if the substitution of PBDEs by chlorinated paraffins or organophosphorus compounds have resulted in increased concentrations of the latter in sludge.

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1. Introduction

Large quantities and a broad spectrum of organic compounds (OCs) currently used in today's society, originating from, e.g. households, hospitals, and industry, may enter municipal sewage treatment plants (STPs). Ideally, most of the harmful OCs would be removed during sewage treatment (e.g. by degradation or evaporation), and therefore would not remain in the sludge or effluent. However, many OCs are highly

lipophilic and have a high affinity for sludge, and thus can concentrate in the sludge, often reaching levels of ng-mg kg⁻¹ dry weight (d.w.) (Bossi et al., 2008; Langdon et al., 2011; Lindberg et al., 2006; Marklund et al., 2005; Stevens et al., 2003; Voulvoulis et al., 2004).

Land application of treated sewage sludge (biosolids) has been adopted worldwide as an option for sludge management. Sludge contains nutrients and organic matter that can help to improve soil productivity. Moreover, land application of

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treated sewage sludge is likely to increase in importance due to the global depletion of phosphorus resources (Steen, 1998) and the need for sustainable nutrient management strategies. Within the European countries, as elsewhere in the world, there are many diverse opinions regarding the suitability of land application of sewage sludge. The Swedish Government does, for instance, encourage this practice, while Switzerland has banned it. Within the European Union (EU), approximately 37% of total annual production of treated sewage sludge is used in agriculture. In the USA, the corresponding figure is 60% (NRC, 2002), and in Sweden 15%. For use on agricultural land, levels of hazardous substances, such as persistent organic pollutants (POPs), toxic metals, pesticides, hormone disrupting chemicals, carcinogens, pathogens, etc., in the sludge should be low. This is especially important to reassure consumers and consumer organizations that products grown on such sludge-amended soil are safe. Today, there are no legal limits for OCs in sludge (destined for agricultural purposes), but EU maximum residue limits (EU, 1986) have been established for six metals (Cd, 20–40; Cu, 1000–1750; Hg, 16–25; Ni, 300–400; Pb, 750–1200; Zn, 2500–4000 mg kg⁻¹ d.w.). Stricter limits have been set in some countries, e.g. Belgium, Denmark, Finland, The Netherlands, and Sweden (In Sweden: Cd, 2; Cr, 100; Cu, 600; Hg, 2.5; Ni, 50; Pb, 100; Zn, 800 mg kg⁻¹ d.w.) (Ministry of the Environment, 1998). In contrast, in the USA, limits are slightly higher (As, 75; Cd, 85; Cu, 4300; Pb, 840; Hg, 57; Ni, 420; Se, 100; Zn, 7500 mg kg⁻¹ d.w.) (U.S. EPA, 1993).

National and international environmental legislations and regulations are crucial to minimize the release of hazardous substances into the environment. National bans of PCBs in the 1970s and the international regulation of POPs, including polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), in the Stockholm Convention (UNEP, 2001) have resulted in a substantial reduction of these substances in the environment. There are also indications that levels of polychlorinated biphenyls (PCBs) and PCDD/Fs have decreased in sewage sludge over the past decades (Clarke et al., 2008, 2010).

An interesting question is whether the increased environmental awareness among consumers and professionals has also contributed to the downward trend in chemical emissions. Triclosan, frequently used as an antibacterial agent in personal care products, has been of public concern due to its negative environmental impact (Crofton et al., 2007; Veldhoen et al., 2006). Similarly, there is an increased awareness among physicians that certain antibiotics, e.g. fluoroquinolones (FQs), have considerable environmental stability and may contribute to the development of antibiotic resistance. This concern may have resulted in decreased use of this type of antibiotic on prescription, and thus decreasing levels in sewage sludge as well as in other environmental matrices. A more complex scenario can be envisioned for the water and dirt repellent perfluorooctane sulfonate (PFOS). Consumer pressure and health problems among workers led to a voluntary phase-out of this chemical in early 2000 by the main manufacturer. PFOS has since been regulated in the Stockholm Convention (UNEP, 2001), and this is expected to lead to even lower emissions. However, mass flow studies have shown that other perfluorinated chemicals (PFCs), which are

used as substitutes for PFOS, can transform into PFOS during sewage treatment (Bossi et al., 2008; Loganathan et al., 2007) and this could generate higher levels in sludge.

Sewage sludge is an attractive matrix for examining time-dependent trends of fluxes of such organic compounds from society. In the last decade, several reviews have compiled global monitoring data of sludge contaminants (Clarke and Smith, 2011; Harrison et al., 2006; Law et al., 2006; Xia et al., 2005). During the same period, the Swedish Environmental Protection Agency (EPA) initiated a program to systematically sample, analyse and bank sewage sludge. In 2010, Germany and Switzerland also started to discuss a routine monitoring and archiving program for sludge samples and STP effluents (Rudel et al., 2010). However, to the best of our knowledge, no systematic studies of the temporal trend of extensive sets of sludge contaminants have been reported.

The purpose of this study was therefore to perform a time-trend analysis of metals, POPs, pharmaceuticals and personal care products (PPCPs), and other OCs in sludge (based on seven years of Swedish EPA data). There were two main aims (i) to determine if the within-year variability in contaminant concentrations in sludge samples from Swedish STPs was sufficiently low to allow time-trend studies over reasonable time-spans (ii) to determine if there were any statistically significant temporal trends in the concentrations of the investigated chemicals. The established time-trends are discussed in relation to society's attempts to reduce the release of harmful substances into the environment, changes in consumer preferences, industry's efforts to find suitable substitutes for regulated or questionable substances, and future design of sludge monitoring programs.

2. Material and methods

2.1. Selection of sludge contaminants

The target compounds (totally 126), some of which are classified as POPs (UNEP, 2001), were selected in 2004, together with the Swedish Environmental Protection Agency (Swedish EPA), from Scandinavian priority lists, the European Union Water Framework Directive (WFD) (EU, 2000, 2008), and the "Working document on sludge" (CEC, 2000). The selection of sludge contaminants was also influenced by: i) the national use statistics; ii) the ability to associate with sludge; iii) representativity (for a group of chemicals); iv) ease and cost of analysis; and iv) should represent a wide range of compound groups. Compounds that are regularly measured by the STPs were not included, e.g. indicator PCBs and PAHs, but the data was collected. Unfortunately, this data was heterogeneous (performed by different laboratories) and not of sufficient quality for time-trend analyses.

The main applications and recorded national annual quantities used in Sweden of the compounds are summarised in Table 1, while the CAS numbers and physicochemical properties are presented in Table S1. It should be noted that the use statistics recorded in the Swedish product register do not include chemicals manufactured or imported in quantities less than 100 kg per company or those present in imported consumer articles.

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