Science of the Total Environment 538 (2015) 750–767

Contents lists available at ScienceDirect

Science of the Total Environment

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

Pharmaceuticals and personal care products in untreated and treated sewage sludge: Occurrence and environmental risk in the case of application on soil — A critical review

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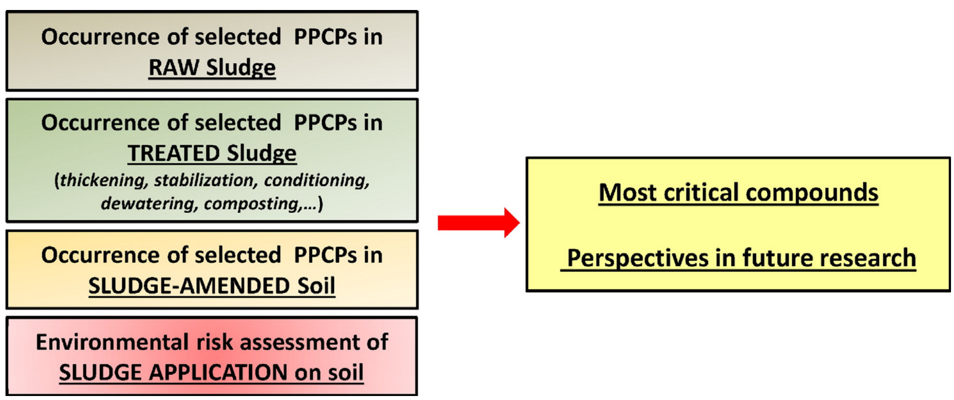
HIGHLIGHTS

- The review refers to concentrations of 169 PPCPs in different kinds of sludge.
- After digestion or composting, concentrations of most compounds reduced.
- K_d are reported for the selected compounds in different kinds of sludge and soils.
- Environmental risk due to PPCPs in case of sludge application on soil was assessed.
- The most critical compounds are triclosan, triclocarban, hormones and antibiotics.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 July 2015
Received in revised form 18 August 2015
Accepted 18 August 2015
Available online xxxx

Editor: D. Barcelo

Keywords:
Sewage sludge
Pharmaceuticals
Personal care products
Environmental risk
Sludge-amended soil

ABSTRACT

This review is based on 59 papers published between 2002 and 2015, referring to about 450 treatment trains providing data regarding sludge concentrations for 169 compounds, specifically 152 pharmaceuticals and 17 personal care products, grouped into 28 different classes. The rationale of the study is to provide data to evaluate the environmental risk posed by the spreading of treated sludge in agriculture. Following discussion of the legislative scenario governing the final disposal of treated sludge in European countries and the USA, the study provides a snapshot of the occurrence of selected compounds in primary, secondary, mixed, digested, conditioned, composted and dried sludge originating in municipal wastewater treatment plants fed mainly with urban wastewater as well as in sludge-amended soil. Not only are measured values reported, but also predicted concentrations based on K_d values are reported. It emerges that in secondary sludge, the highest concentrations were found for fragrances, antiseptics and antibiotics and an attenuation in their concentrations occurs during treatment, in particular anaerobic digestion and composting. An in-depth literature survey of the (measured and predicted) K_d values for the different compounds and treated sludge are reported and an analysis of the influence of pH, redox conditions, sludge type was carried out. The data regarding measured and predicted concentrations of

Abbreviations: AeD, aerobic digestion; AnD, anaerobic digestion; AOX, absorbable organically bound halogens; BAF, biological aerated filter; BNR, biological nutrient reactor; CAS, conventional activated sludge; CEC, cation exchange capacity; CFR, Code of Federal Regulations; CW, constructed wetland; DM, dry matter; D_{ow}, octanol water partition coefficient; E1, estrone; E2, estradiol; E3, estriol; EE2, ethinylestradiol; EQS, environmental quality standard; f_{oc}, fraction of organic carbon; K_a, dissociation constant; k_{biol}, biological degradation rate; K_d, solid liquid partition coefficient; K_{ow}, octanol water distribution coefficient; LAS, linear alkyl sulfonates; MAnD, mesophilic anaerobic digestion; MBR, membrane biological reactor; MEC, measured environmental concentration; NP, nonylphenol; NPnEO, nonylphenol (n) ethoxylates; OM, organic matter; PAH, polycyclic aromatic hydrocarbons; PCB, polychlorinated biphenyls; PCDD, Polychlorinated dibenzo-p-dioxins; PCP, personal care product; PEC, predicted environmental concentration; PhC, pharmaceutical compound; PNEC, predicted no effect concentration; PPCP, pharmaceutical and personal care product; RQ, risk quotient; SRT, sludge retention time; SSD, Sewage Sludge Directive; T, temperature; TAnD, thermophilic anaerobic digestion; TSS, total suspended solids; UASB, upflow anaerobic sludge blanket; WWTP, wastewater treatment plant.

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http://dx.doi.org/10.1016/j.scitotenv.2015.08.108

selected compounds in sludge-amended soil is then analyzed. Finally an environmental risk assessment posed by their occurrence in soil in the case of land application of sludge is examined, and the results obtained by different authors are compared. The most critical compounds found in the sludge-amended soil are estradiol, ciprofloxacin, ofloxacin, tetracycline, caffeine, triclosan and triclocarban. The study concludes with a focus on the main issues that should be further investigated in order to refine the environmental risk assessment.

Contents

1. Introduction

Sludge originates during biological and chemical processes in wastewater treatment plants (WWTPs) and may contain a wide spectrum of organic and inorganic substances as well as microorganisms and viruses which are separated from the liquid phase during treatments.

In Europe, its production is expected to increase from 11.5 M tons of dry matter (DM) (2010) to over 13 M tons of DM by 2020, chiefly due to increased sewerage and treatments in East European countries (Palfrey, 2010; Eriksson et al., 2008). The main disposal routes are incineration, landfill, land application, composting, with the specific percentages varying from country to country. For instance incineration of sludge is about 90% in Belgium, 50% in Germany and 45% in Denmark, while reuse in agriculture reaches 50% in Denmark and 25% in Sweden, where 50% is landfilled or allocated to construction work (Malmborg and Magnér, 2015; Kelessidis and Stasinakis, 2012). Recently Kelessidis and Stasinakis (2012) reported that 53% of sludge in EU-27 is reused in agriculture either directly or after composting, whereas Citulski and Farahbakhsh (2010) reported that more than 40% is spread on land in Canada and according to Venkatesan et al. (2014) more than 50% is spread on land in the USA.

The interest in using sewage sludge in agriculture is due to its nutrient content and soil-conditioning properties that are useful for restoring overexploited land to agricultural use or for improving the humus content and water-holding capacity of light-textured sandy soil as well as in cases where soils are depleted or subject to erosion (Clarke and Smith, 2011; Inglezakis et al., 2014).

Disposal routes of sewage sludge must fulfill specific regulations. With regard to the land application option, in the last 20 years great attention has been placed mainly on the occurrence of heavy metals in sludge and their fate once it is applied on agriculture land, and limits have been set and adopted in different countries (Stasinakis, 2012). Sometimes, additional limits have been implemented for surfactants (mainly linear alkyl sulfonates, LAS), polycyclic aromatic hydrocarbons (PAH), nonylphenol (n)ethoxylates (NPnEO), polychlorinated biphenyls (PCB), phthalates and pesticides in sludge, and studies monitoring their fate once spread on the land are ongoing (Kelessidis and Stasinakis, 2012). No limits have been set for pharmaceuticals (PhCs) and other contaminants of emerging interest, including personal care products (PCPs) in sewage sludge.

Land disposal of sewage sludge is regulated at EU level by the so-called Sewage Sludge Directive (SSD) 86/278/EEC (CEC, 1986) and in each EU country national regulations have also been set in accordance with the SSD. Generally, they set the maximum allowable concentrations of potentially toxic elements in soil after the application of sewage sludge, and maximum annual rates of application. They do not set concentration limits for organic compounds for either sludge or soil. A draft of a working document on sludge and biowaste is under discussion within the EU, where cut-off values are set for other groups of organic compounds (EC, European Commission, 2010).

There is ongoing debate within the scientific community in order to evaluate potential (environmental) risks in this kind of practice, due to the occurrence of toxic and persistent substances in sludge, such as aquifer contamination, the accumulation of pollutants in soil,