Waste Management 34 (2014) 1217–1226

Contents lists available at ScienceDirect

Waste Management

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

Priority and emerging pollutants in sewage sludge and fate during sludge treatment

R. Mailler [a,*], J. Gasperi [a], G. Chebbo [a], V. Rocher [b]

[a] LEESU, UMR MA 102, Université Paris-Est, Agro Paris Tech, 6-8 avenue Blaise Pascal, Champs-sur-Marne, 77455 Marne-la-Vallée Cedex 2, France
[b] SIAAP, Direction du Développement et de la Prospective, 82 avenue Kléber, 92700 Colombes, France

ARTICLE INFO

Article history:
Received 11 October 2013
Accepted 28 March 2014
Available online 3 May 2014

Keywords:
Anaerobic digestion
Thermal drying
Centrifugation
Emerging pollutant
Priority pollutant
Sludge

ABSTRACT

This paper aims at characterizing the quality of different treated sludges from Paris conurbation in terms of micropollutants and assessing their fate during different sludge treatment processes (STP). To achieve this, a large panel of priority and emerging pollutants ($n = 117$) have been monitored in different STPs from Parisian wastewater treatment plants including anaerobic digestion, thermal drying, centrifugation and a sludge cake production unit. Considering the quality of treated sludges, comparable micropollutant patterns are found for the different sludges investigated (in mg/kg DM – dry matter). 35 compounds were detected in treated sludges. Some compounds (metals, organotins, alkylphenols, DEHP) are found in every kinds of sludge while pesticides or VOCs are never detected. Sludge cake is the most contaminated sludge, resulting from concentration phenomenon during different treatments. As regards treatments, both centrifugation and thermal drying have broadly no important impact on sludge contamination for metals and organic compounds, even if a slight removal seems to be possible with thermal drying for several compounds by abiotic transfers. Three different behaviors can be highlighted in anaerobic digestion: (i) no removal (metals), (ii) removal following dry matter (DM) elimination (organotins and NP) and iii) removal higher than DM (alkylphenols – except NP – BDE 209 and DEHP). Thus, this process allows a clear removal of biodegradable micropollutants which could be potentially significantly improved by increasing DM removal through operational parameters modifications (retention time, temperature, pre-treatment, etc.).

1. Introduction

Wastewater treatment plants (WWTPs) produce an important quantity of sludge resulting from total suspended solids (TSS) removal and growth of microorganisms within biological treatments. Actually, about 1 million tons dry matter (DM) of sludge are produced every year by French WWTPs, while Germany and UK produce respectively 2.2 and 1.8 million tons (Kelessidis and Stasinakis, 2012), for a total of 11 million tons DM of sludge in all Europe (EU-27). The management of these sludges is achieved through three principal pathways: agricultural uses (land farming), incineration and disposal/landfilling (Fytili and Zabaniotou, 2008). In 2008, land farming was the main pathway both in France (>60%) and in the European Union (>50%) (Kelessidis and Stasinakis, 2012).

Contamination of WWTP sludges by micropollutants has been reported for several years (Clarke and Smith, 2011; Harrison et al., 2006; Scancar et al., 2000). This results from pollutant sorption during primary and biological treatments because of their hydrophobicity or propensity to be adsorbed on particles (Byrns, 2001). As sludges are mainly land farmed, this contamination is worrying especially considering accumulation of some micropollutants in sludge and their transfer to the environment, like polybromodiphenyl ethers (PBDE) (Eljarrat et al., 2008), metals (Chipasa, 2003), organotins (Craig, 2003) or polychlorobiphenyls (PCB) (Stevens et al., 2002). To limit contamination of the environment by micropollutants, European and national regulations have been established to progressively forbid sludge disposal and regulate land farming. Such regulations concern principally heavy metals, PAHs and PCBs (Table 1). In particular, the Urban Wastewater Treatment Directive (EC, 1986), amended by (91/271/EEC) (EC, 1991), states maximum thresholds and maximum annual flux to land farm for metals.

Despite that, data and knowledge are still missing concerning (i) the quality of treated sludges and (ii) the efficiency of the sludge

* Corresponding author. Tel.: +33 145171328.
E-mail addresses: maillerr@leesu.enpc.fr, gasperi@u-pec.fr (R. Mailler), vincent.rocher@siaap.fr (V. Rocher).

http://dx.doi.org/10.1016/j.wasman.2014.03.028
0956-053X/

Table 1
French (order of 8th of January 1998) and European (EC, 1986) thresholds for PCBs, PAHs and metals in sludges to landfarm.

	Threshold value in sludge (mg/kg DM)		Maximal flux from sludges in last 10 years (g/m^2)	
	General case	Pasture case	General case	Pasture case
Σ7 PCBs[a]	0.8	0.8	1.2	1.2
Fluoranthene	5	4	7.5	6
Benzo(b)fluoranthene	2.5	2.5	4	4
Benzo(a)pyrene	2	1.5	3	2
Cadmium	10 (20–40)		0.015	
Chrome	1000		1.5	
Copper	1000 (1000–1750)		1.5	
Mercury	10 (16–25)		0.015	
Nickel	200 (300–400)		0.3	
Lead	800 (750–1200)		1.5	
Zinc	3000 (2500–4000)		4.5	
Chrome + zinc + copper + nickel	4000		6	

Figures presented are from French regulation (order of 8th of January 1998), while European thresholds from wastewater treatment directive (EC, 1986) are given in brackets.
[a] PCBs 28, 52, 101, 118, 138, 153 and 180.

treatment processes (STPs) for micropollutant removal as well as the mechanisms involved. This paper aims at improving and completing knowledge about Parisian sludges contamination by micropollutants and their fate during four different STPs, i.e. anaerobic digestion, centrifugation, thermal drying and sludge cake production.

As no typical sludge treatment layout can be identified, and different configurations exist depending on the capacity of the treatment plant or the quality of treated sludge expected (regulations), the characterization of each process individually seems to be a relevant strategy.

To achieve that, a large number of micropollutants ($n = 117$) were monitored in these STPs. Contents were measured in raw, digested, centrifuged, thermally dried sludges and sludge cake (cooked then press filtered). Micropollutant removals were calculated, to better understand the behaviors of these compounds and to determine the potential of these processes for controlling the micropollutant contamination of sludge.

2. Material and methods

2.1. Sludge treatment processes (STPs) description and sampling procedure

Three STPs from three WWTPs in Paris were studied (Fig. 1). It should be noted that these WWTPs, run by the Parisian public sanitation service (SIAAP), treat wastewater from the same catchment (downstream Paris conurbation) and the comparison of processes and treated sludges (digested sludge – DS, centrifuged sludge – CS, sludge cake – SC and thermally dried sludge – TS, Fig. 1) is then relevant to underline the differences in micropollutants fate.

The Seine Centre plant treats 240,000 m^3 of wastewater per day. Sludge produced is first centrifuged to achieve a volume reduction, resulting in a production of almost 21,000 tons DM of centrifuged sludge per year (SIAAP source). Then, sludge is incinerated producing ash and smoke, which is specifically treated to minimize odors. The Seine Aval plant receives 1,700,000 m^3 of wastewater per day (biggest in Europe) and produces more than 55,000 tons DM of treated sludge per year (SIAAP source). The first STP consists in a mesophilic (37 °C) anaerobic digestion to transform an important part of organic matter into biogas and eliminating pathogens and parasites. Digested sludge is then dewatered by thickening, thermal conditioning (heat exchange and cooking at 195 °C and 20 bars) and press filtration. These successive treatments allow reducing sludge volume by more than a factor 10 (i.e. DM, Table 1) and producing a dewatered cake called sludge cake which is reused as agricultural fertilizer. The Seine Grésillons plant treats 100,000 m^3 of wastewater per day. Sludge treatment is performed by centrifugation and thermal drying. The thermal drying process can operate at a wide range of temperature, but the facility used in this plant operates at a high temperature (260 °C) compared to conventional dryers (generally 105 °C (Voulvoulis and Lester, 2006)). This allows reducing the water content drastically (i.e. DM content, Table 1) to obtain, after compacting, almost 8000 tons DM of solid pellets per year (SIAAP source) which are stocked in big bags or silos before to be reused in agriculture. More information about WWTPs and treatment processes are presented in Supporting material – Table 1 and on SIAAP website (www.siaap.fr – in French).

Different sampling points have been defined to study both the quality of treated sludges and the fate of micropollutants during treatments: raw sludge (RS), CS, DS, TS and SC – Fig. 1. Thus, inlet and outlet of digestion, centrifugation and thermal drying were sampled, as well as SC.

While six independent campaigns (between October and December 2011) were performed for thermal drying and sludge cake, consecutive day sampling was considered for centrifugation and digestion to throw off the possible lack of homogeneity. Thus, one sample per day was collected within three consecutive days for digestion (October 2011). Similarly, six samples were collected within two periods of three consecutive days (one in October and one in December) for centrifugation. Each sludge sample was manually collected (2 L for TS and SC – 3 L for RS, CS and DS) respecting all guidelines to avoid sample contamination. SC samples are a mix of sludge produced within a week (7 days) while other samples were punctual due to technical issues. For digestion, a period of 16 days has been applied between inlet and outlet samples to take the solid retention time into account.

2.2. General sludge quality parameters

Table 2 displays the general quality parameters for each sample, i.e. dry matter (DM, in % – 1% = 10 g/L) and volatile matter (VM, in % DM). Both criteria are commonly used in sludge management. Minimum, maximum and mean (in italics below) values are given.

Overall, removals of dry matter and volatile matter during anaerobic digestion are about 42% and 56% respectively. This removal is in good agreement with conventional anaerobic digestion removal (Moletta, 2008). VM content in sludge cake is low (42% DM) compared to the other sludges highlighting a removal during the sludge cake production process (thermal conditioning + press filtration, Fig. 1). This is most likely due to