

Contamination of various types of Parisian sewage sludge by a wide range of priority and emerging micropollutants

Romain MAILLER^{1,2}, Johnny GASPERI¹, Dominique PATUREAU³,
Emmanuelle VULLIET⁴, Steven DESHAYES^{1,5}, Sabrina GUERIN², Régis
MOILLERON¹, Ghassan CHEBBO¹, Vincent ROCHER²

¹LEESU, Université Paris-Est, Créteil

²SIAAP, DDP, Colombes

³LBE, INRA, Narbonne

⁴ISA, CNRS, Villeurbanne

⁵LCPP, Paris



Context of the study



- Hydrophobic and adsorbable compounds removed from wastewater by sorption to sludges (Mailler *et al.* 2013; Choubert *et al.* 2012; Clara *et al.* 2007)

➔ Presence of micropollutants in sewage sludge

- > 50% of treated sludge (DM) is landfilled in Europe (Kelessidis and Stasinakis 2012)
- European Directive about sludge management (86/278/CEE) only concerns some metals and PAHs

➔ What about others micropollutants ?



Context of the study

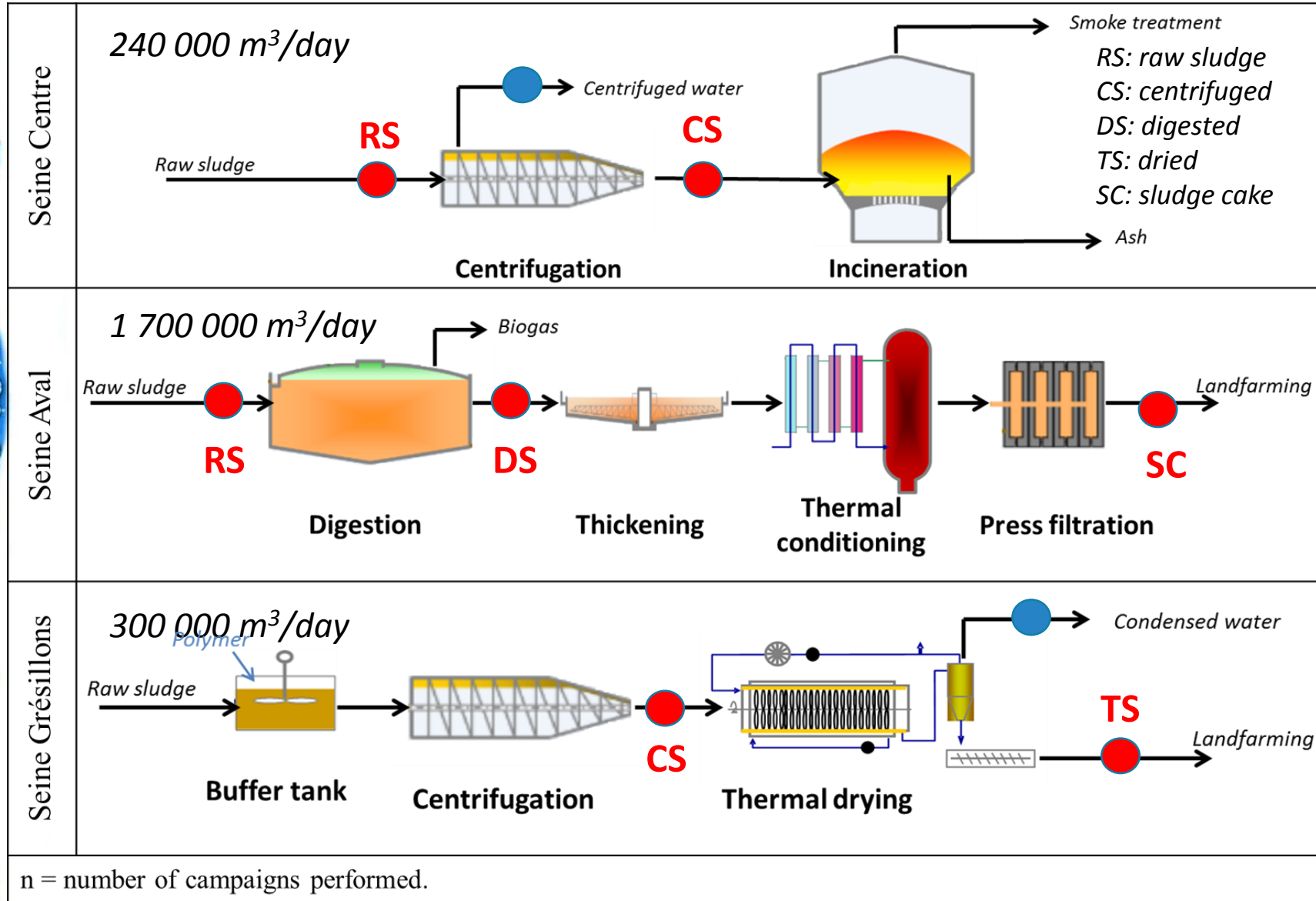


Scientific and operational issues:

- Contamination of Parisian sewage sludges
- Fate during sludge treatments (centrifugation, digestion, thermal drying, press filtration)
- What about centrifuged and condensed waters?



The studied WWTPs



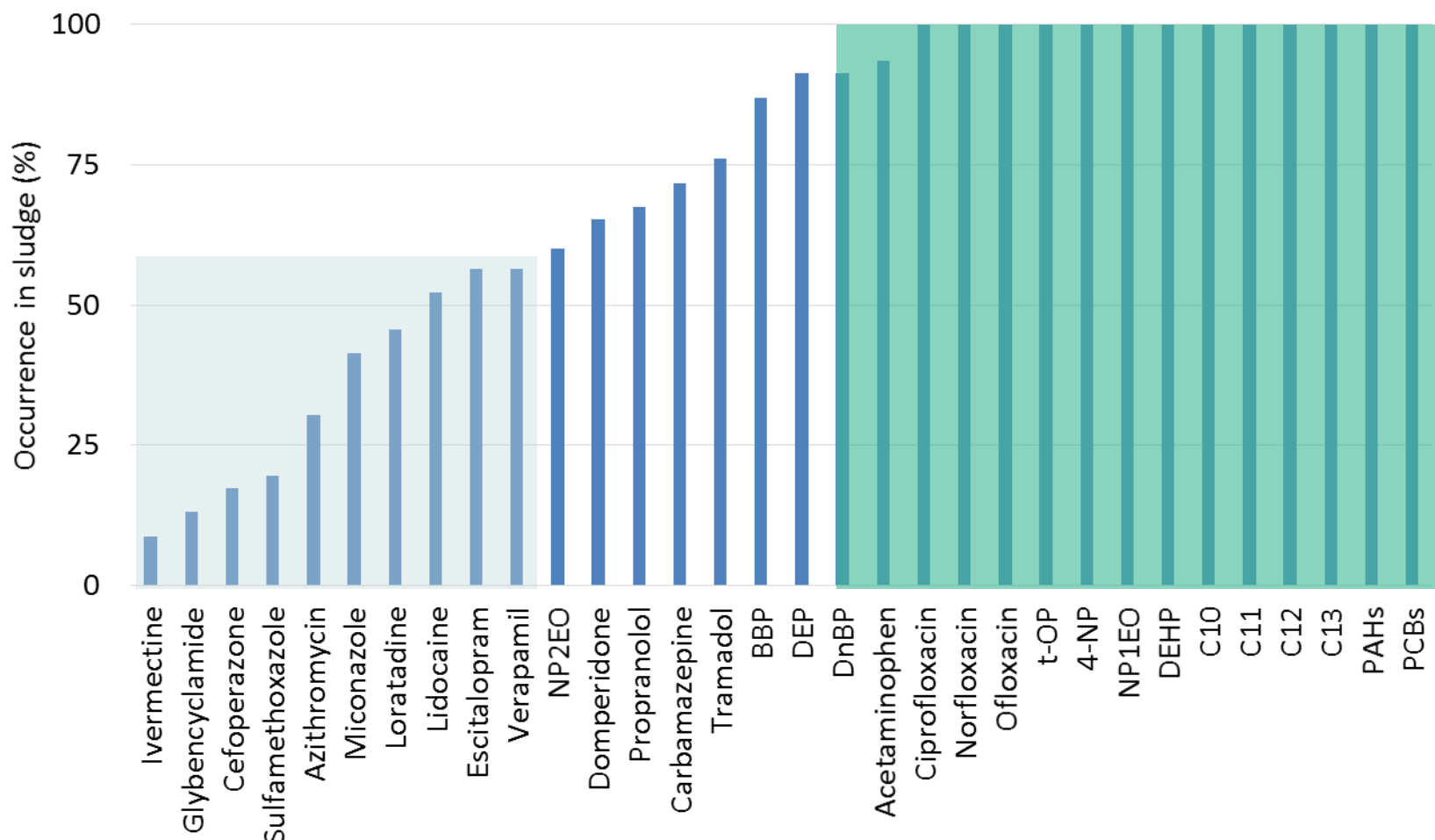
The analytical procedure

- 5 types of sludges: raw, centrifuged, digested, thermally dried and sludge cake
- 7 sampling campaigns (2013 – 2014)
- Punctual sampling
- 71 priority and emerging pollutants:
 - Pharmaceuticals and hormones
 - Perfluorinated acids
 - Alkylphenols
 - LAS
 - PCBs and PAHs
 - Phthalates
- 5 laboratories involved



Results of the study

Occurrence of emerging pollutants in sludge

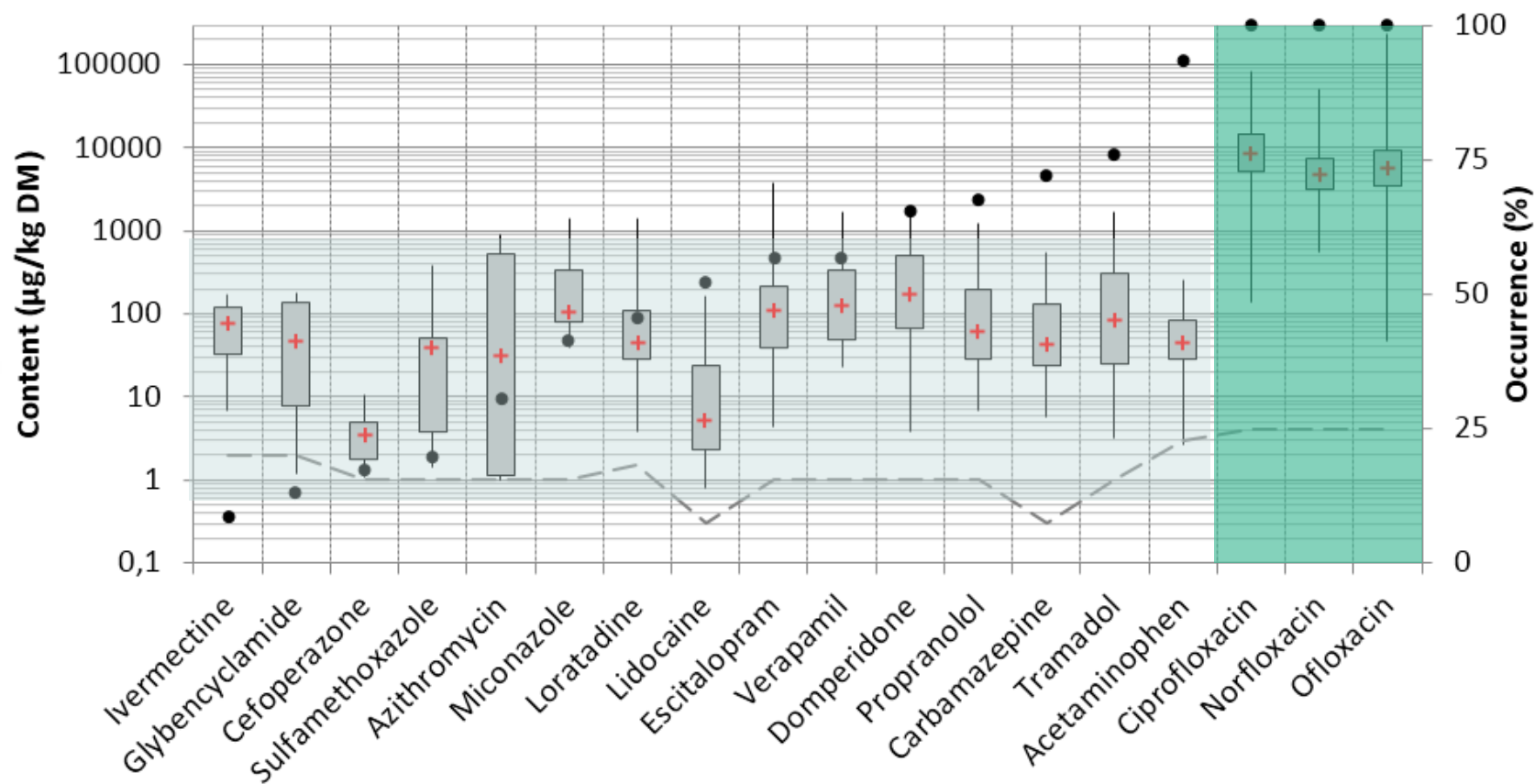


< 50% for some pharmaceuticals

> 75% for fluoroquinolones, phthalates, alkylphenols, LAS, PAHs and PCBs

Results of the study

Contamination of sewage sludge (1/2)

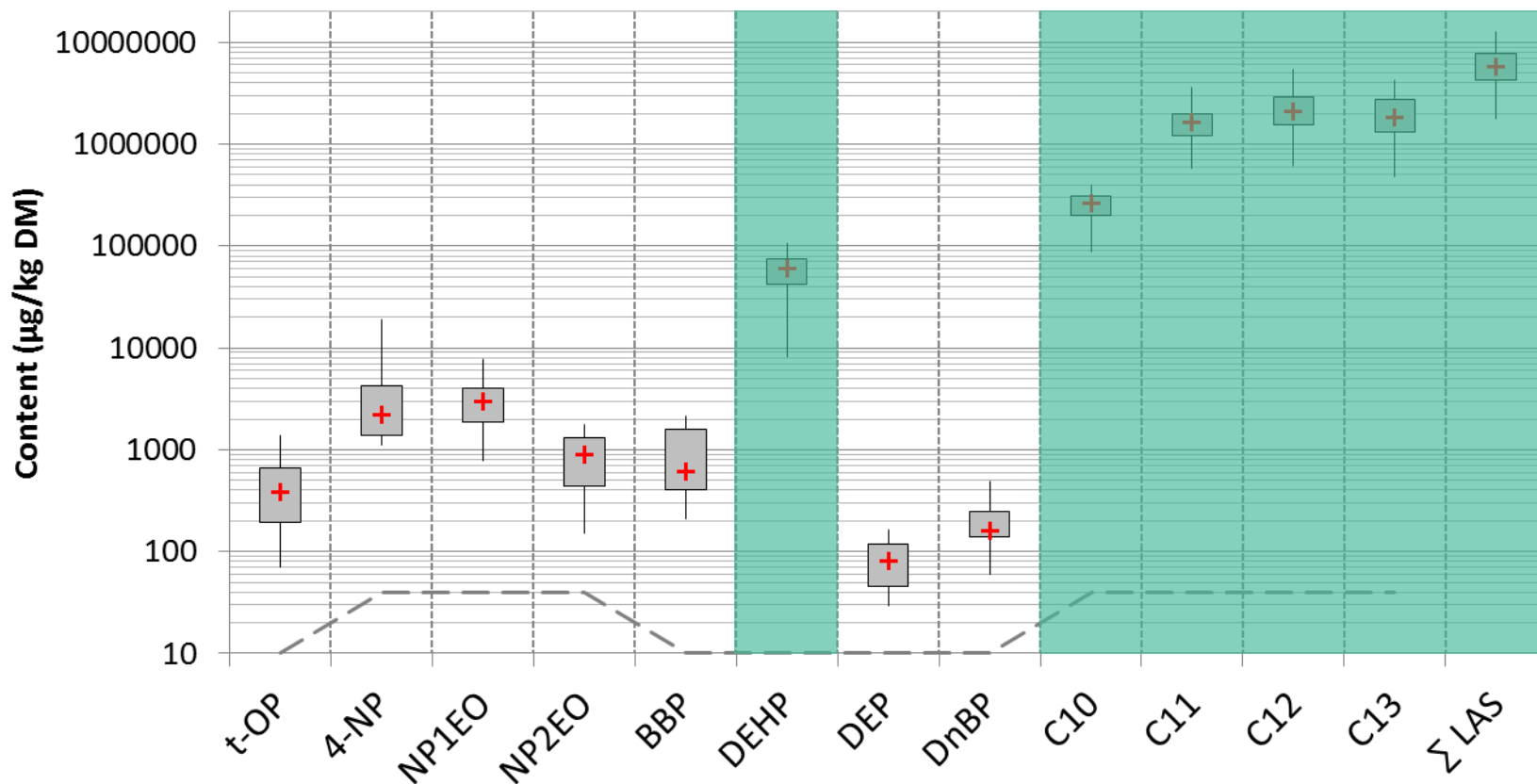


For most pharmaceuticals
10 – 500 µg/kg DM

For fluoroquinolones
up to 200 mg/kg DM

Results of the study

Contamination of sewage sludge (2/2)

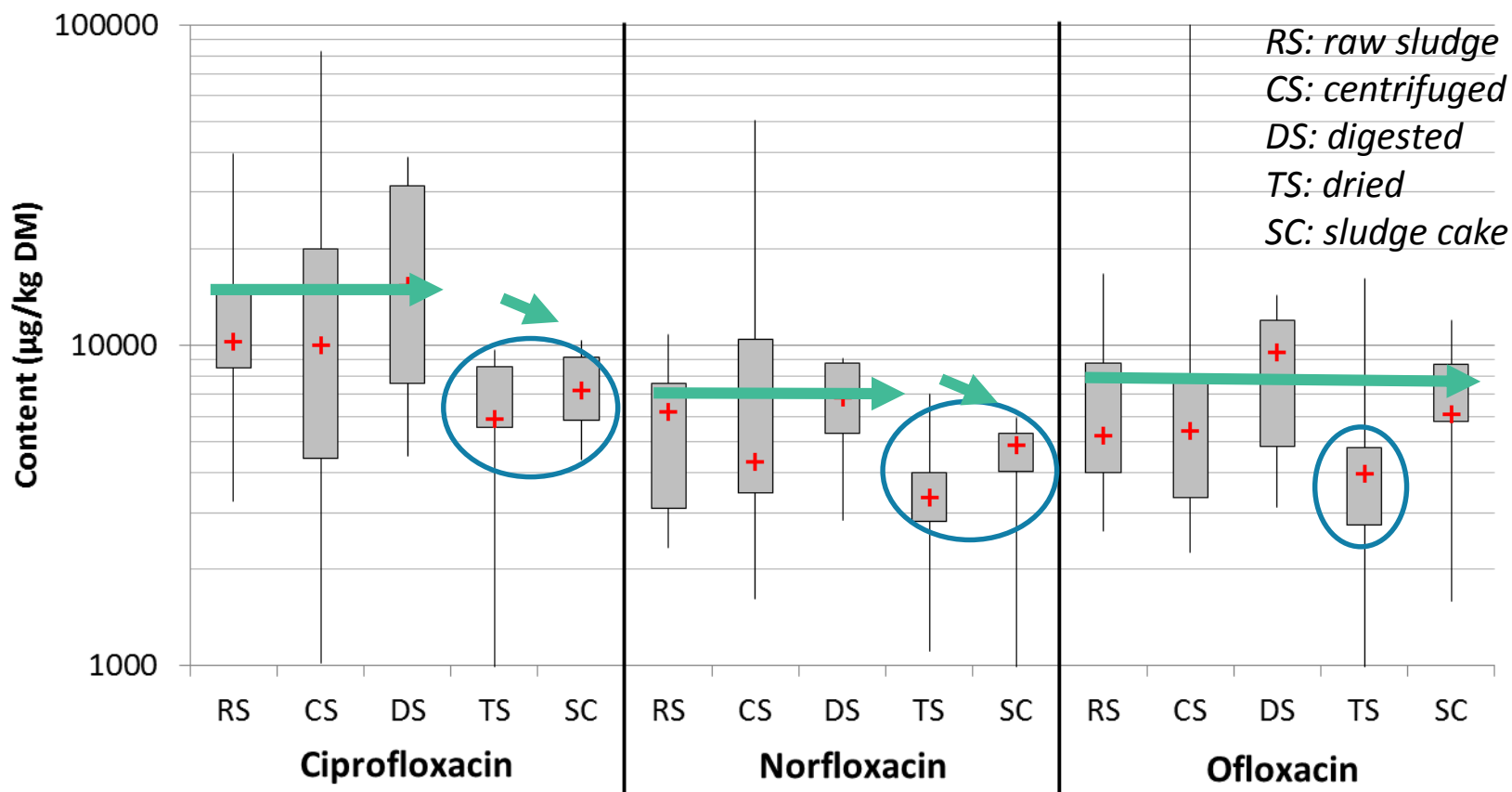


LAS > DEHP ≈ Fluoroquinolones > Alkylphenols ≈
Phthalates > Pharmaceuticals ≈ PAHs ≈ PCBs ≈
perfluorinated acids ≈ hormones

**Very high contents
for DEHP and LAS
LAS up to 5 g/kgDM
!**

Results of the study

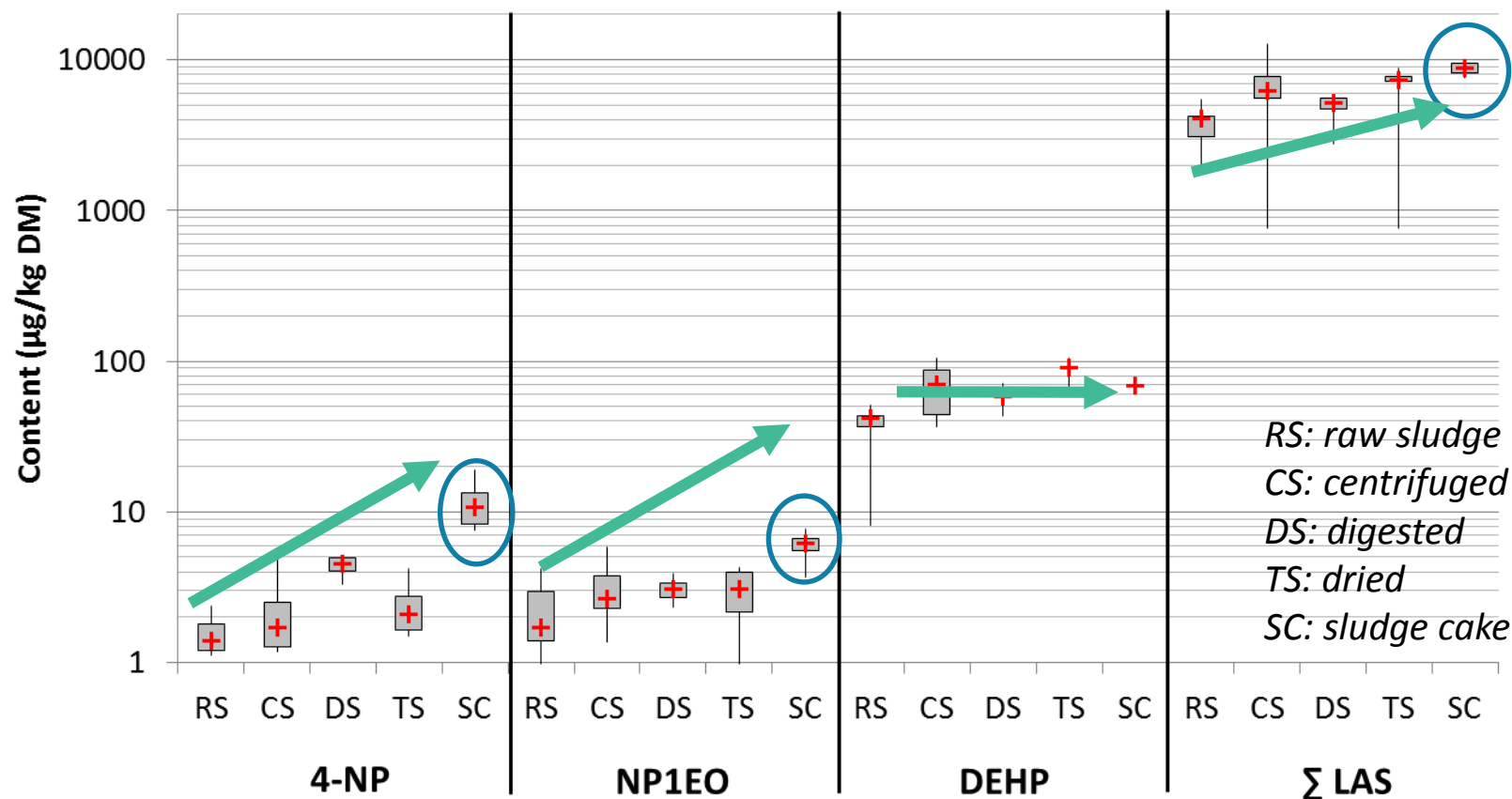
Evolution during sludge treatment (1/4)



Decrease/stability of contents in sludge + decrease of the mass flow
➔ Decrease of the fluoroquinolone fluxes by sludge treatments

Results of the study

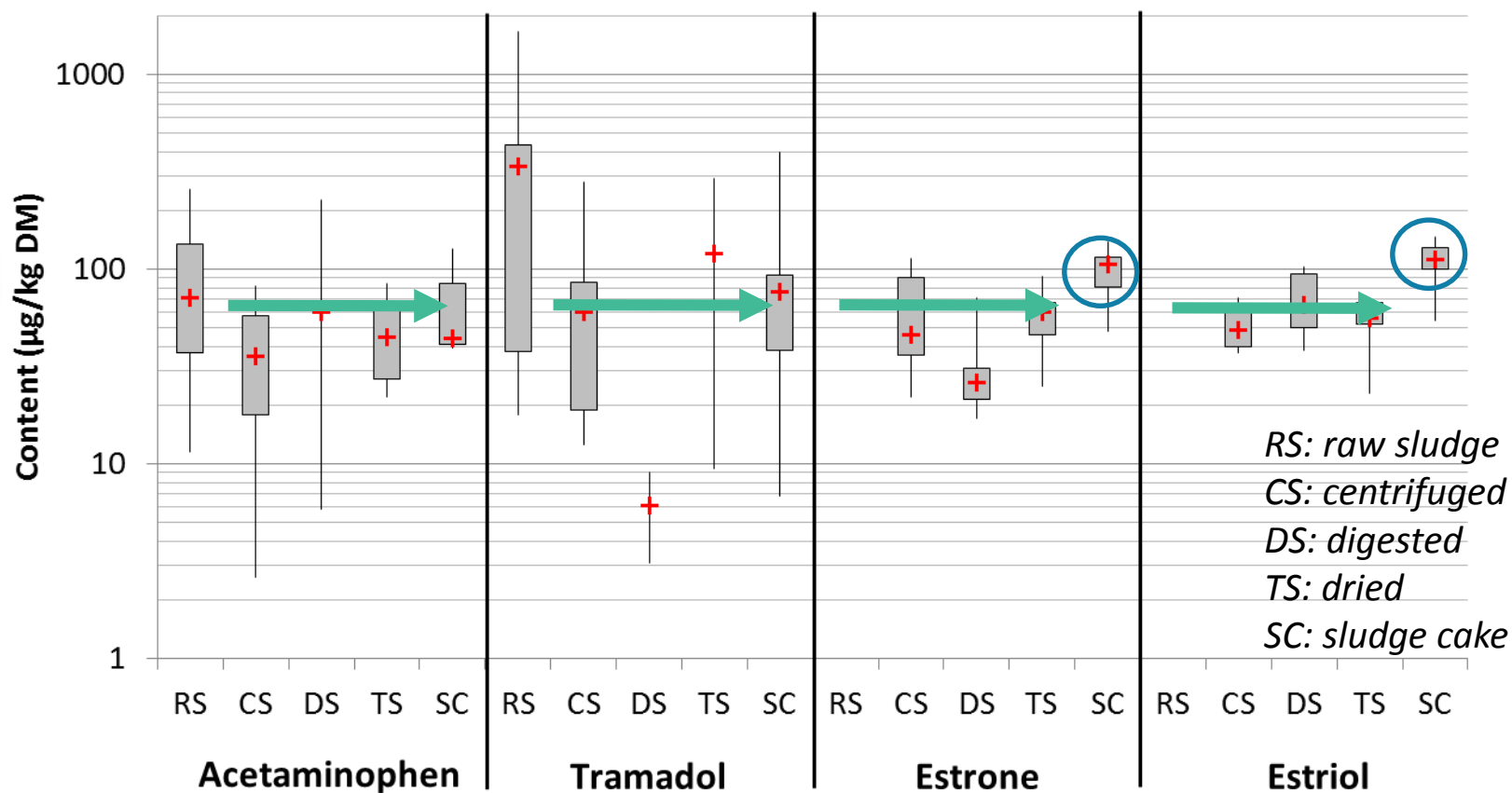
Evolution during sludge treatment (2/4)



Increase of contents in sludge + decrease of the mass flow
→ assesement of pollutant fluxes (next step)

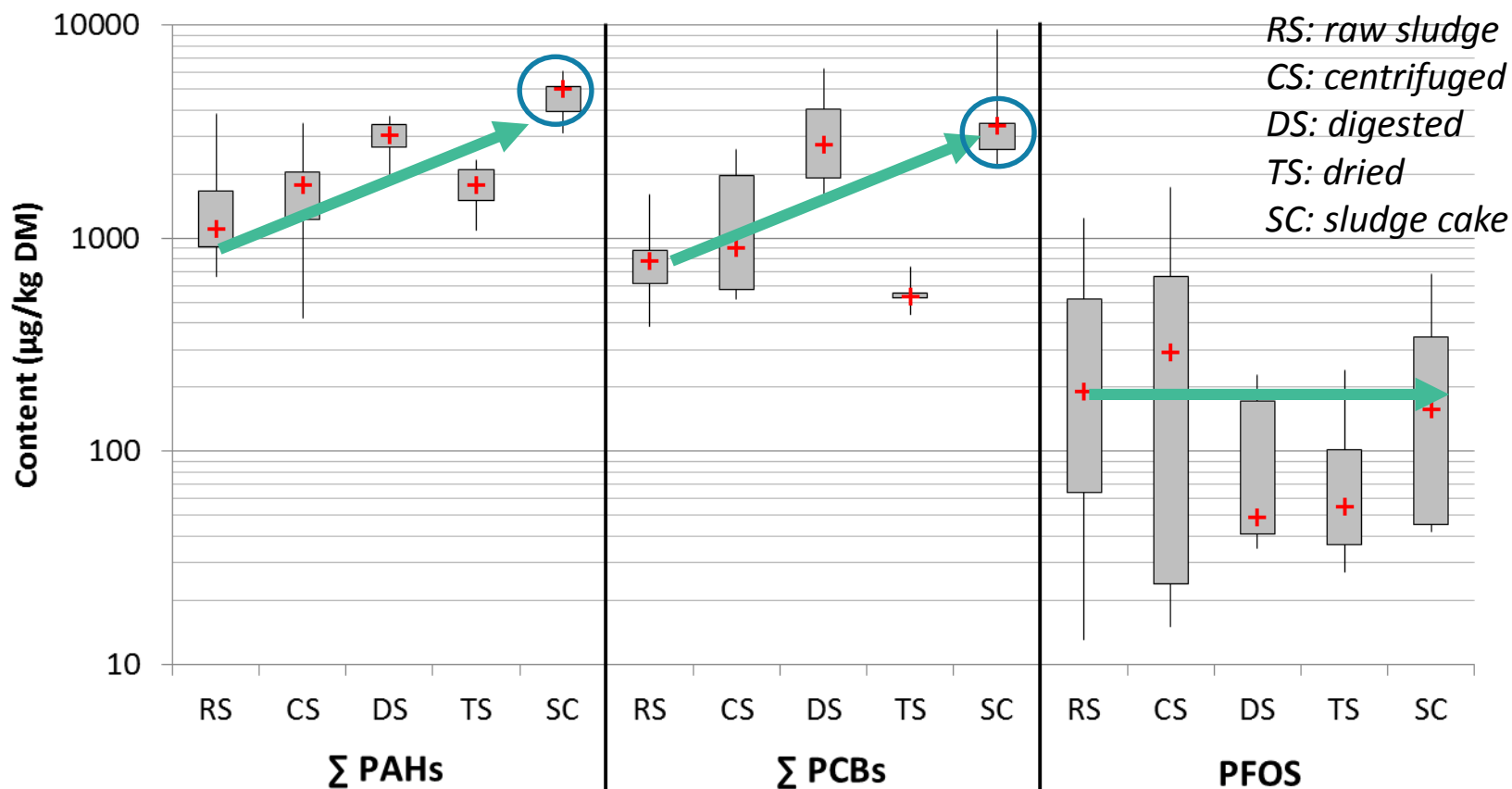
Results of the study

Evolution during sludge treatment (3/4)



Results of the study

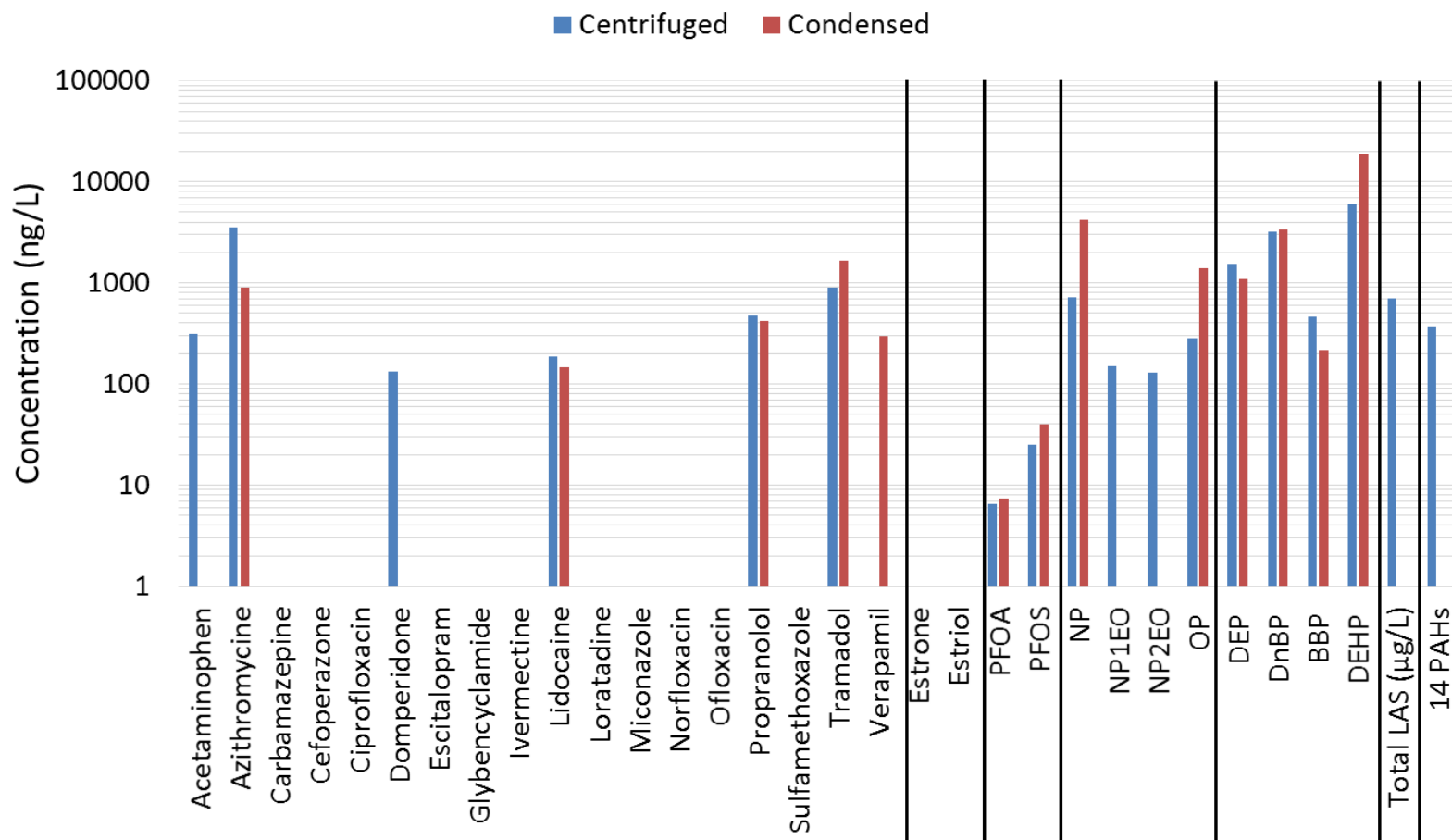
Evolution during sludge treatment (4/4)



Results of the study

The issue of centrifuged and condensed waters

- Centrifugation and thermal drying remove water from sludge and water reinjected to WWTP influents



Average of 3 campaigns



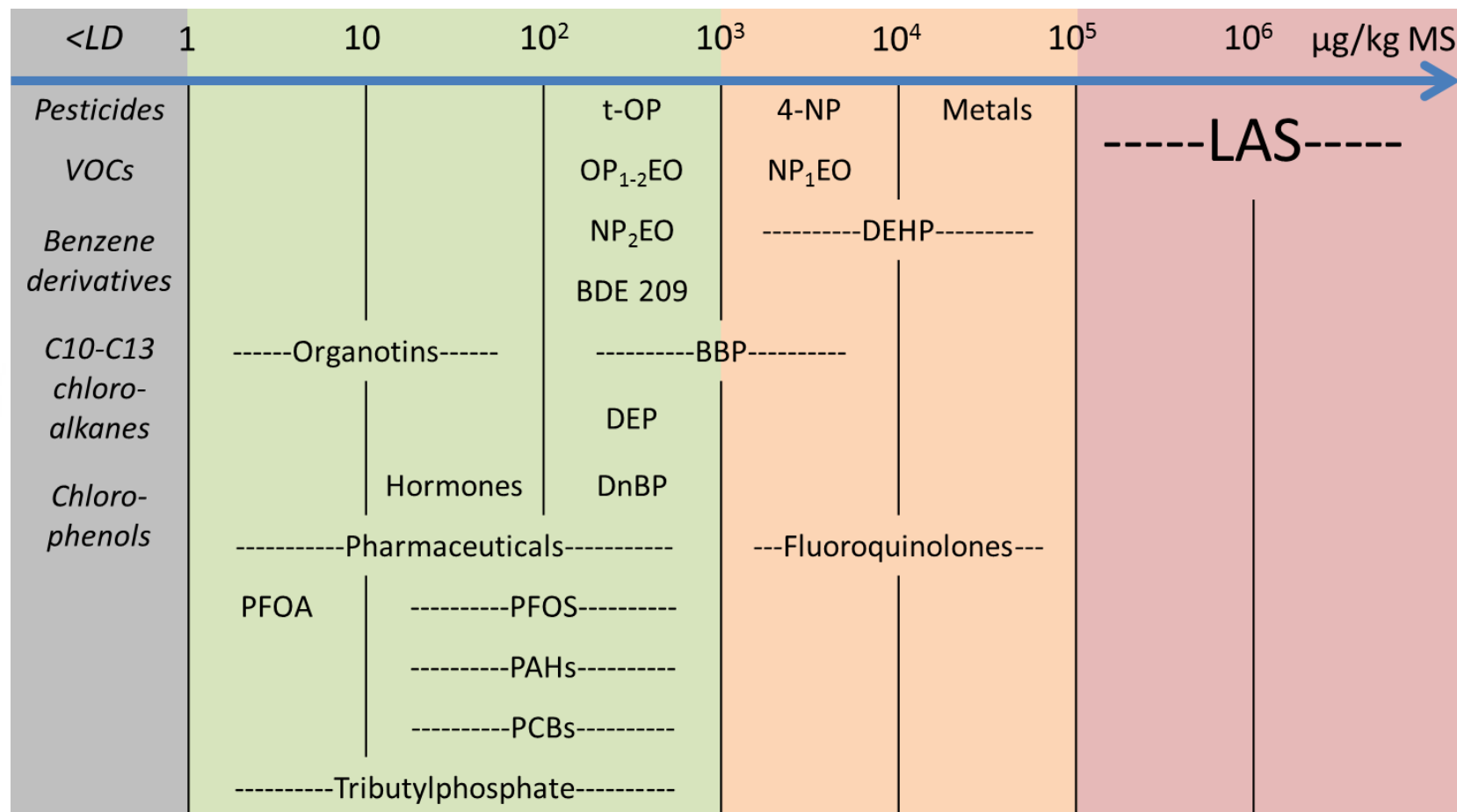
Conclusions

What is the contamination of Parisian sewage sludge by priority and emerging pollutants?

- High occurrence (> 75%) for LAS, phthalates, fluoroquinolones, PAHs, PCBs, alkylphenols, acetaminophen and tramadol
- LAS > DEHP \approx Fluoroquinolones > Alkylphenols \approx Phthalates > Pharmaceuticals \approx PAHs \approx PCBs \approx perfluorinated acids \approx hormones
- The micropollutant pattern is relatively stable in all types of sludge

Conclusions

Emerging pollutants pattern of Parisian sewage sludges



Mailler *et al.* 2014 – Priority and emerging pollutants in sewage sludge and fate during sludge treatment. *Waste Management*, 34, 1217-26

Mailler *et al.* 2015 – Fate of emerging and priority micropollutants during the sewage sludge treatments: case study of Paris conurbation. *Waste Management*, submitted.



Conclusions

What is the contamination of Parisian sewage sludge by priority and emerging pollutants?

- Increase of content during treatment for PCBs, PAHs, LAS, DEHP, hormones and alkylphenols → sludge cakes are the most concentrated
- This trend is not observed for pharmaceuticals and perfluorinated acids
- Presence of most micropollutants in centrifuged and condensed waters → transfer from sludge to water

Perspectives

This study

- Calculation of fluxes by pollutants and by process,
- Removal of micropollutants by anaerobic digestion and thermal drying,
- Mass balances by process and by WWTP,

→ Publication in preparation

Future study

- What about centrifuged and condensed waters treatment?
- What about emerging pollutants with growing concern: PCPs, artificial sweeteners, benzotriazole, Iodinated X-ray agents, surfactants, etc.?

Thank you for your attention !

romain.mailler@siaap.fr

gasperi@u-pec.fr



Mailler et al. 2014 – Priority and emerging pollutants in sewage sludge and fate during sludge treatment. *Waste Management*, 34, 1217-26

Mailler et al. 2015 – Fate of emerging and priority micropollutants during the sewage sludge treatments: case study of Paris conurbation. *Waste Management*, submitted.



Supplementary material

Flux removals observed in this study (1/2)

	Digestion	Thermal drying
Acetaminophen	68 ± 35%	21 ± 25%
Carbamazepine	66 ± 39%	44 ± 40%
Propranolol	88 ± 25%	
Sulfamethoxazole	76 ± 33%	
Lidocaine	90 ± 21%	
Tramadol	98 ± 4%	-48 ± 100%
Domperidone	99 ± 1%	
Ciprofloxacin	43 ± 17%	51 ± 36%
Norfloxacin	43 ± 36%	32 ± 37%
Ofloxacin	49 ± 28%	35 ± 43%
Estrone		
Estriol		

Removal of DM by digestion = 40 ± 6%

Removal of VM by digestion = 54 ± 5%

Supplementary material

Flux removals observed in this study (2/2)

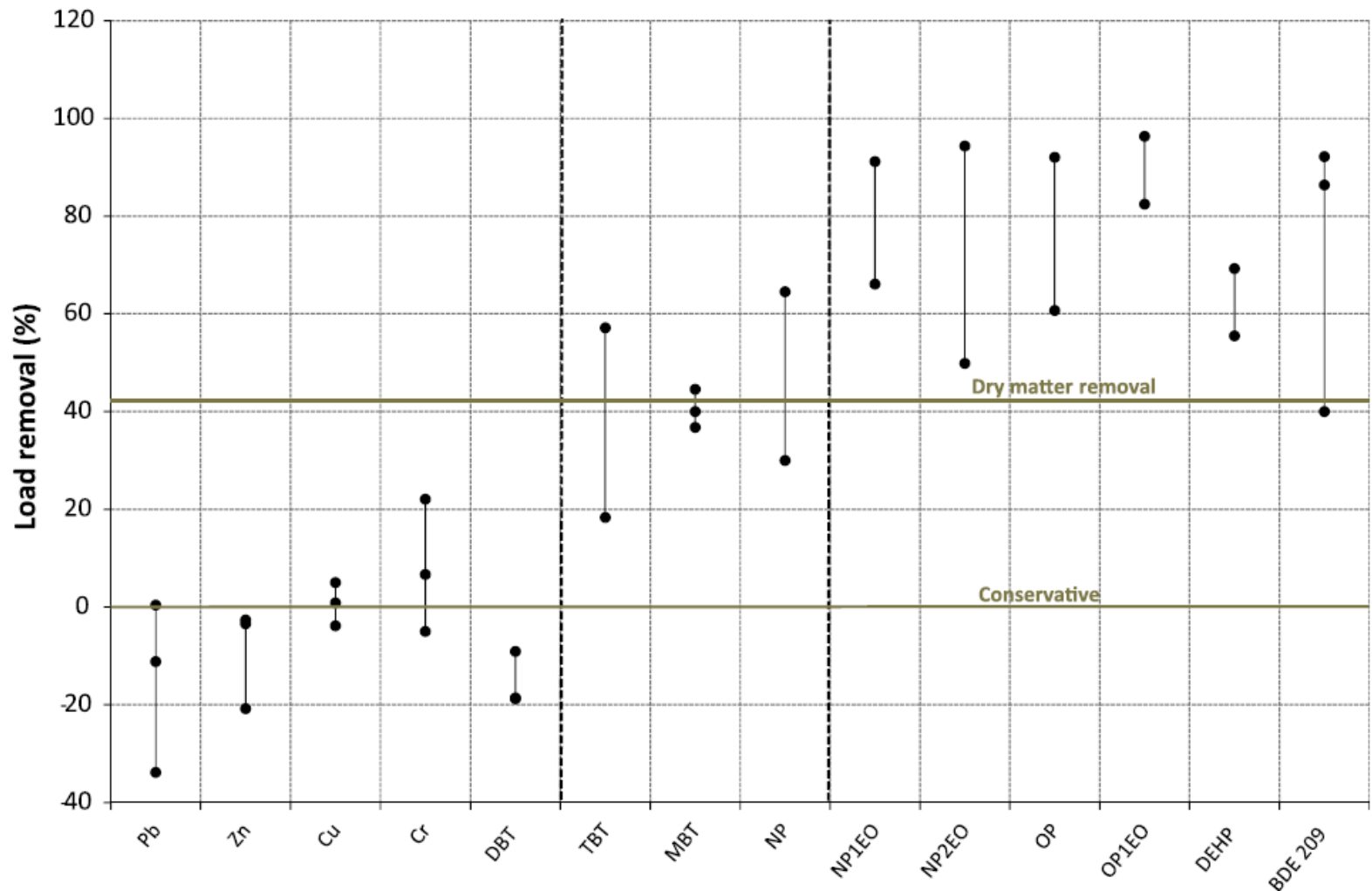
	Digestion	Thermal drying
PFOA	57 ± 8%	93 ± 7%
PFOS	77 ± 8%	45 ± 54%
Σ LAS	15 ± 22%	15 ± 19%
Σ 14 PAHs	-9 ± 23%	16 ± 13%
Σ 20 PCBs	11 ± 27%	26 ± 31%
4-NP	-54 ± 36%	22 ± 10%
NP ₂ EO	32 ± 19%	38 ± 29%
t-OP	-23 ± 37%	43 ± 12%
DEHP	33 ± 42%	-140 ± 108%
BBP	43 ± 14%	-142 ± 71%

Removal of DM by digestion = 40 ± 6%

Removal of VM by digestion = 54 ± 5%

Supplementary material

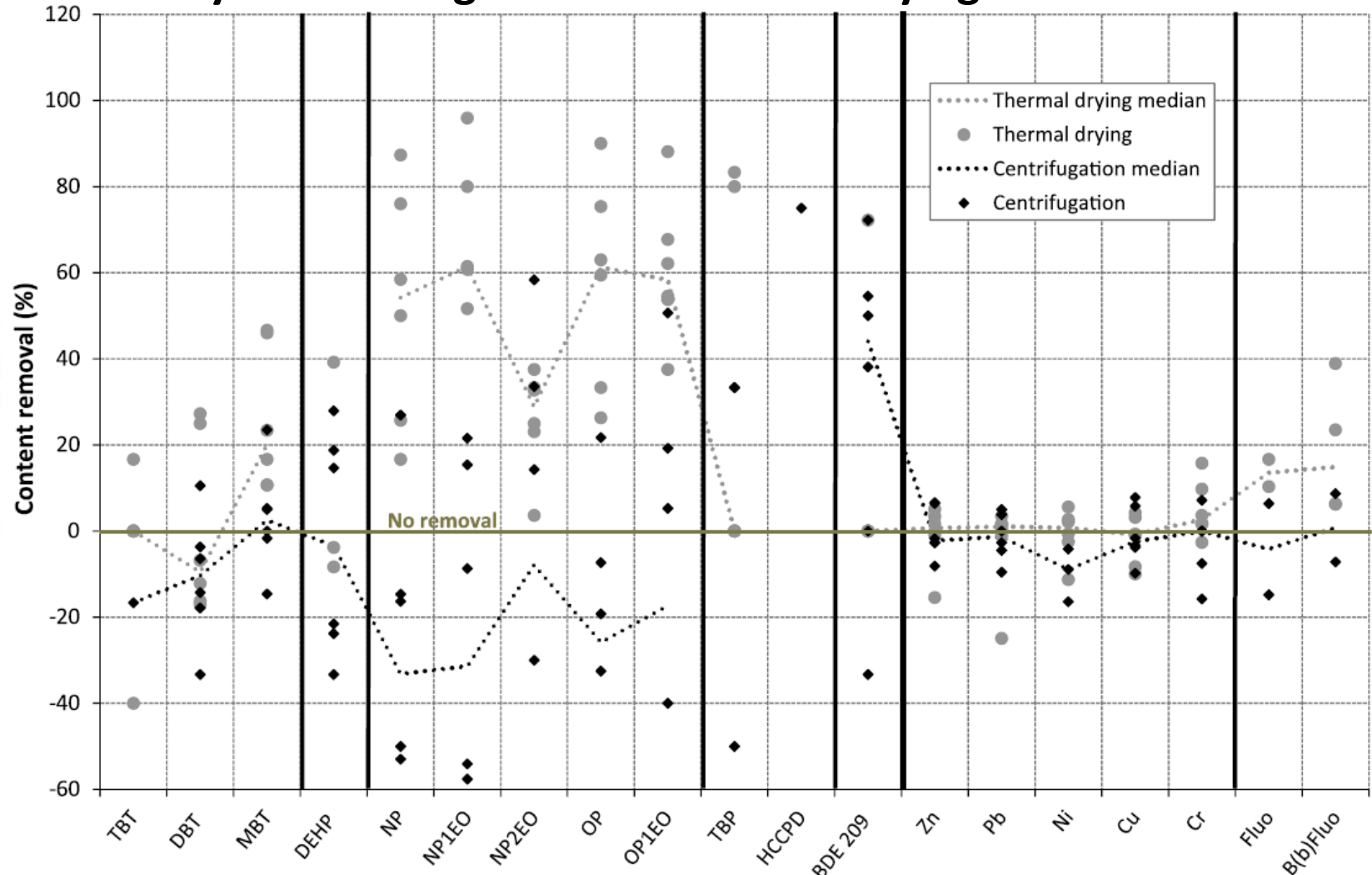
Efficiency of anaerobic digestion



Mailler *et al.* 2014 – Priority and emerging pollutants in sewage sludge and fate during sludge treatment. Waste Management, 34, 1217-26

Supplementary material

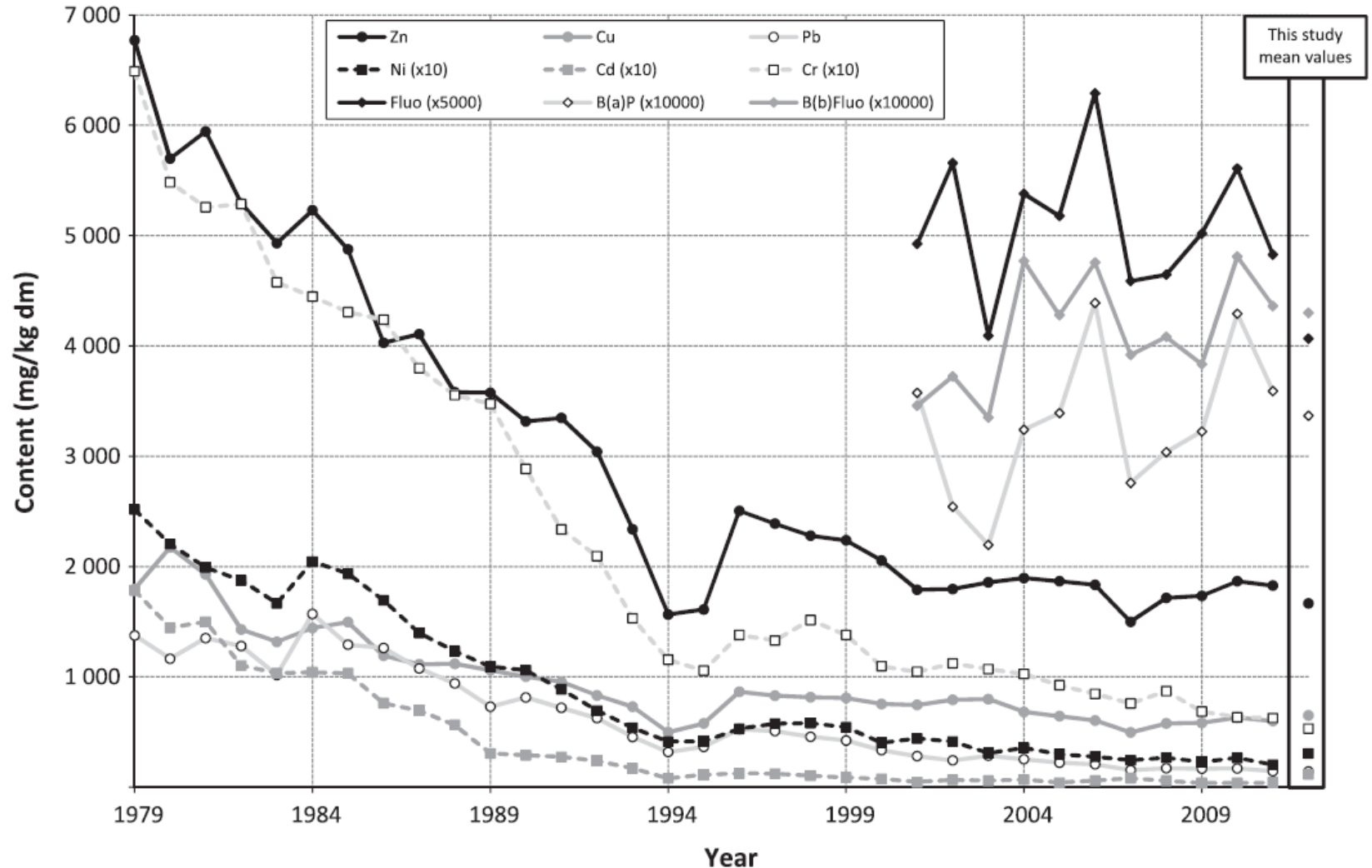
Efficiency of centrifugation and thermal drying



Mailler *et al.* 2014 – Priority and emerging pollutants in sewage sludge and fate during sludge treatment. Waste Management, 34, 1217-26

Supplementary material

Historical evolution of sludge cakes contamination



Mailler *et al.* 2014 – Priority and emerging pollutants in sewage sludge and fate during sludge treatment. Waste Management, 34, 1217-26