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## **Process type is the key driver of the fate of organic micropollutants during industrial scale treatment of organic wastes**

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1 **Process type is the key driver of the fate of organic micropollutants during industrial scale**  
2 **treatment of organic wastes**

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8 **Highlights**

- 9 • New organic micropollutant-organic waste combinations assessed on treatment lines  
10 • Fate driven by process type and compound specific behaviour  
11 • Biological processes were more efficient than physicochemical ones  
12 • Process combination enhanced most organic micropollutants dissipation

13 **Abstract**

14 Organic micropollutants (OMPs) such as polycyclic aromatic hydrocarbons, nonylphenols and  
15 pharmaceuticals products are ubiquitous in organic wastes generated by most human activities.  
16 Those wastes are mainly recycled by land spreading, most often after treatments, such as liming,  
17 dewatering, composting or anaerobic digestion. It has been shown essentially at lab scales that  
18 biological treatments have an effect on the removal of some OMPs. However, less is known on the  
19 role of each step of industrial treatment lines combining physico-chemical and biological treatments  
20 on the OMP-fate and removal. The present study focuses on the impact of waste treatment on the  
21 fate of 53 OMPs along 10 industrial treatment lines treating urban, agricultural wastes or mixtures.  
22 The combination of studying a diversity of organic wastes and of OMPs with different characteristics  
23 (solubility, ionic charges, hydrophobicity etc.), sampling *in situ* industrial sites, quantifying native  
24 OMP concentrations and looking at each step of complete treatment lines allows for a global and  
25 representative view of the OMP fate in the French organic waste treatment sector. Less studied  
26 wastes, i.e. territorial mixtures, revealed intermediate OMP contents and compositions, between

27 urban and agricultural wastes. Dewatering and liming, usually dismissed, had a noticeable effect on  
28 concentrations. Anaerobic digestion and composting had significant effects on the removal of all  
29 pollutant families. Combination of processes enhanced most OMP dissipation. Here we showed for  
30 the first time that the process type rather than the waste origin affects dissipation of organic  
31 micropollutants. Such data could be used to build and validate dynamic models for the fate of OMPs  
32 on solid waste treatment plants.

33

## 34 **Keywords**

35 Anaerobic digestion, composting, dewatering, liming, organic micropollutants, reduction rates

36

37 **Abbreviation list:** ACTC, anhydrochlortetracycline; ANT, anthracene; ATC, anhydrotetracycline; BAA, benzo(a)anthracene; BAP, benzo(a)pyrene; BBF,  
38 benzo(b)fluoranthene; BKF, benzo(k)fluoranthene; BPE, benzo(g,h,i)perylene; CBZ, carbamazepine; CHR, chrysene; CIP, ciprofloxacin; CTC-ECTC,  
39 chlortetracycline and its epimere; DBA, di-benzo(a,h)anthracene; DIC, diclofenac; DM, dry matter; DOX, doxycycline; ENR, enrofloxacin; ERY, erythromycin;  
40 FLO, fluoranthene; FLO, flumequine; FLU, fluorene; FLX, fluoxetine; GEM, gemfibrozil; IBU, ibuprofen; IM, inorganic matter; IND, indeno(1,2,3-cd)pyrene; LOD,  
41 limit of detection; LOQ, limit of quantification; MS, mass spectrometry; NOR, norfloxacin; NP, nonylphenol; NP1EO, nonylphenol monoethoxylate; NP2EO,  
42 nonylphenol diethoxylate; NPs, nonylphenols; OFL, ofloxacin; OMP, organic micropollutant; OTC, oxytetracycline; OW, organic waste; PAHs, polycyclic aromatic  
43 hydrocarbons; PHE, phenanthrene; PP, pharmaceutical product; PYR, pyrene; SPI, spiramycin; TC, tetracycline; TMP, trimethoprim; TRC, triclocarban; TRI,  
44 triclosan; TS, total solid; TYL, tylosin; VS, volatile solid.

45

## 46 **1. Introduction**

47 Organic micropollutants (OMPs) such as pharmaceutical products (PPs), polycyclic aromatic  
48 hydrocarbons (PAHs) and nonylphenols (NPs, coming from the transformation of detergents named  
49 nonylphenol ethoxylates), have been quantified at ppb to ppm concentration levels in the ever  
50 growing quantity of organic wastes (OWs) around the world, e.g. in sewage sludge (Clarke et al.,  
51 2011; Bergé et al., 2012; Verlicchi and Zambello, 2015), in manure (Youngquist et al., 2016; Wohde et  
52 al., 2016; Spielmeyer, 2018; Ghirardini et al., 2020) or territorial mixes (Brändli et al., 2007; Suominen  
53 et al., 2014). OMPs may come from direct human or animal excretion of PPs in waste after  
54 medication, atmospheric deposits, rain wash-off of hydrocarbons, domestic and industrial effluents  
55 loaded with surfactants, detergents etc.

56           Agricultural land application for fertilization is one of the main ways to dispose of those  
57 wastes (Mininni et al., 2015), directly introducing potentially threatening OMPs into soil, that could  
58 be transferred in some cases to surface and/or ground waters. Before application, OWs may undergo  
59 biological treatments such as anaerobic digestion and composting or physico-chemical ones like  
60 dewatering or liming, to stabilise, sanitise the matter and concentrate useful nutrients. Combinations  
61 of these treatment processes are possible. Understanding the fate of OMPs during the OW  
62 treatments is important to evaluate and reduce the threat associated with OW land spreading.

63           All OW treatment processes have an effect on some of the OMPs present in wastes:  
64 anaerobic digestion and composting as biological treatment processes are known to dissipate or  
65 degrade PPs (Ezzariai et al., 2018; Wohde et al., 2016; Spielmeier 2018), nonylphenol ethoxylates  
66 and NP (Zheng et al., 2015; 2018) and PAHs (Brändli et al., 2007; Aemig et al., 2016). The effect of  
67 these treatment processes has been shown in lab and pilot scale experiments (Phan et al. (2018) for  
68 sludge anaerobic digestion; Spielmeier et al. (2015) for manure anaerobic digestion; Geng et al.  
69 (2016) for sludge drying and liming) and in some studies where *in situ* sampling was performed on  
70 industrial sites (Lillenberget al. (2010) for sludge composting; Martin et al. (2015) for sludge  
71 anaerobic digestion and composting; Mailler et al. (2017) for sludge anaerobic digestion), each time  
72 for a small selection of micropollutants. The impact of OW origin and type on the fate of OMPs  
73 during the treatment processes has been poorly documented especially for some OMP families and  
74 for wastes coming from territorial sources, mostly comprising mixes of wastes. To the best of our  
75 knowledge, a wide *in situ* study on OMP fate during OW treatment with unified quantification  
76 methods to identify global trends has not been tried before.

77           This study focuses on the impact of OW treatment on the fate of 53 OMPs along 10 industrial  
78 treatment lines treating urban, agricultural wastes or mixture of both OW types. The combination of  
79 studying a diversity of OWs, a diversity of OMPs with different characteristics (solubility, ionic  
80 charges, hydrophobicity etc.), sampling *in situ* industrial sites, quantifying native OMP concentrations

81 and looking at complete treatment lines allows for a global and representative view of the OMP fate  
82 in the French OW treatment sector.

## 83 **2. Material and Methods**

### 84 **2.1. Organic micropollutants, sites and organic waste treatment processes**

85 Thirty-five pharmaceutical products, 2 biocides, 3 nonylphenols and 13 polycyclic aromatic  
86 hydrocarbons (Table S1; Supplementary Information, SI) with diverse chemical properties in terms of  
87 sorption and transformation (Table S2, SI) were selected to highlight contrasted fate during  
88 treatment processes. Due to the study of various organic wastes, OMPs were also selected based on  
89 their frequent (co)occurrence in sludge, manure, food waste, green waste if data is available.

90 Ten sites located in different parts of France and in La Reunion Island were selected for their  
91 processes and their input feeding waste diversity from agricultural, territorial or urban origins. Sites  
92 were representative of most common types of organic wastes and treatment processes in France, i.e.  
93 anaerobic digestion, composting, drying and liming (Table 1 and Figure S1). Agricultural sites (Agri I-  
94 IV, Table 1) included two farm-size solid batch anaerobic digestion units treating bovine manure, one  
95 continuous liquid anaerobic digestion unit only treating pig manure and one experimental research  
96 site directly composting cow manure. Territorial sites (Terri I-III) were industrial continuous liquid  
97 anaerobic digestion units treating mixtures of agro-industrial waste, liquid pig manure, sewage  
98 sludge, cereal, food waste etc. Urban sites (Urb I-III) were wastewater treatment plants processing  
99 sludge by either anaerobic digestion in mesophilic or thermophilic conditions, composting or both  
100 treatments. A single substrate (the waste undergoing the treatment) could undergo a series of  
101 treatment processes (Process continuity, Table 1 and Figure S1). Samples were taken the same day  
102 along the waste treatment line, before and after each treatment step including drying and phase  
103 separation. For heterogeneous solid matrices (e.g. manure, compost), up to ten sub-samples from  
104 different places were gathered to obtain a composite and representative sample of 5 to 8 kg (dry  
105 mass). For more homogenous liquid samples, 3 to 15 L, depending on total solid content, were

106 sampled in stirred tanks to get a representative sample. All samples were stored at -20°C on sampling  
107 day or the day after.

108

## 109 **2.2. Analytical methods**

110 Dry matter (DM) and volatile solid (VS) contents were measured in duplicates by mass  
111 difference after 24h at 105°C and after additional 2h at 550°C respectively. The remaining ashes were  
112 considered as inorganic matter (IM).

113

### 114 **2.2.1. PAHs and NPs**

115 Raw samples were frozen at -80°C then freeze-dried for a week and grinded (Table S1, SI).  
116 PAHs and NPs were extracted from one gram subsample according to Trably et al. (2004) and Mailler  
117 et al. (2017). Control matrices with known concentration of NPs/PAHs were extracted in parallel.  
118 Solvent extracts were then evaporated and dissolved in either 100% acetonitrile for PAHs or 100%  
119 hexane for NPs. Each extract was analysed with a Waters Alliance 2695 High-Performance-Liquid-  
120 Chromatography with a fluorescence detector. Conditions of separation and detection are specified  
121 in Table S1 (SI). Each sample was extracted twice and each extract quantified twice, giving a total of 4  
122 values. Limits of quantification (LOQs) were estimated from the lowest points in calibration curves: 5  
123 µg/L for PAHs, 100 µg/L for NPs in extracts.

124

### 125 **2.3.2. Pharmaceutical products**

126 All samples were analysed in triplicates. Briefly, solid samples were spiked with a mixture of  
127 internal deuterated-labelled standards, left overnight and extracted as described in Table S1  
128 according to Ferhi et al. (2016). The supernatants were purified after centrifugation with a modified  
129 QuEChERS method (Ferhi et al., 2016). Liquid samples (DM <20%) were centrifuged at 18600 g for 20  
130 min. The obtained solid phase was extracted as mentioned above. The liquid phase was spiked with a  
131 mixture of internal deuterated-labelled standards and purified using the modified QuEChERS method

132 (Bourdat-Deschamps et al., 2014). Pharmaceuticals were analysed with online Solid-Phase-Extraction  
133 and Ultra-High-Performance-Liquid-Chromatography coupled with a triple quadrupole mass  
134 spectrometer (Table S1). The compound concentrations were determined by internal quantification-  
135 isotope dilution in order to correct both extraction/purification losses and potential matrix effects in  
136 mass spectrometry (MS). For this study, 23 compounds were added to the published analytical  
137 method, with MS-MS transitions detailed in Table S3 (SI); trueness recoveries and intra-day precision  
138 are detailed in Table S4 (SI). LOQs for each compound and each extracted matrix were estimated as  
139 the concentration corresponding to a signal-to-noise ratio of ten. Limits of detection (LODs) were  
140 considered equal to LOQ/3. Concentrations between LOD and LOQ values were taken equal to LOQ/2  
141 for statistical analysis. For liquid samples, only the concentrations in the solid fraction were  
142 considered since PP concentrations in liquid phases were often very low or below LOQ or LOD values.

143

## 144 **2.4. Data treatment and analysis**

### 145 **2.4.1. Mean concentrations**

146 Micropollutant concentrations were first expressed in  $\mu\text{g}/\text{kg DM}$ . Results were considered to  
147 follow a normal distribution and the probability of having more than one outlier to be very low with  
148 only 3 or 4 values. Outliers were identified with a two tailed Dixon's Q test (Dean and Dixon, 1951)  
149 with a 95% confidence threshold using Rorabacher et al (1991)'s modifications (for 3 and 4 values,  $Q=$   
150 0.97 and 0.829 respectively) and discarded before mean and standard deviation calculations.

151

### 152 **2.4.2. Correction of output pollutant concentrations**

153 Changes in OMP concentration along a given process were characterised using IM as  
154 comparing point (Zheng et al., 2018), since IM was considered stable along the studied processes  
155 whereas organic matter could evolve during some processes. Output OMP concentrations were  
156 adjusted as follows.

157 
$$[OMP]_{out, corr} = \left( \frac{[OMP]_{out, m}}{IM_{out}} \right) \times IM_{in}$$

158 where  $[OMP]_{out, corr}$ , is the corrected OMP concentration,  $[OMP]_{out, m}$  the measured concentration,  
159  $IM_{in}$  and  $IM_{out}$  the % of IM in input and output respectively. This correction was not applicable for  
160 dewatering processes due to the possible loss of IM in the liquid fractions (not quantified). However,  
161 dissipation rates (section 2.4.3.) were calculated for these dewatering processes assuming a loss of  
162 low amounts of IM. The results on dewatering processes were only compared between themselves  
163 to highlight the partition behaviour of OMPs.

164

### 165 **2.4.3. Pollutant dissipation rates and heat map**

166 For each OMP and process, the input and corrected output concentrations were compared  
167 using a two-sample unequal variance (heteroscedastic) Student's T test, with a two tail distribution  
168 computed with Excel. When different (p-value < 0.05), dissipation rates could be calculated.

169 
$$Dissipation\ rate\ (\%) = \left( \frac{[OMP]_{in} - [OMP]_{out, corr}}{[OMP]_{in}} \right) \times 100$$

170 Four classes of dissipation rates were considered in a heat map gathering all results: no  
171 significant change, up to 30%, 30-70% and over 70%. The "+" sign indicated a higher OMP  
172 concentration in the output, "nc" a non-calculable rate with non-detectable contents or detectable  
173 with no available LOQ value in the output and the input, "\*" the absence of data.

174 For Urb II composting, OMP dissipation rate was calculated considering raw sludge (input)  
175 and final compost (output). Since the composting initial mixture (raw sludge, green waste) was not  
176 sampled, its characteristics were assessed using DM/VS ratio from a generic green waste and a raw  
177 sludge/green waste mass ratio of 33/66. For Urb III liming, the dissipation rate was established  
178 between dried digested sludge (input) and limed pellets (output), considering a 20% mass dilution  
179 through CaO addition.

180 Spiramycin (SPI) was not displayed in Figure 2, its values being only semi-quantitative.

181 The chosen sampling method, i.e. sampling the process input and output on the same day,  
182 without considering the process time (from hours and days for centrifugation, drying, liming to  
183 several days and weeks for anaerobic digestion and composting), implies that the resulting  
184 dissipation rates can be impacted by a temporal variability of the input or output OMP content. This  
185 is particularly true for the biological processes with long running time. There are very few data on the  
186 time variability of the OMP contents in OW treatments by comparison to the one provided for water  
187 treatment and its impact on OMP removal assessment (Verlicchi and Ghirardini, 2019). Mailler et al.  
188 (2017) provided variation coefficients of 50-150% for some OMP (mainly PP) concentrations in raw  
189 sludge sampled 2 times over 4 consecutive weeks at 2 wastewater treatment plants (WWTP). When  
190 considering only one WWTP, these coefficients were fully reduced; they were also lower for digested  
191 sludge and very low for persistent compounds (PAHs, NPs) whatever the type of sludge. This could be  
192 linked to the sludge age entering sludge processes (around 8-10 days depending on water treatment)  
193 and to the presence of storage tanks that are usually present to regulate loads of processes; sludge  
194 age and storage could play the role of buffer media. In the present study, the calculated dissipation  
195 rates will only be considered as apparent and not as formal ones. Furthermore, these rates are  
196 displayed as wide classes, lowering the impact of time variability and only general trends will be  
197 discussed according to process and OW types.

198

### 199 **3. Results and discussion**

200 As expected, all biological and dewatering processes were representative of efficient waste  
201 treatments, given the removal efficiencies of volatile matter (Table 1) and water (Table S5, SI). Urb III  
202 liming led to a 3 fold increase in Ca content following CaO addition.

203 Ten PPs out of 37 were never detected in all samples: 6 antibiotics (sulfamethazine,  
204 sulfamethoxazole, sulfamerazine, sulfachloropyridazine, n-acetyl sulfamethoxazole; tylosin B) and 4  
205 other PPs (ivermectin, eprinomectin, ketoprofene, acetamidophenol). Moreover, five PPs were

206 detected or quantified in a few OWs (marbofloxacin, sulfadiazine, lincomycin, levamisole and  
207 flunixin). The results for these PPs are not presented in this article.

208 In the following parts, the term “setting” is used to characterise the waste origin  
209 (Agricultural, Territorial or Urban) while “site” is used for a single location where several processes  
210 can take place. “Process” refers to only one organic waste treatment at a time, i.e. anaerobic  
211 digestion or composting.

212

### 213 **3.1. Micropollutants in process input substrate**

214 OMP input concentrations are displayed in Table S6 (SI) giving only values above LOQ. (OMP  
215 output concentrations are displayed in Table S7 (SI), as a comparing point).

216 Fluoroquinolones, macrolides and other PPs (including biocides) were more specific to Urb  
217 sites (Figure 1) than tetracyclines. PAHs and NPs were ubiquitous with detection occurrences above  
218 70% whatever the site type.

219

#### 220 **3.1.1. Antibiotics**

##### 221 **3.1.1.1. Quinolones**

222 Fluoroquinolones were weakly quantified in Agricultural (3/25) and Territorial (4/15) settings  
223 (Table S6, SI), in accordance with limited veterinary uses in France since March 2016. Only two  
224 compounds were quantified on Agri I site: a) Enrofloxacin (ENR) in anaerobic digestion substrate and  
225 resulting composting initial mixture and b) Ciprofloxacin (CIP) in anaerobic digestion substrate. ENR is  
226 only authorised for poultry, pets and pigs and CIP for humans. CIP is also a metabolite of ENR  
227 degradation (Martinez et al., 2006), explaining its lower concentration than those in other settings.  
228 Norfloxacin (NOR), Ofloxacin (OFL) and CIP, prescribed in human medicine and only to pet and  
229 equine animals in restricted conditions, were mainly quantified in samples with sewage sludge:  
230 nearly all the Urb anaerobic digestion and composting samples (up to several mg/kg DM) and the  
231 Terri II anaerobic digestion substrate (28% of sewage sludge, Table 1) with concentrations always

232 inferior, strengthening the idea of the sludge contribution to the total amount of these compounds in  
233 initial mixtures. Flumequine (FLQ) was only quantified in small quantities in two Urb composting  
234 substrates which is well correlated with its lower use in human medicine. As already reported (Clarke  
235 et al., 2011; Peysson and Vuillet, 2013; Mailler et al., 2016; Bourdat-Deschamps et al., 2017), the  
236 highest levels of fluoroquinolones and quinolones were quantified in human-related substrates.

237

### 238 **3.1.1.2. Tetracyclines**

239 Among all PPs, tetracyclines were the most abundant and frequently detected compounds in  
240 all settings (Figure 1, table S6, SI). Tetracyclines are widely used in both human and veterinary  
241 medicines. Samples from Agri settings, in particular Agri I site, contained the highest diversity of  
242 tetracyclines and highest contents for some compounds. Samples from territorial setting had  
243 concentrations usually in-between urban and agricultural values, making sense according to the  
244 substrate composition (Table 1). The concentrations of chlortetracycline (CTC-ECTC) in the anaerobic  
245 digestion and composting Agri I substrates reached nearly 15000 µg/kg DM, in line with Spielmeier  
246 et al. (2018, 2014) and Zhou and al. (2013). CTC-ECTC was found on sites where pig manure was  
247 treated (Agri I and Terri I) but not on sites treating waste of human or bovine origin, in accordance  
248 with its wide use in porcine populations for digestive and respiratory infections and its prescription in  
249 very specific conditions otherwise, e.g. cutaneous infections (humans) and calving disorders (cows).  
250 Oxytetracycline (OTC), withdrawn from the health market in France in 2002, was also only quantified  
251 in Agri I and Terri I samples, containing pig manure. Low concentration levels could be explained by a  
252 rapid transformation of OTC reported in other contexts, e.g. a 6.5 day half-life (hydrolysis) at 25°C in  
253 aqueous environment with an optimum at neutral pH (Xuan et al., 2010). Dehydrated degradation  
254 products of tetracycline (TC) and CTC-ECTC, anhydrotetracycline (ATC) and anhydrochlortetracycline  
255 (ACTC), respectively, were often quantified when their parent compounds were also present in the  
256 samples. ATC concentrations were close or superior to TC ones in all settings; this was often the  
257 same for ACTC and CTC. Doxycycline (DOX), used in both veterinary and human medicines, was found

258 in all urban samples with the highest amount at 9281 µg/kg DM, in 2 out of the 3 territorial anaerobic  
259 digestion substrates, with pig manure (2440-4923 µg/kg DM) and only in the Agri I composting  
260 sample, with pig manure (2277 µg/kg DM). This molecule was already found in high quantities in pig  
261 manure (Bourdat-Deschamps et al., 2017; Ho et al., 2014) but was barely reported in sludge samples.

262

### 263 **3.1.1.3. Sulfonamides and Trimethoprim**

264 Sulfonamides were not detected, in accordance with their low prescription, i.e. 1.7% of total  
265 antibiotic human use in France (WHO Report on Surveillance of Antibiotic Consumption, 2018) and  
266 17.9% in animals (ANSES Report on veterinary antibiotics sales, 2019). However, trimethoprim (TMP),  
267 used mainly in conjunction with sulfamethoxazole for a synergistic effect, was quantified in Terri III  
268 anaerobic digestion, Urb I anaerobic digestion and Urb II composting samples in small quantities, 95-  
269 128 µg/kg DM, which is closed to the highest values found by Göbel et al. (2005) for activated sludge.  
270 Its presence in organic waste could be an indicator of a small residual use of sulfamethoxazole.

271

### 272 **3.1.1.4. Macrolides**

273 Comparatively to other antibiotics, macrolides were only detected in small amounts,  
274 mirroring their low use for medicine and veterinary use in France (WHO Report on Surveillance of  
275 Antibiotic Consumption, 2018 and ANSES Report on veterinary antibiotics sales, 2019). Tylosin (TYL)  
276 not authorised for human use was only quantified in samples with pig manure. The low levels of TYL  
277 in samples could be explained by its fast biotic or abiotic degradation observed in other contexts  
278 (Ingerslev et al., 2001; Liu et al., 2010; Kolz et al., 2005). Erythromycin (ERY) was only quantified in  
279 urban sites in small quantities: 16-63 µg/kg DM. Spiramycin (SPI) was quite abundant in Terri settings  
280 even though results were semi-quantitative. Both compounds are used for human medicine and  
281 veterinary use.

### 282 **3.1.2. Other pharmaceuticals and biocides**

283 Of all other PPs, triclosan (TRI), a rather persistent human and veterinary biocide, was the  
284 most abundant compound found in all settings, in particular Agri I site ( $\approx 2300 \mu\text{g}/\text{kg DM}$ ), Urb II site  
285 ( $\approx 5900 \mu\text{g}/\text{kg DM}$ ), and Terri II site ( $\approx 1720 \mu\text{g}/\text{kg DM}$ ) treating waste from activities where TRI could  
286 be used for hygiene purposes. A small quantity of triclocarban (TRC), another persistent antifungal  
287 biocide (Lozano et al. 2018), was detected in the three urban sites. Ogunyoku and Young (2008)  
288 found similar and higher amounts in urban sludge for TRI and TRC; no literature data was available  
289 for agricultural and territorial settings. Fluoxetine (FLX) and carbamazepine (CBZ) were only found in  
290 samples containing sludge, e.g. all Urb sites and Terri II site ( $23\text{-}338 \mu\text{g}/\text{kg DM}$ ). FLX and CBZ are  
291 mostly prescribed to humans as antidepressant and anti-epileptic drugs. Diclofenac (DIC) was only  
292 quantified in urban samples as a compound authorised only for medicine use in France. In  
293 comparison ibuprofen (IBU) was found in samples from all settings, yet in small quantities ( $15\text{-}341$   
294  $\mu\text{g}/\text{kg DM}$ ), due to its wide use as active ingredient in human medicine and veterinary one for pets in  
295 France. Gemfibrozil (GEM), a hypolipidemic agent only prescribed to humans was found in all raw  
296 urban sludge below  $28 \mu\text{g}/\text{kg DM}$ .

297

### 298 **3.1.3. NPs**

299 Concentrations of nonylphenol (NP) and nonylphenol polyethoxylates (NPEOs; Nonylphenol  
300 monoethoxylate, NP1EO; Nonylphenol diethoxylate, NP2EO) were higher than PP ones in all settings,  
301 often above  $1000 \mu\text{g}/\text{kg DM}$  and up to  $176000 \mu\text{g}/\text{kg DM}$  for NP1EO in Terri I anaerobic digestion  
302 input substrate. NP1EO was the most abundant compound in all territorial and urban sites and less  
303 present and concentrated in most agricultural samples except Agri IV input substrate. NPs were often  
304 detected in urban sludge, at similar concentrations (Mailler et al., 2014; 2017). NP2EO was less  
305 present than NP1EO, except in the Agri I input substrate. NP2EO and NP were absent from Territorial  
306 samples. NPEOs as industrial surfactants and detergents (Bergé et al., 2012) may be recovered in  
307 urban and territorial settings. Their presence in agricultural samples was rarely documented  
308 especially at these concentration levels. Mortensen et al. (2003) and Petersen et al. (2003) reported

309 a total NPs content respectively below 0.1 mg/kg DM and 0.5 mg/kg DM in pig manure. Since NP  
310 mainly comes from NPEOs degradation (Ahel et al. 1994) and is more persistent, the relative low  
311 amount of NP or its non-detection in these process input samples seems to indicate a lack of time  
312 and/or inadequate conditions for the degradation of NPEOs.

313

#### 314 **3.1.4. PAHs**

315 Although in smaller quantities than NPs and PPs, most of the 13 PAHs were detected in all  
316 samples with contents up to 340 µg/kg DM. Inputs of urban sites were the most contaminated in  
317 quantity and diversity followed by those of territorial sites and agricultural sites. PAHs are residues of  
318 incomplete combustion of organic fuel (wood, gasoline...) mostly produced by transportation,  
319 housing or industrial activities (Lawal, 2017), explaining the higher quantities in urban and territorial  
320 samples than in agricultural ones. Concentrations were consistent with values reported by Mailler et  
321 al. (2014, 2017) and Stevens (2003). In all sites, two or three aromatic cycle PAHs, mainly  
322 Phenanthrene (PHE), Fluoranthene (FLO), Pyrene (PYR), Chrysene (CHR), were the most abundant  
323 compounds. Concentrations of four or five cycle PAHs were usually 2 to 10 times lower than those of  
324 smaller PAHs. This PAH pattern is commonly found in sewage sludge (Blanchard et al., 2004). Even if  
325 PAHs are very hydrophobic compounds, they were quantified at lower concentrations than those of  
326 some polar PPs (fluoroquinolones) that concentrated in the OWs due to charge interactions.

327

### 328 **3.2. Impact of treatment process and substrate type on pollutant fate**

#### 329 **3.2.1. Anaerobic digestion**

330 Figure 2 contains OMP dissipation rates for each individual process considering the following  
331 scale: Low: 0-30%, Moderate: 30-70%, High: >70%. Anaerobic digestion had a positive effect on  
332 fluoroquinolones and quinolone with the highest dissipation rates (>70%) for urban processes with  
333 high input concentrations, and for Agri I anaerobic digestion. There was more variability in Terri  
334 processes except for CIP that accumulated on all sites which could come from ENR possible

335 degradation (Martinez et al., 2006). CIP is a known ENR degradation product. ENR underwent either  
336 no change or was detected but not quantified, preventing any removal calculation except for Agri I.

337 Tetracyclines were only dissipated in territorial and agricultural settings. In all settings, when  
338 quantified, there was no real trend for TC and CTC-ECTC whereas ATC and ACTC presented high rates  
339 in some Agri and Terri sites. Mesophilic or thermophilic anaerobic digestion was shown to dissipate  
340 TC and CTC-ECTC in swine manure with reduction rates at 75-99% for CTC (Varel et al., 2012; Arikan,  
341 2008; Withey et al., 2016) and 50-99% for TC (Shi et al., 2011; Cetecioglu et al., 2013; Spielmeyer,  
342 2018). To our knowledge, no studies focused on ATC and ACTC fate during anaerobic digestion. While  
343 Urb III anaerobic digestion had no significant impact on the removals of tetracyclines, DOX and TC  
344 accumulated during Urb I anaerobic digestion (185-692%). Such accumulation was already observed  
345 for TC (180%) and CTC-ECTC (1600%) during industrial-scale swine manure anaerobic digestion (Liu et  
346 al., 2018). For some OMPs, a high variability in process input and output contents could partly  
347 explain this result, given our “one day” sampling strategy. Actually, Mailler et al. (2017) observed  
348 moderate time variability, from one to two orders of magnitude over one month for some PPs like  
349 CBZ, CIP, NOR and OFL in raw sludge before treatment. OMP extractability also varied along the  
350 waste transformation. Although extraction and purification losses and MS matrix effects were  
351 assumed to be compensated by adding deuterated-labelled compounds (2.3.2.), the LOQs were  
352 impacted by the variations in extractability. Individual LOQs were identified for each matrix,  
353 impacting dissipation rate calculation when the LOQ/2 value was used. During treatment processes,  
354 degradation products could be transformed back to their parent form (e.g. DIC, CBZ, ERY; Göbel et  
355 al., 2007; Kasprzyk-Hordern et al., 2009) and desorption of OMPs from particulate matter could also  
356 cause an accumulation over time (Köck-Schulmeyer, 2013).

357 TMP dissipated at high rates when present in urban and territorial substrates, as previously  
358 seen (Spielmeyer et al., 2015; Mohring et al., 2009; Feng et al., 2017). TYL mostly disappeared (>-  
359 70%) regardless of the substrate origin in accordance with Withey et al. (2016), Mitchell et al. (2013),

360 Kolz et al. (2005). Even if results were semi-quantitative and had to be taken with cautions, SPI was  
361 removed from Urb I and Terri II (not shown in Figure 2) as observed by Zhu (2014) for sewage sludge.

362 Finally, “other” PPs were only dissipated during Urb I and Urb III anaerobic digestion,  
363 dissipation rates in agricultural and territorial settings were either non-calculable, or when present,  
364 they saw no changes or accumulation during anaerobic digestion. Low to moderate dissipation was  
365 observed for GEM, FLX, CBZ, DIC on Urb I site and IBU on Urb III site, in coherence with Martin et al.  
366 (2012; GEM, CBZ, DIC and IBU), Gonzales-Gil et al. (2016; CBZ and IBU), Bergersen et al. (2012; FLX)  
367 and Malmborg and Magner (2015; FLX) but contradictory to the one of Gonzales-Gil et al. (2016) for  
368 FLX. Low to moderate dissipation occurred for TRI and TRC in Urb sites, in agreement with dissipation  
369 rates in Guerra et al. (2015; < 30%). In Terri II and III, there was either no change or an accumulation  
370 depending on OMPs.

371 Nonylphenol polyethoxylates can be shortened into NP2EO, NP1EO and NP. Mass balances  
372 were difficult to interpret since production and degradation could occur simultaneously. NP1EO  
373 concentrations decreased in nearly all sites except Agri I. NP2EO was highly dissipated in only Agri I  
374 but accumulated in Agri II and Urb III as a possible result of NPnEO (n>2) shortening into NP2EO (or  
375 directly into NP1EO and NP) (Minamiyama et al., 2006). In all anaerobic digestion processes, no  
376 apparent NP degradation was evidenced with even negative rates in Urb I (-56%) and Agri IV (-41%)  
377 processes, illustrating NP accumulation was probably due to NP1EO transformation (Ahel et al., 1994;  
378 Lu et al., 2008; Murdoch and Sanin, 2016; Janex-Habibi et al., 2009). During Urb III and Terri I-II-III  
379 anaerobic digestion, NP1EO was removed with no NP accumulation, reflecting a possible  
380 simultaneous removal observed by Patureau et al. (2008) and Paterakis et al. (2012).

381 PAH dissipation mostly occurred on urban and territorial sites having high inputs but not on  
382 agricultural sites. Low molecular weight PAHs were more dissipated than high molecular weight  
383 ones, like in Chang et al. (2003). The 6 aromatic-cycle PAHs, Benzo(g,h,i)Perylene (BPE) and  
384 Indeno(1,2,3-cd)Pyrene (IND), were unaffected on all sites and even accumulated. These low removal  
385 rates observed on industrial plants are in contradiction with other data from lab-scale anaerobic

386 digestion experiments with spiked sludge (Barret et al., 2010), more controlled process conditions  
387 and homogeneous samples. Accumulation occurred during Urb I and Terri II-III anaerobic digestion  
388 for half of the quantified PAHs with no simple explanation based on site process parameters. As  
389 already stated for PPs, our one-day sampling strategy may have hindered process efficiency even  
390 though for more persistent compounds like PAHs and NPs the time variability of input sludge  
391 concentrations is much lower (Mailler et al., 2017).

392

393 Differences between mesophilic and thermophilic anaerobic digestion processes could not be  
394 assessed due to low OMP detection frequencies in Agri II samples (thermophilic anaerobic digestion).  
395 There seems to be no global influence of the substrate type on the OMP fate during anaerobic  
396 digestion, trends more relying on individual compounds as shown before. Many PPs were not  
397 detected or not quantified in input and/or output samples (Table S6, SI for input samples; Table S7  
398 for output samples), limiting the calculation of removal rates. Low OMP concentrations are more  
399 sensitive to temporal variations often observed in process input matrices, further impeding the  
400 detection of global trends. All sites combined, 17 molecules (NOR, OFL, CIP, ENR, DOX, ACTC, ATC,  
401 TMP, TYL, ERY, NP1EO, NP2EO, PHE, PYR, Benzo(a)Anthracene BAA, Benzo(b)Fluoranthene BBF,  
402 Benzo(a)Pyrene BAP) had high dissipation rates (>70%), 9 molecules (TC, FLX, CBZ, DIC, IBU, TRC,  
403 Fluorene FLU, Anthracene ANT, Benzo(k)Fluoranthene BKF) had moderate rates (30-70%) and 7  
404 molecules (FLQ, CTC-ECTC, OTC, GEM, TRI, FLO, CHR) had low dissipation rates (0-30%) during  
405 anaerobic digestion. Five compounds (NP, Di-Benzo(a,h)Anthracene DBA, BPE, IND) were never  
406 dissipated by anaerobic digestion.

407

### 408 **3.2.2. Dewatering**

409 Dewatering processes after anaerobic digestion differed among the sites. The comparison of  
410 volatile solid (VS)/total solid (TS) content ratios before and after dewatering (Table S5) allowed  
411 assessing if any VS loss occurred (Guerra et al, 2015). Ratios were significantly identical (Student's t-

412 test), except for Terri II and Urb I processes respectively by screw or filter-press: even if the TS  
413 concentration factor seemed correct, dewatering was improper, keeping many solids in the liquid  
414 phase. Drogg et al. (2015) confirmed a lower dewatering efficiency by screw-press than by centrifuge.  
415 Organic matter (or VS) loss in the liquid phase may imply a potential loss of OMPs, especially those  
416 with a high Log Kow or with ionic charges preferentially sorbed to organic matter.

417 A dewatering dissipation rate was calculated from the OMP concentrations in raw matter  
418 before dewatering and after in the dewatered solid. OMP dissipation could happen if: a) a pollutant  
419 either soluble or sorbed to dissolved and colloidal matter was present in the dewatering liquors, b)  
420 suspended solids and potential OMPs migrated to the liquors in case of dewatering inefficiency, or c)  
421 some polymers added during dewatering changed the interactions between some pollutants and  
422 matter making them more “soluble”. In the absence of dissipation, OMPs stayed sorbed on the solid  
423 phase. Accumulation may be explained by heterogeneous sampling. On Terri II and Urb I sites with  
424 low dewatering efficiency, OMPs with high affinity to suspended solids like PAHs, FLX, CBZ, TRI,  
425 NP1EO and NP (hydrophobic interactions), more soluble OMPs like tetracyclines and ERY were  
426 actually dissipated. Comparable amounts of PAHs and PPs per gram of dry matter were found in the  
427 liquid and solid phases of Terri II samples (data not shown), reinforcing the second hypothesis, i.e.  
428 losses by sorption on particulate matter still present in the dewatering liquors.

429 Urb III° thermal dewatering process dissipated most PAHs, CBZ, DIC, IBU, TRI and TRC. High  
430 temperatures (300°C) may increase losses by volatilisation for compounds like PAHs or by chemical  
431 alteration. Mailler et al. (2014) observed 10 to 40% losses in PAH concentrations during thermal  
432 dewatering, which is consistent with our data and a 10-90% dissipation of NPEO concentrations  
433 whereas we observed no significant changes for NP1EO and NP2EO. Tuncal et al. (2011) suggested  
434 volatilisation as a dissipation mechanism for PAHs and Veeken and Hamelers (1999), a possible  
435 hydrolysis in digestate at high temperature (>250°C for a few minutes). Comparison of OMP  
436 dissipation rates for Urb III and Urb III° dewatering processes reveals that PAHs were the most  
437 temperature sensitive OMPs.

438 For the 3 centrifugation processes (Agri I, Terri I, Urb III), very little dissipation occurred, e.g.  
439 for CBZ, DIC, NP, some PAHs especially in Urb III, and accumulation was observed for NP1EO and  
440 some PAHs. The role of centrifugation in the OMP fate is also not well documented. Mailler et al.  
441 (2014) reported no significant dissipation of PAHs, NP and NP1EO during sludge centrifugation. Other  
442 compounds like TMP, macrolides, fluoroquinolones, IBU, GEM, TRC were rather unaffected by  
443 centrifugation whatever the process.

444

### 445 **3.2.3. Liming**

446 The liming process followed anaerobic digestion and thermal dewatering processes (Urban  
447 III). The resulting CaO-sludge mixture was compacted into land-spreadable pellets. Liming increased  
448 pH mixture from 7 to >12, decreased water content through the addition of CaO and the resulting  
449 exothermic reaction, creating unsuitable conditions for microorganisms and stabilising waste matrix.

450 Dissipation occurred for DOX, TC, TMP, FLX, CBZ, TRI and low molecular weight PAHs. High  
451 molecular weight PAHs saw either no changes or accumulated during the liming process.  
452 Accumulation also occurred for NOR and OFL. Given the short time of treatment and its additive  
453 nature, concentration changes were only due to physicochemical phenomena in line with the pH  
454 increase and the exothermic reaction, inhibiting biological reactions and potentially modifying the  
455 interactions between OMPs and their bearing phases. Indeed, pH increase may solubilise organic  
456 matter and thus change OMP solubility and extractability from the matrix, resulting in overall  
457 concentration increase, i.e. for PAHs, NOR, OFL. For positively- (CBZ) or negatively-charged OMPs at  
458 pH 12, strongest interactions may occur through cation exchange and bridging with added Ca<sup>2+</sup>,  
459 reducing extractability interpreted as apparent dissipation as supposed by Ogunyoku and Young  
460 (2014) for the increase of TRI concentration during liming. Changes in sorption strength could also  
461 occur with pH changes.

462

### 463 **3.2.4. Composting**

464 Quinolone and fluoroquinolones were highly dissipated during composting on Urb sites.  
465 Haiba et al. (2013) and Lillenberget al. (2010) found similar results for NOR, CIP and OFL with  
466 reduction rates up to 95%. FLQ concentration sharply decreased in Urb II, which was not reported  
467 before to the best of our knowledge. Agri composting diminished ENR contents. Liu (2018) found a  
468 removal rate of about 80% for ENR when composting swine manure. Biotransformation could be the  
469 main reason behind dissipation. Maia et al. (2014) showed high bacterial biodegradation under  
470 aerobic conditions for OFL, NOR, CIP and Prieto et al. (2011) over 90% decrease for CIP and NOR with  
471 white-rot fungus (*T. versicolor*).

472 Tetracyclines were also dissipated at high rates during composting in both urban and  
473 agricultural settings, either after anaerobic digestion (Urb I, Agri I) or not, in coherence with  
474 published rates: 74%, 92%, and 70% for CTC-ECTC, OTC and TC when composting manure (Wu et al.,  
475 2011; Massé et al., 2014); up to 35% removal for TC when composting digested sludge (Khadra et al.,  
476 2019). Interestingly, lower dissipation rates were obtained on all sites for ACTC and ATC. TC was  
477 rapidly transformed into ATC in other contexts (Halling-Sørensen et al., 2002; Jia et al., 2009); this  
478 could explain a lower dissipation rate for dehydrated forms, with ATC and ACTC molecules  
479 continuously brought by TC and CTC-ECTC transformation.

480 TMP was only dissipated in Urb II at 100% compared to 35% for sewage sludge co-  
481 composting in Khadra et al. (2019). Macrolides highly dissipated (>70%) as previously seen for TYL  
482 and ERY (>95%; Cessna et al., 2011; Ho et al., 2013) during composting of cow/broiler manure.

483 Other PPs, when detected, were strongly dissipated during urban and agricultural  
484 composting, except FLX and CBZ that accumulated in Urb II. Globally, Urb I and II had high dissipation  
485 rates.

486

487 NPs dissipation rates were more site-dependent with medium to high values in Urb I and Agri  
488 I and no changes in Urb II. Zheng et al. (2018) and Lashermes et al. (2010) observed a 50-60%

489 reduction in NP content during sewage sludge composting. No information was available in the  
490 literature concerning NP dissipation during manure composting.

491           Eleven and 12 of the 13 PAHs accumulated during Urb I and II composting, possibly due to  
492 non-extractible PAHs desorbed during organic matter degradation (hence freeing previously trapped  
493 compounds), bad sampling or atmospheric deposition. For Urb II, the addition of green waste co-  
494 substrate (OMP content not quantified) could also explain this increase. There were no changes in  
495 Agri III but ten PAHs (of low to medium molecular weight) were dissipated during Agri I composting,  
496 probably due to the 12 week maturation period. According to Amir et al. (2005), this decrease could  
497 stem from both sorption on non-accessible sites in organic material and biodegradation by  
498 thermophilic microbial populations. A higher dissipation of smallest PAHs could indicate a  
499 preferential degradation by microorganisms whereas biggest compounds, more hydrophobic, could  
500 be more easily sorbed on organic matter and less degraded.

501

502           Overall, total concentrations of PPs were highly decreased during all composting processes.  
503 Results were contrasted for PAHs: total PAH concentrations are expected to decrease during  
504 composting (Amir et al., 2005; Ahtiainen et al., 2002) with smallest compounds preferentially  
505 degraded but this was only observed in Agri I. Results were more process-dependent for NPs with  
506 reduction only observed when composting anaerobically digested inputs.

507           The combination of anaerobic digestion and composting was also evaluated in Agri I and Urb  
508 I. This combination is common to stabilize organic waste depending on the final uses. In this case,  
509 OMP dissipation rates were established considering the input and output concentrations of the  
510 combined processes, normalised with the ash content (2.4.4.). For all pollutant types, both sites were  
511 significantly more efficient in dissipating OMPs, than single processes (Figure 2, "Combined":  
512 dissipation rate from anaerobic digestion input to composting output integrating dewatering).

513           Considering Urb I and Agri I together, tetracyclines were more dissipated by post-anaerobic  
514 digestion composting than by only anaerobic digestion. ACTC and ATC were mainly dissipated during

515 Agri I anaerobic digestion and Urb I composting mainly due to their accumulation during anaerobic  
516 digestion in Urb I. However, ACTC and ATC concentrations in mature composts (per g DM) were the  
517 same on both sites, indicating a complementary effect of processes lowering concentrations even  
518 after an accumulation in Urb I. Reaching the same final concentration could indicate a lower  
519 accessibility of ACTC and ATC left in composts. Agri I presented more tetracyclines and Urb I more  
520 fluoroquinolones, yet both had overall similar contribution of anaerobic digestion and post-anaerobic  
521 digestion composting in the dissipation of tetracyclines and fluoroquinolones.

522 Anaerobic digestion and composting had similar dissipation efficiencies for PAHs and NPs on  
523 both sites. NPs when detected were highly dissipated by combined processes. Process efficiency did  
524 not seem to be linked to initial OMP amount.

525 This dissipation improvement may be explained by the combination of process conditions:  
526 bacterial consortia are fully different; the switch from a reductive environment to an oxidative one  
527 allows for different degradative pathways. Anaerobic digestion stabilises the organic matter matrix  
528 as well as reduces particle size, releasing some OMPs that can be dissipated during composting or  
529 increasing interaction surfaces. It would be interesting to compare the effect of composting alone,  
530 anaerobic digestion and anaerobic digestion + composting on the same substrate in parallel to  
531 decipher more specifically the role of each system.

532

#### 533 **4. Conclusions**

534 *In situ* quantification of 53 OMPs in 10 industrial OW treatment lines was performed. A huge  
535 diversity of pollutants was detected in processes input samples according to OMP uses and OW  
536 types, at concentrations similar to those found in literature. Less studied OWs, territorial mixes,  
537 revealed an intermediary OMP composition, between urban and agricultural settings. For dewatering  
538 processes, particularly those with the lowest efficiency, either soluble and charged (tetracyclines) or  
539 hydrophobic (FLX, PAHs) OMPs could be released in the aqueous fraction which is thus important to  
540 consider when (i) calculating mass balance on the overall treatment line and (ii) assessing the impact

541 of such aqueous fraction if used as fertilizer. Some OMPs like PAHs and DIC were dissipated during  
542 thermal treatment due to either volatilization or temperature sensibility. During liming, either  
543 dissipation (e.g. DOX, TC, CBZ, TMP, TRI, FLU to PYR) or increase of concentration (e.g. NOR, OFL,  
544 DBA, BAP) were observed due to the drastic change in pH, moisture and temperature, modifying the  
545 interactions between OMPs and organic matter. Anaerobic digestion had positive effect mainly on  
546 fluoroquinolones, macrolides, TMP, DOX, ACTC, ATC and NP1EO whereas composting was more  
547 effective on all studied OMPs (except PAHs). Combination of processes enhanced most OMP  
548 dissipation. Overall, dissipation was mostly affected by process type and more compound specific  
549 behaviour was observed. Waste origin had no effect on dissipation.

550           These results are general trends of dissipation for a large OMP diversity and for 8 anaerobic  
551 digestion, 6 dewatering, 1 liming, 4 composting and 2 combined anaerobic digestion + composting  
552 real-scale treatments. Preference was given to the number of processes studied, rather than just one  
553 treatment with its temporal variability and sludge retention time. However, this complete OW  
554 treatment line approach brings *in situ* data that can be used for building or completing global models  
555 of OMP fate during OW treatments, from waste production to field uses, particularly for models of  
556 environmental risk assessment of OW spreading.

557           Investigation of more sites is needed to better understand the role of less used processes  
558 (thermophilic anaerobic digestion, liming...) and establish finer trends for dissipation of individual  
559 compounds.

560

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567

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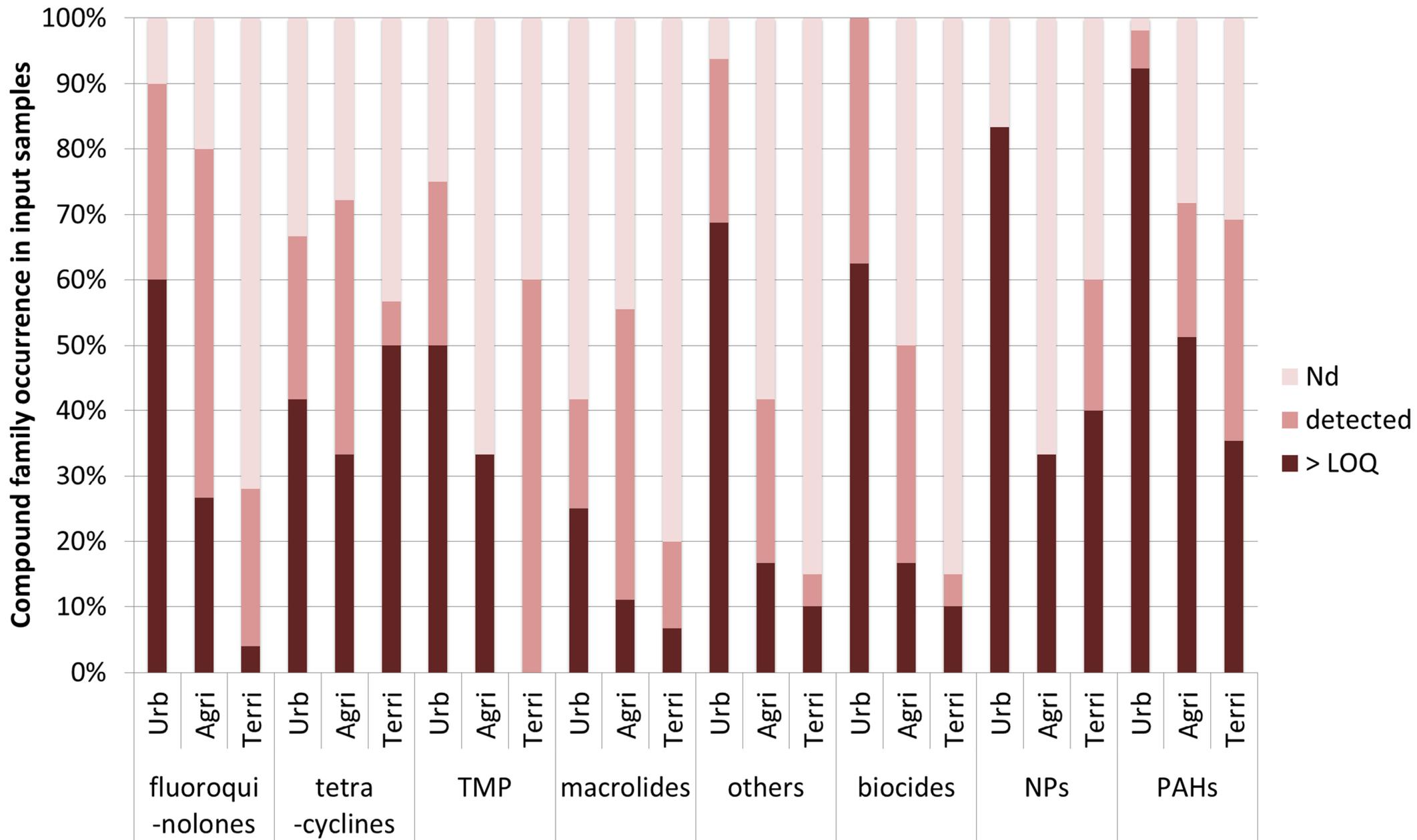
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**Figure 1:** Pollutant occurrences in process input wastes, according to their quantification or detection, ordered by families. Occurrence in % is the number of non-detection, detection or values >LOQ considering all compounds of a given family for all input samples of one setting. Nd = Non-detected.

		Anaerobic Digestion							Dewatering					Liming	Composting			Combined						
		Urb I	Urb III	Agri I	Agri II	Agri IV	Terri I	Terri II	Terri III	Urb I	Urb III	Urb III°	Agri I	Terri I	Terri II	Urb III	Urb I	Urb II	Agri I	Agri III	Urb I	Agri I		
Fluoroquinolones Quinolones	NOR	+	nc		nc	nc	+		nc		nc	+		nc		+					+	+		
	OFL			nc	nc	nc		+	+			nc			+				nc			+	nc	
	CIP		+	+	nc	nc	+	+	+		nc	+							nc	nc				
	ENR	nc	nc	+	nc	nc		nc	nc	nc	nc	nc	+		nc	nc		nc	+	+				
	FLQ	+		nc	nc	nc	+	+	nc				nc			nc			nc	nc		+	nc	
Tetracyclines	CTC - ECTC	nc	nc		nc	nc	+	nc	nc	nc	nc	nc			nc	nc	nc	nc	+	nc	nc	nc	nc	
	DOX	+		+	nc	nc	+	+	+	+	nc	nc			+	+	+	+	+	+	+	+	+	
	TC	+			nc	nc	+	+	nc	nc	nc	+			+	+	+	+	+	nc	nc	nc	nc	
	OTC	nc	nc		nc	nc			nc	nc	nc	+			nc	nc	nc	nc	nc	nc	nc	nc	nc	
	ACTC	+	nc	+	+	nc	+	+	+	+	nc	nc	+			nc	nc	nc	nc	nc	nc	nc	nc	
	ATC	+		+	+	nc	+	+	+	+	nc	nc				nc	nc	nc	nc	nc	nc	nc	nc	
Trimethoprim	TMP	+	nc			nc	nc	nc	+		nc	nc	nc	nc	+			nc	nc	nc	nc	nc	nc	
Macrolides	TYL	nc	nc	+	nc	nc	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	ERY	+	nc	nc	nc	nc	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
Other pharmaceuticals	FLX			nc	nc	nc	nc		nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	CBZ		+	nc	nc	nc	nc		+	+	nc	nc			+	+	+	+	+	+	+	+	+	
	DIC		+	nc	nc	nc	nc	+	+	+	nc	nc			+	+	+	+	+	+	+	+	+	
	IBU	+	+		+	nc	nc		+	+	nc	nc			+	+	+	+	+	+	+	+	+	
	GEM			nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc
	TRI					nc	nc				nc	nc	+											
	TRC			nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc
NPs	NP1EO		+		nc	+	+	+	+	+	+	*									+	+	+	
	NP2EO	nc	+	+	+	nc	nc	nc	nc	nc	nc	nc	*	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	NP	+		nc	nc	+	nc	nc	nc	nc	nc	nc	*	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
PAHs	FLU	+						+	+	+	+			+	+	+	+	+	+	nc	nc	nc	nc	
	PHE	+						+	+	+	+			+	+	+	+	+	+	+	+	+	+	
	ANT	+	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	FLO			nc	+		nc	+	+	+	+													
	PYR	+			nc																			
	BAA	+				nc	nc	+	+	+	+													
	CHR		+	nc			nc				nc	nc												
	BBF		+	nc	nc	nc	nc	nc	+	+	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	BKF	+		nc	nc	nc	nc	nc	+	+	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	BAP	+		nc	+	+	+					+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	DBA	nc	+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	
	BPE				+	nc	nc	+	+	+	+													
	IND		+	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	nc	

*	data missing		no dissipation
nc	non calculable		0-30% dissipation (low)
+	accumulated		30-70% dissipation (medium)
			70-100% dissipation (high)

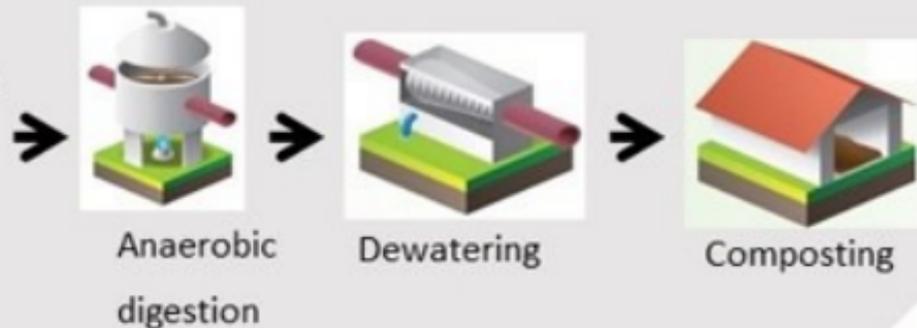
**Figure 2:** Heat map of pollutant dissipation rates. nc: non calculable = both input and output contents were not detected or detected but with no available LOQ. "Combined" means the dissipation rate from the beginning of anaerobic digestion till the end of composting. "°" indicates a thermal dewatering.

Site	Treatment Process	P C	Reactor size (m <sup>3</sup> )	Duration	Input (Substrate)	DM content	Dewatering TS concentration factor	Volatile matter removal	Notes
Agri I	AD		950	38 d	Pig manure	5%	-	48%	<i>Mesophilic AD</i>
Agri I	Dewatering	X	-	-	Digested manure	3%	11.6	-	Centrifuge
Agri I	Composting	X	-	12 w	Dried digested manure	35%	-	57%	No addition of co-substrate
Agri II	AD		740	42 d	Cow manure & straw	29%	-	82%	<i>Thermophilic AD, batch</i>
Agri III	Composting		-	12 w	Cow manure & straw	29%	-	13%	No addition of co-substrate
Agri IV	AD		600	40 d	Cow manure & straw	50%	-	49%	<i>Mesophilic AD, batch</i>
Terri I	AD		6000	30 d	45% AF waste, 45% pig manure, 10% cereal mixture	14%	-	35%	<i>Mesophilic AD</i>
Terri I	Dewatering	X	-	-	(Digested) 45% AF waste, 45% pig manure, 10% cereal mixture	8%	11.5	-	Centrifuge
Terri II	AD		1600	70 d	50% mixed manure, 28% sewage sludge, 22% AF waste mixture	10%	-	55%	<i>Mesophilic AD</i>
Terri II	Dewatering	X	-	-	(Digested) 50% manure, 28% sewage sludge, 22% AF waste mixt.	7%	3.3*	-	Screw-press
Terri III	AD		8000	90 d	50% food waste, 20% cow manure, 30% AF waste mixture	20%	-	60%	<i>Mesophilic AD</i>
Urb I	AD		6200	29 d	Raw sewage sludge	5%	-	35%	<i>Mesophilic AD</i>
Urb I	Dewatering	X	-	-	Digested sewage sludge	4%	5.25*	-	Filter-press
Urb I	Composting	X	-	4 w	Dried digested sewage sludge	45%	-	55%	55% Co-substrate (34% GW and 21% sieve residues)
Urb II	Composting		-	6 w	Raw sewage sludge	18%	-	46%	66% Co-substrate (GW)
Urb III	AD		6000	34 d	Raw sewage sludge	5%	-	38%	<i>Mesophilic AD</i>
Urb III	Dewatering	X	-	-	Digested sewage sludge	3%	6	-	Centrifuge
Urb III°	Dewatering	X	-	-	Dried digested sewage sludge	18%	5.22	-	Thermal drying
Urb III	Liming	X	-	2 h	Thermally dried digested sewage sludge	94%	-	-	Addition of 20% of hydrated lime dw:dw

**Table 1:** Details on each sampled sites and waste treatment processes: Site name, type of ongoing process, link between processes (PC: process continuity), reactor size, process duration (d=days/w=weeks), substrate input, dry matter (DM) content, dewatering efficiency (\* significant loss of volatile solids (VS) during the process; more details in Table S4, SI), volatile matter removal (TS: total solids, AD: Anaerobic digestion, AF= Agri-Food industry, GW = Green Waste).

## 10 industrial organic wastes treatment lines

Organic wastes  
(manure, sludge,  
mixture of manure,  
straw...)



Fate of **53** organic micropollutants all along treatment lines

