

# Organic pollutants in compost and digestate.

## Part 2. Polychlorinated dibenzo-*p*-dioxins, and -furans, dioxin-like polychlorinated biphenyls, brominated flame retardants, perfluorinated alkyl substances, pesticides, and other compounds†‡

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Compost and digestate are important recycling fertilizers and have beneficial effects on soil parameters. However, they can contain significant amounts of organic pollutants. Here, the first comprehensive data set on dibenzo-*p*-dioxins and -furans (PCDD/F), dioxin-like polychlorinated biphenyls (DL-PCB), brominated flame retardants, perfluorinated alkyl substances (PFAS), pesticides, phthalates, nonylphenol and chlorinated paraffins (CP) in source-separated compost and digestate from Switzerland is presented ( $n = 3\text{--}18$ ). The median  $\sum 17\text{PCDD/F}$  and  $\sum 12\text{DL-PCB}$  concentrations were at  $3.2 \text{ ng I-TEQ kg}_{\text{dry weight (dw)}}^{-1}$  and  $3.0 \text{ ng WHO-TEQ kg}_{\text{dw}}^{-1}$ , respectively. Brominated diphenyl ether 209 (BDE 209) accounted for 72% of the total polyBDE content ( $10 \mu\text{g kg}_{\text{dw}}^{-1}$ ). Hexabromocyclododecane (HBCD) and tetrabromobisphenol A (TBBPA) levels were at 100 and  $0.51 \mu\text{g kg}_{\text{dw}}^{-1}$ , respectively. PFAS were identified for the first time in compost and digestate (median concentration  $6.3 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $\sum 21\text{compounds}$ ). Out of 269 pesticides analysed, 30 fungicides, 14 herbicides, eight insecticides and one acaricide were detected. Di-(2-ethylhexyl)phthalate (DEHP) median concentration accounted for  $280 \mu\text{g kg}_{\text{dw}}^{-1}$  and nonylphenol was below the detection limit of  $1 \text{ mg kg}_{\text{dw}}^{-1}$ . The sum of short and medium chain CP was between 90 and  $390 \mu\text{g kg}_{\text{dw}}^{-1}$ . The concentrations observed were at or above the levels found in background soils, which are the main recipient of compost and digestate. Where actually applied, compost can contribute considerably to the total input of organic pollutants to the soil. However, on a total Swiss agricultural area base, inputs seem to be limited.

## Introduction

Composting and digestion (*i.e.*, aerobic and anaerobic degradation of organic wastes) are important waste management

strategies in Europe.<sup>1</sup> The resulting compost and digestate can be used as soil conditioner and fertilizer, recycling nutrients back to agriculture and horticulture. However, if the compost and digestate contain pollutants these are introduced to the soil. Little is known about organic pollutants and pesticides in source-separated compost (*i.e.*, compost solely derived from crude organic kitchen waste and green waste; for reviews see Brändli *et al.* and Buyuksonmez *et al.*;<sup>2,3</sup> for guide and limit values see the Electronic Supplementary Information (ESI), Table S1†). Data on digestate is completely lacking. Emerging organic pollutants, such as brominated flame retardants (BFR), perfluorinated alkyl substances (PFAS) and chlorinated paraffins (CP) have not yet been systematically investigated in recycling fertilizers. These contaminants need to be included in inventory studies, since they have reached similar production volumes to the “classical” persistent organic pollutants and exhibit some similar properties, *i.e.*, ubiquitous occurrence, toxicity, persistence, carcinogenic and/or endocrine disrupting effects.<sup>4–7</sup> In this study polychlorinated dibenzo-*p*-dioxins and -furans (PCDD/F), dioxin-like polychlorinated biphenyls (DL-PCB), BFR (polybrominated diphenyl ethers (PBDE)), hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA)), PFAS, and about 270 pesticides were analysed in 13 compost and five digestate

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samples from commercial plants in Switzerland. Additionally, phthalates and nonylphenol were determined in six, and short and medium chain CP (SCCP and MCCP) in three samples. The 18 samples were part of an extended inventory study on compost where polycyclic aromatic hydrocarbons ( $\Sigma$ 16PAH defined by the US EPA and selected source-diagnostic compounds), PCB (seven Institute for Reference Material and Methods (IRMM) congeners, *i.e.*, PCB 28, 52, 101, 118, 138, 153, 180 ( $\Sigma$ 7PCB), and four chiral PCB), heavy metals and nutrients were determined. Respective results are discussed in a companion paper.<sup>1</sup>

The aim of this paper is: (i) to provide a comprehensive and state-of-the-art overview on the presence of hazardous organic pollutants and pesticides, including emerging contaminants, in source-separated compost and digestate; (ii) to assess variables which determine their concentrations; (iii) to compare the organic pollutant contents in compost and digestate with recipient soils; and (iv) to compare fluxes to soil by compost application to other inputs.

## Materials and methods

### Experimental design, sampling and sample preparation

Samples were selected to cover a wide variety of variables which may influence organic pollutant concentrations in compost and digestate,<sup>2</sup> namely, treatment processes (composting/digestion), origin (urban/rural) and composition of input material (green waste with/without crude organic kitchen waste), and season of collection (spring/summer, autumn, winter). However, it was not possible to account for every combination of these variables due to limited resources.

The sampling procedure was described in Brändli *et al.*<sup>8</sup> Samples were either air-dried at room temperature for 10–30 d in a climate chamber (for PCDD/F, DL-PCB and BFR analysis), dried at 40 °C for seven days (pesticides), frozen at –20 °C (phthalates), lyophilized (CP) or dried chemically<sup>8</sup> (PFAS). Samples dried at ambient temperature and at 40 °C were stored at room temperature in the dark, whereas lyophilized and chemically dried material was stored at 4 °C.

### Analytical procedure

PCDD/F, DL-PCB and BFR analysis was performed as in Zennegg *et al.*<sup>9</sup> PFAS extraction was carried out according to Powley *et al.*,<sup>10</sup> with the following modifications: 7.5 g of chemically dried compost (2.5 g compost and 5 g Na<sub>2</sub>SO<sub>4</sub>) were spiked with perfluoro-3,7-dimethyloctanoic acid used as internal standard for quantification. NaOH and HCl were dissolved in methanol. Before injection, the recovery standard 3,5-bis(trifluoromethyl)phenyl acetic acid was added to the extract. The liquid chromatography-mass spectrometer (LC-MS) separation and detection was described in Berger and Haukås.<sup>11</sup> Two different multi-residue methods (one with LC-MS/MS and the other with gas chromatograph (GC) separation and electron capture detection or nitrogen/phosphorus detection) were used to screen samples for 269 pesticides.<sup>12,13</sup> Phthalate analysis was outlined in Herrchen *et al.*<sup>14</sup> (GC-MS) and nonylphenol was determined according to Hecht *et al.*<sup>15</sup> (LC-MS). CP were analysed and quantified as

outlined in Hüttig *et al.*<sup>16</sup> and Reth *et al.* (GC-MS).<sup>17</sup> The dry matter content was determined at 105 °C. For analytical quality control, we refer to the respective papers.<sup>9–17</sup> Note, however, that compost was considered as a solid matrix similar to soil or sludge, a procedure not uncommon in method guidelines and legislation<sup>18,19</sup> and considered appropriate to get a first overview of the content of organic pollutants in compost and digestate.

### Data processing and statistics

For median calculation, only samples in which a compound was detected (*n* provided in brackets) were taken into account due to the low number of samples. Therefore *n* can differ from the number of analyses. After basic evaluation of the dataset (*i.e.*, median, comparison with literature values and soil data), the variables ‘treatment process’, ‘composition of input material’, ‘origin of input material’ and ‘season of input material collection’ were assessed by the Mann–Whitney-U Test (Statistica 6.1 StatSoft, Inc., Tulsa, USA). This test does not require normal distribution of the data and is robust to outliers. The level of significance was 0.025.

To account for the treatment process, digestates originating from urban areas and containing organic kitchen waste in the input material were tested against composts derived from the same kind of input material. Since only two samples (nos. 17 and 35, see ESI, Table S2†) of green waste composts from urban areas were available, the effects of the origin of input material (urban/rural) and of the input material composition (with/without organic kitchen waste) could not be evaluated separately. Hence, urban composts and digestates containing organic kitchen waste (*n* = 13) were tested against rural green waste composts (*n* = 5) only. Samples with a different congener/compound pattern were identified by principal component analysis (PCA; Statistica 6.1, StatSoft, Inc., Tulsa, USA). Organic pollutant and pesticide contents were tested for (linear) correlation with the duration of the composting process and organic matter content. Correlations between different organic pollutant classes, as well as between heavy metals and organic pollutants, were also assessed. The level of significance for all correlations was 0.05.

### Mass loads of organic pollutants to soil

The input of organic pollutants to Swiss agriculture by compost application and the time to reach Swiss guide or limit values for soil were determined according to Brändli *et al.*<sup>2</sup> Aerial deposition rates and contents in manure and sludge were obtained from the Swiss or European literature (ESI, Table S3†).

## Results and discussion

Organic pollutant concentrations measured in Swiss compost and digestate are first compared with literature values where available. Thereafter, the observed contents are discussed in relation to the hypothesized key determinants and compared with typical concentrations in the soil as the major recipient of these recycling fertilizers. Finally, input *via* compost application to agricultural land is compared with other inputs (*i.e.*, aerial deposition, input *via* manure and sewage sludge application) on

a national and local scale. Common to all compound classes is that no correlations were found either between the contents of organic matter and organic pollutants, or between the content of heavy metals and organic pollutants. For the remaining results it is necessary to refer to the following sections.

## PCDD/F

The median  $\sum 17\text{PCDD/F}$  (sum of the 17 2,3,7,8-chlorosubstituted PCDD/F) concentrations in composts and digestates was  $3.2 \text{ ng I-TEQ kg}_{\text{dry weight (dw)}}^{-1}$ , ranging from  $0.52$  to  $21 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$  (Fig. 1,  $n = 18$ , ESI, Table S2†). One sample was above the Swiss guide value for compost ( $20 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$ ).<sup>20</sup> The median literature value for compost was  $9.3 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$  ( $n = 185$ ),<sup>2</sup> which is almost at the 90% level of this study's dataset (Fig. 1). Median concentrations of the sum of all tetra- to octachlorinated PCDD/F in this work were  $466 \text{ ng kg}_{\text{dw}}^{-1}$  and  $102 \text{ ng kg}_{\text{dw}}^{-1}$ , respectively. Compared with the literature,<sup>2</sup> the concentrations of the higher chlorinated PCDD/F were lower in Swiss compost, whereas lower chlorinated compounds concentrations were in the same range (ESI†, Figs. 1 and 2). This might be explained by the banning of pentachlorophenol (PCP) for wood preservation, which was often contaminated with highly chlorinated PCDD/F, by decreasing emission of higher chlorinated PCDD/F compared with the early 1990s, when most of the literature data was

collected, or by different emission patterns between countries, since most literature data originated from Germany.

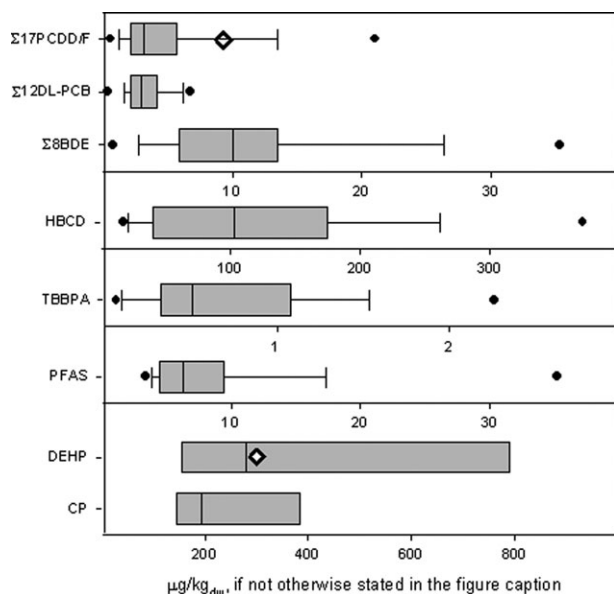
$\sum 17\text{PCDD/F}$  concentrations were not significantly different in the digestate and the compost. Concentrations varied slightly more in rural green waste composts compared with urban samples containing organic kitchen waste, but the median  $\sum 17\text{PCDD/F}$  concentration of the latter was slightly higher ( $3.3$  versus  $2.6 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$ ), which is in line with the literature.<sup>2</sup>

PCA revealed that the ratios of individual 17 toxic congeners to their sum, as well as the ratios of the sum of tetra-, penta-, hexa-, hepta-, and octaPCDD/F to their respective total sum, was different for sample nos. 1, 17 and 35 (ESI†, Fig. 3). Sample no. 1 had higher relative octaCDD (OCDD) content, which might be due to external contamination of the input material or OCDD-formation during composting from precursor compounds (*e.g.*, PCP) as described by Oeberg.<sup>21</sup> Unfortunately, PCP was not included in the pesticide screening and other precursor molecules are also possible. However, the mechanism of OCDD formation during composting seems controversial.<sup>22</sup> Furthermore, there was no indication (*e.g.*, different process, input material) as to why OCDD should be formed in sample no. 1 only. Sample nos. 17 and 35, originating from the same plant, had higher relative HexaCDF contents, as well as higher absolute concentrations. In this plant, small amounts of wood ash were composted which originated from a cogeneration plant burning native wood for energy production. However, PCDD/F levels of bottom wood ash were found to be low<sup>23</sup> and can therefore not account for the relatively high values. The longer the composting process lasted, the higher were the PCDD/F concentrations. This can be attributed to the relative persistence of PCDD/F compared with organic matter. However, for neither of the congeners nor for any of their sums was the increase in concentration significant.

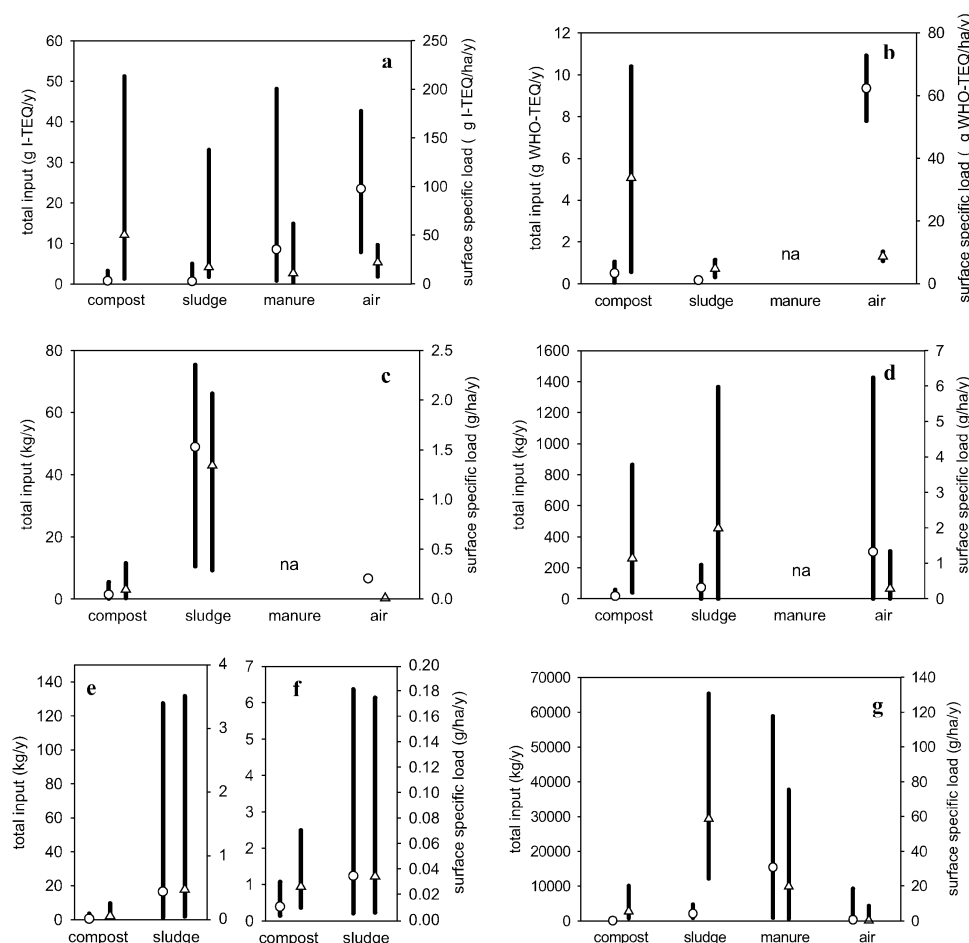
The  $\sum 17\text{PCDD/F}$  contents in Swiss agricultural soil and permanent grassland were at  $1.4 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$ ,<sup>24</sup> which is slightly lower than data for compost and digestate presented above. Atmospheric deposition was the most important input pathway for  $\sum 17\text{PCDD/F}$  to the total agricultural surface of Switzerland ( $24 \text{ g I-TEQ y}^{-1}$ , Fig. 2a). The total loads of sewage sludge and compost application were lower by almost two orders of magnitude. However, considering surface specific loads (*i.e.*, loads per hectare based on a standard fertilization), input by compost accounted for more than double the other inputs (Fig. 2a). The time to reach the Swiss trigger value for soil ( $5 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$  for  $\sum 17\text{PCDD/F}$ ),<sup>25</sup> solely by compost application and aerial deposition (accounting for 0.04% of the total surface specific load) in absence of any removal process was estimated to be 112 years (with the starting level in soil  $1.4 \text{ ng I-TEQ kg}_{\text{dw}}^{-1}$ ,<sup>24</sup> for deposition rate see ESI, Table S3†).

## DL-PCB

The median concentration of the  $\sum 12\text{DL-PCB}$  (sum of PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189) in composts and digestates was  $3.0 \text{ ng WHO-TEQ kg}_{\text{dw}}^{-1}$ , ranging from  $0.4$  to  $6.8 \text{ ng WHO-TEQ kg}_{\text{dw}}^{-1}$  (Fig. 1,  $n = 18$ , ESI, Table S2†). Kerst *et al.*<sup>26</sup> reported comparable median concentration in German compost ( $n = 22$ ). Analogous to PCDD/F and the



**Fig. 1** Concentrations of organic pollutants in compost and digestate (line: median; box: 25th and 75th percentile; lines with whiskers 10th and 90th percentile; dots: outliers; diamonds: median literature value (2); where missing: not available).  $\sum 17\text{PCDD/F}$  (sum of the 17 2,3,7,8-chlorosubstituted PCDD/F in  $\text{ng I-TEQ kg}_{\text{dw}}^{-1}$ ,  $n = 18$ ),  $\sum 12\text{DL-PCB}$  (sum of PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189 in  $\text{ng WHO-TEQ kg}_{\text{dw}}^{-1}$ ,  $n = 18$ ),  $\sum 8\text{BDE}$  (sum of PBDE 28, 47, 99, 100, 153, 154, 183, 209,  $n = 18$ ), HBCD (hexabromocyclododecane,  $n = 18$ ), TBBPA (tetrabromobisphenol A,  $n = 18$ ), PFAS (sum of 21 perfluorinated alkyl substances,  $n = 18$ ), DEHP (di-(2-ethylhexyl)phthalate,  $n = 6$ ) and CP (sum of short and medium chain chlorinated paraffins,  $n = 3$ ). For IRMM PCB and PAH see ref. 1.



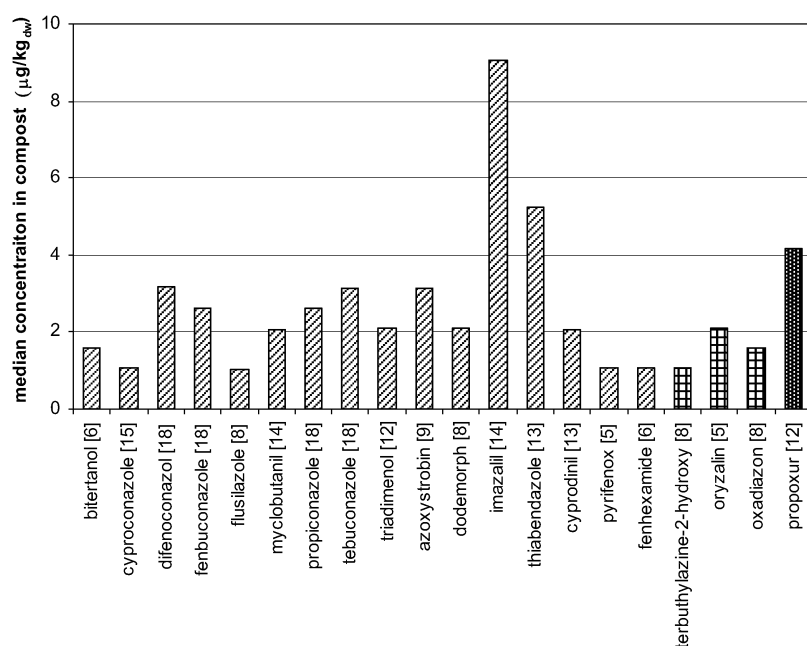
**Fig. 2** Average annual loads of organic pollutants from different sources: application of compost, sewage sludge (sludge), farmyard manure (manure) and atmospheric deposition (air) into agricultural soil: total loads to the Swiss agricultural area ( $\circ$ ), surface specific loads ( $\Delta$ ), bars represent minimum and maximum values, na: not available. (a)  $\Sigma 17\text{PCDD/F}$  (sum of the 17 2,3,7,8-chlorosubstituted PCDD/F), (b)  $\Sigma 12\text{DL-PCB}$  (sum of PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, 189), (c)  $\Sigma 8\text{BDE}$  (sum of PBDE 28, 47, 99, 100, 153, 154, 183, 209), (d) hexabromocyclododecane, (e) perfluorohexane sulfonate, (f) perfluorooctanoate to perfluorododecanoate (g) di-(2-ethylhexyl)phthalate. For assumptions/calculations, see text, for IRMM PCB and PAH see ref. 1. na: not available.

seven IRMM PCB,<sup>1</sup> concentrations of DL-PCB in compost and digestate were similar. Median values for all congeners were higher in urban samples containing organic kitchen waste in the input material compared with rural green waste compost. However, the difference was significant for PCB 126 only. In previous reports, PCB levels were found to be higher in urban composts, whereas the type of input material was not a determinant of their concentrations.<sup>1,2</sup> Consequently, the difference in DL-PCB concentration is rather attributed to the different origin of the input material than to its composition. The  $\Sigma 12\text{DL-PCB}$  correlated well with the  $\Sigma 7\text{PCB}$  (see ref. 1 for  $\Sigma 7\text{PCB}$  data,  $r^2 = 0.05$ ,  $p = 0.001$ ) except for sample no. 76 (green waste, rural). In this sample the  $\Sigma 7\text{PCB}$  was much higher compared with the  $\Sigma 12\text{DL-PCB}$  (ESI, Table S2†). It is assumed that this compost was subject to either PCB input from sources other than aerial deposition, *e.g.*, products containing PCBs in Aroclor-like mixtures, since these contained only small portion of certain DL-PCB congeners,<sup>27</sup> or aerial deposition from a local point source. Also, PCA of DL-PCB singled out sample no. 76 due to its lower contribu-

tion of PCB 105, 118 and 126 and higher ratios of PCB 156, 176 and 189 to  $\Sigma 12\text{DL-PCB}$  (ESI, Table S2†, Fig. 4). Lower PCB 156 and 176 and higher PCB 126 to  $\Sigma 12\text{DL-PCB}$  ratios support the hypothesis of input by Aroclor-like mixtures, but the pattern of PCB 105, 118, and 189 does not agree. No correlation was found between DL-PCB concentrations and the duration of the composting process.

No Swiss soil data for ( $\Sigma 12$ )DL-PCB could be found in the literature. Concentrations in Spanish forest soils were in the same range as in compost (between 1.9 and 8.4 ng WHO-TEQ kg<sub>dw</sub><sup>-1</sup><sup>28</sup>). The input of  $\Sigma 12\text{DL-PCB}$  to the total Swiss agricultural surface was dominated by aerial deposition (mean: 9.4 g WHO-TEQ y<sup>-1</sup>). Compost and manure application accounted for more than one order of magnitude less. As for PCDD/F, surface specific loads to soil by compost application can outweigh input by aerial deposition and manure application (Fig. 2b). However, aerial deposition and sewage sludge data on DL-PCB are sparse and their interpretation requires caution. Since DL-PCB and PCDD/F concentrations in compost were in the same range (assuming WHO-TEQ equals I-





**Fig. 3** Median pesticide concentrations in Swiss compost and digestate for compounds detected more than four times (fungicide ▨, herbicide ▩ and insecticide ▧). Median calculations were based on the number of detections out of 18, which are represented in brackets. Note that the first nine compounds are triazoles.

TEQ), the time for DL-PCB accumulation in soils to a particular level would be similar for both compound classes. Therefore, it would be useful to include DL-PCB in studies and legislation on PCDD/F.

### Brominated flame retardants

The median PBDE concentration (sum of BDE 28, 47, 99, 100, 153, 154, 183, 209,  $\Sigma$ 8BDE) in composts and digestates was  $10 \mu\text{g kg}_{\text{dw}}^{-1}$  (range:  $0.74\text{--}35 \mu\text{g kg}_{\text{dw}}^{-1}$ , Fig. 1,  $n = 18$ , ESI, Table S2†). The dominating compound was decaBDE, *i.e.*, BDE 209 (median  $7.3 \mu\text{g kg}_{\text{dw}}^{-1}$ ). It correlated significantly with the sum of the remaining PBDEs ( $r^2 = 0.58$ ,  $p = 0.0003$ ). Marb *et al.*<sup>29</sup> reported a median value of  $12 \mu\text{g kg}_{\text{dw}}^{-1}$  for the sum of tri- to heptaBDE in compost ( $n = 12$ ). Unfortunately, compound specific comparison is not possible, since only the total sum was reported. Median concentrations of pentaBDE and octaBDE in Swiss composts and digestates, calculated according to Morf *et al.*,<sup>30</sup> were  $1.9 \mu\text{g kg}_{\text{dw}}^{-1}$  and  $0.2 \mu\text{g kg}_{\text{dw}}^{-1}$ , respectively. This is in line with the estimated diffusive emission patterns in Switzerland (emission:  $1.9 \text{ t y}^{-1}$  and  $0.37 \text{ t y}^{-1}$ , respectively).<sup>31</sup> DecaBDE emission data ( $2.1 \text{ t y}^{-1}$ ) was in the same range as pentaBDE,<sup>31</sup> but respective concentrations in compost were much higher. This might reflect increasing production of decaBDE at the beginning of the third Millennium.<sup>32</sup> There was a weak correlation between  $\Sigma$ 8BDE and  $\Sigma$ 12DL-PCB ( $r^2 = 0.35$ ,  $p = 0.01$ ), pointing at similar input pathways to compost, *e.g.*, aerial deposition.

Octa- and decaBDE levels were slightly higher in digestate than in compost. As for  $\Sigma$ 12DL-PCBs, higher  $\Sigma$ 8BDE concentrations were found in urban compost and digestates containing organic kitchen waste compared with rural green waste compost ( $11 \text{ versus } 5.2 \mu\text{g kg}_{\text{dw}}^{-1}$ ). This may reflect higher

PBDE concentrations in urban areas<sup>33</sup> or higher input *via* organic kitchen waste. However, the difference was significant for BDE 183 only. PCA revealed a considerably different BDE pattern in sample no. 28 (ESI, Fig. S5†), which had a high concentration of BDE183 relative to  $\Sigma$ 8BDE. This may be due to contamination by commercially produced octaBDE, in which BDE183 is the major congener.<sup>34</sup> No correlation was found between any of the BDE congeners and the duration of the composting process.

PBDE concentrations in compost were higher than in Swedish soil (pentaBDE:  $122 \text{ ng kg}_{\text{dw}}^{-1}$ , octaBDE: below quantification limit, decaBDE:  $480 \text{ ng kg}_{\text{dw}}^{-1}$ <sup>35</sup>), but congener patterns were similar (no data for Switzerland available).

Even though data are limited, it is suggested that total and surface-specific loads of PBDE to soil by compost application are low compared with other inputs (Fig. 2c). This contrasts with the findings for PCDD/F, PCB and PAH (see above and ref. 1). A possible explanation is additional input sources of PBDE to sewage sludge (*e.g.*, leaching from building materials *via* surface runoff), whereas for the other compounds aerial deposition plays the most important role. However, since PBDE are still released to the environment, increasing levels in most environmental compartments, including compost and soil, may be anticipated.

The median HBCD and TBBPA concentrations in compost were 100 and  $0.51 \mu\text{g kg}_{\text{dw}}^{-1}$ , respectively (Fig. 1,  $n = 18$ , ESI, Table 2†). Even though the European market demand in 2001 of all three classes of BFR was in the same range (PBDE:  $8360 \text{ t y}^{-1}$ , HBCD:  $9500 \text{ t y}^{-1}$ , TBBPA:  $11600 \text{ t y}^{-1}$ ),<sup>32</sup> compost contained significantly more HBCD than TPPBA. This profile has previously been observed in sewage sludge<sup>36</sup> and is explained by the different ways that HBCD and TBBPA are incorporated in products (HBCD as additive, whereas

TBBPA is mainly covalently bound).<sup>30</sup> Concentrations of HBCD and TBBPA were slightly higher in digestate (170 and 1.0  $\mu\text{g kg}_{\text{dw}}^{-1}$ , respectively) than in compost (47 and 0.5  $\mu\text{g kg}_{\text{dw}}^{-1}$ , respectively), contrasting with a laboratory study in which HBCD was found to degrade faster under anaerobic than under aerobic conditions.<sup>37</sup> However, higher concentrations in the digestate input material might have caused this difference

as well. Similar to PBDEs, urban compost containing kitchen waste had significantly higher TBBPA concentrations than rural green waste compost (0.63 *versus* 0.11  $\mu\text{g kg}_{\text{dw}}^{-1}$ ), whereas for HBCD no significant difference was identified. No correlation was found between HBCD or TBBPA concentrations and the duration of the composting process. We are not aware of any soil data on HBCD and TBBPA for comparison.

As for PCDD/F, DL-PCB (see above) and PAH,<sup>1</sup> aerial deposition dominated the input of HBCD to total agricultural soil. The surface specific load of HBCD by compost application was estimated to be between input by sewage sludge (2.0  $\text{g ha}^{-1} \text{y}^{-1}$ ) and aerial deposition (0.3  $\text{g ha}^{-1} \text{y}^{-1}$ , Fig. 2d) as found for PBDE. Data for TBBPA input estimation was not available.

## PFAS

The median PFAS concentration (sum of 6:2 fluorotelomer sulfonate (6:2 FTS), saturated/unsaturated fluorotelomer carboxylates (FT(U)CA), perfluorinated sulfonates (PFS), perfluorinated carboxylates (PFCA), fluorooctane sulfonamides (FOSA), and fluorooctane sulfonamidoethanols (FOSE)) was 6.3  $\mu\text{g kg}_{\text{dw}}^{-1}$ , ranging from 3.4 to 35  $\mu\text{g kg}_{\text{dw}}^{-1}$  (Fig. 1,  $n = 18$ , and ESI, Table S2†). 6:2 FTS, which is the degradation product of recent replacement compounds for perfluorooctane sulfonate (PFOS) in, for example, fire fighting foams, was detected in nine out of 18 samples (median 1.2  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 9$ ). Other FTS and FT(U)CA (degradation products of fluorotelomer alcohols) were not found. Except for perfluorobutane sulfonate (PFBS, another successor of PFOS), all analysed PFS were detected. Their sum concentration varied between 1.0 and 24  $\mu\text{g kg}_{\text{dw}}^{-1}$ , with a median at 2.3  $\mu\text{g kg}_{\text{dw}}^{-1}$  ( $n = 18$ ). Perfluorinated hexa- to dodecanoates were also found in compost (median concentration of the sum of PFCA: 2.8  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 18$ ). Even carbon number PFCA were found in higher concentrations than their odd-numbered homologues, which might be an indication of near-source contamination with input from PFCA manufactured both by telomerisation and electrochemical fluorination. The opposite (odd C number > even C number) was found in Arctic biota,<sup>38</sup> indicating that degradation of fluorotelomer alcohols or other precursor compounds could be the reason for the altered pattern.

No difference in PFAS concentration was found between digestate and compost and between urban organic kitchen waste samples compared with rural green waste composts. No sample was singled out in PCA of the relative contribution of the sums of 6:2 FTS, PFS, PFCA, and perfluorooctane sulfonamide to the total sum (ESI, Fig. S6†). This may indicate a uniform distribution of these compounds. As for

PCDD/F, weak but significant positive correlations of PFS ( $r^2 = 0.27$ ,  $p = 0.03$ ) with the duration of the composting process were observed.

No data on PFAS in compost and soil could be found for comparison. The PFS (perfluorohexane sulfonate, PFOS and perfluorodecane sulfonate) and PFCA (perfluorooctanoate to perfluorododecanoate) concentrations presented here in compost were between the concentrations in sludge and surface sediment from the USA.<sup>39</sup> The total and surface specific loads for PFAS seemed to be lower for compost than for sewage sludge application (Fig. 2 e–f). However, data is sparse or non-existent (deposition data, concentrations in manure) and no final conclusion can be drawn.

## Pesticides

Out of the 269 pesticides (86 fungicides, 86 herbicides, 92 insecticides, five acaricides, one nematicide) that were analysed, 30 fungicides, 14 herbicides, eight insecticides, and one acaricide were detected in compost and digestate (Fig. 3, ESI Tables S2 and S4†). The consumption data on pesticide use available for Switzerland<sup>40</sup> was not reflected in compost. The pesticide half-lives in soil<sup>41</sup> seemed to be related to their occurrence (*i.e.*, compounds with longer half-lives were detected more often).

Fungicides dominated over the other pesticides, not only in terms of incidence of detection (see above), but also of concentrations (median total concentration of fungicides: 42.4  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 18$ ). Highest median concentrations were found for imazalil (9.0  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 13$ ) and thiabendazole (5.3  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 14$ , Fig. 3). These typical post-harvest fungicides are often found on tropical fruits.<sup>42</sup> The median thiabendazole concentration in Swiss compost was slightly lower than in the literature (7  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 23$ ).<sup>2</sup> Triazoles dominated among the fungicides (19 analysed, 13 detected, median of the sum of triazoles: 21.6  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 18$ ). Difenconazole, fenbuconazole, propiconazole, and tebuconazole were detected in all compost samples. These compounds are widely used in viticulture, orchards and horticulture in Switzerland and other countries. They are frequently detected in fruit and vegetables and some of them exhibit half-lives of more than 40 days in soil.<sup>41</sup> Compost and digestate did not differ in fungicide concentrations. The sum of all fungicides was significantly higher in urban composts and digestates containing organic kitchen waste (median: 61  $\mu\text{g kg}_{\text{dw}}^{-1}$ ) compared with rural green waste compost (median: 27  $\mu\text{g kg}_{\text{dw}}^{-1}$ ). Rather than by the origin of the input material, this is explained by higher contamination of organic kitchen waste. In general, numbers and concentrations of fungicides detected in compost decreased with the duration of the composting process. However, respective correlations were weak and not significant ( $p = 0.11$  and 0.15, respectively). Urban samples containing organic kitchen waste exhibited significantly higher median fungicide concentration in winter (90  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 4$ ) compared with summer (64  $\mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 6$ ). This is explained by the higher amount of tropical fruits (often treated with fungicides, especially imazalil and thiabendazole, see above) in kitchen waste in winter.<sup>42</sup>

The median herbicide concentration was 5.8  $\mu\text{g kg}_{\text{dw}}^{-1}$  ( $n = 13$ ). Three herbicides (or herbicide metabolites,

respectively) were detected in more than four compost samples (oxadiazon, median:  $1.6 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 8$ ; terbutylazine-2-hydroxy, median:  $1.1 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 8$ ; and oryzalin, median:  $2.1 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 5$ ). Oxadiazon is not registered in Switzerland, whereas the others are listed in the Swiss phytosanitary product index.<sup>43</sup> The lower abundance of herbicides compared with fungicides in compost can be explained by the fact that herbicides are mainly used at the beginning of the growing season in horticulture and should be mostly degraded by the time of harvest. In contrast, fungicides are applied later during the growth season, or for post-harvest treatment. Other agricultural residues that might contain herbicides (*e.g.*, straw, grass) hardly end up in Swiss commercial composting plants. There was no difference in herbicide concentration in compost and digestate. Median herbicide concentrations in urban samples with organic kitchen waste were significantly higher ( $8.5 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 9$ ) than in rural green waste compost ( $1.1 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 3$ ). Herbicide application is banned in traffic areas in Switzerland, and since it can be assumed that herbicide concentrations in kitchen waste are low (see above), it is concluded that urban green waste is more contaminated with herbicides than the rural equivalent (probably due to higher application rates in, for example, private gardens and public green areas).

The median concentration of the sum of all insecticides was  $4.1 \mu\text{g kg}_{\text{dw}}^{-1}$  ( $n = 18$ ). The only insecticide detected in several composts was propoxur (median  $4.2 \mu\text{g kg}_{\text{dw}}^{-1}$ ,  $n = 12$ ). Propoxur is not used for agricultural purposes, but as a biocide in numerous commercial products to control insect pests in food storage areas, houses, animal shelters, *etc.* The important contribution of propoxur to the total sum of insecticides points to a high consumption, since it is neither particularly hydrophobic ( $\log K_{\text{ow}} 1.56^{41}$ , which is at the lower end of all the compounds detected), nor very persistent (readily degradable<sup>41</sup>). In contrast to former studies,<sup>3</sup> organochlorine insecticides were not detected in composts and digestates. No Swiss data on the detected pesticides in soil are available. Any comparison with soil data from other countries is problematic, since the use of pesticides varies considerably between them. Information on deposition rates obtained from Bucheli *et al.*<sup>44</sup> showed that total and surface specific loads of fungicides by compost application ( $0.001 \text{ kg y}^{-1}$  and  $0.05 \text{ mg ha}^{-1} \text{ y}^{-1}$ ) were at least a factor 20 below aerial deposition ( $1.1 \text{ kg y}^{-1}$  and  $1 \text{ mg ha}^{-1} \text{ y}^{-1}$ ), and several orders of magnitude lower than input from direct application (mean input of fungicides detected in this study:  $200 \text{ g ha}^{-1} \text{ y}^{-1}$ , range  $40\text{--}900 \text{ g ha}^{-1} \text{ y}^{-1}$ , Swiss phytosanitary product index<sup>43</sup>). For herbicides, surface specific loads of aerial deposition and compost application differed by a factor of 1500. However, as for fungicides, application on agricultural crops is considered to be by far the most important input pathway.

### Phthalates, nonylphenol and chlorinated paraffins

The median di(2-ethylhexyl)phthalate (DEHP) concentration was  $280 \mu\text{g kg}_{\text{dw}}^{-1}$  ( $n = 6$ ), corresponding well with literature values ( $300 \mu\text{g kg}_{\text{dw}}^{-1}$ ).<sup>2</sup> This is clearly below the limit of the Danish Statutory Order ( $50 \text{ mg kg}_{\text{dw}}^{-1}$ <sup>45</sup>). The highest value ( $1990 \mu\text{g kg}_{\text{dw}}^{-1}$ ) was detected in an urban digestate sample

containing organic kitchen waste, as well as fruit and vegetable residues from grocery stores, in which also dibutyl phthalate (DBP) was detected ( $105 \mu\text{g kg}_{\text{dw}}^{-1}$ ). These elevated phthalate levels might be due to plastic residues in the input material originating from fruit and vegetable wrapping. DEHP concentrations in agricultural soils were observed to be in the same range as concentrations in compost ( $300\text{--}700 \mu\text{g kg}_{\text{dw}}^{-1}$ ).<sup>46</sup> Total and surface specific loads of DEHP to soil by compost application were estimated to be below the input by sewage sludge or manure application (Fig. 2g).

Even though detected at considerable concentrations in German apples and tomatoes,<sup>47</sup> nonylphenol was not detected in compost. Possible reasons are the relatively high detection limit ( $1 \text{ mg kg}_{\text{dw}}^{-1}$ ) and/or rapid degradation during composting.<sup>48</sup>

CP were analysed and detected in three samples. Concentrations were between  $57\text{--}140 \mu\text{g kg}_{\text{dw}}^{-1}$  for SCCP ( $\text{C}_{10}\text{--}\text{C}_{13}$ ) and  $29\text{--}245 \mu\text{g kg}_{\text{dw}}^{-1}$  for MCCP ( $\text{C}_{14}\text{--}\text{C}_{17}$ ). These concentrations were lower than the qualitative concentrations in Swedish source-separated organic waste<sup>49</sup> and values in sewage sludge,<sup>50</sup> but in the same range as Swiss soils.<sup>51</sup> This resulted in total and surface specific loads of SCCP and MCCP to soil of 5 and 11%, respectively, by compost compared with sewage sludge application.

## Conclusion

This paper presents the most comprehensive dataset on organic priority pollutants in compost, and—for the first time—digestate. Several of the investigated compound classes have not been analysed previously, and most of them could be quantified in concentrations at or above those found in background soils. On a national scale, input of organic contaminants to agricultural soils by compost application seemed to be limited. However, similar to PAH,<sup>1</sup> loads of PCDD/F and DL-PCB to actually treated areas could be of concern. This seemingly negative characteristic of compost application needs to be counterbalanced with its apparent and well-documented beneficial aspects, such as soil improving properties and the sustainable management of natural resources. Further research is needed to appoint relevant sources of organic pollutants in compost, to identify possible stable degradation products and to assess to what extent they are biologically available.

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