

Adaptation of diffusive gradients in thin films technique to sample organic pollutants in the environment: An overview of o-DGT passive samplers

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1	Adaptation of diffusive gradients in thin films technique to sample organic pollutants in
2	the environment: an overview of o-DGT passive samplers
3	
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9	ABSTRACT
10	The adaptation of the diffusive gradients in thin films technique (DGT) to sample organic pollutants in
11	the environment, called o-DGT has been performed since 2011 for various types of organic compounds
12	(e.g. pesticides, pharmaceuticals, hormones, endocrine disrupting chemicals, household and personal care
13	products). To sample these different compounds, configuration of the samplers (mainly receiving phase
14	and diffusive gel) has to be adapted. Up-to-date, sampling of 142 organic compounds by this passive
15	sampler have been tested. This review provides the state-of-art of o-DGT passive sampler development,
16	describing theory and modelling, calibration, configuration of the devices, and field applications. The most
17	used configurations were agarose-XAD-18 and agarose-HLB configuration. o-DGT can be used to
18	sample soils and most of natural waters (range of pH 4-9 and ionic strength 0.001-0.1 M).
19	This review discusses current limitation of o-DGT in light of the feedback of DGT use to sample
20	inorganic contaminants. It mainly concern the low sampling rates currently obtained by o-DGT compared
21	to other passive samplers. This weakness could be compensated in the future with new sampler's design
22	allowing an increase in exposure area.
23	
24	KEYWORDS
25	o-DGT; passive sampler; organic compounds; diffusive gradients in thin films (DGT); monitoring
26	

27 INTRODUCTION

28 Considering its low cost and simplicity, grab sampling is commonly performed to estimate concentration 29 of micropollutants in waters (Allan et al., 2006). However, this technique has some limitations such as the 30 large volume of water required for concentration of trace pollutants in order to comply with analytical 31 sensitivity or the lack of temporal representativeness (Allan et al., 2006). Complementarily to this 32 technique, passive sampling provides in situ pre-concentrated samples and allows access to time-weighted 33 average concentration (TWAC), also called Cw (concentration in water). Passive samplers consist basically 34 in a binding phase able to concentrate targeted compounds within various devices deployed in the 35 environment. Organic contaminants in waters can be sampled by Polar Organic Chemical Integrative 36 Sampler (POCIS) (Alvarez et al., 2004), Chemcatcher® (Kingston et al., 2000), Semi-Permeable Membrane 37 Device (SPMD) (Huckins et al., 1990) or Membrane-Enclosed Sorptive Coating (MESCO) (Paschke et al., 38 2006). However, as shown for some devices (e.g. POCIS or SPMD), passive sampling can be affected by 39 environmental factors (Fauvelle et al., 2017; Harman et al., 2012) such as temperature, biofouling or water 40 flow velocity. Consequently, sampling rates (Rs) needed for TWAC estimations can vary between the 41 studied systems (Alvarez et al., 2004; Buzier et al., 2019; Li et al., 2010; Togola and Budzinski, 2007). 42 Given that sampling rates calibration is expensive and time consuming, their determination is not 43 optimized for each targeted system. Consequently, laboratory-determined sampling rates corresponding to 44 generic conditions are usually used for TWAC estimation and inaccuracies may arise (Buzier et al., 2019). 45 Indeed, Poulier et al., (2014) demonstrated that POCIS passive sampling technique is a semi-quantitative 46 method with an error on TWAC of a factor *c.a.* 2.

47 Similarly to DGT (diffusive gradients in thin films technique) passive sampler for inorganic compounds 48 (Davison and Zhang, 1994), Bondarenko et al., (2011) firstly published the introduction of a diffusive 49 layer in a device to passively sample organic compounds. The presence of a diffusive layer (hydrogel) 50 constrains compounds mass transfer during sampling mostly to diffusion within this layer. Consequently, 51 the device's sampling rate mostly derive from the mass transfer rate imposed by this limiting step. The 52 influence of environmental conditions on sampling rates using such device configuration are therefore 53 limited, compared to standard configurations not using a diffusive layer. The first use of passive sampler 54 devices incorporating a diffusive hydrogel to sample organic compounds in water was reported in 2012 55 (Chen et al., 2012), under the name "o-DGT". This adaptation of DGT to organic compounds sampling 56 mainly consists in changing the binding phase. Since the first adaptation, there is a growing interest for o-57 DGT (Figure 1) and adaptation to various organic compounds have been proposed (pesticides, 58 pharmaceuticals, hormones, endocrine disrupting chemicals and household and personal care products). 59 Among these published articles, 75% of studies concern devices development (tests on binding phases, 60 elution, robustness or analyte conservation). Application of o-DGT in waters or soils is the aim of 17% of 61 other articles and comparison between POCIS and o-DGT samplers is performed by two articles. A 62 review (Gong et al., 2018), published in January 2018, compared the efficiency of three passive samplers 63 for organic compounds: POCIS, o-DGT and Chemcatcher® but with only 12 articles discussed on o-64 DGT.

65 This review proposes an overview of o-DGT passive sampler from its first report in 2011 to the present.
66 Theory, configurations, calibrations, robustness and field applications of the sampler are extensively
67 detailed. This review also discusses its current limitations and future development needed in light of the
68 knowledge already accumulated for inorganic compounds.

69

70 THEORY AND MODELLING

71 Similarly to the initial DGT samplers, o-DGT are usually composed of two hydrogels: a diffusive gel 72 covering a binding gel. A microporous membrane can be added to protect the diffusive gel against 73 particles from the sampled medium. The binding gel is separated from the solution by the diffusive gel 74 and a diffusive boundary layer (DBL) is created at the water/sampler interface due to water viscosity 75 (Figure 2). Mass transfer from solution to the binding gel is constrained to diffusion only and can be 76 modeled using Fick's first law. For simplicity, modelling is commonly made under five assumptions: i) 77 absence of interaction between analyte and diffusive gel, ii) concentration at the interface between the 78 binding and diffusive gel is negligible (i.e. total and irreversible binding within the receiving phase), iii) time 79 to reach steady-state is negligible, iv) diffusive boundary layer thickness is negligible and v) lateral diffusion 80 is negligible. In these conditions, the flux density (φ) can be expressed by Eq. 1 (Davison and Zhang, 81 1994):

$$\varphi = \frac{D \times TWAC}{\Delta_g} \qquad \qquad \text{Equation 1}$$

where D is the diffusion coefficient of the analyte in the diffusive gel (compound and temperature
dependent) and TWAC is the concentration of the targeted analyte in studied environment (water). Flux
density can also be defined by the following equation Eq. 2:

86
$$\varphi = \frac{m}{A \times t}$$
 Equation 2

where *m* is the mass of the analyte in the binding gel, \mathcal{A} is the exposure area between diffusive gel and solution and *t* is the exposure time. Combining equations (1) and (2), the concentration in the studied environment (water) can be quantified by Eq. 3:

90
$$TWAC = \frac{m \times \Delta_g}{t \times D \times \mathcal{A}}$$
 Equation 3

Given that flux density in the sampler will vary proportionally with the analyte concentration variations inthe exposure medium (Eq. 1), any exposure concentration determined with Eq. 3 is in fact a TWAC.

93 Considering exposure area, exposure time and diffusion layer thickness are known parameters and mass of
94 the analyte is determined following elution of the binding gel, diffusion coefficient within the diffusive gel
95 is the only parameter requiring calibration (see next section) for TWAC estimation. Such modelling allows
96 therefore to free TWAC calibration from any environmental condition not affecting diffusion within the
97 sampler such as flow velocity.

98 Eq. 3 should be convenient for most cases. Indeed, assumption i) and ii) are not environment dependent 99 and are usually checked previously during the initial development of the sampler. Assumption iii), iv) and 100 v) have never been validated for organic compounds but their behavior should be similar to inorganic 101 compounds, considering their diffusion coefficient is about one order of magnitude different at worst in most cases (*i.e.* values of $\approx 10^{-7}$ - 10^{-6} cm² s⁻¹ for organics, see **Appendix 1**, *versus* values of $\approx 10^{-6}$ cm² s⁻¹ for 102 103 most inorganics according to DGT Research website). For inorganic compounds, assumption iii) was 104 shown to holds for deployments ≥ 24h (Davison and Zhang, 2012). Warnken et al., (2006) shows that 105 assumption iv) and v) are not strictly valid but the errors from each cancel each other out for standard 106 devices as long as DBL (Diffusive Boundary Layer) thickness remains limited (*i.e.* valid for flow velocity > 107 2cm s⁻¹, (Gimpel et al., 2001)). Therefore, Equation 3 should fail only for a limited number of systems, 108 mostly the ones displaying very low flow conditions. Indeed, Belles et al., (2018) and Buzier et al., (2019) 109 demonstrated for various organic compounds the validity of Eq. 3 in flowing conditions (reported flow 110 velocities ≥ 2.5 cm s⁻¹ for the latter). However, Buzier et al., (2019) reported significant inaccuracies in 111 quiescent conditions (between 30 and 70% depending on the analyte), arising from the formation of a 112 significant DBL. To avoid such inaccuracy, more sophisticated models have been developed for inorganic 113 compounds to consider both DBL thickness and lateral diffusion in order to avoid making assumption iv) 114 and v) (Garmo et al., 2006; Santner et al., 2015). An advanced estimation of TWAC could be calculated 115 using Equation 4 that considers the thickness of the DBL (δ) and lateral diffusion within the sampler 116 (Santner et al., 2015):

117
$$TWAC = \frac{m}{k_{ld}A_g t} \left(\frac{\Delta_g}{D} + \frac{\delta}{D^w}\right) \qquad \text{Equation 4}$$

118 where k_{ld} is the lateral diffusion flux increase coefficient and D^{w} is the diffusion coefficient in water.

However, their use requires deployment of devices with various diffusive gel thickness and adaptation of data treatment. An evaluation of such models for organic compounds is found in Buzier et al., (2019) and suggests that their use currently allows limiting inaccuracies for some compounds in quiescent systems but also increase inaccuracies for flowing systems. Such limitation was attributed to limited accuracy of the additional parameters required compared to Eq. 3 (*i.e.* lateral diffusion, diffusion coefficient in water). Such procedure is therefore promising but still required further developments for organic compounds.

125

126 DETERMINATION OF DIFFUSION COEFFICIENTS

127 Diffusion coefficient accuracy is a key factor for o-DGT as it induces most part of TWAC's uncertainty 128 (estimated at ≈ 23%) (Belles et al., 2018). Experimental determination of diffusion coefficients in the 129 diffusive gel can be performed using three method: i) the diffusion cell method, ii) by fitting Equation 3 130 following devices deployment in controlled solution, iii) the stack of gels method. The first two methods 131 have been used for long for inorganic compounds (Zhang and Davison, 1999) whereas the last one is a 132 method adapted from Rusina et al., (2010).

133 The diffusion cell method was the most use (80%). A diffusion cell device (see Figure 3) is composed of 134 two separated compartments connected with an opening where a diffusive gel is intercalated and allows 135 mass transfer between the two compartments by diffusion. One of the compartments ("source" 136 compartment) is filled with a solution spiked with the analyte of interest whereas the other compartment 137 ("receiving" compartment) is filled with the same solution but not spiked with the analyte. The analyte 138 diffuse through the diffusive gel and a steady state is established after few minutes. Concentration in the 139 receiving compartment is determined over time in order to determine the analyte flux through the 140 diffusive gel and to derive the corresponding diffusion coefficient using Fick's first law (Equation 1).

141

142 The second method (ii) uses time series deployment of o-DGT samplers in known spiked solution. 143 Accumulation of analyte into the binding gel versus time is determined allowing back calculation of the 144 diffusion coefficient using Equation 3. In contrast to the first method, this one allows the use of lower 145 concentrations ($\mu g L^1$ or less) that are more relevant compared to the targeted environmental applications. 146 However, it includes the sorption step on the binding phase and allows determination of an "effective" 147 diffusion coefficient rather than a "physical" diffusion coefficient. Guibal et al., (2017) compared the two 148 methods for four anionic pesticides. They found good agreement between the two methods for two 149 compounds whereas for the two others they measured higher D values (25 and 35%) with the diffusion 150 cell method. Zou et al., (2018) and Guan et al., (2018) found good agreement (4 - 11%) between the two 151 methods for all compounds tested (6 organophosphorus flame retardants and two perfluoroalkyl 152 substances, respectively). When inorganic compounds are considered, Shiva et al., (2015) also reported 153 good agreement between the two methods for nine elements over thirteen. Finally, they advise to favor 154 the second method because of the issue of concentration level relevance.

The last method (iii), adapted from Rusina et al., (2010), was only recently used by Amato et al., (2018) and Belles et al., (2017). Unspiked diffusive gels are stacked with one spiked with the targeted analytes. Analytes diffuse from spiked to unspiked gels and are quantified over time (analyze of unspiked diffusive gels). In contrast, to the two previous methods, the system used do not reach a steady state and Eq. 1 is not valid. Diffusion coefficients are therefore derived using known solutions to Fick's first law for the specific boundary conditions imposed with this method (*e.g.* analyte initially homogeneously distributed across a section of constant surface area, **Equation 5**, (Amato et al., 2018)).

162
$$C(x,t) = \frac{m}{A\sqrt{4\pi D}}e^{-(\frac{x^2}{4Dt})}$$
 Equation 5

163 Where C(x, t) is the analyte concentration in unspiked diffusive gels at a distance x from spiked diffusive 164 gel after a time t. This method allows determination of diffusion coefficient without involving the 165 sorption step similarly to the diffusion cell method but requires no specific material (*i.e.* diffusion cell 166 device). Amato et al., (2018) compared this method and diffusion coefficient measurement by diffusive 167 cell for carbamazepine, diuron and isoproturon. They measured higher D values (30, 18 and 25%, 168 respectively) compared to the diffusion cell method.

Modeling of diffusion coefficient has also been tested. Chen et al., (2013) and Challis et al., (2016) derived diffusion coefficient from molecular weight and diffusive gel porosity but found that modelled values were overestimated compared to measured values. A simple linear relationship between LogP and diffusion coefficient was obtained by Zou et al., (2018) for organophosphorus flame retardants with a good determination coefficient (0.98). However, only 5 diffusion coefficients were used to determine the linear relationship and extrapolation to other compounds is questionable. Modelling of D values, although interesting since it can avoid time consuming laboratory work, still requires some developments.

176 Diffusion coefficients are temperature dependent and can be corrected using the Stoke-Einstein equation177 (Eq. 6):

178
$$\frac{D_1 T_1}{\eta_1} = \frac{D_2 T_2}{\eta_2}$$
 Equation 6

179 where *T* is the temperature and η the water viscosity. When 25°C is taken as the reference condition, Eq. 6 180 is derived into equation (7) and used to calculate diffusion coefficient at the desired temperature (Zhang 181 and Davison, 1995) :

182
$$log D_{T_2} = \frac{1.37023 \times (T_2 - 25) + 8.36 \times 10^{-4} \times (T_2 - 25)^2}{109 + t_2} + log \frac{D_{25} \times (273 - T_2)}{298}$$
 Equation 7

183 Challis et al., (2016) reported for various organic compounds good agreement (typically within 20%)184 between measured diffusion coefficients and corrected ones with Eq. 7.

185

186 STUDIED COMPOUNDS

187 Sampling of 142 compounds from different action families have been tested: pharmaceuticals, hormones,
188 illicit drugs, bisphenol, household products, personal care products, organophosphorus flame retardants,

189 nitrophenols, perfluoroalkyl substances, endocrine disrupting chemicals and pesticides. Pharmaceuticals

190 were the most studied compounds (50% of the studied compounds) followed by pesticides with 20% of 191 studied compounds. The list of compounds is presented in Appendix 1. They display a wide diversity of 192 chemical properties. Acidic (e.g. glyphosate with pKa = - 0.6 (Weng et al., 2019), mecoprop with pKa = 193 3.5 (Guibal et al., 2017)), neutral (e.g. bisphenol (Chen et al., 2018), propranolol (Challis et al., 2016)) and 194 basic (e.g. estriol with pKa = 10.3 (Chen et al., 2018), amphetamine with pKa = 10.0 (C. Guo et al., 2017)) 195 compounds have been sampled by o-DGT. Compounds with a wide range of hydrophobicity (-4.54 < 196 LogP < 7.51) and a wide range of molecular weight (128.17 g mol⁻¹ for naphthalene to 916.11 g mol⁻¹ for 197 tylosin) were investigated; oxytetracycline being the most polar and salinomycin the most hydrophobic 198 compound. Given the wide range of chemical properties displayed by the targeted compounds, it is 199 necessary to check their ability to bind to the receiving phase and to diffuse through the diffusion gel. 200 Moreover, considering the low solubility of the most hydrophobic compounds, adequacy between 201 sampling rate and targeted concentrations has to be considered to allow their quantification.

HLB and XAD18 binding phases were studied for a wide range of compounds with investigations on
sampling of 64 and 52 different compounds, respectively. The most studied compound was the antibiotic
sulfamethoxazole (13 articles), sulfonamide family being the most widely studied with 19 compounds. For
this pharmaceutical family, five receiving phases were found suitable: XAD18, HLB, XDA-1, Sepra ZT
and porous carbon material (PCM).

Sampling of each compound with o-DGT will be characterized by a diffusion coefficient within the diffusive gel. These diffusion coefficients are detailed in **Appendix 1**. Compared to metals, diffusion coefficients of organic compounds are usually lower because of volume difference but are about one order of magnitude different at worst (*i.e.* 10⁻⁷-10⁻⁶ cm² s⁻¹). For a given compound, with an identical sampler configuration, difference between diffusion coefficients determined by two different authors was lower than a factor 1.4 (except for ciprofloxacin with 2.4 factor difference). Average difference for diffusion coefficients of a given compound was about 1.2.

214

215

216 SAMPLER CONFIGURATION

o-DGT samplers are basically prepared with up to three constituents: a binding gel, a diffusive gel and an
optional protective membrane. A broad range of targeted analytes were tested to be sampled by o-DGT
and the sampler configuration mainly depends on the analytes of interest (see appendix 1). All published
configurations are displayed in Table 1 and detailed below.

221

222 Binding phase.

223 For pharmaceutical compounds, 14 binding phases have been tested (listed in Table 2). Only 6 binding 224 phases were finally used: XAD18, HLB, Sepra ZT, PCM, XDA-1 and nanoZnO. The XAD18 was the 225 most used as binding phase for o-DGT to sample pharmaceutical compounds. This binding phase was 226 used in 8 articles (Chen et al., 2015b, 2015a, 2014, 2013, 2012; D'Angelo and Martin, 2018; D'Angelo and 227 Starnes, 2016; Zhang et al., 2018). Among four different binding phases (Oasis® HLB, activated charcoal, 228 MCX and XAD18), XAD18 was selected by Zhang et al., (2018). The others binding phases were not 229 selected because poor adsorptions were obtained for HLB phase and elutions from activated charcoal or 230 from MCX gels were not efficient for some analytes (methcathinone and ephedrine).

231 The second binding phase the most used to sample pharmaceuticals was HLB (6 articles (Amato et al., 232 2018; Buzier et al., 2019; Challis et al., 2018b, 2018a, 2016; Stroski et al., 2018)). Four others binding 233 phases were used by four authors (Sepra ZT (Stroski et al., 2018), PCM (Ren et al., 2018), XDA-1 (Xie et 234 al., 2018a) and nanoZnO (You et al., 2019b)). Before selecting XDA-1, Xie et al., (2018a) have tested 8 235 binding gels: non-polar phases: XDA-1, LX-1180, XDA-600, LX-4027 and XAD18; ion exchange resin: 236 D296; polar phases: NKA-9 and medium polar phases CAD-40. XDA-1 and LX-4027 had greater binding 237 capacity than the others receiving phases. However, LX-4027 did not distribute evenly in the hot agarose 238 was consequently not selected.

For pesticides, 9 binding phases have been tested (Table 2) and 7 were finally used: TiO₂, cyclodextrine
polymer, XDA-1, Strata-X, HLB, activated carbon and Sepra ZT. Only three authors have tested different
binding phase. Guibal et al., (2017) tested two phases (Oasis[®] HLB and Oasis[®] MAX). Oasis[®] HLB phase

- 242 was slightly better than Oasis[®] MAX when o-DGT were deployed in natural waters. Stroski et al., (2018)
- 243 compared HLB and Sepra ZT binding phases and concluded that Sepra ZT binding phase was easy to set
- 244 up because it offered improvements on current-use designs and a more cost effective and widely available

binding resin. The same arguments were used by Bondarenko et al., (2011) to support the use of liquidreceiving phase (cyclohexane) that allowed convenient working conditions.

For hormones, 4 binding phases were selected out of 5 tested (Table 2). Four authors used Oasis[®] HLB as binding phase whereas three authors used three other binding phases. Stroski et al., (2018) concluded, as for pesticides, that the Sepra ZT binding phase was easy to set up. Chen et al., (2018) had tested three binding phases (HLB, XAD18 and Strata-XL-A). Their investigations have demonstrated that the devices with HLB or XAD18 as binding phases can measured hormones with high accuracy, high sensitivity and good precision.

For bisphenols, 4 binding phases were selected out of 5 tested (activated carbon, XDA1, HLB, XAD18
and Strata-XL-A). Only Chen et al., (2018) tested different binding phases to sample bisphenol A and
concluded that both HLB and XAD18 can be efficiently used.

256 Other classes of compounds have been tested: illicit drugs, organophosphorus flame retardants, 257 perfluoroalkyls, household and personal care products, nitrophenols and miscellaneous organic 258 compounds. For illicit drugs and perfluoroalkyl, C. Guo et al., (2017) and Guan et al., (2018) used 259 XAD18. For organophosphorus flame retardants, Zou et al., (2018) used HLB. According to Chen et al., 260 (2017), household and personal care products could be sampled using HLB and XAD18. According to 261 et al., (2018, 2017), endocrine disrupting chemicals (e.g. tris(n-buthyl)phosphate, Belles 262 tris(phenyl)phosphate) could be sampled using strata-X sorbent. According to You et al., (2019a) and 263 Dong et al., (2014), nitrophenols or 4-chlorophenol, could be sampled by HSAC (lignocellulose hazelnut 264 shell-derived activated carbons) and MIP (molecularly imprinted polymers), respectively.

Quantities of binding phase incorporated into binding gel varied from 0.25 to 20% (wet mass:volume) with an average of 13% (Table 1). Protocols were adapted from Zhang and Davison, (1995) where Chelex-100 binding gel was prepared using 2g of resin Chelex-100 in 20 mL of gel solution. The less concentrated was Oasis[®] HLB receiving phase prepared by Guibal et al., (2017). The effective binding capacity calculated with this concentration was sufficient for a long-term deployment (weeks to months).

270 Among the 6 materials successfully used for binding phase preparation, a mixed binding layer combining 2

271 or more material could be developed similarly to what have been done for inorganic compounds (Huynh

- et al., 2012). It could be an interesting way to sample a wider range of organic compounds with a limited
- 273 number of samplers.

274

275 Diffusive layer.

The reported diffusive layers that control mass transfer in the sampler were hydrogels or filter membranes.
This last diffusive layer is the less used since only 10% of the published papers concern filter membranes
(Dong et al., 2014; You et al., 2019b, 2019a). Regarding hydrogels, two different types were used. 80% of
the studies used agarose whereas only 10% used polyacrylamide (**Table 1**). The choice between agarose
and polyacrylamide is based on two criteria: i) no analyte adsorption on the diffusive gel and ii) greater
diffusion coefficient (Chen et al., 2012, 2018, 2017; Fauvelle et al., 2015; Guibal et al., 2017; W. Guo et al.,
2017).

To sample pharmaceuticals by o-DGT, agarose diffusive layer was used by 90% of studies because limited adsorption (<5%) has been observed (Chen et al., 2012; Zhang et al., 2018). However, Stroski et al., (2018) sometimes observed degradation of agarose diffusive gel whereas polyacrylamide shown to be more resistant during field deployment. Agarose degradation was also observed by Challis et al., (2018b) with sometimes a diffusive gel completely destroyed due to aquatic insects grazing. PES membranes could be used also as diffusive gel. This configuration was used by You et al., (2019b) to sample tetracyclines which shown low interaction with PES membranes.

290 For pesticides, agarose was the most used diffusive layer (64% of studies). The second most used diffusive 291 layer was polyacrylamide (29%). Polyacrylamide diffusive gel was chosen by Fauvelle et al., (2015) because 292 higher diffusion coefficients were obtained (higher than a factor 1.5 compared to agarose gels). This 293 difference between diffusion coefficients in polyacrylamide and agarose gels was attributed to the pore 294 size difference between the two types of gels. Such explanation is however surprising since Zhang and 295 Davison, (1999) and Scally et al., (2006) showed that polyacrylamide contains smaller pore sizes compared 296 to agarose gels, reducing diffusion coefficient of metals. Polyacrylamide was also used by Guibal et al., 297 (2017) because they have observed significant adsorption of anionic pesticide on agarose (15%) whereas 298 only 5% adsorption was observed on polyacrylamide. Contrarily, little adsorption (< 5%) was observed 299 two non-ionic pesticides by Xie et al., (2018b) on agarose diffusive gel. The last diffusion layer used to 300 sample pesticide was water Bondarenko et al., (2011). Diffusion coefficients in water were higher 301 compared to hydrogels (agarose or polyacrylamide).

For hormones, agarose gel was used as diffusion layer by all authors, Stroski et al., (2018) using both
agarose and polyacrylamide gels. Authors used agarose gel because poor adsorption of hormones was
observed (<10%) (Chen et al., 2018; W. Guo et al., 2017; Xie et al., 2018b).

305 For bisphenols, illicit drugs, perfluoroalkyl, household and personal care products, and organophosphorus 306 retardant flame, agarose gel was used as diffusive gel. This diffusive gel showed poor adsorption of 307 bisphenols (Chen et al., 2018; Xie et al., 2018b; Zheng et al., 2015), illicit drugs (C. Guo et al., 2017), 308 perfluoroalkyl (Guan et al., 2018), household and personal care products (Chen et al., 2017) and 309 organophosphorus retardant flame (Zou et al., 2018). For Chen et al., (2017), the two type of diffusive gels 310 (agarose and polyacrylamide) showed poor or no adsorption of household and personal care products but 311 agarose had better stability. To sample phenols (4-chlorophenol and nitrophenols), authors used nylon 312 membrane as diffusive layer (Dong et al., 2014; You et al., 2019a).

313 The protocols for the manufacture of agarose and polyacrylamide diffusive gels are the same for all314 authors and were adapted from (Zhang and Davison, 1999).

315

316 Outer protected layer.

Optional addition of membranes beyond the diffusive layer plays the role of physical protection of the
diffusive gel against degradation. Whatever the membranes used, pore size was 0.45 µm. In DGT theory,
membrane is considered as inert regarding the analytes and constitutes only a part of the diffusion path in
the device.

321 Consequently, the choice between different membranes was made after evidencing the absence of 322 adsorption of targeted compounds on the chosen membrane. However, such verification was not 323 systematic and assumption of non-interactions between the analytes and the selected membrane was made 324 by some studies.

A total of 11 types of membranes were tested for o-DGT (list of membranes tested are available in **Table** 3). Finally, polyethersulfone (PES) membranes are the most popular for sampling polar organic compounds and were used in 10 studies. Chen et al., (2012) and Zhang et al., (2018) did not observed adsorption (< 5%) for pharmaceuticals (sulfamethoxazole, methcathinone, ephedrine and tetracyclines). The same observation was obtained by C. Guo et al., (2017) for hormones. However, for four authors, targeted compounds (pesticides (Challis et al., 2016; Chen et al., 2017), pharmaceuticals (Challis et al.,
2016), hormones (Challis et al., 2016), bisphenols (Xie et al., 2018b; Zheng et al., 2015) and
organophosphorus retardant flame (Zou et al., 2018)) were significantly adsorbed (from 10 to 100%) by
this membrane.

334 PTFE (polytetrafluoroethylene) were also used as protective membranes for the sampling of bisphenols
335 (Zheng et al., 2015) and organophosphorus retardant flame (Zou et al., 2018). No significant adsorption
336 (<5%) was observed for bisphenols contrarily to organophosphorus retardant flame (0 to 50%).

337 Other protective membranes were used such as nucleopore track-etch ((bisphenols (Chen et al., 2018) and

household and personal care products (Chen et al., 2017)), PVDF (polyvinylidene fluoride) (hormones (W.
Guo et al., 2017)) and nylon ((pharmaceuticals) (D'Angelo and Martin, 2018)). Adsorption on these
membranes was tested by the authors and, whatever the membrane, no significant adsorption (<5%) was

341 observed.

Glass microfiber and nitrocellulose were also used as outer protective membranes for, respectively, the
sampling of pesticides (Bondarenko et al., 2011; Wei et al., 2019) and nitrophenol (You et al., 2019a).
However, no analyte adsorption test was performed.

To avoid the analyte sorption issue, nine studies (Table 1) use naked o-DGT, making this strategy the second most used. Moreover, Challis et al., (2018b) reported for certain organic compounds a signal suppression induced by PES membranes and then, preconized the use of naked o-DGT. As it was shown with PES membrane used in POCIS, the signal suppression during compounds analysis may be due to release of polyethylene glycol (PEG) by PES membrane (Guibal et al., 2015). Another alternative to conventional membranes to avoid sorption issue was to use aluminum screen (28% open area) as the outer protective layer (Belles et al., 2017).

The use of an outer membrane raises questions. Its use has an undeniable advantage for the protection of the diffusive gel against biofouling or damaging of the diffusive gels by particles and bacteria. However, the impact of using a membrane such as signal suppression during analysis or compound-membrane interaction were shown in some studies. Before using a membrane, two points should be considered: (i) target analytes and their potential interaction with the membrane (several compounds are often targeted and there is no "universally" inert membrane) and (ii) knowledge of the site of field deployment (including

- 358 seasonal changes) to evaluate the risk of biofilm development and the relevance using naked o-DGT.
- 359 Depending on these two points, use of membranes or naked o-DGT should be favored.
- 360

361 INFLUENCE OF ENVIRONMENTAL FACTORS: pH, IONIC STRENGTH, ORGANIC 362 MATTER, BIOFOULING, TEMPERATURE and FLOW EFFECT

363 Considering environmental factors is important because it can influence sampling. Several authors tested
364 the o-DGT robustness over pH, ionic strength, organic matter and flow effect. All these parameters could
365 influence o-DGT sampling by modifying the analyte speciation, diffusion within diffusive gel and/or
366 sorption onto the binding phase.

- 367
- 368 pH.

369 Depending on pH and their pKa, some organic compounds can be neutral or ionic. Change in their 370 protonation will modify their properties and potentially their sampling. A wide range of pH has been 371 tested by the authors (from pH 3 to 11). Considering 20% accuracy is acceptable, no influence of pH on 372 o-DGT sampling is demonstrated for many of the studied compounds. Robustness over pH was 373 demonstrated for 4 hormones (from pH 3.5 to 9.5 (Chen et al., 2018) from pH 7 to 9 (Xie et al., 2018b), 374 from pH 5 to 8 (W. Guo et al., 2017), pH 5 and 8.5 (Stroski et al., 2018)), for some household and 375 personal care products (from pH 3.5 to 9.5) (Chen et al., 2017), for 4-chlorophenol (from pH 3 to 7) 376 (Dong et al., 2014), for nitrophenols (from pH 2 to 7) (You et al., 2019a), for illicit drugs for pH ranged 377 from 4 to 9 (Guo et al. 2017a), for pharmaceuticals (for pH ranged from 5 to 9 (Chen et al., 2012; Ren et 378 al., 2018; Xie et al., 2018a; You et al., 2019b), for pH ranger from4 to 11 (Zhang et al., 2018), for pH 5 and 379 8.5 (Stroski et al., 2018)), for pesticides (from pH 7 to 9 (Xie et al., 2018b), for organophosphorus flame 380 retardants for pH ranged from 3.1 to 9.7 (Zou et al., 2018) and for bisphenols for pH ranged from 4 to 8 381 (Zheng et al., 2015), for perfluoroalkyl substances (from pH 4.2 to 7.8) (Guan et al., 2018). 382 However, sampling of some compounds was found to vary with pH, depending on compounds' pKa. A

383 non-acceptable ratio C_{DGT} / C_{sol} (*i.e.* with more than 20 % of inaccuracy) was obtained by numerous 384 authors. It was the case of Dong et al., (2014) with 4-chlorophenol at pH=8, for some pharmaceuticals 385 (norfloxacin, enrofloxacin, ofloxacin and sulfadimethoxine) when pH < 7.3 Xie et al., (2018a) and for two 386 anionic pesticides (chlorsulfuron and mecoprop) at $pH \ge 7$ (Guibal et al., 2017).

387 The influence of pH on sampler uptake and diffusion was deepened by (Stroski et al., 2018) for 31 388 compounds (pharmaceuticals, hormones and pesticides). In this study, different sampler uptake was 389 obtained for 14 compounds (atenolol, clofibric acid, 2,4-D, fluoxetine, ibuprofen, ketoprofen, naproxen, 390 sulfacholoryridazine, sulfamedimethoxine, sulfamethazine, sulfamethoxazole, sulfapyridine, sulfisoxazole 391 and thiamethoxam) between pH 5 and 8.5. A higher sampling was obtained at pH 8.5 for all compounds 392 except for atenolol where accumulated mass was higher at pH 5. They hypothesized that alteration of 393 robustness over pH was caused by changes in sorption on the binding phase following changed with 394 speciation of analyte, in other words there are a change of analyte-sorbent interaction due to speciation 395 modification of analyte. This hypothesis is in accordance with the study on some anionic pesticides from 396 Guibal et al., (2017) who found, for a given compound, pH dependance for two different binding phases 397 (HLB or MAX). Consequently, Stroski et al., (2018) recommended to consider pH as an important factor 398 for future development and calibration of o-DGT.

399

400 Ionic strength.

401 Ionic strength can affect sampling by the "salting-out" effect reducing the analyte solubility (Togola and 402 Budzinski, 2007; Xie et al., 1997) and can reduces the electrostatic repulsions due to the screening effect 403 of the surface charge of the diffusive gel (Fontecha-Cámara et al., 2007; Joseph et al., 2011). Ionic strength 404 effect sampling by o-DGT was tested by 15 authors by varying ionic strength from 0.0001 to 1 M 405 (imposed with NaCl or NaNO₃). Sampling by o-DGT was found independent on ionic strength usually 406 from 0.001 to 0.5 M (You et al., 2019b, 2019a; Zhang et al., 2018; Zheng et al., 2015; Zou et al., 2018).

Effect of ionic strength was reported for some compounds when it raised to 0.2 – 0.5 M or above. It was
observed for the antibiotic sulfamethoxazole (Chen et al., 2012), for the hormone estrone (Chen et al.,
2018), for the household and personal care products butylated hydroxyanisole and triclosan (Chen et al.,
2017), for 4-chlorophenol (Dong et al., 2014), tetracycline antibiotics and nitrophenols (You et al., 2019b,
2019a). Alteration of sampling at high ionic strength could be attributed to the "salting-out" effect
reducing the analyte solubility (Togola and Budzinski, 2007; Xie et al., 1997).

413 Effect of ionic strength (I) on sampling was also observed at low ionic strength for some compounds. 414 Reduced sampling was reported for perfluorooctane sulfonate at I = 0.0001 M (Guan et al., 2018) and for 415 9 antibiotics (sulfapyridine, sulfadiazine, sulfamethoxazole, sulfathiazole, sulfachloropyridine, norfloxacin, 416 ciprofloxacin, thiamphenicol and florfenicol) at I = 0.01 (Xie et al., 2018a). Conversely, enhanced 417 sampling was reported for three macrolides antibiotics (erythromycin, clarithromycin and azithromycin) at 418 I = 0.01 by the same study (Xie et al., 2018a). Alteration of sampling at low ionic strenght can be explained 419 by modification of electrostatic repulsions with the diffusive gel due to the screening effect of the surface 420 charge as already demonstrated for metals (Fatin-Rouge et al., 2003; Warnken et al., 2005). Indeed, the 421 opposite behavior observed for antibiotics (Xie et al., 2018a) can be linked to their opposite charge at the 422 study's pH (cationic for the three macrolides and anionic for the others at pH 8) which as been shown for 423 trace element to condition reduced (anion) or enhanced (cation) diffusion (Shiva et al., 2015).

To conclude on ionic strength, o-DGT can be used to estimate contamination by organic compounds in most freshwaters (Chen et al., 2012, 2017; Guibal et al., 2017; C. Guo et al., 2017; Zhang et al., 2018; Zheng et al., 2015), providing diffusion coefficient are specifically determined for low ionic strength. This passive sampler can be also used to estimate contamination in waters with high ionic strength such as seawater of some antibiotics with PCM as binding phase (Ren et al., 2018; Xie et al., 2018a) or some endocrine disrupting chemicals with XDA-1 as binding phase (Xie et al., 2018b).

430

431 Dissolved Organic Matter.

432 Dissolved Organic Matter (DOM) can have two type of effects that could alter analyte uptake by o-DGT 433 samplers. First, DOM can cause competition over analyte for sorption to the binding phase and secondly, 434 reactions between DOM and analytes can alter analyte diffusion (Davison et al., 2015; W. Guo et al., 435 2017). Indeed, a significant 40% alteration of triclosan sampling was observed by Chen et al., (2017) for 436 DOM concentration higher than 2 mg DOM L⁻¹. This hydrophobic compounds binds to DOM making 437 diffusion trough diffusive gel more difficult. Dong et al., (2014) similarly observed alteration of 4-438 chlorophenol sampling for DOM concentration ranging from 9.8 to 36.5 mgC L⁻¹. Conversely, sampling 439 of several compounds was found unaltered. This behaviour was for observed for some pharmaceuticals 440 (You et al., 2019b), perfluoroalkyl substances (Guan et al., 2018), hormones (Chen et al., 2018),

441 organophosphorus flame retardants (Zou et al., 2018) and household and personal care chemicals (Chen442 et al., 2017)).

443 It is likely that sampling alteration caused by DOM is compound dependent but also DOM dependent.
444 Until more work is done to investigate DOM effect, interpretation of o-DGT derived concentration
445 should be made with caution when DOM is significantly present.

446

447 Biofouling.

448 Only one article studied biofouling of o-DGT (Challis et al., 2016) and consequences on compounds 449 accumulation during field deployment. Challis et al., (2016) with a long-term deployment (21 days) 450 observed samplers were fouled but do not notice any effect on compounds accumulation. They observed 451 that traditional deployment times (from 2 to 4 weeks) in surface waters allows a linear accumulation in o-452 DGT. Few authors have studied the impact of biofouling on sampling and therefore this topic is poorly 453 documented. However, it is of particular concern given that interference of fouling on inorganic 454 compounds accumulation in DGT was demonstrated in few articles (Devillers et al., 2017; Feng et al., 455 2016; Uher et al., 2012). It clearly needs improvements for organic compounds and is further discuss in 456 the last past of this review "future needs for o-DGT deployment".

457

458 Temperature effect.

Temperature affects mainly water viscosity and molecular thermic agitation (Brownian motion) and as a consequence *D*. As discussed in section "Determination of diffusion coefficients", it is possible to determine a temperature corrected *D* (*cf.* Eq.7). Challis et al., (2016) had measured and calculated (Eq. 7) diffusion coefficients at different temperatures. Relative error was about 20% and the authors concluded that relationship (Eq. 7) is valid to estimate diffusion coefficient. TWAC for field deployments can be calculated based on the average temperature recorded. Commercial devices such as Tynitag can easily perform temperature record with relevant time frequencies (below 1h)

466

467 Flow effect.

468 DGT sampler is known to be poorly sensitive to environmental conditions such as hydrodynamic flow for 469 metals (Gimpel et al., 2001) thanks to the thickness of its diffusive layer. However, DBL (Figure 2) may 470 become significant in low flow conditions and alter DGT sampling by increasing the diffusion path length 471 (Davison and Zhang, 2012). DBL thickness has been estimated by several authors for organic 472 compounds. Values obtained are in the same order of magnitude in well stirred systems (average 473 thicknesses from 0.22 to 0.25 mm) (Belles et al., 2018; Challis et al., 2016; Chen et al., 2013, 2018, 2017; 474 Ren et al., 2018). A higher value was obtained by Challis et al. (2018b) with an estimated median $\delta = 0.34$ 475 mm. Measured DBL thicknesses for organic compounds are very close to those obtained for inorganic 476 compounds in well stirred systems (≈ 0.2 mm, Davison and Zhang, 2012). Without taking into account 477 this boundary layer, the estimate of the concentration should be 20% underestimated by Equation 3. 478 Some authors recommended the use of diffusive gel with a thickness at least 1.0 mm (Chen et al., 2013) to 479 limit the significance of the DBL thickness. A ticker diffusive gel (1.2 mm) was chosen by Belles et al., 480 (2017) to have a gel thickness significantly higher than the DBL one. Other authors propose to include δ 481 ≈ 0.20 mm in calculations to estimate TWAC (Challis et al., 2016). It has been demonstrated for metals 482 (Warnken et al., 2006) that, when using standard devices, the error made by neglecting DBL thickness is 483 cancelled by the error made by neglecting lateral diffusion. This phenomenon is likely to concern also 484 organic compounds and incorporating DBL thickness in concentration calculation for well stirred systems 485 could alter its accuracy. In unstirred solutions, DBL thickness is found to increase up to 0.76 mm (average 486 values (Challis et al., 2016; Chen et al., 2012)). Such increase in the diffusion length will significantly alter 487 sampling and concentration estimation as demonstrated by Buzier et al., (2019) for some pharmaceuticals. 488 In unstirred solutions, increasing diffusive gel thickness or incorporating DBL thickness in concentration 489 calculation should improve accuracy. For some pharmaceuticals, Buzier et al., (2019) estimated that a 2.5 490 mm gel thickness should allow keeping <25% accuracies. It should be noted that such increase in the 491 diffusion length will proportionally decrease the sampling rate and alter the sensitivity.

492

493 APPLICATION & FIELD DEPLOYMENT

494 Different environmental matrixes were tested for field deployments. The first application was made in
495 soils (Bondarenko et al., 2011) but few studies currently concerns such field application (Chen et al.,
496 2015a, 2014; Lin et al., 2018).

497 The majority of studies concerns sampling in freshwaters. The first one was performed in a river in 498 United-Kingdom (Chen et al., 2012) to estimate contamination of sulfamethoxazole with 14-day 499 deployment of o-DGT. Deployments in rivers were also performed by Guibal et al., (2017), C. Guo et al., 500 (2017), Stroski et al., (201), Zhang et al., (2018) and Zheng et al., (2015) for 7 days or in a lake for 12-33 501 days (Guan et al., 2018). These authors concluded that o-DGT devices were suitable to detect organic 502 pollution in freshwaters. Three authors have tested deployment in coastal waters (Ren et al., 2018; Xie et 503 al., 2018b, 2018a). After 3-day deployment of o-DGT (XDA-DGT), endocrine disrupting chemicals were 504 detected. For 3 compounds (estradiol, Bisphenol A and acetochlor), differences between concentration 505 determined by o-DGT and concentrations determined by grab sampling was observed (Xie et al., 2018b). 506 These differences were explained the different nature of sampling, spot sampling being unable to provide 507 TWAC (Xie et al., 2018b, 2018a).

o-DGT were also successfully deployed in non-natural waters. Deployments for 24h to 120h in industrial
wastewaters contaminated with nitrophenolic compounds (You et al., 2019a) showed no significant
difference in estimated concentrations compared to grab samples. Deployments of o-DGT were
performed in WasteWater Treatment Plant (WWTP) influent and effluent from 6h to 33 days (Challis et
al., 2018b, 2016; Chen et al., 2015b, 2013, 2018, 2017; Dong et al., 2014; Guan et al., 2018; C. Guo et al.,
2017; W. Guo et al., 2017; Ren et al., 2018; Zou et al., 2018). A 5-day field deployment in small pound
receiving pig breeding wastewater was performed by You et al., (2019b).

515 Deployment time appears a key factor to ensure accurate results during field deployments in waters. A 7-516 day deployment was recommended by Chen et al., (2013). This duration allows to stay in kinetic uptake 517 regime and to avoid significant biofouling. Biofouling was also observed by Challis et al., (2016) with a 518 long-term deployment (21 days) but accumulation was still linear and indicated that sampler capacity was 519 sufficient for using traditional deployment times (from 2 to 4 weeks) in impacted surface waters.

520

521 FUTURE NEEDS FOR O-DGT DEVELOPMENT

522 This review collected studies which demonstrated the potential of o-DGT. The o-DGT is of particular 523 interest compared to other passive sampler since it has a better robustness over flow variations (Buzier et 524 al., 2019) and allows deployments in soils (Chen et al., 2015a, 2014; Lin et al., 2018). However, a current 525 disadvantage of o-DGT compared to others passive samplers for organic contaminants is its lower ability 526 to concentrate compounds because of lower sampling rates. A detailed comparison of passive samplers 527 efficiency and deployment (*i.e.* o-DGT, POCIS and Chemcatcher®) is proposed by Gong et al. (2018). 528 Compared to sampling rates of POCIS (Polar Organic Chemical Integrative Sampler) o-DGT's ones were about 25 (Challis et al., 2018b) or 50 (Buzier et al., 2019) times lower. These reduced sampling rates are 529 530 mostly due to a reduced exposure area of o-DGT (3.1 cm² versus 41 cm² for POCIS) compared to other 531 passive samplers (Buzier et al., 2019; Chen et al., 2013; Guibal et al., 2017). Therefore, an increase of o-532 DGT sampling area should allow increasing sampling rates and consequently improving sensitivity. Buzier 533 et al., (2019) estimated for 10 pharmaceutical compounds that a 160 cm² sampling area (~7 cm radius) 534 should allow similar sampling rates compared to POCIS. Such a theoretical o-DGT configuration must 535 however be tested in the field, since physical constrains on these larger gels could be significant. A recent study investigated improved design of o-DGT sampler in order to increase sampling rates (Urik and 536 537 Vrana, 2019). Similarly to POCIS, this new o-DGT sampler design has two side and increased size 538 allowing sampling area to be seven times higher than the standard DGT design (22.7 cm²). This new o-539 DGT sampler was successfully tested for polar organic compounds sampling (pharmaceuticals and 540 personal care products). Sampling rates were higher than the one obtained with standard o-DGT in 541 Guibal et al., (2017) (43 versus 13 mL day-1).

542 o-DGT can also be currently impacted by other limitations concerning any passive sampler. In many 543 cases, because of sensitivity and representativeness issues, long deployment times (e.g. several weeks) 544 would be preferred. However, longer deployment in environmental systems will favor biofouling 545 formation in front of the samplers as already observed by Challis et al., (2016) and Chen et al., (2013). 546 Fouling of the device can be due to microorganisms (bacterial, algal or fungi development) or deposition 547 of suspended matter. Although biofouling effect was not studied for organic compounds passive 548 sampling, it is likely to alter analyte sampling as observed for inorganic compounds (Feng et al., 2016; 549 Pichette et al., 2009; Uher et al., 2017, 2012). It was shown by Devillers et al., (2017) that some metals

were adsorbed onto biofouling resulting in decreasing compound concentration at the water/samplerinterface and consequently in smaller diffusion rate into the sampler.

552 Given that several organic compounds have a high affinity for organic matter (Chen et al., 2017), it is not 553 to exclude that some compounds will bind to biofouling and their passive sampling should be 554 consequently altered. However, such behavior still has to demonstrated for organic compounds. Feng et 555 al., (2016) found that the diffusion coefficient of orthophosphate decreased linearly with increasing 556 fouling thickness, allowing a mathematical model to be proposed for diffusion coefficient correction over 557 the formation of the fouling. Similar procedure for organic compounds could be investigated if their 558 sampling was found to be altered by biofouling.

559 Finally, standardization of the calibration procedures (*i.e.* diffusion coefficient determination, see previous 560 sections) would be valuable to favor reproducibility and reliability of o-DGT results. Indeed, 561 determination of diffusion coefficients can be performed by three different methods. The time series 562 deployments method allows using more environmentally relevant concentrations and could be considered 563 as the most relevant method. However, it results from model fitting to not only diffusion process but also 564 to compound binding within the sampler. Rather than a physical diffusion coefficient, it is a calibration 565 parameter usually called "effective diffusion coefficient". Moreover, some authors did not take into 566 account the entire diffusion path (diffusive gel and membrane). Considering that membranes were 567 previously shown to alter diffusion coefficient of some metals (Buzier et al., 2014), it is possible that 568 diffusion of some organic compounds is also altered. Indeed, significant sorption between some organic 569 compounds and polyethersulfone membranes used with POCIS were demonstrated by Endo and 570 Matsuura, (2018). Until it is demonstrated that the membrane used with o-DGT has no influence on the 571 diffusion of the targeted compounds, it is advisable to incorporate the membranes in the calibration 572 experiments.

573

574 CONCLUSION

575 This review investigated the current data available on o-DGT passive samplers. 22 possible configurations 576 were developed (*i.e.* binding gel combined with diffusive gel and membranes) to enable sampling of 577 organic compounds from different chemicals families with a wide range of physico-chemicals properties (*e.g.* hydrophobicity with logP ranged from -4.54 to 7.51) in environmental water bodies or in soil systems.
The two most commonly used configurations were agarose-XAD18 and agarose-HLB. However, it is
important to adapt the configuration of the sampler to the known properties of the targeted compounds.
These two configurations allow to sample organic compounds in a wide range of pH (4-9) and ionic
strength (0.001 to 0.1 M). This robustness indicated that o-DGT can be used in most natural waters.

583 Considering that, compared to other samplers, o-DGT is less influenced by flow variations and allows 584 deployment in several environmental compartments (*e.g.* water, soil and sediment), it seems to have a great 585 potential for monitoring a large class of organic pollutants in environment. However, this sampler cannot 586 currently reach sensitivity offers by other passive samplers (*e.g.* POCIS). Depending on the targeted 587 contamination levels, improvement of o-DGT sensitivity would be desirable.

588

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Table 1. Initial (2007) soil TE concentrations (mg kg⁻¹ DW) for the PHYTOPOP plots, compared with natural pedogeochemical background values (NPBV) and local usual agricultural concentrations (UAC) given as ranges (Lamy et al. 2006).

	NPBV	UAC	PHYTOPOP plots	
			Min	max
Cd	0.014-0.02	0.19 - 0.42	1.98	4.44
Со	2.3 - 3.6	3.0 - 7.7	4	13.5
Cr	13.9 - 21.0	15 - 29	37	89
Cu	2.4 - 5.9	8 - 19	69	218
Hg	0.01 - 0.03	0.08 - 0.15	-	-
Ni	4.2 - 8.2	6 - 20	14	42
Pb	3.7 - 8.3	18 - 43	74.7	484
Zn	8.6 - 19.4	34 - 63	314	692

Table 2. Ammonium nitrate-TE extractable fraction in Pierrelaye soils collected from under the various poplar genotypes. Values presented were measured in 2007 and 2011 and the changes occurring between the two samplings are indicated.

Genotype		Cd			Cu			Zn			pН	
	µg kg	⁻¹ DW	change (%)	µg kg	-1 DW	change (%)	µg kg	⁻¹ DW	change (%)			change (%)
	2007	2011		2007	2011		2007	2011		2007	2011	
Bakan	21.3	16.5	-23	812	690	-15	1970	1620	-18	7.24	7.39	+2
Dorskamp	20.5	16.2	-21	639	525	-18	1620	1370	-16	7.27	7.31	+1
Dvina	39.1	26.2	-33	718	648	-10	2240	1350	-40	7.47	7.47	=
Flevo	24.6	19.3	-21	895	751	-16	2260	1880	-17	7.19	7.36	+2
Fritzi Pauley	17.9	13.9	-23	634	475	-25	165	137	-17	7.36	7.24	-2
1214	29.1	20.9	-28	801	726	-10	2130	1630	-23	7.27	7.52	+3
Koster	19.4	15.6	-20	723	611	-16	1910	1630	-15	7.34	7.34	=
Lena	20.0	15.7	-22	650	532	-18	1590	1280	-20	7.33	7.32	=
Muur	25.7	20.3	-21	839	766	-9	2300	1820	-21	7.29	7.45	+2
Skado	25.9	19.8	-24	758	642	-16	1680	1340	-20	7.38	7.51	+2
Soligo	19.8	16.0	-20	505	433	-15	1490	1210	-19	7.40	7.32	-1
Trichobel	19.0	15.5	-18	620	535	-14	1530	1310	-15	7.38	7.46	+1
Triplo	29.2	21.4	-27	761	726	-5	2010	1390	-31	7.30	7.54	+3
Vesten	22.0	17.1	-23	779	630	-19	1990	1590	-20	7.31	7.38	+1

Table 3. Estimated allometric parameters (standard error) of the power function and statistical outputs (data are for TrunkDW and BranchDW). Logarithm transformation of data and power form $(DW=a^*dbh^{h})$ with a and b as regression coefficients were chosen as the best relevant models. The "a" regression coefficients was the same for all genotypes (a=10.12). The TrunkDW and BranchDW estimates were expressed with the Bakan genotype as the reference. D: *P. deltoides*; DN: *P. deltoides* x *P. nigra*; T: *P. trichocarpa*; TM: *P. trichocarpa* x *P. maximowiczii*. Levels of significance are indicated by asterisks : ***p < 0.001; **p < 0.01; *p < 0.05. ns : not significant.

Genotype	Species	n	TrunkDW	t-	b	BranchDW	t-	b	BranchDW
			estimate	value		estimate	value		Proportion (%)
Bakan (intercept)	ТМ	5	10.12 (2.65)	3.825 (***)	10.12	1.83 (1.65)	1.11 (ns)	1.83	26
Dorskamp	DN	5	-3.704 (3.49)	-1.06 (ns)	6.41	5.79 (2.22)	2.61 (*)	7.62	27
Dvina	D	5	-10.49 (3.48)	-3.01 (**)	-0.37	0.46 (2.22)	0.21 (ns)	2.29	34
Fritzi Pauley	Т	6	-0.0035 (3.34)	-0.00 (ns)	10.11	-2.95 (2.13)	-1.38 (ns)	- 1.12	25
Flevo	DN	5	-5.928 (3.48)	-1.70 (.)	4.19	0.63 (2.22)	0.29 (ns)	2.46	30
I214	DN	5	-9.116 (3.50)	-2.60 (*)	1.00	1.11 (2.23)	0.50 (ns)	2.94	33
Koster	DN	5	-8.78 (3.49)	-2.51 (*)	1.34	-1.58 (2.23)	-0.71 (ns)	0.25	31
Lena	D	6	-14.62 (3.33)	-4.38 (***)	-4.50	2.37 (2.13)	1.11 (ns)	4.19	40
Muur	DN	7	-8.914 (3.26)	-2.73 (**)	1.20	-1.05 (2.08)	-0.50 (ns)	0.78	31
Skado	ТМ	6	0.0242 (3.33)	0.01 (ns)	10.14	2.05 (2.12)	0.97 (ns)	3.88	27
Soligo	DN	7	-5.948 (3.24)	-1.84 (.)	4.17	-2.39 (2.06)	-1.16 (ns)	- 0.56	29
Trichobel	Т	5	-2.827 (3.49)	-0.80 (ns)	7.29	-1.82 (2.23)	-0.82 (ns)	0.00	27
Triplo	DN	6	-14.03 (3.33)	-4.20 (***)	-3.91	-3.43 (2.12)	-1.62 (ns)	- 1.61	37
Vesten	DN	5	-10.65 (3.48)	-3.06 (**)	-0.53	-5.08 (2.22)	-2.29 (*)	3.25	35

Table 4. Macronutrient concentrations (mg kg⁻¹ DW) in wood and branches of the 14 poplar genotypes from the Pierrelaye site. Data represent the mean (\pm SE), resulting from samples taken at three different heights as described in the material and method section (see also Fig. S1). Levels of significance are indicated by asterisks : ***p < 0.001; **p < 0.01. Different letters represent significant differences for element content between the genotypes (one-way ANOVA, Tukey-Kramer HSD test). ns : not significant.

Genotype	Ca	a	ŀ	X	I)	M	g
	Branch	Wood	Branch	Wood	Branch	Wood	Branch	Wood
	***	**	ns	***	***	***	***	***
Bakan	16273 (2171)	1214 (53)	4891 (403)	1519 (152)	1732 (121)	570 (70)	1011 (88)	218 (12)
Dakali	b	ab		ab	ab	ab	ac	ab
Dorskamp	13504 (1861)	1711 (182)	6248 (890)	1989 (112)	2136 (186)	747 (42)	1222 (140)	366 (17)
Dorskamp	ab	b		ac	ab	bc	bc	ef
Dvina	12125 (1337)	1437 (119)	5547 (548)	1550 (120)	1571 (158)	614 (24)	1331 (118)	387 (12)
Dvilla	ab	ab		ac	ab	ab	с	f
Flevo	13787 (532)	1253 (76)	5731 (283)	1904 (118)	1659 (54)	543 (46)	1054 (26)	267 (12)
rievo	ab	ab		ac	ab	ab	ac	bc
Fritzi	14025 (1464)	1250 (118)	5107 (343)	1758 (146)	1985 (185)	576 (60)	897 (39)	248 (11)
Pauley	b	ab		ac	ab	ab	ab	bc
I214	12572 (1569)	1381 (81)	6454 (647)	1795 (111)	1625 (150)	610 (31)	1052 (129)	299 (12)
1214	ab	ab		ac	ab	ab	ac	cde
Koster	9244 (960)	1206 (67)	5233 (418)	1511 (78)	1593 (91)	536 (26)	948 (79)	358 (21)
Koster	ab	ab		ab	ab	ab	ac	df
Lena	10264 (1412)	1445 (105)	5844 (813)	2036 (120)	1613 (199)	557 (34)	935 (96)	302 (11)
Lena	ab	ab		bc	bc	ab	ac	cde
Muur	11812 (1075)	1282 (105)	6047 (529)	1762 (66)	2026 (110)	883 (27)	820 (47)	294 (17)
Wiuui	ab	ab		ac	ab	с	ab	cde
Skado	10184 (1161)	1227 (82)	4796 (466)	1494 (67)	1904 (193)	542 (35)	961 (115)	222 (10)
Skauu	ab	ab		а	ab	b	ac	b
Soligo	14794 (1836)	1647 (136)	6587 (537)	1717 (107)	2216 (137)	643 (45)	1113 (74)	373 (23)
Soligo	b	b		ac	ac	ab	bc	f
Trichobel	11662 (1141)	1306 (122)	6813 (491)	2114 (193)	2345 (119)	746 (53)	785 (35)	266 (13)
TTICHODEI	ab	ab		с	а	ac	ab	bc
Triplo	13348 (1417)	1079 (60)	6610 (625)	1857 (102)	2049 (153)	577 (38)	1303 (132)	279 (7)
тпрю	ab	а		ac	ab	ab	с	bc
Vesten	9150 (403)	1488 (108)	5483 (107)	1780 (91)	1602 (53)	600 (52)	749 (20)	286 (12)
v estell	а	ab		ac	b	ab	a	acd

Table 5. Trace element concentrations (mg kg⁻¹ DW) in wood of the 14 poplar genotypes from the Pierrelaye site. Data represent the mean (\pm SE), resulting from samples taken at three different heights as described in the material and method section (see also Fig. S1). Levels of significance are indicated by asterisks : ***p < 0.001; **p < 0.01. Different letters represent significant differences for element content between the genotypes (one-way ANOVA, Tukey-Kramer HSD test). ns : not significant.

Genotype	Cd	Cr	Cu	Fe	Mn	Pb	Zn
	***	**	***	***	***	***	***
Bakan	1.6 (0.15)	0.1 (0.01)	4.0 (0.3)	12.4 (1.2)	16.1 (2.0)	0.4 (0.04)	132 (13)
Dakali	cd	ab	ab	ab	bd	ab	cdf
Dorskamp	1.7 (0.20)	0.2 (0.02)	6.6 (0.9)	20.7 (3.0)	17.9 (2.6)	0.4 (0.03)	185 (25)
Dorskamp	d	ac	cd	cd	bd	ac	f
Dvina	1.6 (0.18)	0.2 (0.06)	6.6 (0.9)	16.7 (1.7)	20.1 (2.9)	0.4 (0.06)	79 (8)
Dvina	cd	с	с	ad	d	с	abc
Flevo	1.1 (0.25)	0.1 (0.01)	5.8 (0.4)	19.9 (2.1)	19.3 (1.4)	0.6 (0.05)	93 (8)
rievo	ad	ac	ac	bd	cd	ac	ad
Fritzi	1.0 (0.08)	0.2 (0.01)	3.8 (0.3)	10.9 (1.1)	9.4 (0.9)	0.5 (0.09)	118 (11)
Pauley	а	ac	а	а	ab	ac	bd
I214	0.9 (0.11)	0.1 (0.01)	3.8 (0.4)	9.9 (1.1)	7.3 (0.9)	0.4 (0.06)	86 (8)
1214	а	ac	ab	а	а	ac	abc
Koster	1.3 (0.09)	0.2 (0.01)	4.2 (0.4)	13.1 (1.4)	16.8 (2.0)	0.4 (0.04)	91 (8)
KUSICI	ad	ac	ac	abc	ad	ac	ad
Lena	1.3 (0.19)	0.1 (0.01)	5.0 (0.6)	22.5 (3.2)	13.9 (2.7)	0.5 (0.10)	72 (9)
Lena	ad	ac	ac	d	ad	ac	ab
Muur	1.4 (0.09)	0.1 (0.01)	5.2 (0.5)	15.3 (1.4)	15.2 (1.4)	0.3 (0.02)	126 (9)
wiuui	ad	ac	ac	abc	ad	ac	cde
Skado	1.09 (0.10)	0.1 (0.01)	5.7 (0.3)	12.3 (1.3)	13.7 (1.8)	0.3 (0.02)	143 (14)
Skauu	abc	ac	bc	ab	ad	ac	df
Soligo	1.7 (0.13)	0.2 (0.02)	5.6 (0.3)	13.4 (1.5)	13.5 (1.4)	0.3 (0.03)	101 (7)
Songo	bd	bc	ac	abc	ad	bc	ad
Trichobel	1.2 (0.10)	0.1 (0.01)	4.1 (0.2)	10.5 (1.0)	9.6 (1.2)	0.5 (0.05)	172 (13)
TTICHODEI	ad	ac	ab	ab	abc	ac	ef
Triplo	1.4 (0.15)	0.1 (0.01)	4.7 (0.4)	14.4 (1.3)	16.6 (2.8)	0.3 (0.02)	71 (8)
	ad	а	ac	abc	bd	а	a
Vesten	1.2 (0.03)	0.1 (0.01)	4.7 (0.1)	14.5 (0.4)	13.4 (0.4)	0.3 (0.03)	82 (3)
v esten	ac	ac	abd	abc	ad	ac	ab

Table 6. Trace element concentrations (mg kg⁻¹ DW) in branches of the 14 poplar genotypes from the Pierrelaye site. Data represent the mean (\pm SE), resulting from samples taken at three different heights as described in the material and method section (see also Fig. S1). Levels of significance are indicated by asterisks : ***p < 0.001; **p < 0.01. Different letters represent significant differences for element content between the genotypes (one-way ANOVA, Tukey-Kramer HSD test). ns : not significant.

Genotype	Cd	Cr	Cu	Fe	Mn	Pb	Zn
	***	ns	***	***	***	ns	***
Bakan	0.5 (0.04)	0.1 (0.01)	2.2 (0.2)	2.1 (0.2)	2.5 (0.1)	0.4 (0.08)	33 (2)
Dakali	bdf		abc	ac	bd		bd
Dorskamp	0.7 (0.05)	0.1 (0.02)	4.1 (0.3)	2.9 (0.3)	2.3 (0.2)	0.4 (0.05)	51 (2)
Dorskallip	f		e	bc	ad		e
Dvina	0.6 (0.05)	0.1 (0.01)	2.6 (0.2)	3.0 (0.3)	2.7 (0.3)	0.4 (0.05)	25 (2)
Dvilla	ef		ad	с	cd		ab
Flevo	0.4 (0.05)	0.1 (0.01)	3.2 (0.3)	2.8 (0.2)	2.0 (0.1)	0.4 (0.04)	27 (2)
rievo	ad		cde	bc	abc		ab
Fritzi	0.2 (0.02)	0.1 (0.01	1.7 (0.1)	1.9 (0.2)	1.6 (0.1)	0.4 (0.05)	28 (1)
Pauley	а		а	ab	а		ab
I214	0.4 (0.03)	0.1 (0.01)	2.0 (0.1)	1.6 (0.2)	2.1 (0.2)	0.4 (0.04)	36 (2)
1214	ad		ab	а	abc		cd
Koster	0.5 (0.05)	0.1 (0.02)	2.0 (0.1)	2.4 (0.2)	2.9 (0.1)	0.4 (0.05)	32 (1)
KUSIEI	bcde		ab	ac	d		bd
Lena	0.5 (0.06)	0.1 (0.01)	2.5 (0.2)	4.2 (0.5)	2.1 (0.3)	0.4 (0.05)	21 (2)
Lena	bcde		ad	d	ad		а
Muur	0.4 (0.02)	0.1 (0.01)	2.0 (0.1)	2.3 (0.1)	2.1 (0.2)	0.3 (0.03)	32 (1)
wiuui	bcd		а	ac	abc		bc
Skado	0.3 (0.03)	0.1 (0.01)	2.9 (0.2)	2.1 (0.1)	1.8 (0.1)	0.5 (0.04)	31 (1.2)
Skauu	ac		cd	ac	а		bc
Soligo	0.5 (0.03)	0.1 (0.01)	3.2 (0.3)	2.4 (0.2)	1.6 (0.1)	0.3 (0.03)	32 (2)
Soligo	df		de	ac	а		bc
Trichobel	0.3 (0.04)	0.1 (0.02)	2.5 (0.3)	2.0 (0.3)	1.6 (0.1)	0.4 (0.05)	40 (2)
TTICHODEI	ab		ad	ac	а		d
Triplo	0.4 (0.05)	0.1 (0.01)	2.0 (0.1)	2.1 (0.2)	1.9 (0.1)	0.3 (0.03)	23 (1)
	ad		а	ac	ab		а
Vesten	0.4 (0.03)	0.1 (0.02)	3.0 (0.3)	2.9 (0.2)	1.7 (0.1)	0.4 (0.05)	26 (2)
v esten	bcd		bd	bc	ab		ab

	Ca	K	Р	Mg	Mn	Cu	Fe	Zn	Cd	Cr	Pb
Bark	17326	5923	1109	679	13.4	3.4	16.0	183.1	1.23	0.15	0.64
	(471)	(256)	(57)	(23)	(0.4)	(0.2)	(1.7)	(9.5)	(0.08)	(0.03)	(0.16)
Ratio bark/wood	14	4	2	3	8	1	8	6	4	1	1
Ratio branch/wood	8	3	4	4	8	2	6	5	3	1	1

Table 7. Element concentrations (mg kg⁻¹ DW) in bark of the poplar Skado genotype from the site of Pierrelaye. Data represent the mean $(\pm SE)$. The ratio bark/wood and branch/wood (data from tables 4 and 5) are indicated.

Fe Zn Genotype Ca K Р Mg Cd Cr Mn Cu Pb kg⁻¹ ha⁻¹ year⁻¹ g⁻¹ ha⁻¹ year⁻¹ 9.00 3.01 0.21 Branch 29.94 3.19 1.86 29.60 7.41 22.90 241.94 0.82 Bakan 7.96 2.98 Wood 6.36 13.00 11.65 10.96 172.38 2.71 0.54 2.33 1.14 11.35 5.25 1.79 15.00 5.52 155.48 1.45 0.13 Branch 1.03 17.43 0.31 Dorskamp Wood 4.54 3.90 1.71 0.83 5.24 9.38 6.62 1.64 0.30 0.92 116.39 7.14 15.61 2.02 1.71 25.88 8.55 21.50 101.54 2.12 0.26 0.48 Branch Dvina 3.53 3.81 1.51 6.32 7.47 0.33 Wood 0.95 6.54 61.52 1.56 0.92 13.17 5.47 1.58 1.01 18.47 5.59 89.11 1.07 0.14 0.54 Branch 18.98 Flevo Wood 2.82 4.28 1.22 7.22 0.82 0.25 0.84 0.60 4.42 6.40 61.38 20.04 7.30 2.84 1.28 13.37 5.36 15.62 168.02 1.43 0.22 0.74 Fritzi Branch Pauley 5.35 7.53 2.47 1.06 6.87 7.24 8.05 0.89 0.52 Wood 118.90 1.81 22.31 2.88 12.91 11.45 6.72 17.58 152.62 1.62 0.21 Branch 1.87 0.75 I214 Wood 5.01 6.52 2.22 7.56 7.41 5.92 129.25 1.32 0.33 1.09 1.41 13.99 7.92 2.41 1.44 25.49 19.77 137.22 1.91 0.24 0.58 Branch 6.43 Koster Wood 4.01 5.03 1.78 1.19 9.55 6.59 8.01 107.47 1.56 0.39 1.28 Branch 21.70 12.36 3.41 1.98 29.29 10.59 47.56 153.24 2.74 0.28 1.11 Lena 4.57 Wood 6.44 1.76 0.96 6.79 7.81 13.40 65.88 1.54 0.38 1.22 13.89 7.11 2.38 17.92 1.69 0.15 0.34 Branch 0.96 6.07 17.98 148.14 Muur Wood 3.36 4.61 2.31 5.39 1.05 0.28 0.77 5.27 6.05 84.17 0.68 8.96 25.68 2.04 0.25 0.53 Branch 19.03 3.56 1.80 10.68 23.02 267.66 Skado Wood 6.06 7.38 2.68 1.09 8.69 10.35 1.59 2.38 14.64 154.45 0.61 17.31 7.71 2.59 6.56 15.67 117.83 1.97 0.22 1.30 15.80 0.38 Branch Soligo Wood 4.76 4.96 1.86 4.50 93.05 1.54 0.33 0.90 1.08 9.32 6.89 16.08 9.39 3.23 1.08 13.22 5.59 14.51 236.51 1.63 0.18 0.65 Branch Trichobel Wood 4.95 8.02 2.83 1.01 6.12 9.44 7.50 152.33 1.19 0.47 1.42 14.49 36.45 2.99 0.22 0.62 10.30 155.50 Branch 29.26 4.49 2.86 31.49 Triplo Wood 2.18 7.45 8.07 87.36 0.36 4.08 7.02 1.06 7.26 1.44 1.16 27.52 16.49 4.82 2.25 40.26 14.20 43.59 247.02 3.54 0.41 0.78 Branch Vesten 3.28 9.53 16.45 143.53 2.34 Wood 8.13 9.73 1.56 16.00 2.33 0.67

Table 8. Element export by wood and branches of the 14 poplar genotypes grown at the Pierrelaye site for 7 years. Data are means for the number of replicates, calculated from element content (mg kg⁻¹ DW) provided in tables 4 and 5, and growth data provided in fig. 3 (Odt ha⁻¹ year⁻¹).

Binding phase	Targeted analytes	Diffusive phase	Membranes	Thickness of diffusive gel (mm)	Binding phase concentration in binding gel (% mass:volume)	References
Activated charcoal	Bisphenols	Agarose	PTFE	0.75	5	Zheng et al., (2015)
Activated charcoal	Naphtalene	Water	Glass microfiber	10.50	п.с.	Bondarenko et al., (2011)
Activated charcoal	Nitrophenols	Nylon membrane	Nitrocellulose	0.16	1	You et al., (2019a)
Cyclodextrine polymer	Biocides	Agarose	Glass microfiber	1.00	n.c.	Wei et al., (2019)
MIP	4-chlorophenol	Nylon membrane	n.i.	0.18	30	Dong et al., (2014)
n.i.	Atrazine	n.i.	n.i.	n.i.	n.i.	Lin et al., (2018)
NanoZnO partcicles	Pharmaceuticals	PES membrane	No membrane	0.16	0.25	You et al., (2019b)
Oasis HLB	Endocrine disrupting chemicals	Agarose	Nucleopore track-etch	0.35 – 2.00	20	Chen et al., (2018)
Oasis HLB	Household and personal care products	Agarose	Nucleopore track-etch	0.80	20	Chen et al., (2017)
Oasis HLB	Organophosphorus flame retardants	Agarose	PTFE	0.75	20	Zou et al., (2018)
Oasis HLB	Pesticides	Polyacrylamide	No membrane	0.77	3	Guibal et al., (2017)
Oasis HLB	Pharmaceuticals	Agarose	No membrane	0.16 - 0.84	7	Buzier et al., (2019)
Oasis HLB	Pharmaceuticals and pesticides	Agarose	n.i.	0.75	10	Amato et al., (2018)
Oasis HLB	Pharmaceuticals and pesticides	Agarose	No membrane	0.75	7	Challis et al., (2018a)
Oasis HLB	Pharmaceuticals, hormones and pesticides	Agarose	No membrane	1.00	7	Challis et al., (2016)
Oasis HLB	Pharmaceuticals, hormones and pesticides	Agarose	No membrane	0.75	8	Stroski et al., (2018)
Oasis HLB	Pharmaceuticals, hormones and pesticides	Agarose	No membrane	0.75	7	Challis et al., (2018b)

Table 1: Configuration of o-DGT (*n.c.* not concerned, *n.i.* not indicated).

Oasis MAX	Pesticides	Polyacrylamide	No membrane	0.75	3	Guibal et al., (2017)
PCM	Pharmaceuticals	Agarose	PES	0.80	1	Ren et al., (2018)
Sepra ZT	Pharmaceuticals, hormones and pesticides	Polyacrylamide	No membrane	0.75	7	Stroski et al., (2018)
Strata-X	Pesticides, endocrine disrupting chemicals and others	Agarose	No membrane	1.20	10	Belles et al., (2017)
Strata-X	Pesticides, endocrine disrupting chemicals and others	Agarose	No membrane	2.00	0.5 to 10	Belles et al., (2018)
TiO ₂	Glyphosate	Polyacrylamide	PES	0.91 (with prefilter)	8	Weng et al., (2019)
TiO ₂	Herbicides	Polyacrylamide	PES	0.80	10	Fauvelle et al., (2015)
XAD-18	Endocrine disrupting chemicals	Agarose	Nucleopore track-etch	0.35 – 2.00	20	Chen et al., (2018)
XAD-18	Hormones	Agarose	PVDF	0.75	20	W. Guo et al., (2017)
XAD-18	Illicit drug	Agarose	PES	0.80	20	C. Guo et al., (2017)
XAD-18	Perfluoroalkyl substances	Agarose	PES	0.75	20	Guan et al., (2018)
XAD-18	Pharmaceuticals	Agarose	Nylon	0.80	20	D'Angelo and Starnes, (2016)
XAD-18	Pharmaceuticals	Agarose	PES	0.80	20	Chen et al., (2013)
XAD-18	Pharmaceuticals	Agarose	PES	0.80	20	Chen et al., (2014)
XAD-18	Pharmaceuticals	Agarose	PES	0.80	20	Chen et al., (2015a)
XAD-18	Pharmaceuticals	Agarose	PES	0.80	20	Chen et al., (2015b)
XAD-18	Pharmaceuticals	Agarose	PES	0.80	n.i.	Zhang et al., (2018)
XAD-18	Sulfamethoxazole	Agarose	PES	0.80	20	Chen et al., (2012)
XAD-18	Tetracycline	Agarose	Nylon	0.80	20	D'Angelo and Martin, (2018)
XDA-1	Endocrine disrupting chemicals	Agarose	No membrane	0.80	10	Xie et al., (2018b)
XDA-1	Pharmaceuticals	Agarose	PES	0.80	10	Xie et al., (2018a)

Compounds	Binding phase	Authors
Bisphenols	Activated carbon	Zheng et al., (2015)
Bisphenols	XAD18, HLB and Strata-XL-A	Chen et al., (2018)
Bisphenols	XDA-1	Xie et al., (2018b)
Hormones	HLB	Challis et al., (2018b, 2018a, 2016); Chen et al., (2018); Stroski et al., (2018)
Hormones	Sepra ZT	Stroski et al., (2018)
Hormones	Strata-XL-A	Chen et al., (2018)
Hormones	XAD18	Chen et al., (2018); W. Guo et al., (2017)
Hormones	XDA-1	Xie et al., (2018b)
Household and personal care produc	XAD18, HLB and Strata-XL-A	Chen et al., (2017)
Illicit Drug	XAD18	C. Guo et al., (2017)
Organophosphorus flame retardants	HLB	Zou et al., (2018)
Other	HSAC	You et al., (2019a)
Other	MIP	Dong et al., (2014)
Other	Strata-X	Belles et al., (2018, 2017)
Other	XAD18	Chen et al., (2018)
Perfluoroalkyl	XAD18	Guan et al., (2018)
Pesticides	Activated carbon and cyclohexane	Bondarenko et al., (2011)
Pesticides	CDPM	Wei et al., (2019)
Pesticides	HLB	Amato et al., (2018); Challis et al., (2018b, 2018a, 2016); Guibal et al., (2017); Stroski et al., (2018)
Pesticides	MAX	Guibal et al., (2017)
Pesticides	n.i.	Lin et al., (2018)
Pesticides	Sepra ZT	Stroski et al., (2018)
Pesticides	Strata-X	Belles et al., (2018, 2017)
Pesticides	TiO ₂	Fauvelle et al., (2015); Weng et al., (2019)
Pesticides	XDA-1	Xie et al., (2018b)

Table 2: List of binding gels tested by authors.

Pharmaceuticals	HLB	Amato et al., (2018); Buzier et al., (2019); Challis et al., (2018b, 2018a, 2016); Stroski et al., (2018); Zhang et al., (2018)
Pharmaceuticals	MCX and activated carbon	Zhang et al., (2018)
Pharmaceuticals	nanoZnO	You et al., (2019b)
Pharmaceuticals	PCM	Ren et al., (2018)
Pharmaceuticals	Sepra ZT	Stroski et al., (2018)
Pharmaceuticals	Strata-X	Belles et al., (2017)
Pharmaceuticals	XAD18	Chen et al., (2015b, 2015a, 2014, 2013, 2012); D'Angelo and Martin, (2018); D'Angelo and Starnes, (2016); Xie et al., (2018a); Zhang et al., (2018)
Pharmaceuticals	XDA-1, LX-1180, XDA-600, LX-4027, D296, NKA-9 and CAD-40	Xie et al., (2018a)

Compounds	Utilisé	References
Bisphenol	No membrane	Xie et al., (2018b)
Bisphenol	Nucleopore track- etch	Chen et al., (2018)
Bisphenol	PES	Xie et al., (2018b)
Bisphenol	PTFE	Xie et al., (2018b)
Bisphenols	Mixed cellulose ester (MCE)	Zheng et al., (2015)
Bisphenols	Nylon	Zheng et al., (2015)
Bisphenols	PES	Zheng et al., (2015)
Bisphenols	PTFE	Zheng et al., (2015)
НСРР	Cellulose nitrate	Chen et al., (2017)
НСРР	Cyclopore track- etch	Chen et al., (2017)
НСРР	Nucleopore polycarbonate	Chen et al., (2017)
НСРР	Nucleopore track- etch	Chen et al., (2017)
НСРР	Nucleopore track- etch	Chen et al., (2017)
НСРР	PES	Chen et al., (2017)
Hormones	No membrane	Challis et al., (2018b, 2018a, 2016); Stroski et al., (2018); Xie et al., (2018b)
Hormones	Nucleopore track- etch	Chen et al., (2018)
Hormones	PES	Challis et al., (2016); Xie et al., (2018b)
Hormones	PTFE	Xie et al., (2018b)
Hormones	PVDF	W. Guo et al., (2017)
Illicit drug	Mixed cellulose ester (MCE)	C. Guo et al., (2017)
Illicit drug	Nylon	C. Guo et al., (2017)

Table 3: Outer protected layer tested by authors.

Illicit drug	PES	C. Guo et al., (2017)
Illicit drug	PTFE	C. Guo et al., (2017)
Organophosphorus flame retardants	PTFE	Zou et al., (2018)
Other	No membrane	Belles et al., (2017)
Other	Nucleopore track- etch	Chen et al., (2018)
Perfluoroalkyl	PES	Guan et al., (2018)
Pesticides	Glass-fiber	Bondarenko et al., (2011); Wei et al., (2019)
Pesticides	No membrane	Amato et al., (2018); Belles et al., (2018, 2017); Challis et al., (2018b, 2018a, 2016); Guibal et al., (2017); Stroski et al., (2018); Xie et al., (2018b)
Pesticides	PES	Challis et al., (2016); Fauvelle et al., (2015); Weng et al., (2019); Xie et al., (2018b)
Pesticides	PTFE	Xie et al., (2018b)
Pharmaceuticals	Mixed cellulose ester (MCE)	Zhang et al., (2018)
Pharmaceuticals	No membrane	Amato et al., (2018); Belles et al., (2017); Buzier et al., (2019); Challis et al., (2018b, 2018a, 2016); Stroski et al., (2018); You et al., (2019b)
Pharmaceuticals	Nylon	D'Angelo and Martin, (2018); D'Angelo and Starnes, (2016); Zhang et al., (2018)
Pharmaceuticals	PES	Challis et al., (2016); Chen et al., (2015b, 2015a, 2014, 2013, 2012); Ren et al., (2018); Xie et al., (2018a); Zhang et al., (2018)
Pharmaceuticals	PTFE	Zhang et al., (2018)
Phenols	Nitrocellulose	You et al., (2019a)

