Diffusive gradients in thin films: devices, materials and applications

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REVIEW

Diffusive gradients in thin films: devices, materials and applications

Cai Li[1,2] · Shiming Ding[2] · Liyuan Yang[1] · Yan Wang[2,4] · Mingyi Ren[1,2] · Musong Chen[2] · Xianfang Fan[2] · Eric Lichtfouse[3]

Abstract

A large number of ex situ sampling techniques have been used traditionally to investigate the impact and fate of pollutants in soil, sediment and waters. However, the distribution and form of chemical species present are often altered prior analysis, due to the alterations during sampling and transfer to the laboratory. Alternatively, a robust in situ passive sampling technique, diffusive gradients in thin films (DGT), has been developed for the measurement of labile concentrations, species and distribution of various solutes in soil, sediment and waters. Here we review the recent developments in DGT device configurations and components, e.g., binding agents, diffusive phases and filter membranes. We highlight new configurations for effectively reducing the measurement errors and the disturbance of environmental media. We discuss DGT applications for the analysis of soil, sediment and water, such as evaluation of bioavailability and toxicity, measurement of nutrients and organic substances, and assessment of relationships between multiple solutes. We also present the coupling of DGT with other in situ measurement techniques such as dialysis samplers (Peeper), diffusive equilibrium in thin films (DET) and planar optodes (PO).

Keywords Diffusive gradients in thin films (DGT) · Device · Passive sampling technique · Biogeochemical processes

Abbreviations

BPs	Bisphenols
BPA	Bisphenol A
BPF	Bisphenol F
BPB	Bisphenol B
DBL	Diffusive boundary layer
DGT	Diffusive gradients in thin films
DIFS	DGT induced fluxes in sediments or soils
DET	Diffusive equilibrium in thin films
HR-Peeper	High-resolution Peeper
PO	Planar optode
CID	Computer imaging densitometry
LA-ICP-MS	Laser ablation inductively coupled plasma mass spectrometry
ICP-MS	Inductively coupled plasma mass spectrometry
ICP-OES	Inductively coupled plasma optical emission spectrophotometry
GFAAS	Graphite flame atomic absorption spectroscopy
Zr-oxide	Zirconium oxide
SPR-IDA	Suspended particulate reagent-iminodiacetate
APA	Agarose cross-linked polyacrylamide
MBL	Mixed binding layer
SRP	Soluble reactive phosphorus
SWI	Sediment–water interface
DRP	Dissolved reactive phosphorous
ROL	Radial oxygen loss
1D, 2D	One-dimensional, two-dimensional
EDCs	Endocrine disrupting chemicals
HPCPs	Household and personal care products
4-CP	4-chlorophenol
PSS	Poly (4-styrenesulfonate)

✉ Shiming Ding
smding@niglas.ac.cn

✉ Liyuan Yang
youngliyuan@126.com

[1] School of Water Conservancy and Environment, University of Jinan, Jinan 250022, China

[2] State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing 210008, China

[3] CNRS, IRD, INRA, Coll France, CEREGE, Aix Marseille University, 13100 Aix-en-Provence, France

[4] Nanjing EasySensor Environmental Technology Co., Ltd, Nanjing 210018, China

PA	Polyacrylate
PEI	Poly (ethyleneimine)
Py-PEI	Polymer-bound Schiff base
SH-CNP	Thiol-modified carbon nanoparticle
Fe_3O_4NPs	Fe_3O_4 nanoparticle aqueous suspensions
PQAS	Polyquaternary ammonium salt
PVDF	Polyvinylidene fluoride
CN	Cellulose nitrate
PES	Polyethersulfone

Introduction

With industrial development and urbanization, increasing levels of discharge of environmental pollutants such as heavy metals, nutrients and organics, not only result in severe environmental problems, but also pose a great risk to human health (Henner et al. 1999; Dsikowitzky and Schwarzbauer 2014; Heim and Schwarzbauer 2013; Li et al. 2017). A large number of ex situ active sampling techniques have been used to investigate the impacts and fates of compounds in soils, sediments and water, such as direct sampling, centrifugation and filtration, and displacement (Henner et al. 1997; Gimbert et al. 2005; Wang et al. 2015). However, the distribution and form of the chemical species present are often altered prior to analysis, due to the processes involved in sampling and transfer to the laboratory. Traditional sampling and extraction techniques give access to the total extractable analyte concentrations, which usually include some biologically unavailable fractions of inert organic species and large colloids (Divis et al. 2005). However, full assessment of the potential risk of a target analyte requires analysis of the labile fraction rather than total analyte concentrations (de Paiva Magalhães et al. 2015; Divis et al. 2005; Fernandez-Gomez et al. 2011). It is crucial to understand the complex interaction of biogeochemical processes responsible for speciation and distribution of analytes, as well as the resupply kinetics from solid phase to solution phase in sediments or soils (Wu and Wang 2017).

In situ passive sampling techniques are considered an ideal alternative method to these conventional sampling techniques, with many advantages including low environmental disturbance, having low cost and time requirements, and avoiding the need for active media transport (Chen 2015; Zarrouk et al. 2013). Diffusive gradients in thin films (DGT) is an in situ passive sampling techniques commonly used in environmental chemistry, which has been applied to the detection of elements and compounds in natural environments, including water, sediment and soil. This technique uses a specially designed passive sampler that typically houses a binding layer and diffusive layer. Labile fractions, including free ions, labile complexes which can dissociate and mobile fractions released from solids, diffuse through

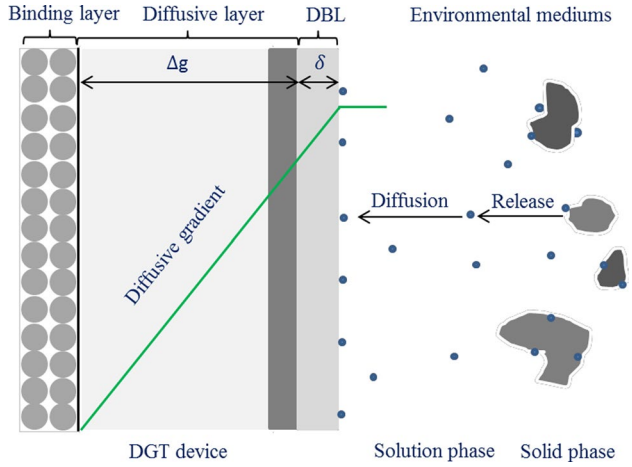

Fig. 1 Diffusive gradients in thin-film (DGT) device and mechanisms

diffusive layer and are then rapidly and irreversibly assimilated by the binding layer. Therefore, a diffusive gradient is established within the diffusive layer, between the internal medium and the binding layer interface (Fig. 1) (Ernstberger et al. 2002; Sun et al. 2013). Based on Fick's first law of diffusion, the time-weighted average concentration (C_{DGT}) of labile species during the deployment time (t) can be obtained using DGT, according to Eq. (1) (Davison and Zhang 1994):

$$C_{DGT} = \frac{M(\Delta g + \delta)}{DAt} \quad (1)$$

where Δg is the diffusion layer thickness; δ is the diffusive boundary layer thickness, which calculated based on previously reported methods (Davison and Zhang 2012; Galceran and Puy 2015; Scally et al. 2003); A is the exposed surface area of the DGT device; M is the accumulated mass of analyte in the binding gel; D is the analyte diffusion coefficient in the diffusive layer, determined by diffusion cell or DGT time-series deployment methods (Ding et al. 2016c; Pan et al. 2015; Zhang and Davison 1999).

Furthermore, DGT induced fluxes in sediments or soils (DIFS) modeling was developed to investigate the kinetics resupply of solutes to porewater from soil/sediment solids (Harper et al. 1998, 2000; Sochaczewski et al. 2007). Resupply mechanisms were described and evaluated using the resupply parameter from solid phase to solution phase R (C_{DGT}/C_{SOL}), the distribution coefficient between solid and porewater K_d (C_S/C_{SOL}), the solid phase adsorption rate constant k_1, the desorption rate constant k_{-1} and the response time T_C needed for the perturbed system to reach a 63% equilibrium level. DGT and DIFS therefore pose a significant advantage as compared to traditional measurement methods, as they can reflect the mobility, bioavailability and resupply

kinetics of solutes in environmental media (Gu et al. 2017; Naylor et al. 2006; Xu et al. 2018).

The DGT technique was developed in 1994 by William Davison and Hao Zhang with use for the determination of trace metals in water most common, during the early stages of development (Davison and Zhang 1994; Zhang and Davison 1995). Since then, the DGT technique has been modified and expanded to allow measurement of a significant number of elements and compounds, including multiple metals (Altier et al. 2016; Gao et al. 2009; Gimpel et al. 2003; Yabuki et al. 2014), nutrients (Cai et al. 2017; Menzies et al. 2005), organic chemicals/compounds (Chen et al. 2012a, 2013; Dong et al. 2014), radioactive elements (Drozdzak et al. 2015; Leermakers et al. 2009), oxyanions (Stockdale et al. 2008, 2010) and rare earth elements (Yuan et al. 2018). The simultaneous measurement of metals and oxyanions via DGT also provides an opportunity for analysis of the complex relationships between various elements (Mason et al. 2005; Panther et al. 2013; Wang et al. 2017a). Furthermore, the combination of DGT with other techniques has provided novel insight into different aspects of biogeochemical processes at high resolution, such as dialysis samplers (Peeper) (Xu et al. 2012a), diffusive equilibrium in thin films (DET) (Gao et al. 2007; Pradit et al. 2013) and planar optodes (PO) (Hoefer et al. 2017; Lehto et al. 2017). Obtaining a two-dimensional (2D) sub-mm high-resolution distribution of analytes in heterogeneous environments, such as sediments and the rhizosphere, is possible using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) (Stockdale et al. 2010; Warnken et al. 2004b) or computer imaging densitometry (CID) (Ding et al. 2013; Teasdale et al. 1999).

This study provides a comprehensive review of DGT development in recent years, including: device components, improved device configurations, and their application in soils, sediments and water. Moreover, the advantages and challenges of DGT technique are discussed.

Devices and components of diffusive gradients in thin films

To date, two types of DGT samplers have been reported, solid binding phase devices and liquid binding phase devices, with both types having different holder configurations. Solid binding phase devices consist of four types, including the piston-type and dual-mode devices used for analysis of solutions and dry soils (Luo et al. 2014; Pan et al. 2015), and the traditional and new flat-type probes used for analysis of sediments and flooded soils (Ding et al. 2016b; Wu et al. 2014; Zhang et al. 2002). DGT devices typically contain a binding layer which rapidly and irreversibly accumulates the solute species, a diffusive gel layer which allows solute species to pass through, a protective filter membrane which prevents adherence of particles to gels, and a front plate with a window which clips to the backing plate and holds all the layers firmly together. Recently, the improvements in traditional DGT device construction, as well as the developments in liquid binding phase device, have significantly expanded the potential applications.

DGT configuration

Measurement in solutions and dry soils

The piston-type device incorporates two components: the DGT piston and DGT cap (Fig. 2a). The binding gel, diffusive gel and filter membrane were sequentially laid together on the DGT piston and fixed by the DGT cap (exposure area: 3.14 cm^2), and the surface of the filter membrane is exposed to waters or soils during measurements. The typical piston-type device can be deployed into soils by hand pressing after the soil is moistened (Luo et al. 2010; Oporto et al. 2008; Santner et al. 2010; Zhang et al. 2004). The density of the soil, different thicknesses of soil layers, and the diffusive characteristics of target analytes may be potentially altered during pressing, which may result in measurement errors (Ding et al. 2016b).

In order to reduce the potential artificial interference for DGT measurement in soils, Ding et al. (2016b) developed the dual-mode DGT device, which has two different forms (Fig. 2b), with installation of the core on the top of the base for the measurement in water, or with the core inserted into the base to form an open cavity for measurements in soil. The deployment setup of dual-mode device differs significantly for soil measurements, as compared with piston-type device. Soil samples filled up to the open cavity with a height of 10 mm (i.e., the thickness of the soil layer is unified to 10 mm) after soils have been moistened, with devices then gently shaken by hand, to ensure complete and homogeneous contact between the DGT exposure surface and soils. The accuracy and variability of dual-mode and piston-type DGT devices were evaluated by determining the amounts of P, As, Cd and Pd with each device. Results indicated that accumulated masses established by both devices were significantly similar with correlation coefficients R^2 of 0.99 or 1.00. In addition, total measurement variability decreased from 5.57% in the piston-type DGT, to 3.37% in dual-mode DGT measurements (reduced by 42%).

Measurement in sediment and flooded soils

The flat-type probe is composed of a flat base with raised bottom edge for accommodation of the binding gel, diffusive gel and filter membrane, fixed by windowed frame (exposure area: 27 cm^2) (Fig. 3a), which is vertically deployed in

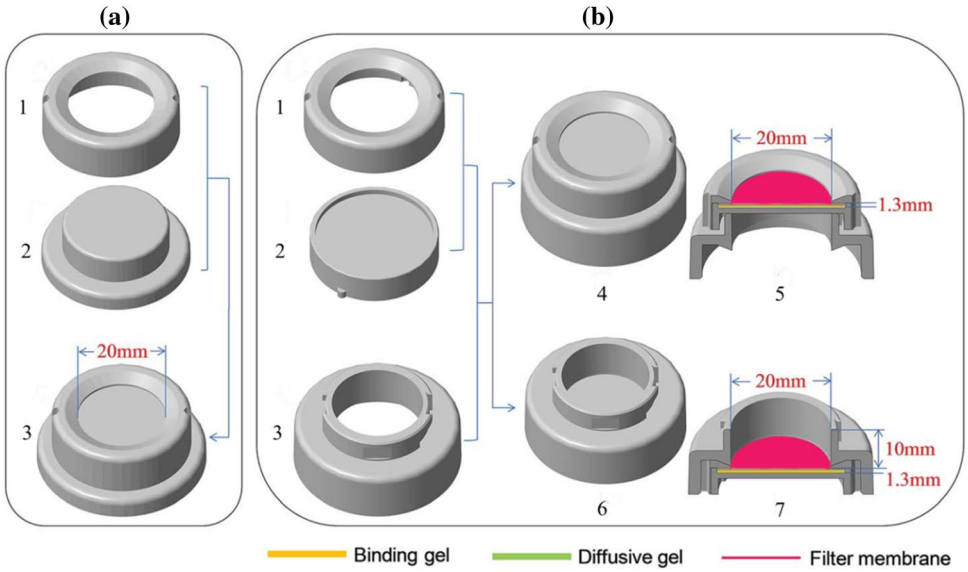

Fig. 2 Structure of the piston-type diffusive gradients in thin-film (DGT) device (**a**) and the dual-mode DGT device (**b**). **a** 1, the cap; 2, the piston and 3 their assembled form in the traditional holder system; **b** 1, the "O-shape" ring; 2, the recessed base of the DGT core in the new holder and 3, the hollow base (open cavity) for accommodation of the DGT core; 4 and 6 are the two different assembled forms of the DGT core and the open cavity for measurements in water and soil, respectively; 5 and 7 are the sectional drawing of 4 and 6, respectively (Ding et al. 2016b). Reprinted with permission from Ding et al., Copyright (2016b), The Royal Society of Chemistry

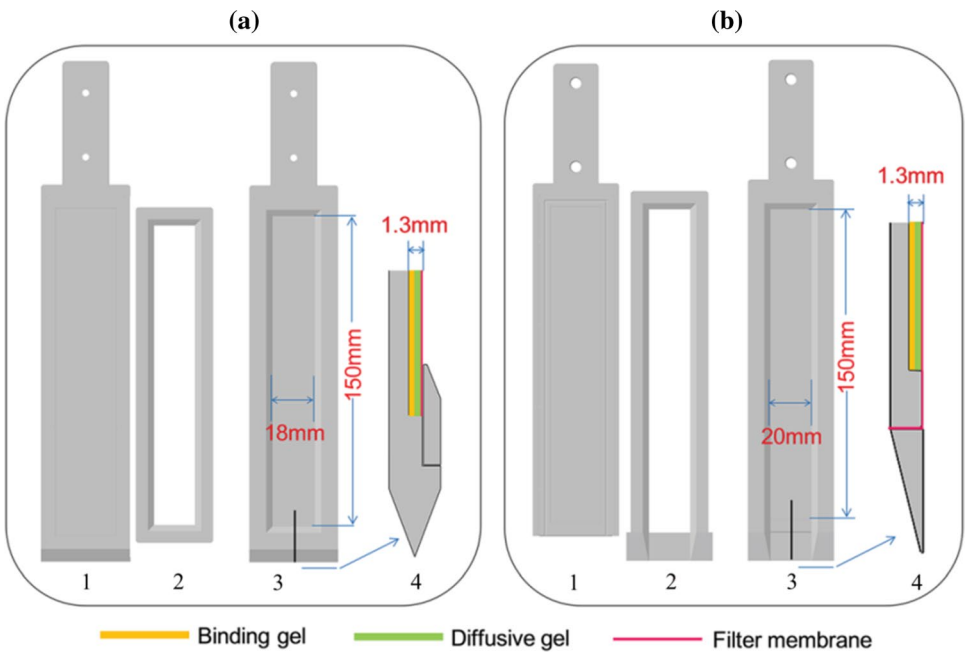

Fig. 3 Structure of the traditional flat-type diffusive gradients in thin-film (DGT) device (**a**) and the new flat-type DGT device (**b**): 1, the flat base; 2, the flat base and 3, their assembled form for the two types of probes, respectively; 4 is a cutaway view of the bottom of 3 loaded with binding gel, diffusive gel and a filter membrane (Ding et al. 2016b). Reprinted with permission from Ding et al., Copyright (2016b), The Royal Society of Chemistry

sediments or soils. Due to raised bottom edge, the typical flat-type probe can cause larger particles from the top layer of sediments to be transported down the sample profile and deeper layers during the insertion process (Santner et al. 2015). Additionally, the gap formed between the surface of flat-type probe and sediments due to raised bottom edge has been found to induce increased exchange in the overlying water (Ding et al. 2016b).

Furthermore, the flat-type probe for sediment measurements have been improved, with the significant differences being the removal of the raised bottom edge, as well as ensuring the window frame is flat at the front and has a beveled back (Fig. 3b). The performances of new and traditional flat-type DGT were compared for S(II) measurement, combining with ZrO–AgI binding gel and CID technique, The similar S(II) distribution trends were observed with both

devices from the 2D distribution of S(II). However, the new flat-type probe without a raised bottom edge, induces much less disturbance of solid particulates and overlying water from sediment surfaces, reducing transfer into the deeper layers during deployment, as compared to the traditional design. The penetration depths of dissolved sulfide and sulfide-enriched sands in sediments have been reduced to 33% and 20% of those measured using the traditional holder. Because the seepage of oxic overlying water into the sediment has been effectively inhibited using new holder, the distribution of sulfide with low concentration appeared at a much shallower depth (2.5 mm vs. 6 mm). The improved dual-mode DGT and new flat-type DGT devices have been successfully used for the measurement of various solutes in soils and sediments (Lin et al. 2017a; Xu et al. 2017; Zhang et al. 2017a).

Liquid binding phase device

The liquid binding phase device has mainly been applied to the measurement of solutes in water and soils, with a configuration similar to piston-type devices. As shown in Fig. 4, the typical liquid binding phase device is composed of two parts: the base with a reservoir for the placement of the binding phase solution; and the cap with a 29.5-mm open window which can be clipped onto the base (Liu et al. 2016; Wu et al. 2017). In order to avoid leakage of the liquid binding solution, a silica rubber spacer was fitted onto the base before the dialysis membrane was placed on the top of the spacer. As compared with solid binding phase DGT, liquid binding phase DGT provides excellent contact between the polymer solution and diffusive layer, overcoming fragility and swelling problems associated with solid gel. In addition, liquid binding phase DGT require simpler analytical procedures which directly measure analyte concentrations from polymer solution without complex elution steps (Chen et al. 2011; Fan et al. 2009b). However, the measurement using this type of DGT cannot reflect the spatial change of the analyte in environmental medium, due to that the binding agent is mobile in the binding solution.

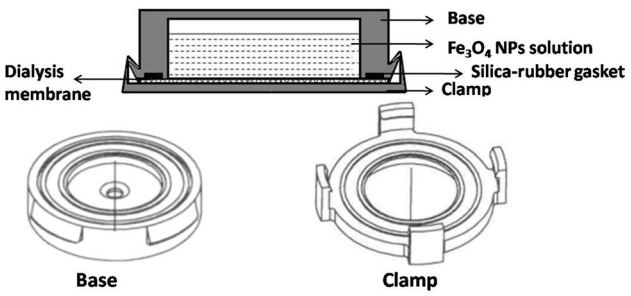

Fig. 4 The construction of diffusive gradients in thin-film (DGT) devices for liquid binding phase systems, comprised of a base with reservoir, for the placement of the binding phase solution and a cap with a 29.5-mm open window which can be clipped onto the base (Liu et al. 2016). *NP* nanoparticles. Reprinted with permission from Liu et al., Copyright (2016), Elsevier. N

In summary, the development of DGT configuration, including new solid and liquid binding phase devices, has expanded the application of this technique and reduced the disturbances on environmental media and induced measurement errors.

Binding agents

A large number of studies have suggested that labile species concentrations measured by different binding phases for same target analyte vary significantly (Ding et al. 2010b; Panther et al. 2011; Zhang et al. 2018a). Moreover, labile species concentrations are dependent on the binding strength and capability of the binding agent, providing possibility of using different DGT devices to determine the speciation and bioavailability of solutes, by choosing various suitable binding agents (Fan et al. 2013; Li et al. 2005a). To date, two different binding phases have been developed, including solid binding phases and liquid binding phases. Tables 1 and 2 summarize various solid binding agents and liquid binding agents, respectively, as well as their target analytes and applied conditions.

Single solid binding agents

Single solid binding phases are prepared by impregnating single binding agents (e.g., resin) into polyacrylamide or agarose gels (Guan et al. 2017; Zhang et al. 2004; Zheng et al. 2015). In addition, some ion exchange membranes are directly used as single solid binding phases for measuring metals with DGT (Li et al. 2002, 2006).

Chelex-100 resin is the most widely used solid binding adsorbent and has been applied for the measurement of trace metals in sediments, soils and waters (Divis et al. 2007; Song et al. 2018a; Zhang et al. 1998b). The most notable advantage of Chelex-100 resin is that it can determine a large number of metals, to date, the Chelex-100 is typically used to measure cationic metals with DGT, including Fe, Mn, Cd, Co, Cu, Ni, Pb, Zn and Al (Garmo et al. 2003; Leermakers et al. 2016; Song et al. 2018b; Turner et al. 2012; Wu et al. 2011). Other available materials have also been reported for the determination of metals, such as adsorbent suspended particulate reagent-iminodiacetate (SPR-IDA) and activated carbon (Davison et al. 1997; Lucas et al. 2012, 2014). SPR-IDA and activated carbon DGT can measure fewer analytes than Chelex-100 but activated carbon DGT can be applied for Au and bisphenols (BPs) measurement over a wide range of pH and ionic strength conditions (Guan et al. 2017; Lucas et al. 2012; Warnken et al. 2004a). Both

Table 1 Binding agents for preparation of solid binding phases in diffusive gradients in thin films (DGT)

Solid binding agents	Target analytes	Analytical techniques	Applied condition	References
Chelex-100	Zn, Cd, Co, Ni, Cu, Al, Pb, Cr, Mn, Fe, As, Hg, U	ICP-MS[b], ICP-AES[c] LA-ICP-MS	pH: 5.0–8.5; I: 0.01 mM–0.75 M $NaNO_3$	Divis et al. (2007), Gao et al. (2009), Leermakers et al. 2016), Song et al. (2018a), Wu et al. 2011)
SPR-IDA[a]	Co, Ni, Cu, Zn, Cd, Pb	LA-ICP-MS[d]	pH: 4.0–8.0	Davison et al. (1997), Gao and Lehto (2012), Warnken et al. (2004a, b)
Activated carbon	Au, As, Sb, bisphenols (BPs)	ICP-MS, HPLC[e]	pH: 2.0–9.5 I: 0.001–0.5 M NaCl	Lucas et al. (2012, 2014), Zheng et al. (2015)
Saccharomyces cerevisiae	Cd, Pb, MeHg	CV-AFS, ICP-OES[f]	pH: 4.5–8.5I: ≥ 0.005 M $NaNO_3$	Menegário et al. (2010), Pescim et al. (2012), Tafurt-Cardona et al. (2015)
Amberlite IRA 910	As, V	HG-AFS[g]	pH: 3–9 I: 0.001–0.05 M $NaNO_3$	Rolisola et al. (2014), Luko et al. (2017)
Whatman P81 membrane	Cd, Cu, Pb, Zn, Co, Ni, Mn, Ba, Hg, U species	FAAS[h], ICP–MS	pH: 4–9	Colaço et al. (2014), de Oliveira et al. (2012), Larner and Seen (2005), Pedrobom et al. (2017)
Whatman DE 81 membrane	Cr, U species	ICP-OES, ICP-MS	pH: 4–9 I: 0.01–0. 5 M $NaNO_3$	Li et al. (2006), Suárez et al. (2016)
AgI	S	CID[i]		DeVries and Wang (2003), Teasdale et al. (1999), Widerlund and Davison (2007)
Ferrihydrite	As, Mo, Sb, V, W, Se, P	ICP-SFMS[j]		Osterlund et al. (2010), Price et al. (2013), Zhang et al. (1998a)
Metsorb	As, V, Sb, Mo, W, Se, P, U, Glyphosate and Aminomethyl Phosphonic Acid	ICP-MS	pH: 4.0–8.3; I: 10^{-3}–0.7 M $NaNO_3$	Bennett et al. (2010), Drozdzak et al. (2015), Fauvelle et al. (2015), Panther et al. (2010, 2013), Price et al. (2013)
Zr-oxide	P, As, Cr, Mo, Sb, Se, V, W	ICP-MS, CID	pH: 3–10 I: 0.01–750 mM $NaNO_3$	Ding et al. (2010b, 2011, 2016c), Guan et al. (2015), Sun et al. 2014)
MnO_2	Ra	ICP-MS		Gao et al. (2010), Leermakers et al. (2016)
Spheron-Thiol	Hg	HPLC–ICP-MS[k]		Cattani et al. (2008), Docekalova and Divis (2005), Fernandez-Gomez et al. (2011)
Tulsion® CH-95	Hg, MeHg	CVA-FS[l]	pH: 4.10–10.03/4.10–8.10 I: 0.1 mM–1 M NaCl	Ren et al. (2018a)
3-mercaptopropyl functionalized silica gel	MeHg, MMHg	GC-ICP-MS[m] GC-Py-AFS[n]		Clarisse et al. (2009, 2012), Fernandez-Gomez et al. (2014), Liu et al. (2012)
Chelex–Metsorb	Mn, Co, Zn, Ni, Cu, Cd, Pb, V, As, Mo, Sb, W, P	ICP-MS	pH: 5.03–8.05 I: 0.001–0.7 M	Arsic et al. (2018), Panther et al. (2014)
Chelex–Ferrihydrite	Zn. Pb, Cd, Cu, Mn, Mo, P, As	ICP-MS	pH: 3–8 I: 0.001–0.01 M NaCl	Huynh et al. (2012), Mason et al. (2005)
AgI–Ferrihydrite	P, V, As, Mo, Sb, W and U	LA-ICP-MS		Stockdale et al. (2010)
ZrO–AgI	P, S, As	CID, HG-AFS[o]	pH: 4–9; I: 0.01–750 mM NaCl	Ding et al. (2012), Xu et al. (2017)
ZrO–SPR-IDA	P, As, Co, Cu, Mn, and Zn	LA-ICP-MS	pH 4–8 I: 1–100 mM $NaNO_3$	Kreuzeder et al. (2013)

Table 1 (continued)

Solid binding agents	Target analytes	Analytical techniques	Applied condition	References
ZrO–Chelex	Fe, Mn, Co, Ni, Cu, Zn, Pb, Cd, P, As, Cr, Mo, Sb, Se, V, W	CID, ICP-MS	pH: 5–9 I: 1–750 or 2/3–750 mM $NaNO_3$	Ding et al. (2016a), Wang et al. (2016, 2017a)
Micro-sized zeolite	NH_4^+–N	ICP-AES	pH: 3–8 I: 0.001–10 mM $NaNO_3$	Feng et al. (2015)
Microlite PrCH cation exchange resin	NH_4^+–N	Automated colorimetric method	pH: 3.5–8.5 I: ~0.012 M NaCl	Huang et al. (2016c)
Purolite A520E anion exchange resin	NO_3–N	Automated colorimetric method	pH: 3.5–8.5 I: 0.0001–0.008 M NaCl	Huang et al. (2016a)
AMI-7001 anion exchange membrane	NO_3–N	Automated colorimetric methods	pH: 3.5–8.5 I: 0.0001–0.014 M NaCl	Huang et al. (2016d)
CMI-7000 cation exchange membranes	NH_4^+–N	Automated colorimetric methods	pH: 3.5–8.5 I: 0.0003–0.012 M NaCl	Huang et al. (2016d)
SIR-100-HP	NO_3–N	Flow injection analysis	pH: 3–8 I: 0–0.018 M Na_2SO_4	Cai et al. (2017)
XAD-18 resin	Antibiotics/drugs/perfluoroalkyl substances (PFASs)/endocrine disrupting chemicals	HPLC–UV[p] RRLC–MS/MS[q]	pH: 6–9/4–9 I: 0.001–0.1 M NaCl	Chen et al. (2012a, 2013, 2018c), Guan et al. (2018), Guo et al. 2017a)
XDA-1 resin	Antibiotics/endocrine disrupting chemicals (EDCs)	UPLC-MS/MS[r]	pH: 7–9 I: 0.5/0.4–0.8 M NaCl	Xie et al. (2018a, b)
Oasis® HLB or Oasis® MAX	Anionic pesticides/endocrine disrupting chemicals/polar organic contaminants	HPLC-TOF[s]	pH: 3–8 I: 0.01–1 M $NaNO_3$	Challis et al. (2016), Chen et al. (2018c), Guibal et al. (2017)
Bondesil® C_8 silica particles	Organotin compounds	PTV-LVI-GC/MS[t]	pH: 4–9 I: 0.01–1 M, NaCl	Cole et al. (2018)
Hydrophilic–lipophilic-balanced	Household and personal care products (HPCPs)	LC-MS/MS[u]	pH: 3.5–9.5 I: 0.001–0.1 M NaCl	Chen et al. (2017b)

[a]Suspended particulate reagent-iminodiacetate (SPR-IDA)

[b]Inductively coupled plasma mass spectrometry (ICP-MS)

[c]Inductively coupled plasma atomic emission spectrometry (ICP-AES)

[d]Laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS)

[e]High-performance liquid chromatography (HPLC)

[f]Inductively coupled plasma optical emission spectroscopy (ICP-OES)

[g]Hydride generation-atomic fluorescence spectrometry (HG-AFS)

[h]Flame atomic absorption spectroscopy (FAAS)

[I]Computer imaging densitometry (CID)

[j]Inductively coupled plasma sector-field mass spectrometry (ICP-SFMS)

[k]High-performance liquid chromatography inductively coupled plasma mass spectrometry (HPLC–ICP-MS)

[l]Cold vapor atomic fluorescence spectrometry (CVA-FS)

[m]Gas chromatography coupled inductively coupled plasma mass spectrometry (GC-ICP-MS)

[n]Gas chromatography coupled to atomic fluorescence spectrometry via a pyrolytic reactor (GC-Py-AFS)

[o]Hydride generation coupled with AFS (HG-AFS)

[p]High-performance liquid chromatography ultraviolet (HPLC–UV)

[q]Rapid resolution liquid chromatography-tandem mass spectrometry (RRLC–MS/MS)

[r]Ultra-performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS)

[s]High-performance liquid chromatography time-of-flight (HPLC-TOF)

[t]Pressure temperature vaporization-large volume injection gas chromatography-mass spectrometry (PTV-LVI-GC/MS)

[u]Liquid chromatography-tandem mass spectrometry (LC-MS/MS)

Table 2 Binding agents used for preparation of the liquid binding phase in diffusive gradients in thin films (DGT)

Liquid binding phase	Target analyte	Analytical techniques	Applied condition	References
Poly(4-styrenesulfonate)	Cu, Cd, Co, Ni	FAAS[a]ICP-MS[b]	pH: 4–8 Not suitable for high ionic strength	Chen et al. (2012b), Li et al. (2003, 2005b)
Polyvinyl alcohol	Cu	FAAS	pH: 5.6–8.6 I: 10^{-4}–0.7 M $NaNO_3$	Fan et al. (2009a)
Polyacrylate	Cu, Cd	FAAS	pH: 4–8 Not suitable for high ionic strength	Fan et al. (2009b)
Polymer-bound Schiff base	Cu, Cd, Pb	FAAS	pH: 4–8.5 I: 10^{-4}–0.1 M $NaNO_3$	Fan et al. (2013)
Poly (ethyleneimine)	Cu, Cd Pb	FAAS	pH: 4–8 I: 10^{-4}–0.1 M $NaNO_3$	Sui et al. (2013)
Thiol-modified carbon nanoparticle (SH-CNP)	Hg	ICP-MS	pH: neutral value I: 0.001–0.5 M $NaNO_3$	Wu et al. (2017)
Fe_3O_4 NPs	As/P	ICP-MS ICP-OES[c]	pH: 4.5–9/3–10 I: 0.001–0.5 M $NaNO_3$	Liu et al. (2016), Zhang et al. 2018a)
Polyquaternary ammonium salt (PQAS)	P	AMS[d]	pH: 3–10 I: 10^{-4}–1 M $NaNO_3$	Chen et al. (2014b)
Zr-based metal–organic frameworks	P		pH: 6.5–8.5 I: 10^{-5}–0.1 M $NaNO_3$	Qin et al. (2018)

[a]Flame atomic absorption spectroscopy (FAAS)

[b]Inductively coupled plasma mass spectrometry (ICP-MS)

[c]Inductively coupled plasma optical emission spectrophotometry (ICP-OES)

[d]Ammonium molybdate spectrophotometry (AMS)

SPR-IDA (bead size 0.2 μm) and ground Chelex-100 resin (bead size ~ 10 μm) allow obtaining high-resolution imaging of trace metals in sediments and soils in combination with the analysis of LA-ICP-MS (Zhou et al. 2018). Taking the expensive prices and limited binding capacity of SPR-IDA into consideration, the ground Chelex-100 resin can become a promising binding material for high-resolution 2D imaging analysis. Menegário et al. (2010) fixed baker's yeast (Saccharomyces cerevisiae) in agarose gel to form a DGT binding phase, and the labile Cd(II) measured by this type of DGT showed excellent agreements with that of Chelex DGT. This type of DGT has been developed for measuring Pb and MeHg in seawaters and freshwaters (Pescim et al. 2012; Tafurt-Cardona et al. 2015). Rolisola et al. (2014) and Luko et al. (2017) immobilized Amberlite IRA 910 into polyacrylamide gel to form a DGT binding phase capable of measuring As(III) and V, and the DGT performance was independent of pH 5–9 and pH 3–9 for the two analytes, respectively.

In addition, several binding materials have been developed for the measurement of Hg species. Spheron-Thiol resins were developed to determine Hg(II) and MeHg, showing a stronger affinity for mercury species than Chelex-100 (Docekalova and Divis 2005; Fernandez-Gomez et al. 2011). Three-mercaptopropyl functionalized silica (Clarisse et al. 2012; Fernandez-Gomez et al. 2015), the ion exchange resin Duolite GT73 and Ambersep GT74 as well as the novel functionalized macroporous cross-linked polystyrene (Tulsion® CH-95) can measure several mercury species (Pelcova et al. 2014, 2015; Ren et al. 2018a). Further, we found the Tulsion® CH-95 binding agent has higher adsorption capacity and elution efficiency for Hg(II) and MeHg than other adsorbents (Ren et al. 2018a).

The use of ion exchange membranes as DGT binding phases has advantages of excellent mechanical strength, flexibility, reuse and easy preparation, in comparison with other types of binding phases (Huang et al. 2016d; Li et al. 2002, 2005a). Li et al. (2002) utilized commercially available solid cellulose phosphate membranes (Whatman P81) as DGT binding phase to determine Cu and Cd, and this type of DGT has been extended to measurement of other metals, such as Cd, Cu, Pb, Zn, Co, Ni, Mn, Ba, Hg and U species (Colaço et al. 2014; de Oliveira et al. 2012; Larner and Seen 2005; Mengistu et al. 2012; Pedrobom et al. 2017). In addition, the Whatman DE 81 with amino functional groups has been applied for the measurement of U species and Cr(VI) in waters (Li et al. 2006; Pedrobom et al. 2017; Suárez et al. 2016).

Titanium dioxide-based adsorbent (Metsorb), iron oxyhydroxide-based adsorbents (Ferrihydrite) and zirconium oxide (Zr-oxide)-based adsorbents have been employed to measure anionic analytes. Metsorb DGT, Ferrihydrite DGT and

Zr-oxide DGT can measure similar anionic analytes including As, Mo, Sb, V, W, Se, P in a similar range of pH (Ding et al. 2016c; Osterlund et al. 2010; Panther et al. 2010, 2013; Zhang et al. 2017a, b). Compared with Ferrihydrite, the capacity of Metsorb DGT was higher, with superior results found when deployment times were greater than 1 day or in seawater deployment. Both Metsorb and Ferrihydrite DGTs were found to be independent of Cl^- and SO_4^- for the measurement of dissolved reactive phosphorous (DRP) in water, whereas Ferrihydrite DGT was more susceptible to the effects of HCO_3^- (Mason et al. 2008; Panther et al. 2011). A high phosphate binding capacity was observed for Zr-oxide DGT, up to 223 μg P cm^{-2}, which was significantly higher than Ferrihydrite DGT (6.94 μg P cm^{-2}), Metsorb DGT (12.7 μg P cm^{-2}) (Ding et al. 2010b). The capacities of Zr-oxide DGT for oxyanions are ~ 29 to > 2397-fold that of Metasorb and approximately 7.5 to 232-fold that of Ferrihydrite DGT in freshwater and seawater, respectively (Ding et al. 2016c). Teasdale et al. (1999) initially developed a new DGT method for 2D measurement of S(II), based on the use of AgI as DGT binding phase. S(II) reacts specifically with the pale yellow AgI to form black Ag_2S substance. Therefore, the accumulation amount of S(II) can be quantitatively related to the grayscale intensity of the gel surface measured by the computer imaging density (CID) (Robertson et al. 2008; Teasdale et al. 1999).

Recently, DGT methods have been widely developed to measure the inorganic nutrient P (Ding et al. 2010b; Price et al. 2013; Zhang et al. 1998a), while there were relatively few reports about N. Feng et al. (2015) determined NH_4–N concentrations using DGT, based on the use of a novel binding material containing micro-sized zeolite in natural waters, effective between pH 3–8 and 0.001–10 mM ionic strengths. Huang et al. (2016a, c) used Purolite A520E anion exchange resin and Microlite PrCH cation exchange resin and as DGT binding agents for measurement of NO_3–N and NH_4–N in freshwaters, respectively, with neither A520E DGT or PrCH DGT affected in the pH range of 3.5–8.5. Huang et al. (2016d) also used AMI-7001 and CMI-7000 cation exchange membranes as binding layers for DGT measurements of NO_3–N and NH_4–N in freshwaters, respectively. They have the advantages of easy preparation, low detection limits and the use of non-toxic chemicals (i.e., acrylamide) in comparison with the uses of A520E and PrCH, enable the two DGTs more promising for measurement of N. Moreover, a novel styrene divinylbenzene-based absorbent with amine functional groups (SIR-100-HP) has been developed for the measurement of NO_3–N in soils, with performance found to be effective at a pH range of 3–8 and ionic strengths of 0–0.018 M (Cai et al. 2017).

The binding agents used for the measurement of metals, oxyanions and nutrients have been considerably developed, while there is a tendency for the development of DGT in measurement of organic compounds. XAD-18 resin is to date, the most commonly used adsorbent for measuring antibiotics and it has been developed to simultaneously several drugs and perfluoroalkyl substances (PFASs) in water and soil (Chen et al. 2014a, 2015a; Guan et al. 2018; Guo et al. 2017a). Some studies have proven that XAD-18 DGT is effective in situ tools to monitor multiple antibiotics and illicit drugs in the wide range of pH 4–9, with ionic strengths of 0.001–0.1 M NaCl (Chen et al. 2015b; Zhang et al. 2018b). In addition, Xie et al. (2018a, b) used the novel XAD-1 resin as a binding phase for the determination of antibiotics and endocrine disrupting chemicals (EDCs) in seawater. Compared with XAD-18 resin, the XAD-1 resin was suitable for application in high ionic strength environments (0.4–0.8 M), such as seawater. Activated charcoal DGT has also been employed for the simultaneous determination of bisphenols in water and soil, with performance not affected by pH within the range of 5–8, or ionic strengths between 0.001 and 0.5 M (Guan et al. 2017; Zheng et al. 2015). Ren et al. (2018b) developed a novel binding phase using porous carbon material (PCM) as binding agent for measuring 20 antibiotics in water samples, and its measurement was independent of pH 4.2–8.4 and ionic strength 0.001–0.5 M NaCl. Guibal et al. (2017) utilized both Oasis® HLB and Oasis® MAX sorbent in DGT devices, to monitor anionic pesticides in rivers, with accuracies better than 30% found under the conditions of pH 3–8 and ionic strength 0.01–1 M $NaNO_3$. The application of hydrophilic–lipophilic-balanced (HLB) binding agents has been reported for the measurement of household and personal care products (HPCPs) in waters, being effective in the pH range of 3.5–9.5, and ionic strengths of 0.001–0.1 M, with dissolved organic matter levels of 0–20 mg L^{-1} (Chen et al. 2017b). Chen et al. (2018c) further compared the performances of three different resins [HLB, XAD18 and Strata-XL-A (SXLA)] for measuring endocrine disrupting chemicals, showing that the measurements with HLB and XAD18 DGT were more stable than that of SXLA-DGT, with the tolerant pH range of 3.5–9.5 and ionic strength of 0.001–0.5 M. The use of DGT for measurement of organometallics have been optimized, with Cole et al. (2018) using Bondesil® C_8 silica particles as DGT sorbents for measurement of five organotin compounds in sediment, with performances effective within a range of pH 4–9 and ionic strength of 0.01–1 M NaCl.

Hybrid solid binding agents

Both direct and indirect interactions occur between different analyte species in biogeochemical processes. Consequently, in order to further understand potential coupled relationships, the hybrid DGT incorporating different adsorbents in a single gel has been developed. The concentrations of Zn, Pb, Cd, Cu, Mn, Mo, P and As in water and soil, were

determined using mixed binding layer (MBL) DGT, consisting of Ferrihydrite and Chelex-100 (Ferrihydrite–Chelex DGT) (Huynh et al. 2012; Mason et al. 2005). The added Chelex-100 does not interfere with the absorption capacity of Ferrihydrite for As and P, although the coating of Ferrihydrite on to the surface of Chelex-100 can affect the capacity of Chelex-100 to absorb trace metals. In particular, Ferrihydrite–Chelex DGT cannot be used to measure Fe. The simultaneous measurement of six labile cations (Mn, Co, Ni, Cu, Cd, and Pb) and six oxyanions (V, As, Mo, Sb, W, and P) was performed using Metsorb–Chelex DGT both in freshwater and seawater (Arsic et al. 2018; Panther et al. 2014). Both the diffusion coefficients and the measurement concentrations of cations and oxyanions agreed well with Metsorb–Chelex DGT, compared with Metsorb DGT and Chelex DGT, respectively. Both the number of analyte species measured and the accuracy were higher with Metsorb–Chelex DGT than Ferrihydrite–Chelex DGT.

In addition, AgI–Ferrihydrite DGT (for simultaneous measurement of S and oxyanion), and ZrO–AgI (for As, S and P measurement) have been developed in combination with some high-resolution analyte techniques, such as computer imaging densitometry (CID) (Ding et al. 2012; Stockdale et al. 2008, 2010; Xu et al. 2017). ZrO–SPR-IDA DGT was developed for 2D imaging of P, As, Co, Cu, Mn, and Zn distributions in combination with LA-ICP-MS, by embedding Zr-oxide and SPR-IDA into an ether-based urethane polymer hydrogel (Kreuzeder et al. 2013). Furthermore, Wang et al. (2016, 2017a) obtained the concentrations and distributions for eight cations (Fe, Mn, Co, Ni, Cu, Zn, Pb, and Cd) and eight anions (P, As, Cr, Mo, Sb, Se, V, and W) in water and sediments, using ZrO–Chelex DGT. ZrO–Chelex DGT was shown to have high capacities for all analytes, and although the capacity for As was lower than Zr-oxide DGT, it still exhibits significantly better responses than other reported DGT (Sun et al. 2014).

The main disadvantage of MBL DGT is the limited binding capacity, due to having a single layer of mixed gel and therefore reduced amounts of adsorbents, as well as the mutual interference between different adsorbents (Huynh et al. 2012). It is to note, that adding increased concentration of binding agents into mixed gels were demonstrated to be useful for elevating the binding capacity of MBL DGT (Wang et al. 2017a).

Liquid binding agents

Liquid binding phases are typically polymer solutions containing functional groups, with recent applications including homogeneous suspension solutions with nano-particulates, the homogeneous liquid binding phase with nano-particulates offers the advantages of a large surface area, rich in functional group, with high mobility (Liu et al. 2016).

The use of poly (4-styrenesulfonate) (PSS) and sodium polyacrylate (PA) as the liquid binding phase for DGT, have been developed for the measurement of Cu and Cd (Fan et al. 2009b; Li et al. 2003), but PSS DGT is not suitable for long-term deployment under high ionic strength condition (Li et al. 2003, 2005b). Several metals (Cu, Cd, Co and Ni) have also been measured simultaneously using PSS-E6 and PSS-7E4 DGTs (Chen et al. 2012b), respectively. The results showed that the use of large molecular weight PSS-E6 was more suitable than PSS-7E4 due to lower pretreatment depletion rate, and lower cost and pollution. In addition, polymer-bound Schiff base (Py-PEI) and poly (ethyleneimine) (PEI) were employed as DGT liquid binding agents for the measurement of labile Cu, Cd and Pb (Fan et al. 2013; Sui et al. 2013). Py-PEI DGT has higher uptake percentages for metals than PA DGT and PSS DGT. Labile Hg species were determined using thiol-modified carbon nanoparticle (SH-CNP) suspensions as the DGT liquid binding agent, with performance effective in ionic strength of independence of 0.001–0.5 M $NaNO_3$, as well as other interfering ionic Cd, Cr, Cu and Pb (Wu et al. 2017). In addition, Fe_3O_4 nanoparticle aqueous suspensions (Fe_3O_4NPs) have been applied as liquid binding phases in DGT measurement of DRP concentrations and As species (Liu et al. 2016; Zhang et al. 2018a). Furthermore, polyquaternary ammonium salt (PQAS) aqueous solutions have also been applied as the liquid binding phase in DGT systems, to monitor the concentration of DRP in water, with effective performance within pH range of 3–10 and at ionic strengths of 10^{-4}–1 M (Chen et al. 2014b). Qin et al. (2018) used Zr-based metal–organic frameworks (MOFs, UiO-66) as liquid binding agent for DGT measurement of P (UiO-66 DGT), and good agreements were obtained between the concentrations measured by DGT and in waters (C_{DGT}/C_{sol}: 0.9–1.1). The capacities of different DGTs in measurement of P are in the order of Zr-oxide DGT (223 μg P cm^{-2}) > UiO-66 DGT (20.8 μg P cm^{-2}) > Fe_3O_4NPs DGT (15.4 μg P cm^{-2}) > Metsorb DGT (12.7 μg P cm^{-2}) > PQAS DGT (9.9 μg P cm^{-2}) > Ferrihydrite DGT (6.94 μg P cm^{-2}) (Qin et al. 2018; Zhang et al. 2018a).

In summary, a large number of binding agents have been developed, and their uses have made DGT to be a commonly used sampling technique for measurement of a wide range of analytes.

Diffusive phase

A series of materials have been applied as the diffusive gel in DGT (Table 3), including agarose cross-linked polyacrylamide (APA) gel (Shiva et al. 2017; Zhang and Davison 1995), agarose gel (Colaço et al. 2012; Docekalova and Divis 2005; Wang et al. 2016), dialysis membranes (Li et al. 2003), nylon membranes (Panther et al. 2008) and

Table 3 Types and characteristics of diffusive gels used in diffusive gradients in thin films (DGT)

Diffusive gel	Pore size	Thickness	Moisture content	Analytes	References
APA gel	> 5 nm	Typically 0.8 mm	> 95%	Oxyanions (including PO_4^{3-}), trace metals except of Hg and NH_4^+, and organic pollutants	Chen et al. (2011), Wang et al. (2016), Zhang and Davison (1999)
Agarose gel	> 20 nm	Typically 0.8 mm	> 99%	Various cations and anions, organic pollutants, Hg, NH_4^+, and the analytes capable and incapable of measurements with the APA gel	Divis et al. (2005), Wang et al. (2016)
Dialysis membrane	< 5 nm	50–85 μm	–	Cu, Cd, Co, Ni and As	Chen et al. (2012b), Li et al. (2003, 2005b)
Nylon membrane	0.22 μm	0.18 mm	–	4-chlorophenol (4-CP)	Dong et al. (2014)
3MM chromatography paper		0.46 mm		Various cations	de Almeida et al. (2012), de Oliveira et al. (2012)

Table 4 Some reported diffusion coefficients for Hg species, NH_4–N and some organic species in agarose gel ($D \pm$ standard deviations; $\times 10^{-6}$ cm^2 s^{-1}, 25 °C)

Element	*D*	References
Hg	9.07 ± 0.23/7.65 ± 0.60	Pelcova et al. (2014), Ren et al. (2018a)
MeHg	9.06 ± 0.30/9.94 ± 0.49	Pelcova et al. (2014), Ren et al. (2018a)
NH_4–N	15.2/17.1	Feng et al. (2015), Huang et al. (2016c)
BPA/BPB/BPF	5.03/2.64/4.44	Zheng et al. (2015)
HPCPs	3.36–7.30	Chen et al. (2017b)
Anionic pesticides	2.7–4.6 (22 °C)	Guibal et al. (2017)
17β-estradiol (E2)	4.65 ± 0.37	Guo et al. (2017b)
Methcathinone/ephedrine	7.60/6.62	Zhang et al. (2018b)
Ketamine/methamphetamine/amphetamine	8.13 ± 0.12/8.55 ± 0.14/7.72 ± 0.18 (22 °C)	Guo et al. (2017a)

paper-based diffusion layer, such as 3MM chromatographic paper (de Almeida et al. 2012; de Oliveira et al. 2012) as well as chromatography paper grade 1 Chr and 17 Chr (Larner and Seen 2005).

APA gel is the most widely used diffusive layer and has been employed for the measurement of various cations and anions, with advantage of retaining stability in the range of pH 2–9 after hydrogel (Osterlund et al. 2010; Wu et al. 2014). Nevertheless, some studies have shown that the interactions between APA gel functional groups and target analytes, can result in inaccurate measurements, such as with Hg (Divis et al. 2005; Pelcova et al. 2015; Tafurt-Cardona et al. 2015), Cu (Garmo et al. 2008b), NH_4^+ (Huang et al. 2016c), and some organics (Guan et al. 2017). APA gel possesses positive charge, therefore electrostatic interactions can interfere with metal diffusion at low ionic strengths according to the Donnan potential (Warnken et al. 2005). In addition, APA gel has relatively poor mechanical resistance, poor reproducibility and sophisticated preparation processes (Fan et al. 2013). An alternative diffusive material agarose gel, had been applied for the measurement of almost all DGT analytes, such as various cations, anions, organic pollutants, Hg and NH_4^+, as well as many analytes which cannot be measured with APA gel (Chen et al. 2012a; Divis et al. 2005; Huang et al. 2016d; Tafurt-Cardona et al. 2015; Wang et al. 2016); and Table 4 summarizes some reported diffusion coefficients of these species in agarose gel. The main binding sites on agarose gel are negative charges, which have attractive and repulsive interactions with cations and anions, respectively. Compared with the APA gel, the agarose gel (typically containing 1.5% agarose) is more stable in gel thickness, showing no swelling on hydration (APA gel showed 3.2-fold swelling) (> 99%) (Wang et al. 2016). It was also found that the thickness of agarose gel remained stable after hydration and storage under 4–40 °C, 0–1.0 M ionic strength, and 2–11 pH conditions, with the storage time extending to 300 days.

The reported diffusion coefficients for cations and anions in APA gel and agarose gel determined by diffusion cells (D_{cell}) and DGT time-series deployment methods (D_{DGT}), respectively, are presented in Table 5. The results indicated a considerable variation of the diffusion coefficients in APA

Table 5 Diffusion coefficients for some elements in agarose cross-linked polyacrylamide (APA) gel and agarose gel (1.5%) determined using diffusion cells D_{cell} and diffusive gradients in thin-film (DGT) time-series deployment methods D_{DGT}, respectively ($D \pm$ standard deviations; $\times 10^{-6}$ cm^2 s^{-1}, 25 °C)

Analyte	Agarose cross-linked polyacrylamide gel												Agarose gel
	D^a_{cell}	D^b_{cell}	D^c_{cell}	D^d_{cell}	D^e_{DGT}	D^f_{DGT}	D^g_{DGT}	D^h_{DGT}	D^i_{DGT}	D^j_{DGT}	D^k	RSD[l] (%)	D^m
As	5.26	5.36	5.54	5.21	6.02	8.13	5.96		6.78	5.54	6.09	**14.9**	6.38
Mo	5.18	5.58	6.28	5.96	6.33	6.43	7.24		6.81	5.70	5.94	9.9	6.53
Sb	4.90	5.50	6.04	5.55	6.22	6.16	6.74		6.86	5.25	6.23	**10.7**	6.59
V	3.75	3.88	7.14	6.72	7.98	8.13	8.92		8.02	6.69	5.85	**26.3**	5.91
W	6.22	4.28	6.89	5.45	6.88	7.06	6.50		6.26	5.82	5.77	**13.7**	6.54
Cd	5.52	5.36			5.56	5.72	5.56	5.50			8.03	**16.1**	8.41
Co	5.17	5.03			5.29	5.38	5.88	5.80			6.08	7.2	6.89
Cu	5.75	5.27			5.61	5.47	5.34	5.50			5.50	2.9	6.56
Mn	4.88	4.95			4.68	4.44	5.11	6.40			6.00	**13.8**	6.90
Ni	5.21	5.13			5.13	5.29	5.65	6.00			5.50	6.0	6.69
Pb	7.75	6.80			8.03	8.17	7.70	9.40			6.70	**11.7**	8.64
Zn	5.47	5.39			6.22	6.37	5.78	4.60			5.50	**10.5**	7.47

[a]Conditions: 0.01 M $NaNO_3$, pH 4.01(Panther et al. 2014)

[b]0.01 M $NaNO_3$, pH 4.00 (Shiva et al. 2015)

[c]0.01 M $NaNO_3$, pH 7.1 (Panther et al. 2013)

[d]0.01 M $NaNO_3$, pH 6.4 (Osterlund et al. 2010)

[e]0.01 M $NaNO_3$/0.004 M $Mg(NO_3)_2$, pH 6.06, using Chelex–Metsorb DGT (Panther et al. 2014)

[f]0.01 M $NaNO_3$/0.004 M $Mg(NO_3)_2$, pH 6.06, using Chelex DGT for anions and Metsorb DGT for cations

[g]0.01 M $NaNO_3$/0.004 M $Mg(NO_3)_2$, pH 4.01, using Chelex–Metsorb DGT (Shiva et al. 2015)

[h]pH = 5.90, using Chelex DGT (Garmo et al. 2003)

[i]0.01 M $NaNO_3$, pH 7.1; using Metsorb DGT (Panther et al. 2013)

[j]0.01 M $NaNO_3$, pH 6.2, using ferrihydrite DGT (Osterlund et al. 2010)

[k]Obtained from DGT Research Ltd.

[l]Relative standard error (RSD%) of the diffusion coefficients in APA gel using the data of a–k, with the error > 10% marked in bold

[m]The analyte diffusion coefficients in agarose gel 0.01 M $NaNO_3$, pH 5.50 for cations, Ph 6 for anions, using ZrO–Chelex DGT, obtained from EasySensor Ltd.

gel for the same analytes, even that the detection was performed in similar measurement conditions, using the same methods or by the same persons. Except for Cu, the errors (RSD%) of all other elements exceed 5%, and the errors of 8 elements even exceed 10%. These different reports have caused confusion to the users in calculation of DGT concentration and the difficulty in comparing the measuring results in different studies. The errors should be caused by differences in raw materials and production batches in preparation of the diffusion gel and binding gel. The diffusion coefficients measured by diffusion cells and DGT time-series deployment methods are also different, and the D_{DGT} are slightly higher than D_{cell} (Table 5). The reason is likely the presence of a diffusive boundary layer on each side of the diffusion gel in diffusion cell, even under fast stirring rate condition (Shiva et al. 2015). The measurement conditions of DGT time-series deployment method are closer to that of DGT deployment, compared to diffusion cell method. In addition, the diffusion coefficients for analytes in agarose gel are higher than that in APA gel, possibly due to the larger pore size of agarose gel (> 20 nm) than APA gel (> 5 nm) (Zhang and Davison 1995).

Dialysis membranes and nylon membranes have widely been employed in liquid binding phase DGT for the measurement of heavy metals. No strong interactions exist between metals and dialysis membranes; therefore dialysis membranes do not concentrate Cd and Cu ions from solutions in a wide range of ionic concentrations (Li et al. 2003). Moreover, the diffusive boundary layer is not significant when using dialysis membranes as compared with APA hydrogel or agarose gel, even in poorly mixed waters. The nylon membrane presents excellent mechanical strength and flexibility and has been developed to determine 4-chlorophenol (4-CP) concentration; due to it is inertness with organic compounds (Dong et al. 2014).

In addition, ultra-thin diffusion layer has been applied for DGT measurements, such as the use of a 0.01-mm Nuclepore membrane or a 0.1-mm Durapore® PVDF membrane (Ding et al. 2016a; Lehto et al. 2012). It is of note that the measured C_{DGT} level was significantly lower than the pore water concentration when using a thin diffusion layer

(10–100 μm thicknesses). Therefore, to avoid erroneous date-interpretation, DGT-measured result should be interpreted as the time-averaged flux F_{DGT} (Teasdale et al. 1999; Widerlund et al. 2012):

$$F_{DGT} = \frac{M}{At} \quad (2)$$

Compared with the conventional 0.80 mm thickness APA gel and agarose diffusive gel, ultra-thin diffusion layers can reduce the diffusion distance, diffusion time and vertical diffusion of analytes in diffusive layer. Therefore, they are suitable for investigating the distribution of mobile chemical species in soils and sediments at extremely high spatial resolutions.

In summary, the considerable variation in diffusion coefficients in APA gel is worthy of concern. The uses of agarose gel and ultra-thin membrane have improved the performance of DGT in expanding the range of analytes and increasing the spatial resolution. Particularly, the agarose gel can be used as a promising standard diffusive layer in DGT measurements of various inorganic and organic analytes.

Filter membrane

Filter membranes (usually 0.45 μm pore sizes) have been applied as protective layer, preventing some large particles from contaminating or damaging diffusive gels outer surface. Polyvinylidene fluoride (PVDF) membranes, cellulose nitrate membranes (CN), cellulose acetate membrane and hydrophilic polyethersulfone filter membranes (PES) are commonly used as DGT filter membrane (Desaulty et al. 2017; Kalkhajeh et al. 2018; Rolisola et al. 2014; Wang et al. 2016; Yabuki et al. 2014). Scally et al. (2006) reported the diffusion coefficients obtained by diffusion gels plus filter membranes were higher than those determined by the diffusion gel alone. Wang et al. (2016) further demonstrated that different filter membranes had a significant effect on the diffusion rates of anions and cations, showing the ranked order of diffusion rates to be: agarose < agarose plus PVDF < agarose plus PES < agarose plus CN. Compared the agarose gel, the larger pore size of filter membrane may be a reason for the increased diffusion rates. Some studies have indicated that different filter membranes have different affinities for solutes; and therefore, different filter membranes could result in alteration of diffusion coefficients (Garmo et al. 2008a, b; Weltje et al. 2003). In addition, the filter membrane thickness can also affect diffusion coefficients, while this aspect is rarely considered in DGT research.

A long deployment time is typically required for DGT deployment when measuring trace metals and other analytes with low concentrations in natural waters. However, the biofilm attachment on the surface of filter membrane after long deployment time could alter the diffusional properties of the DGT device (Li et al. 2005a). To overcome this problem, Pichette et al. (2007) utilized antibiotics, copper and silver to pretreat the filter membrane, respectively. The results showed that copper and silver could prevent algal colonization within 14 days after DGT deployment. Pichette et al. (2009) also suggested an optimized deployment time of 4 days, since good measurement results and reduced biofilm effects were obtained. Other treatment methods have been applied, including the uses of additional polycarbonate membrane on the surface of DGT devices to limit biofilm growth (Uher et al. 2012), corrected diffusion coefficient to revise the DGT measurement (Feng et al. 2016). For the liquid binding phase, the dialysis membrane can be replaced by a fresh membrane regularly, which can reduce the biofouling effects without disturbing the liquid binding phase (Li et al. 2005a).

DGT deployment and sample analysis

In general, DGT measurement needs to undergo four setups, including diffusive and binding gels preparation, DGT assembly and storage, DGT deployment and retrieval, and sample analysis and date treatment. For assembly of the DGT device, the binding gel is placed on DGT base bottom, which is covered in order by a diffusion gel and a filter membrane and fixed in the DGT holder. The DGT device is generally sealed in a clean plastic bag contains a small volume of 0.01 M $NaCl/NaNO_3$ solution, to keep a humid and ionic strength condition. Prior to deployment in anoxic soils and sediments, the loaded DGT device must be put into certain concentration of $NaCl/NaNO_3$ solution, with nitrogen gas filled into the solution for at least 16 h to remove the oxygen in the probe. This avoids oxygenation of the sediments or soils and the changes in the lability of solutes during deployment.

DGT deployment in sediments

The flat-type probe is usually deployed in flooded soils and sediments (Fig. 3). In laboratory, the flat-type probe can be inserted vertically and slowly into the sediment in a core or other containers by hands, with 2–4 cm length left in the overlying water (the probe has a totally 15 cm effective length). When applying in the field, some researchers have inserted the flat-type probes into sediments by diver or lander (Brodersen et al. 2017). The lander has relatively big volume and needs additional equipment (camera) to identify the sediment–water interface (SWI). Ding et al. (2015) developed a man-pack, small volume releasing device. The probe can be inserted into the sediment vertically utilizing the gravity of releasing device. After that, the device can be retrieved from the water column, leaving the DGT probe in the sediment. The releasing device can be applied in shallow

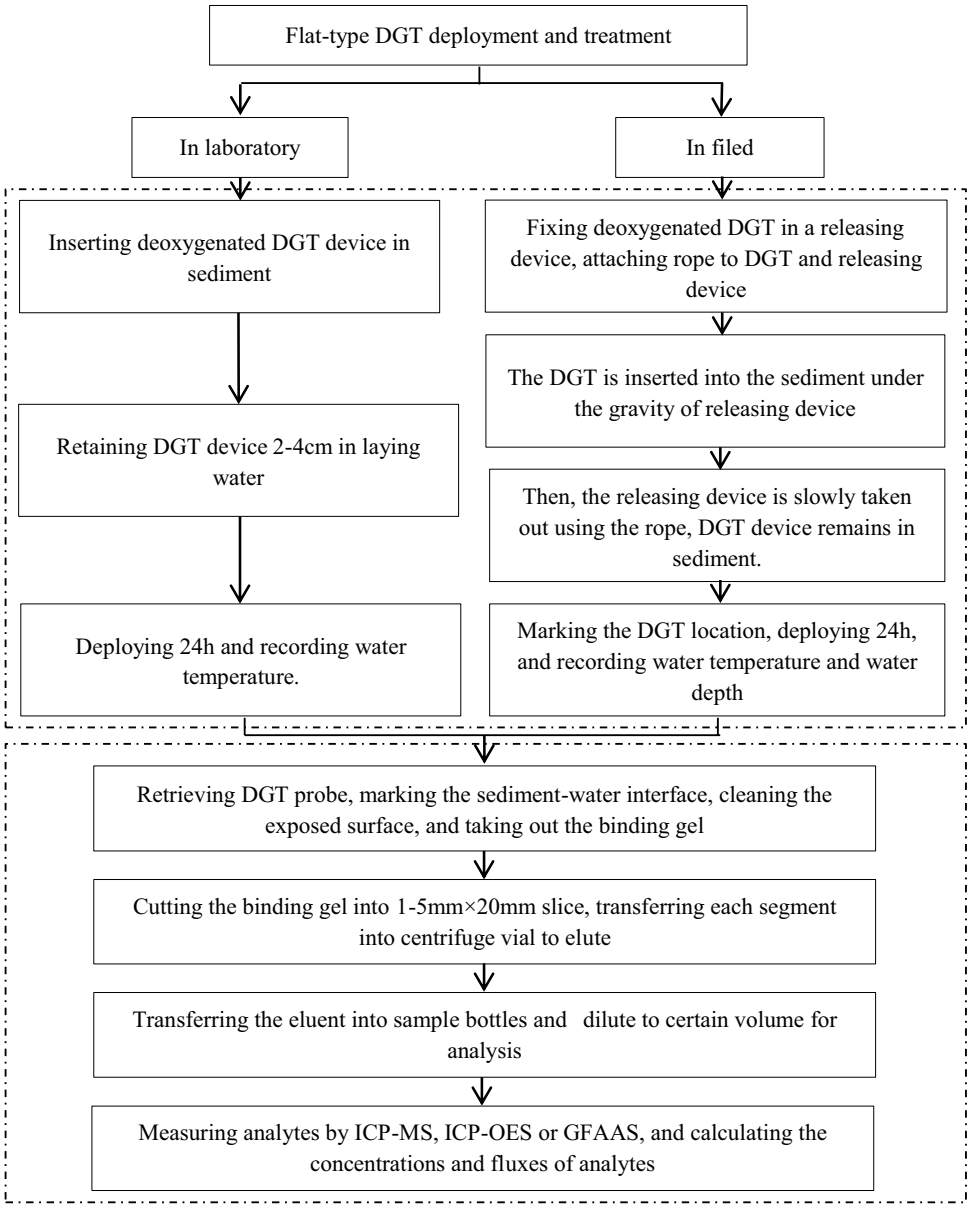

Fig. 5 Operational procedures for in situ deployment of the flat-type diffusive gradients in thin-film (DGT) device in sediment and treatment for analysis

water bodies (< 10 m depth of water). Ding et al. (2015) further developed a new method to identify the sediment–water interface after retrieval of the DGT probe. The back of the flat-type probe is attached by a sheet of sponge. There are numerous small holes on the surface of the sponge where the sediment particles will be squeezed in when inserting the probe, giving a clear mark of the sediment–water interface position on the sponge. Meanwhile, a sheet of plastic membrane is fixed on the top of the device prior to deployment. Once the probe is pulled out from the sediment, the plastic membrane will immediately cover both the front and back sides of the probe. This measure can preserve the sediment–water interface mark from water flow impact during pulling out the probe. The deployment of flat-type DGT and sample analysis is summarized in Fig. 5.

DGT deployment in soils

Dryland soil must be moisturized prior to DGT deployment, but the water content has not been standardized in the literature (Kalkhajeh et al. 2018; Luo et al. 2014). Some researchers adjusted the water content at a high level (over 90% of the maximum water holding capacity) (Guan et al. 2016; Gao et al. 2017; Kalkhajeh et al. 2018; Zhang et al. 2004). This may result in anoxic condition in soils and cause the bias of the measurement from the field. Other researchers adjusted the water content to around 70% of the maximum water holding capacity (Zhang et al. 2017a). This content approaches the level in the field dryland soils, while the deployment operation can be preceded smoothly under most cases. The water content can be increased step-by-step on

a 5% interval if it is difficult to operate the deployment for some types of soils containing high content of organic matter or calcite. Adjustment of the water content is traditionally made using a glass rod by hands. It is tedious if a large number of samples are treated. Another good method is that using an electric non-metallic stirrer to mix the soil sample after addition of deionized water (www.easysensor.net). This method can significantly save in labor, while the mixing efficiency can be greatly improved comparing to the use by hands. After adjustment of the water content, the soil sample is incubated at a certain temperature for 48 h for sufficiently moist prior to DGT deployment. For flooded soils, the DGT device can be directly deployed with the soils without adjustment of the water content. They can also be dried after sample collection and treated according to the procedure of dryland soil. It is of note, that air drying of flooded soil should be avoided prior to DGT deployment because that the properties of the soil may be changed (Zhang et al. 2017a).

The deployment procedure is different between using the piston-type and cavity-type (dual-mode) DGT device. For the use of piston-type DGT device, the device is deployed into wetted soil sample by hand pressing,a good contact should be ensured between the filter membrane and soil sample. For the use of cavity-type DGT device, the wetted soil sample is filled up to the open cavity of the device directly, and the device is gently shaken to ensure good contact between the filter membrane and soil sample. Then, DGT device is transferred to semi-closed zip lock bag prefilled with a small amount of deionized water for 24 h. The deployment of DGT and sample analysis in soil is summarized in Fig. 6.

DGT deployment in waters

The piston-type DGT device can be deployed in freshwater, seawater, and wastewaters for obtaining time-averaged concentrations of solutes, including organic chemicals/compounds, nutrients and multiple cations and anions (Guo et al. 2017a, b; Huang et al. 2016c; Wang et al. 2016; Xie et al. 2018a). The piston-type DGT device has been in situ deployed in water by several ways. The DGT devices can be attached to a buoy with fishing line (Pichette et al. 2007); multiple devices can be fixed into a polyhedral structure (the device exposure windows outward) (Guo et al. 2017a; Zheng et al. 2015); the devices can be fixed on plastic mesh strips or in the middle of two plexiglass plate (Lucas et al. 2014; Uher et al. 2017).

Many studies used a constant thickness diffusion layer to directly measure the time-averaged concentrations of solutes in water (Gimpel et al. 2003; Zheng et al. 2015). However, a thin solution layer is adjacent on the surface of DGT device to form a diffusive boundary layer when the device is deployed in water, which can affect the solute diffusive rate (Warnken et al. 2006; Zhang et al. 1998a). The diffusive boundary layer thickness (δ) in water can be calculated by simultaneously using various thicknesses of diffusive gel layer (Garmo et al. 2006; Scally et al. 2006; Zhang et al. 1998a):

$$\frac{1}{M} = \frac{\Delta g}{DC_{\mathrm{DGT}}At} + \frac{\delta}{DC_{\mathrm{DGT}}At} \tag{3}$$

where Δg is the thickness of diffusive layer; C_{DGT} is the solute concentration measured by DGT; A is the exposed surface area of the DGT device; M is the accumulated mass of analyte in the binding gel; D is the analyte diffusion coefficient in the diffusive layer; t is the deployment time.

Davison and Zhang (2012) have reviewed that the thickness of diffusive boundary layer is typically ~ 0.2 mm in well-stirred solutions, and varies from 0.26 mm in fast-flowing waters, 0.31 mm in lake epilimnion, to 0.39 mm in stagnant pond. Huang et al. (2016b) recently found that the thickness of diffusive boundary layer in some pond and wetland sites are approximately 100% of the standard DGT diffusive layer thickness ($\Delta g = 0.09$ cm). Therefore, a substantial error will be introduced in DGT concentration calculation if ignoring the diffusive boundary layer thickness.

Analysis of DGT sample

The DGT samples from the use of liquid binding phases can be determined directly without elution step. In comparison, the sample analysis from the use of solid binding phases generally requires an elution step. The elution agencies vary with the types of binding gels. The elution steps of common used binding gels are summarized in Table 6.

For sample analysis, the eluent containing target analytes can be determined by multiple analysis methods, such as inductively coupled plasma mass spectrometry (ICP-MS), inductively coupled plasma optical emission spectrophotometry (ICP-OES) or graphite flame atomic absorption spectroscopy (GFAAS). For the measurements in sediments and flooded soils, the 1D solute profile distributions with the use of flat-type DGT probes can be obtained by section-elution-analysis procedure. Several cutting tools have been applied for the section of the binding gel, including Teflon coated razor blade (Zhang et al. 2002), Plexiglass gel cutter (Gao et al. 2006) and multi-bladed ceramic cutter (Ding et al. 2015). Compared with other cutting tools, the use of multi-bladed ceramic cutter not only greatly increases the sample treatment efficiency, but also avoids the risk of metal contamination during the cutting. Furthermore, the spatial resolution of the cutting can be varied from 1.0 mm to 5.0 mm through adjusting the interval between the adjacent single ceramic blades.

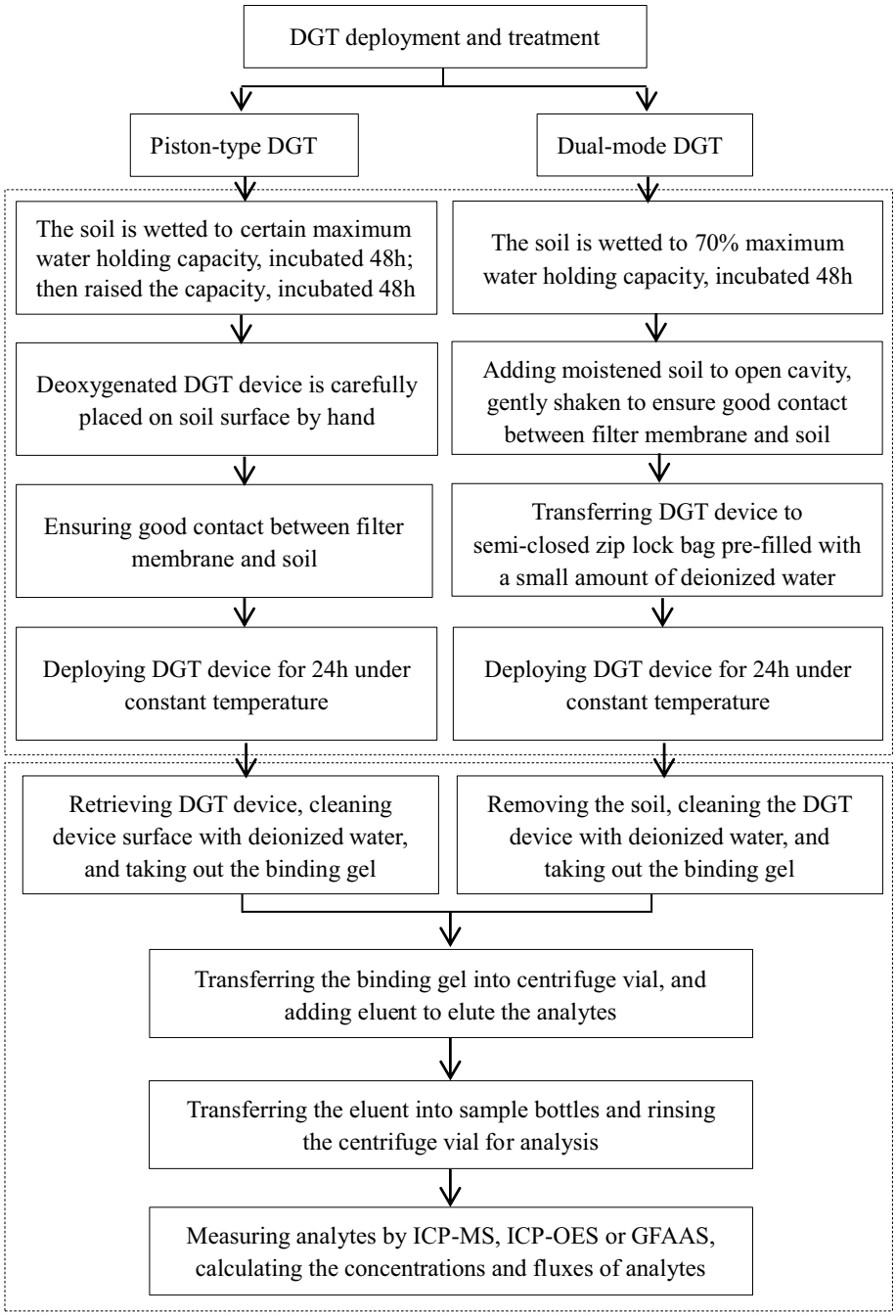

Fig. 6 Operational procedures for piston-type and dual-mode diffusive gradients in thin-film (DGT) deployment and treatment

In addition, some high-resolution techniques have been developed to obtain 2D solute distributions at sub-mm resolution, including 2D slicing-elution-microcolorimetric method for the determination of P (Ding et al. 2011, 2012), the staining technique coupled with computer imaging densitometry (CID) for the measurement of S(II), P and Cr(VI) (Ding et al. 2013; Teasdale et al. 1999; Yao et al. 2016), as well as laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) for the determination of cations and anions (Kreuzeder et al. 2013; Stockdale et al. 2010; Warnken et al. 2004b). The CID technique is established based on a relationship between the solute accumulation mass and the grayscale intensity, which simplifies the analysis procedures, improves the analysis efficiency and spatial resolution compared with traditional section-elution-analysis method. In comparison, the LA-ICP-MS requires a sophisticated instrument, complex analytical and expensive cost.

Table 6 Elution steps for some commonly used binding gels

Binding gel types	Elution steps	References
Chelex-100	1 M or 2 M HNO_3 for 24 h to extract any metals	Song et al. (2018a), Zhang and Davison (1995)
Ferrihydrite	1.4 M HNO_3 and 0.1 M HF or 1 M HNO_3 for 24 h to extract anions	Osterlund et al. (2010), Panther et al. (2013)
Metsorb	1 M NaOH or the mixture of 1 M NaOH and 1 M H_2O_2 for 24 h to extract anions	Bennett et al. (2010), Panther et al. (2013)
Zr-oxide	The mixture of 0.2 M NaOH and 0.5 M H_2O_2 for 3–5 h to extract anions at 4 °C	Ding et al. (2016c)
Chelex–Metsorb	First 1 M HNO_3 for 24 h to extract cations, then wash the binding gel surface by deionized water, followed by elution in 1 M NaOH for 24 h to extract anions	Panther et al. (2014)
Chelex–Ferrihydrite	3 M HNO_3 for 24 h to extract for any analytes	Huynh et al. (2012)
ZrO–Chelex	First 1 M HNO_3 for 16 h to extract cations, then add deionized water to rinse for 2 h, finally mixture of 0.2 M NaOH and 0.5 M H_2O_2 for 3–5 h to extract anions at 4 °C	Wang et al. (2017a)

Application of the diffusive gradients in thin films

The measurement of concentration, speciation and distribution of analytes in soil, sediment and water using DGT, has been widely developed, since DGT was initially developed for the measurement of Zn concentrations in environmental water (Davison and Zhang 1994). A large number of studies have demonstrated the superiority of DGT for the measurement of species distribution and bioavailability, concentrations of nutrients and organics, and multiple elements, compared with other traditional chemical extractions methods (Dai et al. 2018; Uher et al. 2017). Moreover, the research progresses in chemical fractionation, speciation and bioavailability of analytes using DGT measurement and analysis, have been discussed in detail in the literature (Davison 2016; Menegario et al. 2017; Zhang and Davison 2015). Here, we review the recent advances in the application of DGT to the analysis of soil, sediment and water environments.

Evaluation of bioavailability and toxicity

Currently, DGT techniques have been successfully applied for assessing the bioavailability and toxicity of solutes in soils and sediments. Zhang et al. (2014) provided an overview of the DGT assessment of P bioavailability in sediments and soils; therefore, this review focused on the recent development of DGT for assessing bioavailability and toxicity of metals. The bioavailability of As in different soil types were evaluated, comparing DGT and other traditional techniques (Dai et al. 2018; Zhang et al. 2017a), and the results indicated that the concentration of As measured by DGT exhibited a better correlation with the concentration absorbed by plants than those measured by other conventional extraction methods. Song et al. (2018a) demonstrated the potential application of DGT techniques for evaluating the bioavailability of heavy metals in sediments of land–freshwater interfaces, compared with the sequential extraction method. A significant positive correlation was observed between C_{DGT} and exchangeable and weak acid soluble fraction of Cd, as well as a significant negative correlation between C_{DGT} and residual Cd fraction, which confirmed the DGT technique is a suitable method to assess the bioavailability of metals.

Plant uptake mechanisms for metals were explored using DGT, with results suggesting that the uptake of Ni by (*Raphanus sativus*) was controlled by diffusion and plant limiting uptake mechanisms, while the uptake of Cd was mainly controlled by plant limiting uptake mechanisms (Luo et al. 2014). Peng et al. (2017) further used DGT to prove that the distribution and bioavailability of Se and plant uptake processes were associated with plant species, growth cycles and differences in soil fractions. Song et al. (2018b) predicted the level of metal bioaccumulation and bioavailability in river sediments using DGT, and the accuracy of results was verified by comparing the DGT-measured results, to the results of direct sediment analysis and plant sample analysis, respectively. The strong correlation in bioaccumulation concentrations of Cr, Cu, Zn, and Cd and the DGT measurements, suggests that DGT can be an effective tool for the assessment of metal bioaccumulation and bioavailability in plants. However, a weak correlation for As, as well as negative correlation observed between Ni and both measured concentrations by DGT and measured levels in plant tissues. This may because that the DGT measurement is limited to a thin layer (a few millimeters thickness) from the surface of the DGT device, while plant uptake covers a much larger volume of soils in the field (Tian et al. 2018; Williams et al. 2012). Therefore, the uptake and accumulation mechanisms of metals in plants need to be further explored in combination with other analysis techniques.

DGT has been proved to be a useful monitoring tool to reflect the accumulation extent of toxic elements in living organisms (Luider et al. 2004; Tusseau-Vuillemin et al. 2004). Gu et al. (2017) mimicked the biological responses of *Eisenia fetida* to Cd in soils using DGT. The results indicated that DGT measurement results can be more powerful for prediction of the accumulation of Cd in *Eisenia fetida*, as well as the induced adverse effects in comparison with total Cd concentration in soils. He et al. (2018) utilized DGT to predict trace metal bioavailability to chironomids (*C. tentans*) in sediments, and strong correlations were obtained between DGT-measured metal fluxes in the surficial sediment and metal bioaccumulation in *C. tentans*. Therefore, DGT-measured fluxes provide new surrogate to assess bioavailable fraction of toxic element accumulation in benthic invertebrates in sediments. In addition, Philipps et al. (2018) verified the performance of the DGT for predicting the Cu bioaccumulation in fish in waters containing low concentrations of natural organic matter.

Measurement of time-averaged concentrations

An important application of DGT is to measure DGT-labile concentrations of nutrients, metal(loid)s and organics in environment. Menegario et al. (2017) have summarized and discussed this application by focusing on metal(loid)s in waters. Therefore, this review paid attention to nutrients and organics. The high level of agreement has been observed between the NH_4–N concentrations obtained by DGT and those measured in bulk solution (C_{DGT}: $C_{SOL} = 0.922 \pm 0.070$–$1.030 \pm 0.060$) (Feng et al. 2015). Huang et al. (2016a, b) compared labile concentrations determined by DGT and grab sample for NH_4–N and NO_3–N concentrations in freshwater, respectively. Results showed C_{DGT}: $C_{SOL} = 0.83$–1.3 for NH_4–N, and C_{DGT}: $C_{SOL} = 1.03$–1.04 for NO_3–N. Huang et al. (2017) further measured dissolved inorganic nitrogen (PrCH resin for NH_4–N and A520E resin for NO_3–N) and phosphorus (Metsorb for PO_4–P) simultaneously in freshwater using a hybrid DGT. The ratio C_{DGT}/C_{SOL} was 0.83–1.3 for NH_4–N, NO_3–N and PO_4–P, which demonstrated a high level of measurement accuracy for the new DGT. In addition, Cai et al. (2017) have applied DGT techniques to the determination of NO_3–N concentrations in soil, showing excellent agreement with pore water concentrations. Moreover, the heterogeneity of NO_3–N distribution in soil can be clearly observed from 2D distribution plots.

The toxicity and potential risk of organics are of wide global concern; therefore numerous studies have applied DGT techniques to the in situ measurement of organics in various environments (Cizmas et al. 2015). Chen et al. (2013, 2014a, 2015a, b) used XAD-18 as a DGT sorbent for the in situ sampling of multiple antibiotics in water and soil, combined with the DIFS model to investigate the supply kinetics of antibiotics from soil solid phase into solution. Estrogenic activity was measured by XAD-18 DGT and conventional grab sampling, with good agreement between conventional sampling methods and DGT sampling results (Guo et al. 2017b). However, the DGT method provided the additional benefits of being in situ technique and more efficient in terms of cost, time, and labor. Moreover, XAD-18 DGT has been employed to measure the concentration of illicit drugs in wastewater and rivers, showing no significant difference in detected concentrations between DGT and active sampling methods; although the concentrations obtained by DGT were slightly lower than with active sampling (Guo et al. 2017b; Zhang et al. 2018b). Xie et al. (2018a) found that antibiotics concentrations in seawater were significantly different using XAD-1 DGT and active sampling. This difference may be due to the fact that active sampling missed the effects of discharge, tide and dilution. The ratio of EDCs concentrations obtained by XDA-1 DGT and direct sampling in seawater, were all between 0.8 and 1.2, which demonstrates that XDA-1 DGT is suitable for in situ measurement of EDCs in seawater (Xie et al. 2018b). In addition, the concentration of household and personal care product (HPCP) compounds in water, were measured using DGT, in comparison with auto and grab sampling methods (Chen et al. 2017b). Results suggested that concentrations of most HPCPs obtained by DGT are similar to those measured by auto sampling; however, they were not always consistent with the results of grab sampling, due to the grab sampling method ignoring some peak, point source, rain or discharge events.

Dong et al. (2014) used a novel molecularly imprinted polymer (MIP) as the binding agent for DGT sampling of 4-chlorophenol (4-CP) in water, with performances of MIP DGT tested. The concentration of free form 4-chlorophenol obtained by MIP DGT decreased with the increasing concentration of dissolved organic carbon due to electrostatic interaction between 4-chlorophenol dissolved organic carbons. The concentrations of bisphenols (BPs) bisphenol A (BPA), bisphenol B (BPB) and bisphenol F (BPF) were measured in water and soils (Guan et al. 2017; Zheng et al. 2015). After 7 days of deployment, the BPs concentrations in water obtained by DGT were in good agreement with active sampling results. Results also showed almost all free 4-chlorophenol was sampled by DGT with a C_{DGT}: C_{SOL} ratio of around 1.01 ± 0.05 in synthetic solutions, while only 39.1% of the total concentration of 4-chlorophenol was collected by DGT in intermediate untreated industrial wastewater, due to the high concentrations of iron oxyhydroxide colloids and organic matter in the wastewater. It is worth noting that the toxicity of organometallics tended to be higher than inorganic metals, therefore accurate measurement of organometallics is critical. Fernandez-Gomez et al. (2015) measured and predicted the concentration and

bioavailability of MeHg in water using thiol-functionalized resin gel DGT, confirming the DOM can enhance the MeHg photodegradation process. DGT based on silica-bound sorbents was successfully employed for the measurement of five organotin compounds in sediments (Cole et al. 2018). The obtained 1D profile distributions demonstrated spatial distribution heterogeneity for five organotin compounds, with DGT fluxes ranging between 4.3×10^{-8} and 1.6×10^{-5} ng cm^2 s^{-1} in sediments.

Investigation of coupled relationship between different elements

DGT simultaneously determinates the concentrations of multiple metals and oxyanions and has been recently used to further understand the potentially coupled relationships between different elements. Wu et al. (2014) used Chelex-100 DGT to measure 14 metals (Fe, Mn, Cd, Co, Cu, Ni, Pb, Zn, Al, As, Sr, V, Cr and Sb) at the sediment–water interface (SWI). The coupled relationships between metals were observed, such as the similarities in concentration distributions between Cd, Co, V, Sb, Cu, Pb, As, Sr, Cr, Al and Fe, as well as Ni, As, Sr, Zn and Mn. Wang et al. (2017a) simultaneously measured eight cations (Fe, Mn, Co, Ni, Cu, Zn, Cd and Pb) and eight oxyanions (P, V, Cr, As, Se, Mo, Sb and W) in sediments using ZrO–Chelex DGT. Similar distributions were observed between six elemental groups: Fe, Mn, P, and As (1), Co and Ni (2), Zn and Pb (3), V and Cr (4), Se and Mo (5), and Sb and W (6). The coupled relationship and co-remobilization observed between trace metals and Fe/Mn was verified using DGT, where the redox cycle of Fe/Mn played an important role in the distribution and speciation of some trace elements (Parker et al. 2017). Naylor et al. (2004, 2006) simultaneously measured sulfide with Fe, Cu, Mn, Ni and Zn concentrations using DGT–CID techniques in marine sediments, while the simultaneous release of sulfide with Fe, Mn, Ni and Zn has been observed from distribution imaging. The coupled relationship between P and Fe(II) was observed at a millimeter scale by ZrO–Chelex DGT, with the mechanism identified for Fe-redox controlled P release from Fe-bound P in sediments (Ding et al. 2016a; Wu and Wang 2017; Xu et al. 2013). The phenomenon of simultaneous release of DRP and dissolved sulfide from the SWI was initially established by combining ZrO–AgI DGT and CID technique (Ding et al. 2012). Furthermore, the 2D distribution of DRP and dissolved sulfide, as well as the corresponding Fe(II) distribution, provided high-resolution information on the mechanism of microbial iron reduction (MIR) dominated sulfate reduction and Fe-coupled P mobilization, using ZrO–Chelex DGT, and AgI DGT, combined with CID analysis techniques (Ma et al. 2017).

Combined use of DGT with DIFS

DIFS models were applied to describe the resupply capacity from solid phase to solution phase, when analytes were transferred to or consumed by DGT. The principles of DIFS and calculation methods have been well established, and DIFS has been widely applied in soil and sediment analysis, to investigate resupply processes (Harper et al. 1998, 2000; Sochaczewski et al. 2007). Zhang et al. (2017a) compared the As availability and release capacity from solids to soil solutions by DGT–DIFS in different soil types, showing greater availability of As in flooded soils, with higher values of R, K_d, k_1, k_{-1} and lower T_C values than dry soils. The resupply capacities of three BPs (BPA, BPB and BPF) from soil solids to solutions were evaluated, with K_d values obtained by DIFS indicating that most BPs in soils could participate in labile exchange (Guan et al. 2017). Gao et al. (2018) predicted the Ni mobilization and release capacity from solid phases in riparian reservoir soil by DGT–DIFS. The small R (<0.25) and obviously high K_b suggested a slow rate of resupply of Ni from the solid phase. The lower Ni resupply capacity may be caused by the adsorption of Fe/Mn oxides and organic matter, according to the positive correlations between Ni concentrations and Fe, Mn and TOC concentrations. Xu et al. (2018) combined DGT and DIFS model to evaluate the dynamic release mechanism of Zn in the riparian soils. The results showed that labile Zn can irreversibly and rapidly release into soil solution from solid phase, indicating the release and pollution risk of Zn in riparian reservoir soils.

In addition, DIFS models can be applied to the prediction of resupply capacities from sediment solids to porewater. Chen et al. (2018a) studied the P resupply kinetics from sediment solids to porewater using DIFS, after dredging for 6 years. Simulation results indicated that the P release capacity decreased after sediment dredging in April, due to increased R, K_d and k_1 values. Moreover, DIFS was used to evaluate the resupply capacities of P in sediments after aluminum sulfate (ALS) and lanthanum modified bentonite amendment, respectively (Lin et al. 2017a; Wang et al. 2017b). Both the increase in K_d and k_1, as well as the decrease in T_C, suggested that release of P from sediment solids was difficult after amendment. Lin et al. (2017b) combined with DGT and DIFS to demonstrate that ALS could also effectively immobilize As in sediments. K_d and k_1 consistently increased, indicating the As release capacity from sediment solids was reduced by ALS amendment. Moreover, the transfer, uptake and bioavailability of P, Cu, Cd, Pb, and Zn were assessed in the macrophyte rhizosphere in sediments using DGT and DIFS (Wang et al. 2018; Wu et al. 2018). It verified that the diffusion and resupply characteristics of metals at the rhizosphere interface were controlled by multiple factors, including the solid phase resupply capacity

and the labile pool size in the sediments. Furthermore, the observed depletion of labile P pool size in rhizosphere sediments indicated the essential role of macrophyte in controlling the release of P from sediments.

Combined use of DGT with DET

The combination of DGT and other in situ passive sampling techniques has become a recent focus in DGT research and development. The diffusive equilibrium in thin films (DET), contained only the diffusive gel and the membrane, with a similar device setup to DGT, relying on the diffusion equilibrium established between DET gel and pore water (Davison et al. 1991; Ding et al. 2010a; Harper et al. 1997). DET measured the total dissolved species concentration, while DGT obtained information on labile species concentration; therefore, the concentrations acquired from DET were often higher than DGT (Gao et al. 2006, 2007).

The reported application of combined DGT and DET systems, have involved insertion of both devices in sediments in a back-to-back manner (Gao et al. 2012; Gregusova and Docekal 2013; Lesven et al. 2008). The 1D distribution profiles of Fe, Mn, Co, Cd, Cu and Zn in sediment have also been obtained using back-to-back DGT and DET probes (Yang et al. 2012). The concentration distributions of Fe, Mn and Co were in good agreement for both DGT and DET measurement. In addition, the ratio *R*: DGT/DET or DGT/centrifugation was applied to assess the supply capacity from the solid phase into solution, showing that Mn and Fe were more readily released from the solid phase. Various coupling relationships have been observed between Fe and As, Co, Ni and Mn, using the DGT and DET technique (Pradit et al. 2013). The maximum concentration of Cu, Zn, Cd and Pb detected at the SWI, was probably the result of organic matter degradation and similar results were also observed by Yang et al. (2012). The As concentration in soil porewater was measured using direct sampling, DGT and DET (Garnier et al. 2015). Results suggested that profile distributions obtained by all three sampling methods were similar in magnitude and pattern of concentrations. In addition, Menezes-Blackburn et al. (2016) established the P mobility and resupply capacity in soil, using DGT and DET combined with the DIFS model, showing that DGT induced inorganic P flux, occurred mainly from soil solids resupply.

In addition, DGT and DET have also been combined into a single device. Briefly, the DET–DGT probe consisted of three layers: the membrane filter, the DET gel as a diffusive gel and a DGT binding gel (Robertson et al. 2008, 2009). The relationship between Fe(II) and S(II), P, as well as the high-resolution (sub-mm) 2D imaging of the distributions of these species in seagrass sediments, were obtained using the hybrid probes (Pages et al. 2011). The heterogeneity distributions of Fe(II) and S(II), P, as well as the coupled relationships between Fe(II) and P, as well as Fe(II) and S(II) were observed from 2D distribution images. Furthermore, Cesbron et al. (2014) developed a hyperspectral imaging method combined with a DET–DGT probe for the measurement of S(II), Fe(II) and DRP, at a sub-mm resolution in sediments, with iron and sulfide interactions in deeper sediment profiles. Moreover, the co-distribution of dissolved Fe(II) and DRP was obtained, providing evidence for Fe-coupled mobilization of P, with Pages et al. (2014) further confirming the results using a DET–DGT probe.

Pages et al. (2012) used a DET–DGT probe to measure S(II) and Fe(II) at high-resolution (sub-mm) and P and NH_4^+ at low-resolution (mm), in the rhizosphere of *Zostera capricorni* under both light and dark conditions. Under light conditions, low S(II) and Fe(II) concentrations were observed around the rhizosphere; in contrast, high Fe(II) was observed in the near-surface sediment and high S(II) concentrations in the deep sediment. In addition, the effects of photosynthetic activity and radial oxygen loss (ROL) on the regulation of plant uptake of P and NH_4^+ were demonstrated. Arsic et al. (2018) compared the speciation and distributions of Fe, As and Sb under oxic and anoxic conditions using a DET–DGT probe. Results suggested that Fe(II), As(III) and As(V) were more mobile under anoxic conditions, while Sb(III) and Sb(V) were released into water from sediments under oxic conditions. In addition, a decoupling relationship was observed between the Fe(II) cycle and Sb mobilization, which indicated that other processes may play a more important role in Sb mobilization, such as sulfur and/or organic carbon mechanisms.

Combined use of DGT with dialysis sampler (Peeper)

Dialysis sampler (Peeper) systems are an in situ passive sampling technique, that rely on a diffusive equilibrium between porewater and the Peeper sampler, which is similar to DET in principle (Azcue et al. 1996). Peeper probe measures the concentration of total dissolved species; however, the significant difference with DET was the sampling medium for Peeper, which was a solution instead of a hydrogel. The Peeper sampler incorporates two fractions: with a chamber containing equilibrium solution and a dialysis membrane covered chamber. A high-resolution Peeper (HR-Peeper) sampler commonly has a total length of 15 cm with 75 chambers on a base plate (18 mm (length) × 1 mm (width) × 1 mm (height)) and a volume in each chamber of about 15 μL, with each pair of two adjacent chambers separated vertically by a 1-mm-thick wall, allowing a vertical resolution of 2 mm. Xu et al. (2012b) used a Peeper device to collect pore water samples for the simultaneous analysis of DRP and soluble Fe. The HR-Peeper was further developed to enable measurement of more analytes, enlarging the chamber column and decreasing the vertical resolution to 4 or 5 mm (Fig. 7)

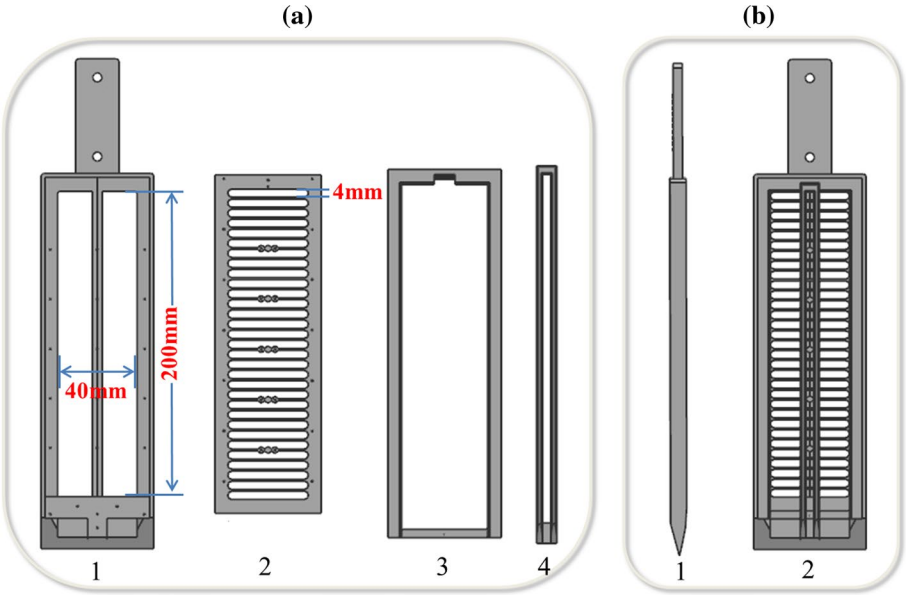

Fig. 7 Schematic representation of the high-resolution (HR) Peeper device, **a** 1, the flat base to support the Peeper champers (2); 3 and 4 the window and fixed strip to fix the Peeper champers and dialysis membrane; **b** 1 and 2, the side and front of HR-Peeper device

(Ding et al. 2018b). Moreover, the combination of DGT and HR-Peeper devices provides insight into biogeochemical processes for P (Ding et al. 2018a; Lin et al. 2017a).

Chen et al. (2015c, 2016a, b) investigated the effects of different levels of bioturbation on P release and kinetics in sediments, with the relationship between Fe(II) and P mobilization investigated using HR-Peeper and DGT at a millimeter resolution. Results suggest that macrozoobenthos bioturbation resulted in a decrease in the concentrations of soluble/labile P in sediments, with labile P concentrations being much lower around the worm burrow. In contrast, bivalve bioturbation increased the concentrations of soluble/labile P. These different results may be caused by different biological types. Macrozoobenthos induced an increase in DO in worm borrows, causing Fe(II) to be oxidized to Fe(III) oxyhydroxides, which increased the adsorption for P and further inhibited P release. However, bivalve bioturbation shortened the O_2 penetration depth and caused the depletion of O_2 in sediments, resulting in the reduction of Fe(III) oxyhydroxides and an increase in soluble/labile Fe(II), as well as P release from sediment. The effects of elevated P in water on the release of heavy metals from sediments were investigated in a microcosm experiment, using DGT and HR-Peeper (Chen et al. 2017a). Results showed that DGT-labile metal concentrations decreased from the 10th to 20th days and dissolved metal concentrations decreased on the 20th day of incubation with elevated phosphorus levels in the water, due to the metal-P precipitate; on the 40th day, the degree of decrease in DGT-labile metals was reduced. In addition, Sun et al. (2017) combined the DGT and HR-Peeper technique, with the DIFS model to investigate the competitive effect of P on As mobility in sediments. Results indicated that soluble/labile P and As increased, while the soluble/labile Fe(II) fraction decreased with rising P content in water, due to competitive effects of P and As. DIFS simulation results showed an increased As resupply capacity from the solid phase to pore water, with elevated P concentrations in water.

Using DGT and Peeper systems, the mechanisms of P release during algal blooms were investigated on an hourly scale (Chen et al. 2018b). Both the occurrence of algal blooms and their decomposition were found to increase the concentration of SRP in porewater. During algal blooms, increasing soluble Fe(II) concentrations were the main reason for the release of SRP, suggesting Fe-redox cycling controlled P mobilization. Furthermore, Ding et al. (2018b) verified the same Fe–P coupling mechanism in sediments, providing further insights into the contribution of internal P loading to N limitation. During the pre-bloom and bloom period, TP concentrations increased and significantly negatively correlated with sediment SRP flux and water column TN/TP concentrations, which demonstrates the contribution of internal P to N limitation. Liu et al. (2018) studied the effects of temperature on the release of P using DGT and Peeper, which simultaneously determined labile P/Fe and soluble P/Fe concentrations at different temperatures. Results showed that concentrations of DGT-labile P and SRP increased with rising temperatures, which may be caused by the increase in the microbial reduction of Fe(III) oxyhydroxides and algal degradation. Xing et al. (2018) utilized DGT to measure DGT-labile P and HR-Peeper techniques to measure soluble Fe(II), SRP in the rhizosphere. Exploring further plant uptake mechanisms for P in sediments, it was established that root-mediated radial O_2 loss processes caused the formation of Fe plaques on the root surface, with Fe plaques absorbing and accumulating P from

sediments and porewater adjacent to the roots. Finally, the root excreted oxalic acid and other LMWOAs, which were found to increase the release of P in Fe plaques and absorption by the root. Similar mechanisms of plant-mediated P uptake have been obtained by combining DGT and PO techniques (Brodersen et al. 2017).

Combined use of DGT with planar optode

Some changes in physicochemical parameters, such as pH and O_2, are associated with changes in metal species dynamics and distributions. Nevertheless, sophisticated experimental setups and complex data analysis processes have provided only a limited understanding of these processes. Planar optode (PO) is a robust 2D imaging technique that has been widely applied for the measurement of pH, O_2, pCO_2 and temperature in sediments and soils (Moßhammer et al. 2016; Santner et al. 2015). Both DGT and planar optode are in situ measurement techniques that can provide high-resolution information on analyte fluxes and distributions (Davison and Zhang 1994; Glud et al. 1996). Hoefer et al. (2015) studied the effects of sulfur (S^0) application on metal solubility in the rhizosphere of *Salix smithiana*, with the 2D distribution of Mn, Zn and Cd fluxes obtained by DGT–LA-ICP-MS and the O_2 2D distribution by planar optode imaging. Results indicated that S^0 oxidation caused depletion of O_2 and local acidification along single roots, inducing an increase in Mn, Zn and Cd solubility at the root–soil interface. To investigate seagrass-derived phosphorus and Fe(II) solubilization, Brodersen et al. (2017) utilized DGT–LA-ICP-MS for assessment of the 2D distribution of Fe(II) and Ca^{2+}, DGT–CID for 2D distribution of sulfide and phosphate and planar optode for 2D distribution of O_2 and pH, demonstrating the relationship between nutrient mobilization and radial O_2 loss. Kreuzeder et al. (2018) combined the DGT and planar optode techniques to study the biogeochemistry of P and metals in the rhizosphere of three plants (wheat, buckwheat and white lupine). The results showed that the root apices were more active for P solubilization, and plants could response to P deficiency by inducing acidification or alkalization in the rhizosphere.

Furthermore, DGT–PO hybrid sensor has been developed for simultaneous 2D imaging of O_2/pH and trace metal species dynamics at a sub-mm resolution. Stahl et al. (2012) used a novel DGT–PO hybrid sensor to simultaneously obtain 2D imaging distributions of O_2 dynamics and trace metal fluxes. As shown by the 2D imaging distribution of O_2 and corresponding metals, irrigated burrows induced enhanced O_2 levels, resulting in highly localized mobilization of Ni, Cu and Pb, as well as a decrease in dissolved Fe(II) around the burrow system. Lehto et al. (2017) used a DGT–PO sensor to investigate the influence of deposition of particulate organic matter (POM) on O_2 dynamics and metals mobilization at SWI. Results indicated that the deposition of POM resulted in the depletion of O_2 concentrations, with anoxic conditions at the SWI also increasing mobilization of Fe and Mn, while decreasing Zn, Ni and Co mobilization. The combined single-layer gel consisted of the DGT with PO applied for analysis of rhizosphere biogeochemical processes (Hoefer et al. 2017; Williams et al. 2014). Results showed that ROL-derived acidification of the local rhizosphere environment, resulted in elevated mobilization of trace metals, such as Al, Co, Cu, Fe, Mn, Ni and Pb. In addition, 2D imaging of O_2/pH and trace metals showed that Fe-redox cycles regulate the plant uptake of trace metals.

In summary, the combination of DGT with other high-resolution, imaging techniques allows the simultaneous measurement of multiple elements, enhancing our understanding of the relationship between labile species and dissolved species. This has greatly broadened the application scope of DGT and allows its function in revealing the biogeochemical processes for various solutes. In terms of analysis of the heterogeneous nature of sediments and flooded soils, the combined method looks very promising for future application.

Advantages and challenges

Advantages of DGT development

Over the last two decades, DGT has developed into an important measurement method, because of its notable advantages: (1) DGT can perform in situ measurements, with accurate interpretation of the measurement results possible. Furthermore, DGT measurements do not ignore certain events, such as discharge, rain and tide episodes, and are independent of soil properties and water flow conditions; (2) DGT can measure a great number of labile metal species, oxyanions, organics and other solutes, even at low concentrations due to pre-concentration capacity; (3) DGT can be applied to the measurement and evaluation of concentrations, distributions, speciation and bioavailability of analytes, allowing interpretation and prediction of resupply mechanisms from sediment and soil solid phase to solution phase, in combination with DIFS; (4) DGT can provide high-resolution (mm and sub-mm) information in combination with other in situ measurement techniques, such as DET, Peeper and PO.

DGT development challenges

The DGT technique has been continually developed for more than 20 years, and its effective application in sediments, soils and water have been well established. However, significant challenges remain for the future development of DGT.

1. Overcoming environmental interference to DGT measurement. In addition to biofilms which could affect analyte diffusion, electrostatic interactions and ion competition can also interfere with solute diffusion in the diffusive layer, affecting the adsorption performance of the binding gel. Besides, the adsorption of organics by the filter membranes could affect the DGT measurement of organics. There are currently no effective methods to resolve these problems.
2. Expanding the capability of DGT measurement. With development of the mixed binding layer, hybrid DGT devices have provided new insights into the complex relationships between trace metals and oxyanions in various environments. The measurement of multiple targets using a mixed binding layer is required, such as for the simultaneous measurement of heavy metals and organics, or nutrient. Furthermore, in order to study the biogeochemical processes for a specific target under heavily polluted conditions, highly selective binding agents need to be further developed. The currently available DGT binding agents are too large (mm–μm size) to match the combined use of other 2D imaging techniques, such as PO. Smaller binding agents in the nano-size range should be developed and applied in the future.
3. Unify the DGT operational procedure. To date, many researchers have applied the DGT technique to the measurement of various solutes in sediment, soil and water using different operational procedures. The use of varying operational procedures results in a greater potential for error, as well as reducing the comparability among measurement results. Therefore, the unification of operational procedures for DGT device assembly, deployment and sample treatment is required. Standardized systems, such as types of diffusion gel and filter membrane would significantly facilitate comparison among different studies. Furthermore, water content should be optimized to a similar level for the measurement of dryland soils.
4. Improvement of the interpretation of DGT measurement results. DGT has been demonstrated to be a useful tool to indicate the lability and bioavailability of nutrients and metals in soils and sediments. It has the potential to be applied in environmental quality and ecological risk assessments. However, the uptake of solutes by organisms are not only controlled by diffusive mechanism in soils or sediments, but also influenced by organism types and other factors. It is still far away from establishing environmental criteria based on DGT measurement. More research should be performed in this aspect. Furthermore, DIFS modeling has been successfully employed for the evaluation and prediction of the resupply capacity from soil or sediment solid phase. Further development of the interpretation of liquid–solid interactions is required, in combination with other models.

Conclusion

A great number of studies have demonstrated that the DGT technique is not only a versatile in situ passive sampling tool for the measurement of metals and oxyanions, but also a potential tool for the determination of inorganic nutrients and organics. In this paper, the principles of DGT technique and research progress are described, with the performances of tradition DGT devices and improved DGT devices reviewed, as well as commonly used binding and diffusion materials discussed in detail. The novel applications and development of DGT techniques in recent years are discussed, as well as the potential capabilities of DGT, that have yet to be fully exploited. Overall, more research attention should be given to the investigation of biogeochemical processes by DGT.

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