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Sodium polyacrylate as a binding agent in diffusive gradients in thin-films technique for the measurement of Cu^{2+} and Cd^{2+} in waters

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ABSTRACT

An aqueous solution containing sodium polyacrylate (PA, 0.0030 M) was used in diffusive gradients in thin-films technique (DGT) to measure DGT-labile Cu^{2+} and Cd^{2+} concentrations. The DGT devices (PA DGT) were validated in four types of solutions, including synthetic river waters containing metal ions with or without complexing EDTA, natural river water (Hun River, Shenyang, China) spiked with Cu^{2+} and Cd^{2+}, and an industrial wastewater (Shenyang, China). Results showed that only free metal ions were measured by PA DGT, recovery = 98.79% for Cu^{2+} and recovery = 97.80% for Cd^{2+} in solutions containing only free metal ions, recovery = 51.02% for Cu^{2+} and recovery = 51.92% for Cd^{2+} in solution with metal/EDTA molar ratio of 2:1 and recovery = 0 in solutions with metal/EDTA molar ratio of 1:1 and 1:2. These indicated that the complexes of Cu–EDTA and Cd–EDTA were DGT-inert or not DGT-labile. The DGT performance in spiked river water (recovery = 8.47% for Cu^{2+} and recovery = 27.48% for Cd^{2+}) and in industrial wastewater (recovery = 14.16% for Cd^{2+}) were also investigated. Conditional stability constants ($\log K$) of PA–Cu and PA–Cd complexes were determined as 6.98 and 5.61, respectively, indicating strong interaction between PA and the metals.

1. Introduction

Diffusive gradients in thin-films (DGT) technique has widely been used as an *in situ* sampler for trace metals in waters, sediments and soil since its invention in the mid-1990s [1–5]. Traditional DGT devices comprise a polyacrylamide hydrogel (PAM) as a diffusion layer and a chelex-100 resin as a binding agent. Since then, varying binding agents have been developed to measure various analytes [6] and analyte species [7], e.g. Thiol-Speron resin for mercury [8], diethylaminoethyl-substituted cellulose polymer membrane for uranium [9] and ferrihydrite for phosphorus [10].

DGT-labile concentrations measured by DGT devices with these different binding agents [11] may provide information on bioavailability of metals in varying biological environments [12–14] which are determined by the interactions between the analytes and these binding agents with different binding strengths. This is similar to the sequential extraction methods which can measure the bioavailability of metals for the plants with different extractants, such as 0.1 M $NaNO_3$, 0.01 M $CaCl_2$ and 1 M NH_4Ac [15].

Besides the solid form binding agents mentioned above, a poly(4-styrenesulfonate) (PSS) aqueous solution coupled with a dialysis membrane was also used in DGT devices. The advantages of this solution binding phase DGT include, better reproducibility with the excellent contact and mobile characteristics and better accuracy and precision with the direct measurements of metal concentrations in the polymer solutions without elution steps [11,16,17].

This work studied the possibility of water soluble polyacrylate (PA) with carboxylic groups ($-COO^-$) as an aqueous solution binding phase in DGT devices (PA DGT) for the measurement of DGT-labile Cu^{2+} and Cd^{2+} in waters. The binding properties of PA DGT for Cu^{2+} and Cd^{2+} were investigated under conditions of pH and ionic strength. PA DGT was evaluated under laboratory and natural river water conditions.

2. Experimental

Unless otherwise stated all laboratory measurements were performed by triplicate. Stirring of all the laboratory solutions was achieved by recirculating the solution using an aquarium pump at a rate of $15\,L\,min^{-1}$ [18]. All the tests were performed at room temperature (25 °C).

2.1. Preparation of the dialysis membrane and purification of polyacrylate and poly(4-styrenesulfonate)

Cellulose acetate dialysis membranes (Sigma, M_w 12,000 or greater retained) were pretreated following the procedures rec-

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ommended by Li et al. [16] to remove attached chemicals during production.

Sodium polyacrylate (PA, average $M_w \sim 2 \times 10^7$) from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China, was purified following the procedures described by Li et al. [16]. A 20 g portion of PA was dissolved in 200 mL of deionized water. The solution was then transferred to a dialysis membrane bag prepared as described above. The bag was immersed in deionized water for 120 h with the water frequently replenished. This process effectively removed all of the low molecular weight PA that passed through the dialysis membrane. The dialyzed PA was then filtered with a 0.45 μm pore size cellulose nitrate filter membrane (Dikma, Tianjin, China) to remove any undissolved particles. After purification, the concentration of PA was determined gravimetrically. A PA stock solution of 0.0050 M (concentration of carboxylic groups) was prepared for use in the experiments.

2.2. Measurement of diffusion coefficients of Cu^{2+} and Cd^{2+} in the dialysis membrane

Before DGT measurements, the diffusion coefficients, D_m, of Cu^{2+} and Cd^{2+} through the dialysis membrane in synthetic river water and in synthetic industrial wastewater were determined according to the procedures recommended by Li et al. [16]. A diaphragm diffusion cell comprising two compartments was used, each with an interconnecting 2.0 cm diameter opening. A 3 cm diameter and 85 μm thickness disc of freshly pretreated dialysis membrane was placed on the openings between the compartments, ensuring that the membrane was the only mass transport medium. Both compartments were filled with 75.0 mL matrix solution of the same concentrations. One compartment (A, source) initially contained 10.0 mg L^{-1} of Cu^{2+} or Cd^{2+} and the other compartment (B, receiving) contained 0.0030 M PA. PA in compartment B is aimed to bind the diffused metal ions and reduce their concentrations. Both compartments were stirred continuously during the experiment using an overhead stirrer at a constant speed. Samples were taken from both compartments at 0.5 h intervals up to 3 h and measured by FAAS. The diffusion coefficients (D_m) were calculated using equation [16]:

$$D_m = \frac{M\Delta g}{ACt} \tag{1}$$

where M is the mass transported from compartment A to compartment B, Δg the thickness of the membrane, A the area of the openings on the cells, C the concentration of metal ions in compartment A and t the diffusion time. Average concentrations in compartment A at the sampling intervals were used to correct concentration changes in the bulk solution.

2.3. Binding of Cu^{2+} and Cd^{2+} to PA

To assess the binding properties of PA for Cu^{2+} and Cd^{2+}, DGT devices designed to hold PA according to Li et al. [16] were used. Concentration of PA in the device was optimized to achieve maximum binding by examining binding capacities of the devices with varying PA concentrations of 0.0010, 0.0020, 0.0030, 0.0040 and 0.0050 M in a well-stirred solution containing 5.0 mg L^{-1} Cd^{2+} for 24 h.

The DGT devices with optimized PA concentration were then placed in 0.10, 0.20, 0.50, 1.0, 2.0, 5.0, 10.0 and 20.0 mg L^{-1} Cu^{2+} or Cd^{2+} solutions for 72 h to examine the binding capacity of PA DGT. The effects of varying pH (2–11 in 10 increments) and $NaNO_3$ concentration (0.001, 0.01, 0.1 and 0.7 M) on the binding capacities of Cu^{2+} and Cd^{2+} to PA were studied by immersing the DGT devices in 5 mg L^{-1} Cd^{2+} or Cu^{2+} solutions for 24 h. Solution pH was adjusted using 2% HCl or NaOH (analytical grade, Sinopharm Chemical Reagent Co., Ltd.). Solution ionic strength was adjusted with appropriate addition of $NaNO_3$ at pH ~ 7.

The conditional stability constants of the complexation were also estimated by immersing the DGT devices in solutions containing 0.20, 0.40, 0.60, 0.80 and 1.0 mg L^{-1} Cu^{2+} or Cd^{2+} for 240 h (extra time for the system to reach equilibrium). The metal concentrations in the PA solutions and remaining in the solutions outside the devices were measured [16].

2.4. Validation of PA DGT

Well-stirred synthetic river water solutions were used to test the performance of this DGT set-up. The solutions were prepared using salts, 0.4696 g KCl, 4.080 g $NaNO_3$, 4.328 g $CaCl_2$, and 2.492 g $MgSO_4$ corresponding to the cation concentrations of river water, in 30 L solution. The solutions spiked with 0.1 mg L^{-1} of Cu^{2+} or Cd^{2+} were allowed to equilibrate overnight before the DGT devices were deployed in the solution for time intervals of 24–96 h. Large volume of solution (30 L) was used to ensure that the depletion of the metals in the solution was negligible. All salts used were of analytical grade and were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China).

2.5. Measurement of DGT-labile metals by PA DGT

To measure DGT-labile metals by PA DGT, DGT devices were deployed at time intervals from 24 to 96 h in three 50 L solutions, including a synthetic river water containing 0.1 mg L^{-1} Cu^{2+} and Cd^{2+} ion with spiked ligand of EDTA (analytical grade, Sinopharm Chemical Reagent Co., Ltd.) at molar ratios of 1:2, 1:1 and 2:1 [17] at pH 7.5, natural river water containing only 0.1 mg L^{-1} Cu^{2+} and Cd^{2+}, and filtered industrial wastewater containing Cd^{2+}. Industrial wastewater and river water were collected from the surface of local industrial wastewater canal on September 25, 2007 and Hun River, Shenyang, China, on July 16, 2007 and immediately filterated on site through a 0.45 μm cellulose nitrate membrane. Major cation concentrations of river water was described above, dissolved organic carbon (DOC) and pH river water was 10.43 mg C L^{-1} and pH 7.8. The industrial wastewater contained the following composition: $[Mg^{2+}] = 0.87$ mM, $[Ca^{2+}] = 2.3$ mM, $[Na^{+}] = 2.7$ mM, $[K^{+}] = 0.22$ mM, DOC = 40.97 mg C L^{-1} and pH 7.2.

2.6. Instrument

Metal concentrations were measured by flame atomic absorption spectrometry (FAAS) (AA-700, PE) after appropriate dilutions and acidification to pH ~ 2 using HNO_3.

DOC concentration was measured by a DC-190 TOC analyzer (Dohrmanne, USA).

3. Results and discussion

3.1. Optimization of the concentration of PA solution

To achieve the maximum binding of the metals to PA, PA concentration was optimized. PA with various concentrations (0.0010, 0.0020. 0.0030, 0.0040 and 0.0050 M) hosted in the DGT devices [16] were immersed in a well-stirred Cd^{2+} solution of 5.0 mg L^{-1} for 24 h. The results showed that the accumulated mass of Cd^{2+} in the PA solutions increased with the concentrations of the PA solution up to 0.0030 M (38.94 μg for 0.0010 M PA solution, 41.48 μg for 0.0020 M PA solution, 44.76 μg for 0.0030 M PA solution). The increase of binding capacity was due to the more binding agents contained in the solutions. Then there was no notable change for accumulation of Cd^{2+} in the range of 0.0030–0.0050 M (44.04 μg for 0.0040 M PA solution, and 44.98 μg for 0.0050 M PA solution).

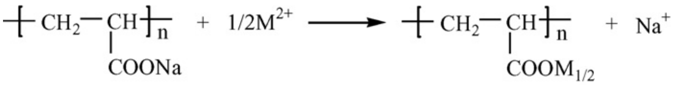
Therefore, a concentration of 0.0030 M PA solution was chosen for Cd^{2+} binding and used in DGT device. It was also found that this concentration was relatively easy to handle with less viscosity and less bubbles between the dialysis membrane and binding solution.

3.2. *Diffusion coefficients of Cu^{2+} and Cd^{2+}*

Diffusion coefficients of Cu^{2+} and Cd^{2+} in dialysis membrane at 25 °C were determined as $0.91 \times 10^{-6}\,cm^2\,s^{-1}$ in synthetic river water and $0.89 \times 10^{-6}\,cm^2\,s^{-1}$ in synthetic industrial wastewater, respectively, for Cu^{2+}, and $1.17 \times 10^{-6}\,cm^2\,s^{-1}$ in synthetic river water and $1.11 \times 10^{-6}\,cm^2\,s^{-1}$ in synthetic industrial wastewater, respectively, for Cd^{2+}. Temperature effects on the diffusion coefficients of the metals were corrected according to Stokes–Einstein equation $D_1\eta_1/T_1 = D_2\eta_2/T_2$ [19] where D_1 and D_2 are diffusion coefficients at absolute temperature T_1 and T_2, respectively. η_1 and η_2 are viscosities of water at T_1 and T_2, respectively.

3.3. *Metal binding properties of PA DGT*

This PA DGT working mechanism for M^{2+} (Cu^{2+} and Cd^{2+}) is based on the formation of stable complexes of PA and the metals [20–25]. The metals were complexed by the carboxylic groups on the polymer.



$$\left[\mathrm{CH_2-\underset{\displaystyle COONa}{CH}}\right]_n + 1/2\,\mathrm{M^{2+}} \longrightarrow \left[\mathrm{CH_2-\underset{\displaystyle COOM_{1/2}}{CH}}\right]_n + \mathrm{Na^+} \qquad (2)$$

The conditional stability constants of metal PA complexes were calculated to be $\log K = 6.98$ for Cu^{2+} and $\log K = 5.61$ for Cd^{2+} using a previously reported approach [16]. Although the stability constants determined in literature by different techniques under different conditions were not always in agreement [26,27], the relatively high values of $\log K$ for Cu^{2+} and Cd^{2+} indicated the strong interactions between PA and the metals. The previous research showed that the other relatively abundant metal ions tended to form weaker complexes with PA (e.g. $\log K\,(Zn^{2+}) = 3.3$ [26], $\log K\,(Co^{2+}) = 2.6$ [26], $\log K\,(Mn^{2+}) = 3.4$ [26], $\log K\,(Ca^{2+}) = 0.87$ [27], and $\log K\,(Mg^{2+}) = 0.55$ [27]) comparing to Cu^{2+} and Cd^{2+}.

To estimate the capacity of PA DGT, the DGT devices were immersed for 72 h in well-stirred solutions in a range of concentrations of Cu^{2+} or Cd^{2+} (0.10, 0.20, 0.50, 1.0, 2.0, 5.0, 10.0 and 20.0 mg L^{-1}). Fig. 1 showed that the binding of the metals to PA reached a plateau at metal concentration of 1.0 mg L^{-1} for Cd^{2+} and 2.0 mg L^{-1} for Cu^{2+}, which indicated that the binding capacities of PA were 0.416 μmol mL^{-1} for Cu^{2+} and 0.498 μmol mL^{-1} for Cd^{2+}, respectively.

The pH influences the chemical forms of PA and the metals, hence affects the binding capacity of PA. Fig. 2 showed that at natural water pH range, the metals were bound efficiently to PA. Small amount of metal were bound at pH > 9 due to the formation of insoluble hydroxide forms of metals. At pH < 4, the mass bound decreased rapidly with increase of pH as a result of the formation of acidic forms of the carboxyl groups (COO^-).

The effect of ionic strength on the binding capacity of PA DGT was also investigated. Fig. 3 showed that the accumulated mass of Cu^{2+} and Cd^{2+} by PA decreased with enhancement of the ionic strengths in the range of 0.001–0.7 M for Cu^{2+} and 0.001–0.1 M for Cd^{2+}. The binding of Cu^{2+} and Cd^{2+} to PA was likely through ion exchange mechanism, i.e. Na^+ in PA was replaced by Cu^{2+} or Cd^{2+}. According to le Chatelier's principle, low concentrations of Na^+ would favor the complexation of Cu^{2+} or Cd^{2+} (Eq. (2)). This became more apparent in the $NaNO_3$ concentration range of 0.1–0.7 M, in which the binding capacity of PA was greatly reduced (Fig. 3). Hence, the use of PA as

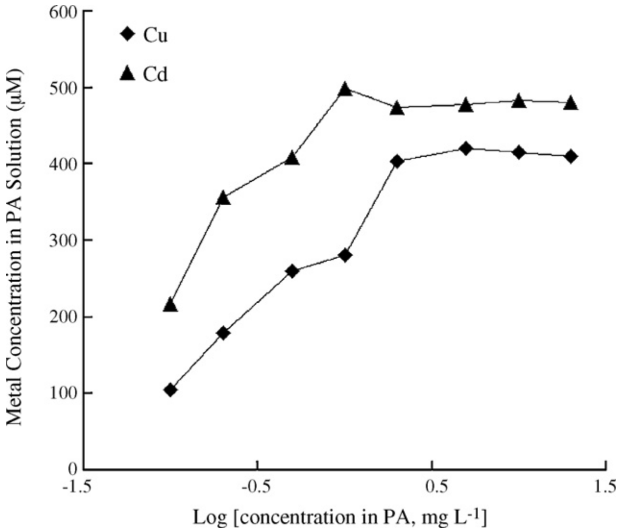


Fig. 1. Mass of Cu^{2+} (◆) and Cd^{2+} (▲) bound in 0.0030 M PA solution with varying concentrations of Cu^{2+} or Cd^{2+}, 0.10, 0.20, 0.50, 1.0, 2.0, 5.0, 10.0 and 20.0 mg L^{-1} at temperature 25 °C.

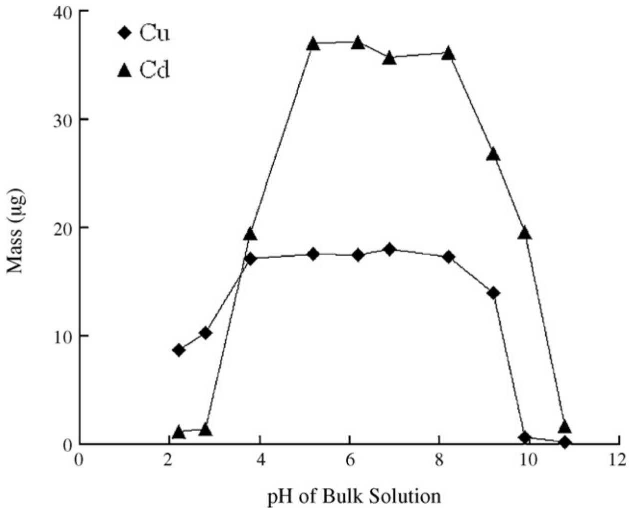


Fig. 2. Effect of pH on the binding capacity of PA DGT for Cu^{2+} (◆) and Cd^{2+} (▲): concentration of Cu^{2+} = 5 mg L^{-1} and Cd^{2+} = 5 mg L^{-1}, time = 24 h, temperature = 25 °C.

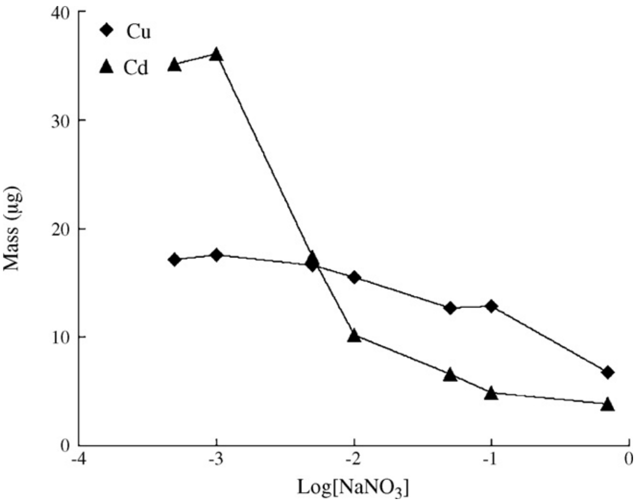


Fig. 3. Effect of ionic strengths on the binding capacity of PA DGT Cu^{2+} (◆) and Cd^{2+} (▲): concentration of Cu^{2+} = 5 mg L^{-1} and Cd^{2+} = 5 mg L^{-1}, time = 24 h, temperature = 25 °C.

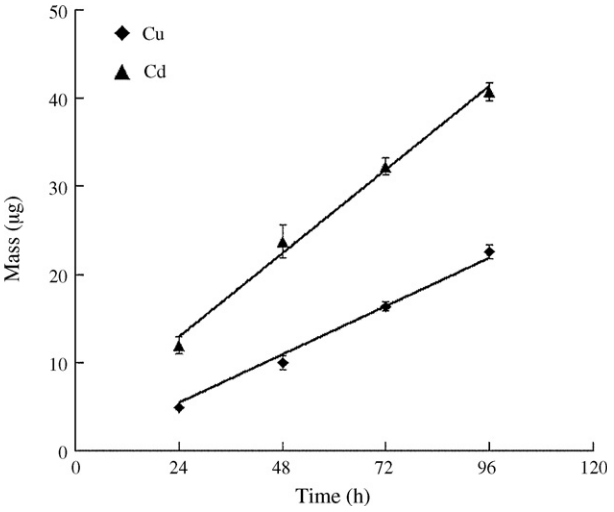


Fig. 4. The mass of Cu^{2+} and Cd^{2+} accumulated by PA DGT vs. time. DGT devices were suspended in well-stirred synthetic river water of known concentrations for different time periods. For (♦) $C = 0.1\ mg\ L^{-1}\ Cu^{2+}$; for (▲) $C = 0.1\ mg\ L^{-1}\ Cd^{2+}$; $\Delta g = 85\ \mu m$, $A = 3.14\ cm^2$.

a DGT binding phase is not suitable for long-term deployment in waters with high ion strengths such as sea water or wastewater.

3.4. Validation of PA DGT

The PA DGT was validated in synthetic river water [28] by examining the abeyance of the DGT equation, i.e. proportional mass accumulation in the device (M) to deployment time and the recoveries. Recovery here is defined as the DGT-labile metal concentration (C_{DGT}) divided by the total metal ion concentration in the solution directly measured by FAAS (C_{FAAS}). Fig. 4 showed that the uptake of Cu^{2+} and Cd^{2+} in synthetic solution increased linearly with time up to 96 h ($r^2 = 0.9895$ for Cu^{2+} and $r^2 = 0.9865$ for Cd^{2+}) with the recoveries of 98.79% for Cu^{2+} ($n = 9$, RSD (%) = 3.86) and 97.80% for Cd^{2+} ($n = 9$, RSD (%) = 4.12) (Table 1).

3.5. Measurement of DGT-labile $Cu^{2+}$and Cd^{2+}

Different molar ratios of metals/EDTA in synthetic river water were used to measure DGT-labile Cu^{2+} and Cd^{2+}. It was found that the DGT-labile Cu^{2+} and Cd^{2+} dramatically decreased when more EDTA was added (the recoveries of 51.92% for Cd^{2+} and 51.02% for Cu^{2+} in 2:1 molar ratios of metals/EDTA solutions and virtually 0 in the 1:1 molar ratios was or lower). This indicated that PA DGT measured unbound metal ion concentrations in the EDTA solutions. Although the complexation of the metals was also through the interaction between the carboxylic groups on EDTA, exchanging EDTA with PA from the complexes was not thermodynamically favored, due to the high stability constant of EDTA/metals [29]. In other words, Cu^{2+} and Cd^{2+} EDTA complexes were DGT-inert.

In the Hun River solution with spiked Cu^{2+} and Cd^{2+}, the recoveries of PA DGT for DGT-labile Cu^{2+} (recovery = 8.47%) and Cd^{2+} (recovery = 27.48%) were low (Table 1). These were because of the presence of natural organic matter (DOC 10.43 mg C L^{-1}) which complexed a significant fraction of the added Cu^{2+} and Cd^{2+} [16]. However, a linear relationship between the accumulated mass and time ($r^2 = 0.9872$ for Cu^{2+} and $r^2 = 0.9820$ for Cd^{2+}) was observed which confirmed that the DGT equation still held and it measured the unbound metal concentrations.

In the filtered industrial wastewater, a 14.2% recovery for Cd^{2+} was obtained (Table 1) which indicated a lower DGT-labile Cd^{2+} concentration due to the higher concentration of DOC. Cu^{2+} was not measured in this experiment because its concentrations were below the detection limit of the FAAS instrument.

Table 1
The DGT-labile concentrations of heavy metals by PA DGT in synthetic river water, in spiked river water and in industrial wastewater.

	C_{DGT}[a] (μM)	C_{FAAS}[a] (μM)	Recovery (%)[b]
Synthetic river water			
Cu^{2+}	1.532	1.549	98.79
Cd^{2+}	1.113	1.138	97.80
Spiked river water			
Cu^{2+}	0.1261	1.489	8.47
Cd^{2+}	0.2773	1.009	27.48
Industrial wastewater			
Cu^{2+}	–	–	–
Cd^{2+}	0.02520	0.1780	14.16

[a] C_{DGT} and C_{FAAS} are the DGT-labile metal ion concentration and the total filterable metal ion concentration measured by FAAS respectively. Data presented here are the mean values of three replicates.

[b] Recovery is defined as the DGT-labile metal concentration (C_{DGT}) divided by the total filterable metal ion concentration (C_{FAAS}).

4. Conclusions

It has been demonstrated in this study that 0.0030 M PA solution can be used as a new binding agent for DGT applications in waters with varying matrixes. This work also confirmed that, in solutions containing different concentrations of EDTA complexing ligand, this DGT measured the same fraction of the metals, free and inorganic metal ions, i.e. metals not complexed by EDTA which agreed with previous DGT work [17]. This new DGT device would be a plus in striving to provide useful information on the speciation of metals with its different binding strength from the traditional DGT.

Future work for this DGT will focus on more complicated solutions containing more types of complexing ligands to find out exact fractions measured by this DGT. This could provide in depth information on the interactions of different types of complexing ligands with metals, and possibly link bioavailability of the metals with DGT-labile concentrations. More work may also be done to confirm the better mass transport of this mobile liquid DGT set-up.

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