



Applications of Diffusive Gradients in Thin Films (DGT) in Soil studies

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Mini Review

Soil is the key component in surface-Earth system providing food, fiber and other ecosystem services, and controlling element biogeochemical cycles. Reliable and easy-to-use techniques play critical roles in understanding environmental processes such as soil conservation and land reclamation that directly affect soil function and agricultural productivity. Diffusive gradients in thin-films (DGT) technique is one of such tools and has become increasingly powerful in soil science research. DGT was developed in 1993 by Davison and Zhang at Lancaster University, UK [1] for measuring trace elements in natural waters, but was soon extended to sediments [2,3] and soils [4,5]. Conceptually, the key components of a DGT device are the well-defined diffusive layer/gel where ions diffuse following Fick's first diffusion law and diffusion gradients are formed, and the binding layer/gel for efficiently binding the diffused ions. In last two decades, DGT measurements and DGT-derived information have contributed greatly to elucidating biogeochemical mechanisms and dynamics in soils.

Similar to measurements in waters and sediments, DGT is useful in soil studies through acquiring mobile or labile concentrations and fluxes of nutrients, metal/metalloids, radionuclides, organics, and others (e.g. rare earth elements, nano ZnO) in soils [6]. For example, Kalkhajah et al. [7] examined P availability in 75 topsoil samples from five representative Chinese PGVPs (plastic-covered greenhouse vegetable production systems) covering a wide range of physiochemical soil properties and cultivation history using several soil P tests, and found that DGT-measure P concentration correlated well with soil solution P demonstrating that DGT P is a versatile measure of P mobility regardless of soil type. More recently, some attempts have been made to use DGT for selective determination of chemical species. By controlling the binding specificity in DGT, selectively measurement of methylmercury [8], Se(IV) [9] and Sb(III) [10] is fulfilled in paddy soils growing with rice plants.

Furthermore, if complexes of a metal dissociate rapidly (fully labile) during the diffusion process in the diffusive layer, they can be captured by DGT [11]. These studies reveal that DGT measurement not only provide a concentration or flux information of a chemical or species in soils, but also reflect the kinetic information of metal complexes.

DGT has great potential in bioavailability study in soils. In a broad sense, DGT provides a better prediction of the uptake of nutrients and metal/metalloids (e.g., P, Cd, Cu, Pb, As, Se, Zn, and Ni) by plants (e.g. wheat, lettuce, and lupin) and soil organisms (e.g. earthworms) in comparison to conventional chemical extraction methods, such as CaCl₂ and EDTA extractions. For example, Tandy et al. [12] found that the concentrations of Cu, Zn and P in the youngest leaf of barley in a pot trial with fourteen agricultural soils can be best predicted by DGT, compared to other tests, such as EDTA and DTPA extraction for Cu and Zn, NaHCO₃ extraction for P and soil solution concentrations. Relative to other soil tests, DGT method best mimicked bioavailability of Cd to earthworm (*Eisenia fetida*) [13], although the same quality observation was not made for uranium uptake by plants [14]. Recently, DGT has been extended to study the bioavailability of organic contaminants to terrestrial plants, but progress remains to be seen. It should be pointed out that these studies only focus on contaminants of herbicides (glyphosate, atrazine and its metabolites) and methylmercury without thoroughly investigating the underlying uptake mechanisms. In addition, compared to some conventional soil tests, the database for soil monitoring using DGT is still small so far, indicating that more studies are needed to expand the application. When a DGT device is deployed in wetted soil, chemical concentration in the solution phase of the thin soil layer (~ 1 mm) contiguous to the DGT sampling window gets depleted quickly. To respond, there will be a resupply or desorption of chemical from

the soil phase to the solution phase. As such, DGT measurement not only captures the solution phase concentration, but can provide information concerning the desorption kinetics from soil solid to solution phase as well. Based on DGT data, models such as DIFS (DGT-Induced Fluxes in Soils) [15] can be used to derive parameters of desorption kinetics of nutrients and contaminants, including labile distribution coefficient (K_{dl}) between solid phase and soil solution, soil response time (T_c , time needed to bring the DGT-soil interfacial concentration from 0 to 63% of its pseudo steady state value) to depletion, and the desorption rate constant ($k-1$). Such studies were initially carried out for cationic metals (e.g. Cd, Ni, Zn), and later extended to include anions (e.g. As and P). Ernstberger et al. [16] used DGT devices containing a Chelex binding gel to acquire a series of time-dependent DGT results ($R(t)$, the ratio of DGT-measured to soil solution concentration at time t) in five different soils, and modelled these nonlinear $R(t) \sim t$ relationships using DIFS. As reflected by the derived T_c values, desorption of Zn and Cd from soil solids was quick, whereas desorption of Ni was slow [16]. The result indicates that the mobilities of metals are strongly related to their element characteristics, such as chalcophile (S-loving, e.g. Zn and Cd) and siderophile (Fe-loving, e.g. Ni), which may be further effected by edaphic factors (e.g. pH, organic matter). Menezes-Blackburn et al. [17] adopted a holistic approach based on DGT, DET (Diffusive Equilibration in Thin-films, similar to DGT but without the binding gel) and DIFS to evaluate the desorption kinetics of phosphorus in 32 soils. Results showed that DGT-measured inorganic P flux responses in the first hour was mainly a function of soil water retention and organic carbon, at longer times it became a function of the P resupply from the soil solid phase. Quite recently, DGT has been further used to study the desorption kinetics of organic contaminants, i.e. antibiotics and bisphenols, in soils [18-20]. Modelling of the time-dependent DGT results using DIFS revealed that the supply of three bisphenols in five spiked soils to DGT was limited by their desorption rate, but soils with more binding sites (e.g. higher organic mater and iron oxides) could resupply BPs more quickly, highlighting the danger of just considering partition effects [19].

Soils are generally of high heterogeneity, especially at the interfaces with water and biota (e.g. plant roots) [21]. DGT has greatly facilitated the understating of solute heterogeneous distribution and dynamic mobility across key environmental interfaces, such as plant rhizosphere [22,23]. With easy-to-access equipment of laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) and easy-to-operate method of colorimetry, high-resolution information of inorganic solutes, such as phosphorus [24], metals [25], and sulfide [26], at sub-millimeter scale is acquired through mapping the binding layer after retrieval. At the root tips of rice (RIL 46) where was characterized by O_2 enrichment and low pH, a new geochemical niche of greatly enhanced As, Pb, and Fe(II) mobilization into solution was clearly captured by DGT gels [27]. Using DGT, increase of Fe and as fluxes

in root vicinity of lupin (*Lupinus albus L.*) was also captured, and As and Fe (II) co-solubilization by root exudates was found, which was likely as a response to P deficiency [28]. Under P-limited conditions, plant root activities and related processes of P mobilization, as reflected by DGT measurement, are typically localized around root apices, but also expand towards the extension/root hair zone, reflecting the complex interaction of plant P acquisition strategies with rhizosphere soils [24].

Collectively, DGT has promising applications in concentration/flux and speciation measurements, bioavailability studies, desorption kinetics explorations and interfacial process investigations in soils. More work should be done to relate kinetic parameters from solid desorption or diffusion process with element/compound properties, soil properties, and ultimately with biota uptake or response. To provide a holistic view in element/chemical mobility and availability related to processes of oxidation-reduction, adsorption-desorption, biota (e.g. plant roots, benthic fauna, and microbiology) activity at millimeter or even higher resolution, researchers can combine DGT with other diffusion-based techniques, such as planar optodes for solutes (e.g. pH, O_2 , pCO_2) and soil zymography for enzymes.

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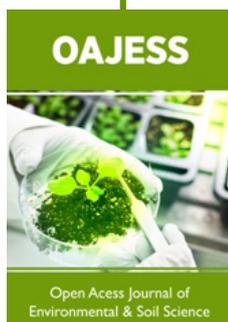
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