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Editorial

Mobilization, Speciation, and Transformation of Organic and Inorganic Contaminants in Soil–Groundwater Ecosystems

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The delicate balance of our ecosystems is under threat from the unrelenting release of contaminants into the environment. Among the most concerning are organic and inorganic pollutants that infiltrate the soil and permeate into the groundwater, posing significant risks to both environmental health and the wellbeing of humans who use groundwater as drinking water. To address this pressing issue, we present our Special Issue on the topic of "The Mobilization, Speciation, and Transformation of Organic and Inorganic Contaminants in Soil–Groundwater Ecosystems". This collection of research articles and studies seeks to shed light on these critical processes and foster innovative solutions for safeguarding our soil–groundwater ecosystems.

The common types of inorganic pollutants in soil and groundwater environments include inorganic salts, toxic metals, radioactive substances, etc. [1–4]. The common types of organic pollutants in soil and groundwater environments include polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs), chlorinated solvents, polychlorinated biphenyls (PCBs), pesticides, and other emerging contaminants [5–8]. Biological contaminants have also raised significant concerns [9]. Both organic and inorganic pollutants are propagated through soil–groundwater systems and eventually enter the food chain, posing health risks to humans. Long-term rock–water interactions, different groundwater recharge patterns, and intensive human activities have resulted in a complicated enrichment of those pollutants [10,11]. The mobilization, speciation, and transformation of these pollutants in the soil and groundwater ecosystem vary greatly depending on the specific hydro-biogeochemical processes and environments.

Understanding the factors influencing the mobilization of contaminants is crucial to implementing effective strategies to prevent their spread and mitigate their impacts. Unforeseen climatic events, human activities, and land-use changes all contribute to the release of harmful substances into subsurface environments. Climate change poses a threat to groundwater by affecting various aspects of the physical, chemical, and biological characteristics of soil and surface water bodies and aquifer recharge patterns. For instance, when heavy precipitation causes flooding, soil erosion occurs, and pollutants such as heavy metals, organic compounds, nutrients, and pathogens are transported from the soil into surface water bodies [12]. In regions where geomorphic units facilitate frequent interactions between surface water and groundwater, pollutants are carried into the groundwater aquifers as surface water infiltrates in large quantities, subsequently deteriorating the groundwater environment [13].

The hydraulic connections and frequent exchanges between surface water and groundwater (SW–GW) constitute a widespread phenomenon [14]. The variations in the water quality and quantity in these bodies of water are significantly influenced by their mutual interactions on both the time and space scales [15–17]. Once one of them becomes polluted, the other is inevitably under serious threat. During the process of pollutants infiltrating and recharging groundwater from surface water, the migration process and flux of pollutants are notably influenced by the local hydrodynamic characteristics and biogeochemical processes within the hyporheic zone [18,19]. Differences in flow pathways, velocities, residence times, and the exchange between surface water and groundwater control the reactive transport of, and affect the flux in, pollutants.

In the process of pollutants from the vadose soil environment entering the groundwater system, their adsorption, migration, and transformation within the geological media are complex and take forms such as surface runoff, leaching, erosion, desorption, and dissolution [19–21]. This complexity encompasses factors including the soil lithology, porosity, moisture content, pollutant type and concentration, oxidation–reduction potential (ORP), pH, organic matter, cation exchange capacity, microbial communities, etc. [22,23]. It is worth noting that certain elements (e.g., phosphorus, iron, fluoride, arsenic, iodine, uranium, and molybdenum,) are naturally abundant in the form of minerals/rocks in soils and groundwater sediments [3,24–26].

Although elevated levels of these inorganic constituents are often attributed to geogenic processes such as weathering, leaching, and water–rock interactions, anthropogenic activities can be expected to introduce and further intensify the contaminations [27–29]. These contaminants are primarily derived from domestic, agricultural, and industrial sources, including the application of fertilizers and pesticides in agricultural production, irrigation with sewage (or reclaimed water), leakage from domestic sewage networks and landfill site wastewater, waste residues generated during mining, or oil and gas field exploitation. [1–8,18–20]. For instance, fluctuations in groundwater levels caused by either natural or anthropogenic processes accelerate the leaching of organic matter, nitrogen, and heavy metals from the soil into shallow groundwater [30] and affect the aquifer redox cycling and microbial activity [31]. Intensive groundwater pumping can draw recently recharged contaminated modern groundwater into deeper aquifer systems [32]. Petroleum and natural gas exploitation causes petroleum hydrocarbon and salinity contamination in the vadose zone [33] and shallow groundwater [34]. Furthermore, the role of metals (e.g., Hg, Zn, Cu, and Cd) in co-selecting antibiotic resistance might be important in the spread of antibiotic resistance genes [20].

The speciation of contaminants adds another layer of complexity to the soil–groundwater environment. As pollutants interact with the soil matrix and microbial communities, they undergo chemical transformations, altering their properties and toxicity. Contaminants can undergo different chemical transformations, resulting in various species with different properties and toxicities. Metals can exist in different oxidation states, and organic contaminants can undergo degradation or transformation into metabolites and greenhouse gases. The speciation of contaminants affects their mobility, bioavailability, and toxicity, and an understanding of these forms is crucial to accurately assessing their environmental impact and risk.

For instance, arsenic (As) primarily exists as As(V) and can transform into more mobile As(III) under reducing conditions, affecting the groundwater quality [35]. Chromium (Cr) exists as both the Cr(VI) and Cr(III) forms, with Cr(VI) becoming soluble and migrating into groundwater under oxidizing conditions [36]. Uranium (U) in soil and sediments is typically found as less mobile U(IV) compounds, while, in groundwater, it exists as more mobile uranyl U(VI) and ${UO_2}^{2+}$ complexes, which are strongly influenced by various groundwater physicochemical factors [37]. Redox conditions and natural organic matter play key roles in these transformations [24,29,38,39].

Fluoride often exists in nature in the form of F^- and forms soluble salts with monovalent alkali metals in groundwater, such as fluoride salt (NaF) and fluoride potassium salt (KF) [3]. Meanwhile, fluorite (CaF_2), sellaite (MgF_2), and fluoride-bearing silicate minerals such as mica, amphibole, tourmaline, and fluorapatite are commonly found in soils or rocks. Therefore, the weathering of fluoride minerals could be a natural source of fluoride in water [3]. In the groundwater systems, I commonly exists in the form of iodide (I^-), iodate (IO_3^-), and organic iodine (OI), and I^- is the dominant species in groundwater, whereas IO_3^- and OI are the common species in the soil/sediments [29].

Organic contaminants include a range of conventional pollutants such as petroleum hydrocarbon, phenolic compounds, and pesticides, while concerns have also extended to persistent organic pollutants, such as perfluoroalkyl and polyfluoroalkyl substances (PFAS), pharmaceuticals and personal care products (PPCPs), antibiotics, and microplastics [2,5,8]). Perfluorinated compounds (PFCs) including PFAS are persistent organic pollutants that can linger in soil and impact groundwater long after their use [40]. They are highly toxic and include chemicals such as PFOA and PFOS. The antibiotic pollution in soils and groundwater is diverse, with substances such as ciprofloxacin and sulfamethoxazole found in high concentrations [8,20]. Microplastics include small plastic particles including polyethylene and polyvinyl chloride, etc., resulting from the breakdown of larger plastic waste [41,42].

Pollutants in soil and groundwater pose significant risks to both ecosystems and human health [39,43]. These pollutants enter the human body through pathways including bioaccumulation, the food chain, and drinking water, leading to different types of toxicity, including chemical toxicity, radiation toxicity, and carcinogenicity. Common pollutants, such as nitrogen, when present in excess, can cause various cancers and other health issues [21]. Heavy metals, such as arsenic, chromium, and lead, can lead to cancer, organ damage, and nervous system disorders [29,36]. Radioactive isotopes in groundwater can be highly toxic, affecting various bodily systems. Additionally, non-degradable organic pollutants, such as antibiotics and microplastics, pose emerging health risks due to their persistence and limited microbial degradation [20,42].

The transformation of contaminants within soil–groundwater ecosystems can lead to either an exacerbation or attenuation of their effects. The processes of degradation, redox reactions, and other chemical transformations can significantly impact the persistence and fate of pollutants. An in-depth exploration of these mechanisms together is vital to developing sustainable solutions for remediating contaminated sites. The range of strategies for alleviating the impact of contaminants in soil–groundwater systems includes the monitoring and assessment of pollutant levels, the implementation of containment measures, the promotion of natural attenuation through bioremediation, the incorporation of remediation materials, and the employment of engineered solutions such as permeable reactive barriers or pump-and-treat systems to capture and treat contaminated groundwater, as well as phytoremediation to treat contaminants before they seep into aquifers ([44–49]). Before these techniques are used, their cost and resulting environmental impacts should be considered.

Bioremediation emerges as a particularly promising, sustainable, environmentally friendly, and cost-effective strategy. To start with, understanding the assembly of microbial communities, their driving forces (e.g., their pH, salinity, nutrients, and metals) and their role in transforming pollutants in the vadose-zone–groundwater ecosystem is instrumental to unlocking the full potential of bioremediation [22,50–52]. While advanced techniques have been developed for characterizing microbes, significant gaps remain in our understanding of dynamic subsurface microbial communities and the biogeochemical processes in the environments they inhabit [53], which hinder the effective use of biostimulation or bioaugmentation as an in situ remediation strategy. For example, the complex coupled carbon and iron cycling at multiple redox interfaces across subsurface environments can be expected to positively and negatively affect both microbial and extracellular enzyme activity [54]. Dissimilatory nitrate reduction to ammonia is enriched in the downgradient along the groundwater flow path [55]. Sulfate reduction accelerates groundwater arsenic contamination in aquifers with replete iron oxides [56]. The intrusion of produced water enhances the salinity and petroleum hydrocarbon levels in shallow groundwater, causing a transformation in the composition and functionality of bacterial and archaeal communities [34]. Moreover, hydrodynamic disturbance is another possible driving force for microbial community assembly and biogeochemical processes in sedimentary environments [57].

The mobilization, speciation, and transformation of contaminants in soil–groundwater ecosystems represent an urgent call for action in the face of environmental degradation. The task of preserving the integrity of our soil–groundwater ecosystems is a complex one that requires a multidisciplinary approach. There is still a gap in our understanding of the mechanisms that connect mobilization, speciation, health impacts, and microbial processes related to groundwater contaminants. To effectively address soil pollution sources and manage the risks of groundwater pollution, it is essential to comprehend how complex geological and hydrogeological factors in soil–groundwater systems interplay with climate change and human activities. This understanding can significantly inform the development of remediation strategies.

As we present this Special Issue on "The Mobilization, Speciation, and Transformation of Organic and Inorganic Contaminants in Soil–Groundwater Ecosystems", we call upon researchers to come together, share knowledge, and work toward a shared vision of a cleaner and greener future. Through the insightful research and innovative solutions showcased in this Special Issue, we are reminded of our collective responsibility to safeguard the delicate balance of nature.

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