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Uranium accumulation in environmentally relevant microplastics and agricultural soil at acidic and circumneutral pH

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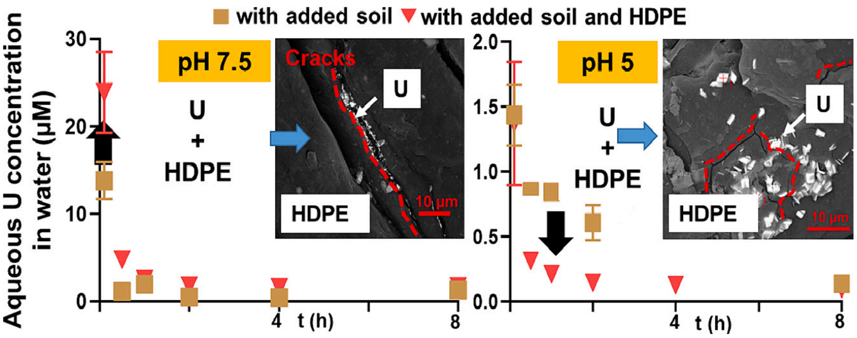
HIGHLIGHTS

- Accumulation of U on HDPE was confirmed by microscopy and spectroscopy.
- HDPE modified the accumulation and mobilization of U in soil as a function of pH.
- Uranium precipitation onto the surface of weathered MPs in water.
- Exposure to U-bearing plastic solids is a critical understudied pathway.

GRAPHICAL ABSTRACT



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ABSTRACT

The co-occurrence of microplastics (MPs) with potentially toxic metals in the environment stresses the need to address their physicochemical interactions and the potential ecological and human health implications. Here, we investigated the reaction of aqueous U with agricultural soil and high-density polyethylene (HDPE) through the integration of batch experiments, microscopy, and spectroscopy. The aqueous initial concentration of U (100 μM) decreased between 98.6 and 99.2 % at pH 5 and between 86.2 and 98.9 % at pH 7.5 following the first half hour of reaction with 10 g of soil. In similar experimental conditions but with added HDPE, aqueous U decreased between 98.6 and 99.7 % at pH 5 and between 76.1 and 95.2 % at pH 7.5, suggesting that HDPE modified the accumulation of U in soil as a function of pH. Uranium-bearing precipitates on the cracked surface of HDPE were identified by SEM/EDS after two weeks of agitation in water at both pH 5 and 7.5. Accumulation of U on the near-surface region of reacted HDPE was confirmed by XPS. Our findings suggest that the precipitation of U was

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facilitated by the weathering of the surface of HDPE. These results provide insights about surface-mediated reactions of aqueous metals with MPs, contributing relevant information about the mobility of metals and MPs at co-contaminated agricultural sites.

1. Introduction

The growing production of plastics and the lack of appropriate waste disposal strategies have globally spread plastic pollution raising potential ecological and health concerns (Alimi et al., 2018; Jin et al., 2022; Machado et al., 2019; Sun and Wang, 2023). Each year, microplastics (MPs) are loaded into the land with an estimated quantity exceeding the current presence of MPs in ocean water (Chen et al., 2022; Horton et al., 2017). As a result, soil is likely the long-term sink of MPs developing an important threat to the food safety and ecosystems (Corradini et al., 2021). This widespread distribution of plastics is alarming as they act as pollutants but also as carriers of other harmful contaminants, such as potentially toxic metals, causing risk to organisms (Li et al., 2023; Ren et al., 2021).

The environmental pollution of plastic includes various sizes ranging from macro- to micro- and nanoplastics (Mitrano et al., 2021; Schefer et al., 2023). Plastic debris can be generated under mechanical abrasion, photodegradation, or chemical oxidation among other mechanisms (Galloway et al., 2017; Reichert et al., 2024). Extended exposure to environmental biotic and abiotic processes enhances the reactivity and toxicity of plastic polymers (Duis and Coors, 2016; El Hayek et al., 2023; Hidalgo-Ruz et al., 2012). For instance, weathering of plastics caused by UV radiation, chemical oxidation, or mechanical abrasion can increase surface roughness and oxygen-containing groups, stimulate surface chemical reactivity, and thus enhance the sorption capacity and the formation of inorganic coatings in water and soil (Pincus et al., 2023; Ren et al., 2021). Such processes are of concern in environments where toxic metals co-occur with plastic pollution (Purwiyanto et al., 2020; Tang et al., 2020; Wang et al., 2019). We recently detected the co-occurrence of MPs with uranium (U) and other metals in freshwater collected from rural and urban areas in New Mexico, U.S. (Quiambao et al., 2023). Interaction of MPs with U in natural environments (Chen et al., 2021) may represent a critical understudied exposure pathway where the maximum contaminant limit (30 $\mu g\ L^{-1}$) set by the U.S. Environmental Protection Agency is exceeded (Elseblani et al., 2023; Girlamo et al., 2023; Hashemi et al., 2023; Hoover et al., 2018; Ravalli et al., 2022). Therefore, the effect of the interfacial interaction of U and MPs on the accumulation of U in agricultural soil serves as the basis for this study.

Metals can accumulate on surface of plastics through adsorption and precipitation mechanisms (Quiambao et al., 2023; Rochman et al., 2014; Xiong et al., 2024). Such phenomena depend on surface specific area and the degree of hydrophilicity of plastics that provide active ionic sites to react with metals (Mao et al., 2020). The development of these surface reactions differs with respect to two key factors: (1) the environmental conditions (e.g., the chemistry, the elemental and mineral composition of water and soil) and (2) the physicochemical properties of the polymers and metals themselves (Ren et al., 2021). For example, Godoy et al., 2019 showed that polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) have a specific high affinity to adsorb chromium (Cr), copper (Cu), cobalt (Co), and lead (Pb) that depended on the water chemistry of seawater, urban wastewater, and irrigation water. Different chemical interactions can occur between MPs and metals, including electrostatic interactions, van der Waals interactions, the hydrophobic and hydrogen bonding, and π-π interactions (Li et al., 2022; Liu et al., 2022b; Xiong et al., 2024).

The effect of pH and aqueous chemistry on the interfacial interactions of U with MPs in the environment is still largely overlooked. At circumneutral pH, the negatively charged uranyl carbonate (U-CO_3) complexes can form in natural water and soils depending on U and calcium (Ca) concentrations (El Hayek et al., 2018; Rout et al., 2024). At acidic pH, the positively charged UO_2^{2+} and U hydrolysis species can predominate (Laurette et al., 2012). The interaction of poly(methylmethacrylate) PMMA, PE, and PS pristine MPs with aqueous U in acidic experiments performed at pH 3 was not detected (Quiambao et al., 2023). However, at circumneutral pH under similar laboratory conditions, MPs served as substrate surface for the adsorption and nucleation of U precipitates (Quiambao et al., 2023). These reactions have been mostly investigated in aqueous environments in which the role of soils has not been considered.

Existing studies on the combined pollution of MPs and heavy metals suggest the potential of MPs to increase metals solubility in soils (An et al., 2023; Li et al., 2021). All these studies considered various metals other than U. For instance, a recent meta-analysis by An et al., 2023 showed that both the MPs characteristics (i.e., the polymer type and size of plastics) and the soil physicochemical properties (i.e., soil organic matter, sand content, and soil pH) influence the extractable fractions of heavy metals (mainly Cu, Pb, cadmium (Cd), iron (Fe), and manganese (Mn) from soil. Another experimental study showed that a high concentration of PE MPs in soil (10 % in weight) decreases the adsorption capability of soil to Pb^{2+} and Zn^{2+} (Li et al., 2021). Electrostatic interactions and surface complexation are the main reported influencing factors on the mechanisms of PE and heavy metals interaction in soil (Liu et al., 2024). The interaction of metals with MPs varies depending on the weathering processes that MPs undergo in the environment (Schefer et al., 2023). Different studies reported that metal interaction with aged MPs is enhanced in comparison to non-weathered plastics following the increase of cracks, specific surface area, and oxygen-containing functional groups (Dong et al., 2019; Ren et al., 2021; Tang et al., 2023; Tziourrou et al., 2021). This phenomenon was described in marine and estuarine environments where hydrous metal oxides were detected on plastics surface (Holmes et al., 2014). The inorganic coatings can affect the interaction of MPs surface with toxic metals (Pincus et al., 2023). These findings emphasize the importance of considering the interfacial interaction of U with MPs over time under environmentally relevant conditions and its effect on U accumulation in soil (Jin et al., 2022).

The objective of this study is to determine the effect of surface adsorption and precipitation of U with coexisting MPs on the accumulation of U in agricultural soil by the integration of batch experiments, microscopy, and spectroscopy analyses. Aqueous U (100 μM) was mixed with soil and high-density-polyethylene (HDPE) at environmentally relevant nominal concentration of plastics in systems containing Ca and bicarbonates at circumneutral and acidic pH (De Souza MacHado et al., 2018; Huerta Lwanga et al., 2017; Machado et al., 2019). A novel aspect of this study is the identification of the role of MPs in affecting the concentration of aqueous U in agricultural soil mixed with HDPE depending on pH. Our findings contribute relevant information to understand the interaction and precipitation of U on plastics surface in agricultural environment, which may increase their potential to co-enter the terrestrial food chain.

2. Materials and methods

2.1. Preparation and characterization of HDPE plastic particles

High-density polyethylene (HDPE) bottles were purchased from the United States Plastic Corporation. We focused on HDPE in this study due to the high-production volume and large abundance of PE in both terrestrial and aquatic environments, and because HDPE is highly used in plastic packaging and food-service plastics (Pincus et al., 2023).

Furthermore, PE was recently detected as the most prevalent polymer in human placenta, indicating its wide occurrence and bioaccumulation (Garcia et al., 2024). Bottles were cut into small pieces and then blended using a coffee grinder. This mechanical breakdown protocol mimics the natural mechanical abrasion that can potentially produce micro- and nano-sized particles from plastic debris in the environment (Dawson et al., 2018; Gao et al., 2022). The size, shape, polymer chemistry, surface composition of the generated HDPE particles were studied using scanning electron microscopy energy-dispersive spectroscopy (SEM-EDS), Attenuated Total Reflection-Fourier Transform Infrared (ATR-FTIR) spectroscopy, and X-ray photoelectron spectroscopy (XPS). ImageJ software was used to estimate the size range of HDPE particles from the SEM images. More information on the solid characterization is provided in the Supplementary material.

2.2. Soil characterization

A sandy loam (USDA Texture) characterizing 80 % sand, 8 % silt, and 12 % clay was provided by Agvise Laboratories (US). The range of soil pH was 7.2–7.5 and contained 4 % organic matter. Bulk elemental composition was determined using an X-Ray Fluorescence (XRF) Rigaku ZSX Primus II Wavelength dispersive instrument. The concentration of acid-extractable U and other metals in soil was measured using inductively coupled plasma mass spectrometry (ICP − MS, PerkinElmer NexION 300D) and inductively coupled plasma optical emission spectrometry (ICP − OES, PerkinElmer Optima 5300DV), respectively. The soil digestion was conducted by adding 2 mL of HNO_3, 4 mL of HCl, and 2 mL of HF and then samples were heated in a Digi prep MS SCP Science block digester at 95 °C for 4 h. The acid digested solutions were filtered using 0.45 μm syringe filters and diluted with 18 ultrapure water to 25 mL. Information on soil properties and chemical composition are summarized in Table 1 and Table S1.

2.3. Batch experiments for aqueous U interaction with HDPE and agricultural soil

Batch experiments were conducted to study U adsorption and precipitation with HDPE and soil particles in simplified plant nutrients solution containing 3 mM Ca and 1 mM HCO_3^- with pH maintained at 7.5 and 5. These pH conditions are environmentally and ecologically relevant as well as they are in the range of optimal pH for the growth and nutrition of most plants (Läuchli and Grattan, 2017; Min et al., 2014). In addition, the acidic pH 5 will allow an understanding of the surface interaction of U and HDPE that can occur near the root surface in the rhizosphere where it is reported that plant root surface has acidic properties (Nye, 1982; Rudolph et al., 2013). Our selected systems contained constant concentrations of Ca and carbonate and other simplified plant nutrients ($MgSO_4$, 0.5 mM; NH_4NO_3, 2 mM; KCl, 1 mM) to mimic the water chemistry of plant watering solution and the environmental conditions where uranyl-carbonate complexes are prevalent (El Hayek et al., 2019). The concentration of HDPE (0.5 wt%) was selected according to previous studies reporting the environmentally relevant concentration of MPs (de Ruijter et al., 2023; Lenaker et al., 2019; Redondo-Hasselerharm et al., 2023; Xia et al., 2021). All experimental units were run in quadruplicates. All glassware was rinsed with 10 % (*v*/*v*) nitric acid and DI water and then was sonicated for 30 min with ultrapure water in a Cole-Parmer CPHX Series sonicator. The selected concentration of U for this study is based on concentrations detected at mine wastes, in situ leaching, and other sites affected by U contamination (Blake et al., 2017; Gonzalez-Estrella et al., 2020; Ruiz et al., 2016; Saunders et al., 2016). A 50 mL stock solution of 4 mM of uranyl acetate $UO_2(CH_3COO)_2{\bullet}2H_2O$ was prepared as a source for soluble U in the batch experiments. A volume of 50 mL carbonate solution was added into each 125 mL Erlenmeyer flask with 100 μM of U and/or 0.05 g of HDPE particles at pH 5 and pH 7.5 both with and without soil. 0.5 M HCl or 0.1 M NaOH was used to adjust the pH over time. Controls represent flasks without U or HDPE. Table S2 outlines the experimental matrix of the batch reactors. The flasks were agitated at 150 rpm for 336 h (14 d) at room temperature. Aliquot volumes were collected at various time points (from 0.1 to 336 h) to measure the concentration of aqueous U and were filtered with a Pall Laboratory Acrodisc 0.45 μm syringe filter and diluted with 2 % nitric acid for U analyses using ICP-MS. After 336 h, all the remaining solutions in each batch reactor were filtered using a 0.5 μm glass microfiber filter and glass frit filter units. The collected filtrate at each pH (i.e., reacted HDPE, reacted HDPE-soil, and reacted soil) was analyzed using SEM-EDS for the identification of U precipitation in the solids and to determine any physiochemical changes to the soil and plastic particles. X-ray computed tomography (CT) scanning was used to check on MPs agglomeration with soil particles in the collected filtrate. Zeta potential measurement was conducted using a Malvern Zetasizer Nano-ZS equipped with a He—Ne laser (633 nm) and non-invasive backscatter optics to measure the surface charge of soil particles in the collected filtrate. XPS analysis was conducted to determine U content and the chemical changes on the near-surface region for reacted HDPE with U from experiments with only added HDPE. Further detail is provided in the Supplementary material.

Table 1

[a]Soil characterization performed by Agvise Laboratories including textural class of the soil and its properties. The concentration of U in acid digested soil was measured in our laboratory using ICP-MS as described in the text. The concentration of trace elements and phosphorus was measured using ICP-OES.

Soil properties	
% Sand [a]	80
% Silt [a]	8
% Clay [a]	12
USDA Textural Class (hydrometer method) [a]	Sandy loam
% Organic carbon [a]	2
% Organic matter (Walkley-Black method) [a]	4
pH (water) [a]	7.4
% Total nitrogen (Analyzer) [a]	0.182
Olsen phosphorus mg Kg^{-1} [a]	24
P mg Kg^{-1} (acid extracted)	487.2 ± 43
U mg Kg^{-1}	1.3 ± 0.05
Cu mg Kg^{-1}	7.7 ± 0.3
Cr mg Kg^{-1}	40.2 ± 4.2
Pb mg Kg^{-1}	7.2 ± 0.9
Zn mg Kg^{-1}	27.6 ± 2

2.4. Chemical equilibrium modeling

The effect of U speciation on the interaction of U with HDPE and agricultural soil in function of pH was investigated by determining the aqueous U species using Visual MINTEQ under atmospheric equilibrium. The chemical equilibrium calculations were based on the chemical composition of the water used for this study which contained simplified plant nutrients solution ($CaCl_2$, 3 mM; $MgSO_4$, 0.5 mM; NH_4NO_3, 2 mM; KCl, 1 mM) at pH 7.5 and pH 5 in 1 mM HCO_3^- at 25 °C. Given that phosphorus (P) has a critical role in U solubility and speciation, the concentration of water soluble P was measured in the solutions collected from batch experiments at each pH using ICP-OES in order to be included in the chemical equilibrium calculations (Table S3).

2.5. Statistical analysis

For the different aqueous U concentrations between experiments with added soil and experiments with added soil and HDPE, each condition consisted of 4 replicates and a 2-way ANOVA considering the time and the addition of HDPE as factors was used with Šídák's multiple comparisons testing for individual time points. The statistically significant level was confirmed at *p* value lower than 0.05 for all the statistical tests.

3. Results and discussion

3.1. Characterization of HDPE plastic particles

ATR-FTIR indicated a polymer match (89 %) to PE (Fig. S1). SEM-EDS of the unreacted plastics identified Ca, silicon (Si), titanium (Ti), sulfur (S), sodium (Na), and magnesium (Mg) on the surface of HDPE along with carbon (C) (Fig. 2A-C and Fig. S2). These elements represent additives incorporated into the plastic polymer during the manufacturing process of the plastic bottles and are consistent with additives previously identified in PE polymers (Cherif Lahimer et al., 2017; Hahladakis et al., 2018; Hermabessiere et al., 2017). HDPE particles size ranged from 0.3 μm to 5 mm and presented a heterogeneous morphology (Fig. S3) making them relevant to the reported plastic fragments and particles in global sediments and agricultural soils (Fig. 2A-C) (Chamas et al., 2020).

3.2. Aqueous uranium removal in water mixed with HDPE and agricultural soil at pH 5 and pH 7.5

High sorption capacity of the sandy loam soil to U was detected at both pH conditions, resulting in >99 % U removal after 24 h of reaction at both pH 7.5 and 5 (Fig. 1A, B). This is a relevant characteristic of the common mineral phases of sandy loam soil (Table 1) that were confirmed by XRD analysis indicating the presence of mainly quartz followed by feldspar minerals (Fig. S4). The effect of high sorption capacity of these silica phases (Massey et al., 2014) along with quartz (Ilton et al., 2012), feldspar (Blake et al., 2015), iron (hydr)oxides (Lefebvre et al., 2023), and clay minerals (Schlegel and Descostes, 2009; Zhang et al., 2022) on the solubility of aqueous U was reported at various aqueous geochemical conditions.

The addition of HDPE to soil increased significantly ($P < 0.05$) the initial removal of U at pH 5 as compared to experiments with only soil (Fig. 1D). For example, the concentration of aqueous U decreased from 100 to 0.85 μM (a decrease of 99.2 %) after 0.5 h in experiments with only soil, while it decreased to 0.3 μM (a decrease of 99.7 %) in experiments with added HDPE and soil. This decrease corresponds to the significant ($P < 0.05$) 2.8-fold reduction in the remaining aqueous U concentration with the addition of HDPE. The decrease in the concentration of aqueous U remained significantly ($P < 0.05$) higher in experiments with added HDPE and soil than in experiments with only soil until 4 h of reaction (Fig. 1D). Lower U removal was observed at pH 7.5 compared to pH 5 with 86.2 % decrease of aqueous U in experiments with soil and 76.1 % decrease in experiments with added HDPE and soil within 0.1 h of reaction (Fig. 1 and Table 2). Unlike experiments at pH 5, the decrease of aqueous U at pH 7.5 was detected 1.2 to 10.1 % lower in experiments with added HDPE and soil than in experiments with only

Table 2
Summary of the percentage decrease in aqueous U concentrations from soil reactions with and without HDPE at pH 7.5 and pH 5.

	pH 7.5		pH 5	
Time (h)	% decrease for experiments with added soil	% decrease for experiments with added soil and HDPE	% decrease for experiments with added soil	% decrease for experiments with added soil and HDPE
0.1	86.2	76.1	98.6	98.6
0.5	98.9	95.2	99.2	99.7
1	98	97.5	99.2	99.8
4	99.6	98.4	99.3	99.9
24	99.1	98.5	99.8	99.9
336	>99.9	>99.9	>99.9	>99.9

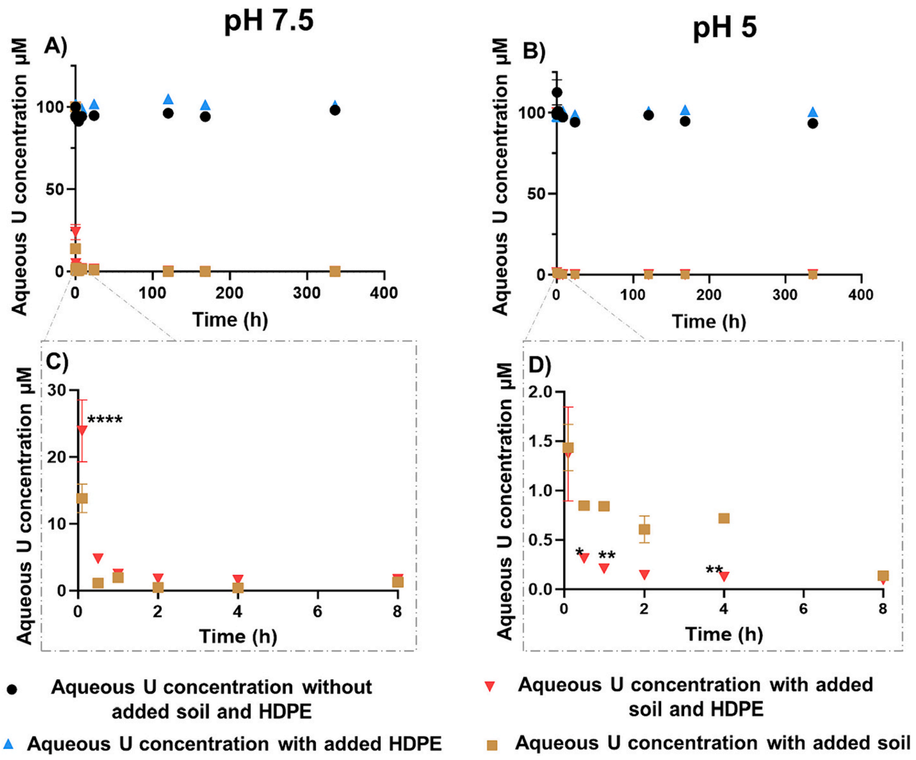

Fig. 1. Aqueous U concentration (μM) as a function of time in solutions containing 1 mM HCO_3 and 3 mM Ca at pH 7.5 and pH 5 with 100 μM initial U_i concentration studied alone, or mixed with soil, HDPE, or with soil and HDPE. Data points represent the mean and the standard error of mean from experiments with four replicates. The absence of appearance of certain error bars corresponds to their short size compared to the size of the symbol. The results of the total experimental time points from 0.1 to 336 h (A-B). A zoom into the variation of aqueous U concentration between experiments with added soil and experiments with added soil and HDPE during only the first 8h of experiments (C-D). Asterisks in figures C and D indicate significant difference between experimental conditions by 2-way ANOVA considering the time and the addition of HDPE as factors (* P<0.05; ** P<0.001; **** P<0.0001).

soil within the first 4 h (Fig. 1C). In experiments containing only HDPE, the concentrations of aqueous U remained constant at 101 ± 2 μM and 99 ± 2 μM for the extent of the experiments at pH 7.5 and 5 respectively. These levels of aqueous U are in the range of concentrations that was measured in the control experiments without added soil and HDPE (Fig. 1A, B).

pH can change the speciation of U as well as the charge and protonation level of silica and other soil minerals (El Hayek et al., 2018; Gabriel et al., 2001; Ilton et al., 2012; Shen et al., 2020). Chemical equilibrium modeling based on the experimental conditions (Table S3) used in this study suggests that neutrally and positively charged aqueous U complexes predominantly occur at pH 5 (mainly $UO_2HPO_{4\ (aq)}$ with 73 % and the positively charged uranyl oxides species with 19 %). In experiments at pH 7.5, negatively and neutrally charged aqueous U complexes are predicted to predominantly increase with the main presence of $(UO_2)_2CO_3(OH)_3^-$ (46 %), $CaUO_2(CO_3)_3^{2-}$ (42 %), and $Ca_2UO_2(CO_3)_{3\ (aq)}$ (11 %). The higher removal of U observed at pH 5 as compared to pH 7.5 could be attributed to the (1) occurrence of uranyl phosphate complexes and the possible precipitation of $(UO_2)_3(PO_4)_{2(s)}$ at pH 5 as determined by chemical equilibrium modeling (saturation index = 0) which can enhance the immobilization mechanisms of U in soil (Meza et al., 2023; Wen et al., 2018), and to the (2) electrostatic attraction of the predominant positively charged uranyl oxides onto the negatively charged soil particles such as quartz (Kar et al., 2012). Additionally, U oxides participated in cationic exchange mechanisms as seen with heavy metals in sandy loam soils, or to substitute the cations that are present in the soil minerals (Gong and Donahoe, 1997; McLemore, 2014). However, a circumneutral pH was detected in previous studies to increase the surface negative charge on soil minerals in the presence of carbonate as compared to acidic condition (Ilton et al., 2012; Saleh et al., 2018). In our experiments, the negative zeta potential values of soil particles were 2 times higher at pH 7.5 (− 23 mV ± 2) than at pH 5 (− 12 mV ± 1), confirming the increase of negative charge on the surface of soil particles at pH 7.5. Ilton et al., 2012 reported that the increase in pH can deprotonate soil silanol groups, creating a more electrostatic repulsion for uranyl carbonate aqueous species. Such mechanisms can explain the detected lower removal of U in our experiments at pH 7.5 as compared to pH 5. Interestingly, for experiments with only added HDPE, the quantitative aqueous analysis of U did not reveal a noticeable interaction of HDPE with U at both pH conditions. However, the findings presented in this study suggest that adding HDPE particles to soil can modify the mechanisms of U accumulation depending on pH as follows. Under acidic pH, the occurrence of neutrally charged phosphate uranyl complexes and the positively charged U complexes (UO_2^{2+} and UO_2OH^+) resulted in a significant ($P < 0.05$) increase in the initial removal of U in the mixture of soil and HDPE as compared to soil with no plastics. However, under circumneutral pH, the high electrostatic repulsion between the negatively charged uranyl carbonate complexes and the mixture of soil and HDPE, resulted in a significant ($P < 0.0001$) decrease in the initial removal of U as compared to soil with no plastics. These effects of HDPE in soil were predominantly observed within the first hours of experiments as shown in Fig. 1C and 1D. Phosphate was reported in many other studies to increase U immobilization in soil through metal complexation, precipitation, redox and adsorption mechanisms (Dong et al., 2023). Under our experimental conditions, the precipitation of U in soil was not detected by electron microscopy at both pH 5 and 7.5 given that the accumulated U concentrations in soil were below the detection limit (<100 mg kg^{-1}). Further research should investigate the role of the speciation of U and phosphate levels in soil on U immobilization and interaction with the coexisting plastics.

3.3. Detection of soil inorganics on the surface of HDPE particles following two weeks of agitation in water

The sorption of micro-sized soil minerals with the surface of HDPE particles after two weeks of batch experiments was confirmed by SEM-EDS at both pH conditions, with no detectable precipitation of U (Fig. 2). SEM images show plastic particles in micrometer and millimeter scales interacting with the large soil particles (> 1 mm) in the collected filtrate at pH 7.5 (Fig. 2D). Various particles from the fine fraction of soil (14–60 μm) were found (Fig. 2) on the surface of HDPE. Image 2E shows a quartz mineral between the cracks of HDPE surface. The EDS spectra corresponding to the soil particles observed on the surface of HDPE identified Si, Al, Ca, K, Ti, Fe, and Mg. These elements constitute the typical essential minerals of sandy loam soil (quartz, feldspar, and clay silicates) as identified by the XRD analysis (Fig. S4). The examination of the surface of HDPE using SEM-EDS analysis (Fig. 2F) also revealed that the surface of some HDPE particles was coated with inorganic elements, including Si, K, Ti, Ca, and Mg. Such observation confirms that two weeks of agitation in carbonate water was sufficient to induce noticeable chemical and physical changes on the surface of HDPE particles. This phenomenon was replicated at acidic pH. Fig. 2G-I show the surface of HDPE particles in the collected filtrate from experiments at pH 5 covered by soil minerals as compared to the HDPE particles from experiments at pH 7.5 (Fig. 2D—F). This observation is consistent with previous studies showing that the degree of MPs interaction with soil minerals can differ depending on the pH of media (Jin et al., 2022; Luo et al., 2020). For example, Luo et al. suggested that the electrostatic and hydrophobic interactions are the dominant mechanisms determining MPs sorption capacity with soil minerals and organic matter respectively (Luo et al., 2020). With the increase in pH, the deprotonation of functional groups of soil particles increases, resulting in a higher electrostatic repulsion between plastic particles and the soil as compared to a more acidic pH (Luo et al., 2020). In addition, the detection of fine soil particles along with the formation of inorganic coating on the HDPE surface in our experiments is in agreement with Wang et al., 2023 study showing that plastic debris in soil can form plastic-rock complexes. It should be noted, that under our experimental conditions, we did not detect in the collected filtrates any significant changes in soil density or in soil aggregation in the presence or absence of HDPE using the CT scanning analysis (Figs. S5 and S6 and Table S5).

The findings presented in this study provide relevant insights about the interaction of soil minerals with HDPE surface and highlights the need to further understand the specific mechanisms of U accumulation in soil in the presence of HDPE. Pincus et al., 2023 documented that the complexation of inorganic coatings to plastics, such as aluminosilicates and ferrous oxides/oxyhydroxides, can considerably affect the sorption of toxic inorganic contaminants to plastics (Ashton et al., 2010; Violante and Pigna, 2002). The examination of HDPE-soil mixtures using electron microscopy did not reveal any information regarding U precipitation or interaction with the solid mixture. However, the quantitative measurement of aqueous U removal in this study demonstrated that HDPE-soil mixtures significantly changed the initial removal of U as compared to only soil particles. Additional microscopy and spectroscopy analyses were conducted to better understand the interaction of U with HDPE particles.

3.4. Uranium precipitation on HDPE surface at pH 5 and pH 7.5

U-bearing precipitates on the surface of reacted HDPE particles at both pH 5 and 7.5 were detected using SEM-EDS and by fitting of high-resolution XPS U 4f spectra. The examined HDPE particles were obtained from experiments with only added HDPE. SEM images (Figs. 3, 4 and Fig. S7) show U-bearing precipitates on the cracked surface of HDPE after 14 d of agitation in water, suggesting that the weathered sites on the MPs surface facilitated the nucleation of the U-bearing precipitates. This observation is consistent with previous studies showing that the weathered sites in MPs can act as a substrate for adsorption through electrostatic interaction or inner sphere complexation (Gao et al., 2021; Liu et al., 2022a). The EDS spectra of these precipitates contained U, Ca, Cl, and K (Fig. 3C, F, and Fig. S7). Titanium peak co-occurred in the same

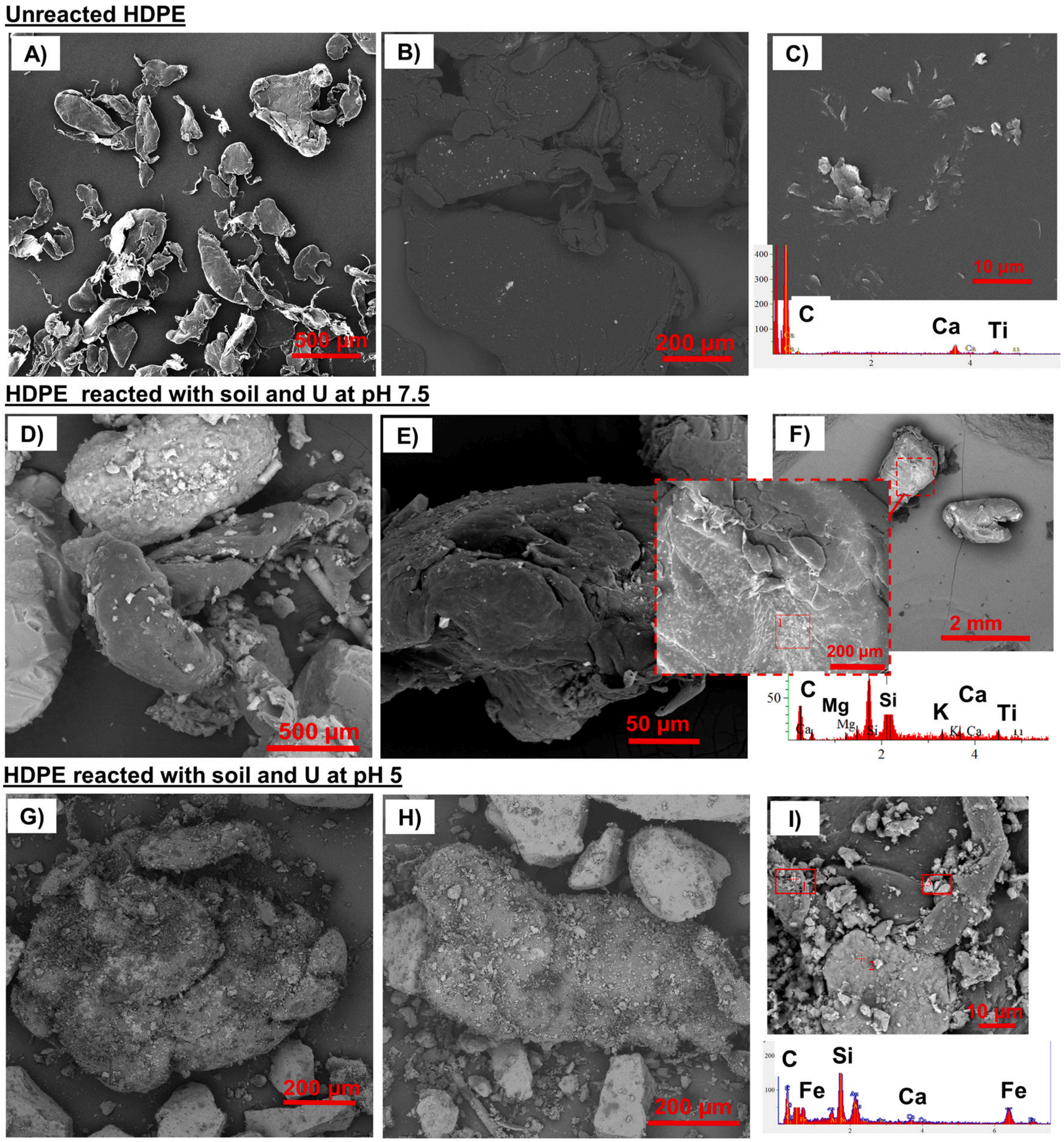

Fig. 2. SEM images and EDS spectra for unreacted HDPE (A-C) and the collected filtrate from batch experiments containing HDPE reacted with soil and aqueous U at pH 7.5 (D-F) and HDPE reacted with soil and aqueous U at pH 5 (G-I). All experiments were conducted in systems containing 100 μM initial U_i concentration, 1 mM HCO_3^- and 3 mM Ca.

area where U-bearing precipitates were detected at pH 7.5 (Fig. 4F). These elements were also identified on the surface of unreacted HDPE (Fig. 2A-C and Fig. S2) and may correspond to the chemical impurities in HDPE structure as reported in the literature. For example, $TiCl_x$ species are used as catalyst to modulate the grade of PE production (Piovano et al., 2017). The increase in the appearance of these elements in reacted HDPE agrees with previous studies reporting that weathering can expose inorganic additives as the surface properties of plastics are altered (Holmes et al., 2014; Liu et al., 2020). The EDS spectrum of the U—K bearing solid phases on HDPE surface and the X-Ray maps show principally U, K, and O (Fig. 4B—F).

The precipitation of U onto the surface of pristine MPs was observed at pH 7 in ultrapure water but not at acidic pH (Quiambao et al., 2023). In the present study, U precipitation occurred on the surface of HDPE at pH 7.5 despite the presence of bicarbonate in solution. The precipitation of U with Ca was previously reported in solution without bicarbonate at pH 7 (Gonzalez-Estrella et al., 2020; Laurette et al., 2012). Gonzalez-Estrella et al., 2020 observed a partial or complete dissolution of U precipitates at pH 7 in reactors containing 10 mM bicarbonate and Ca. This observation is in accordance with the present study, as the quantitative data shows that U mostly remained soluble in carbonate water in batch experiments with added HDPE (Fig. 1A, B). However, U precipitation on HDPE surface was still detected after 14 d of experiments, suggesting that the active weathered sites of HDPE facilitated the mechanisms of nucleation and formation of U-bearing precipitates. Such formation of U-plastic solids as an organic particulate can affect U toxicity and bioavailability in agricultural soil, as evidenced in our previous study (El Hayek et al., 2021). Additional analyses using XPS were conducted to examine the chemical associations of U to HDPE by measuring the signal of U 4f, C 1 s and O 1 s on the near-surface region of reacted and unreacted particles.

Results from the XPS survey scan revealed that 0.55 % and 0.43 % U 4f were accumulated in the reacted HDPE at pH 7.5 and pH 5 respectively, indicating the chemical association of U at the top 5–10 nm of the

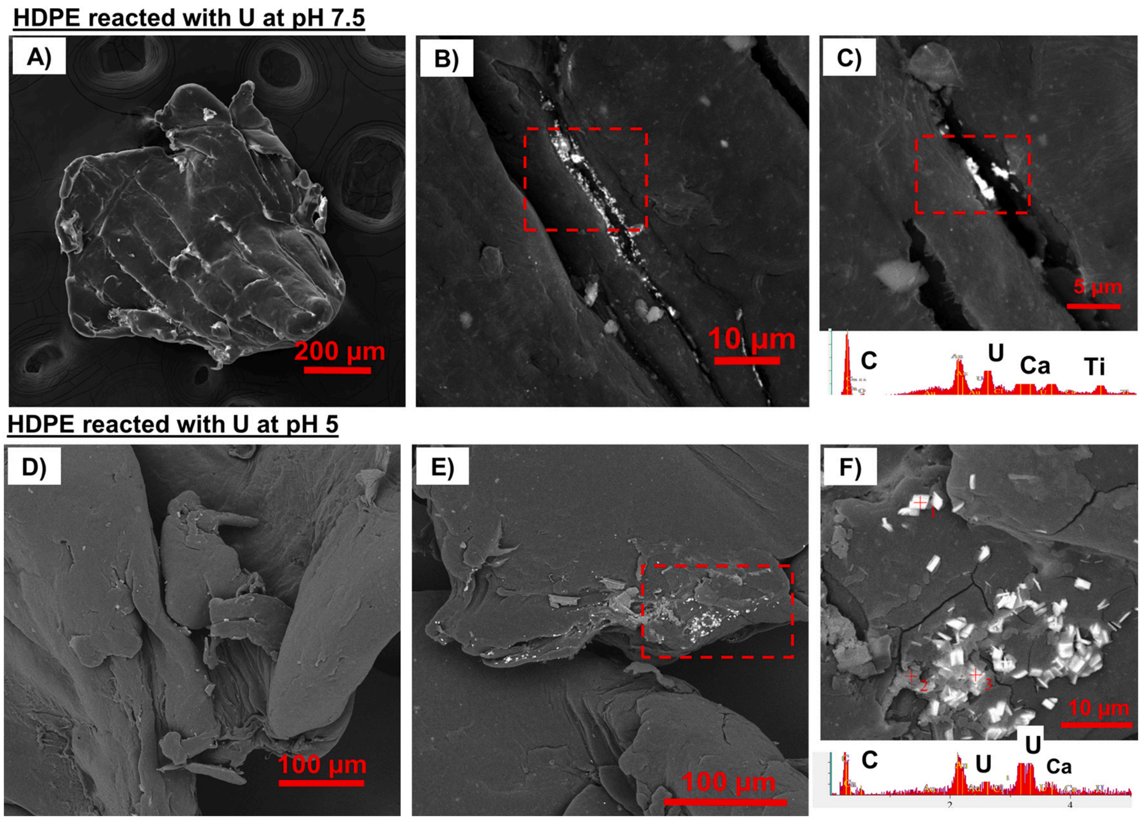

Fig. 3. SEM images and EDS spectra for the collected filtrate from batch experiments for HDPE reacted with aqueous U at pH 7.5 (A-C) and HDPE reacted with aqueous U at pH 5 (D-F). All experiments were conducted in systems containing 100 μM initial U_i concentration, 1 mM HCO_3^- and 3 mM Ca.

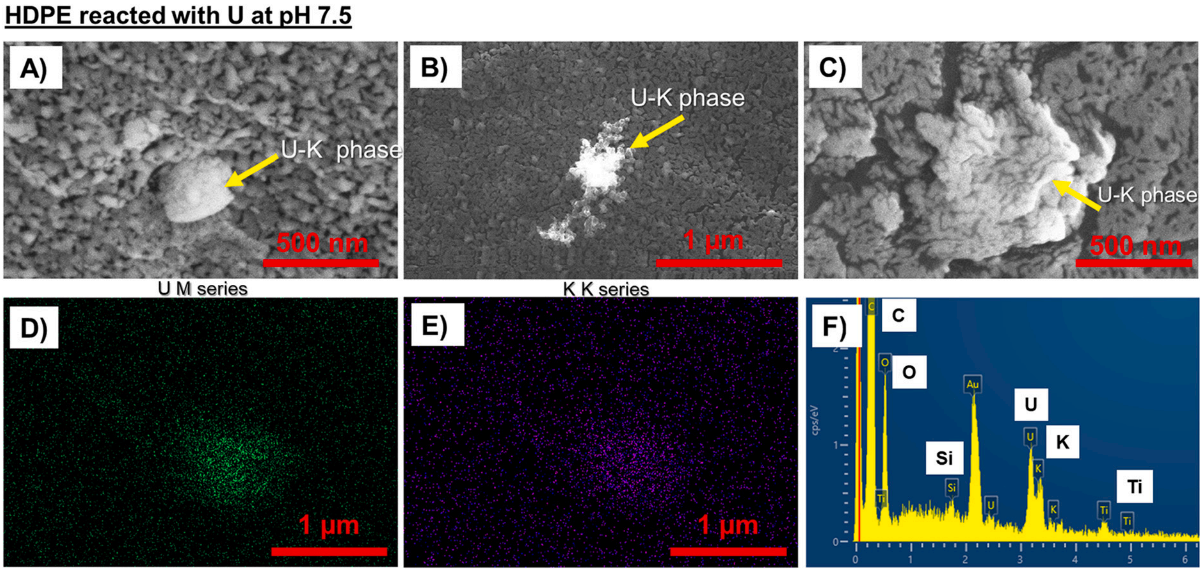

Fig. 4. Top row-Examples of U-bearing particles on the surfaces of HDPE at pH 7.5: U-K bearing solid phases (A-C). U-M X-ray map of particle shown in Figure C (D). K-Kα X-ray map of particle shown in Figure C (E). EDS X-ray spectrum of particle in Figure B (taken at different scale) showing the presence of U, K, and O (F). The Au peak is from the gold coating applied to the sample to prevent charging. All experiments were conducted in systems containing 100 μM initial U_i concentration, 1 mM HCO_3^- and 3 mM Ca.

near-surface region of HDPE (Fig. 5). An increase in the overall oxygen content was identified using XPS analysis for all reacted HDPE as a function of pH, with reacted HDPE at pH 5 having 1.2-fold higher oxygen content than those reacted at pH 7.5. In addition, at pH 5, the relative oxygen content increased 72 % at the near-surface region of reacted HDPE in the absence of U as compared to unreacted HDPE.

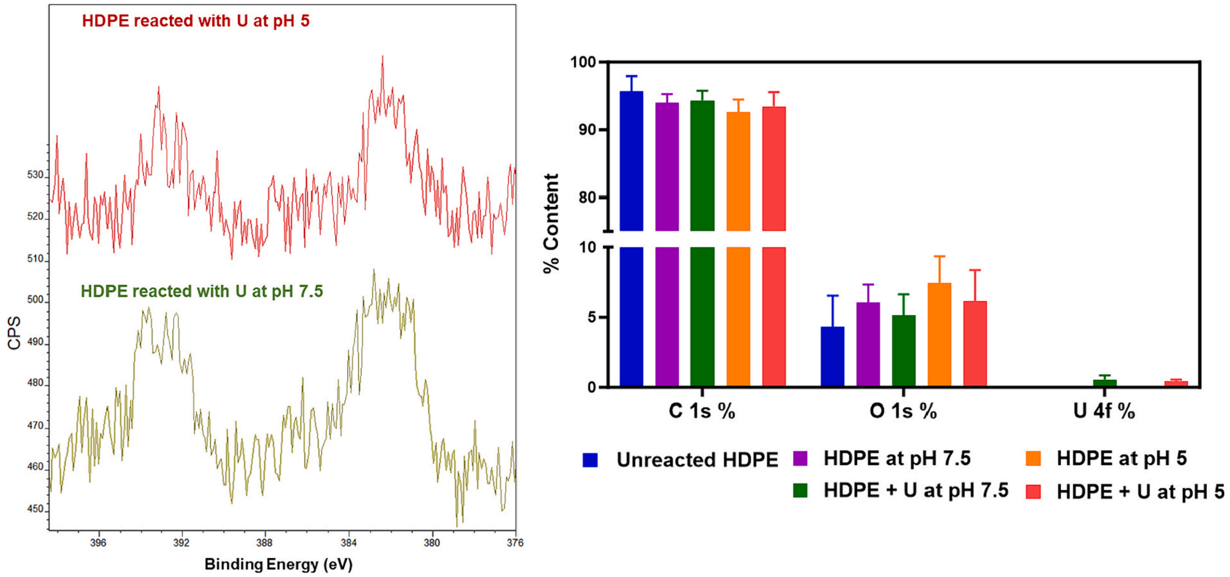

Fig. 5. U 4f high-resolution spectra for the reacted HDPE with aqueous U at both pH 7.5 and 5 with the percent composition of C 1s, O 1s and U 4f spectra. All experiments were conducted in systems containing 100 μM initial U_i concentration, 1 mM HCO_3 and 3 mM Ca.

However, when U was detected at the near surface, the relative oxygen content increased only 42 % as compared to unreacted HDPE. Similarly, at pH 7.5, the relative oxygen content increased 40 % in controls with no U while it increased 19 % when U was accumulated on the surface of HDPE. These results suggest that the two weeks agitation of HDPE in carbonate water augmented the level of oxygen-containing groups at the near-surface region of particles. This oxidation process was enhanced at pH 5 as compared to pH 7.5. The mechanical abrasion and agitation in water can lead to the formation of surface cracks and make the MPs surfaces gain oxygen, Si, and other impurities (Bullard et al., 2023; Song et al., 2017). Such a phenomenon explains the high occurrence of oxygen, Ti, and other elements near the U—K bearing solid phases. Previous studies reported that the presence of oxygen functional groups could serve as reactive surface sites for metals adsorption, enabling the nucleation and precipitation of metals (Gao et al., 2021). For instance, the sorption capacity of weathered MPs to metals (e.g., Cr, Co, nickel (Ni), Cu, Zn, Cd, Pb, and strontium (Sr)) was higher than the sorption of pristine particles due to the increased surface polarity and charges (Gao et al., 2021; Holmes et al., 2012; Johansen et al., 2019; Liu et al., 2020; Wang et al., 2020). In the present study, the decrease in the relative content of oxygen occurred when U level increased, suggesting an interaction of U with the oxygen polar groups on the surface of particles.

4. Conclusion

This work indicates that the surface chemistry of HDPE can mediate the rapid accumulation and precipitation of U on the near-surface region of HDPE particles in water containing Ca and carbonate at pH 5 and pH 7.5. These findings have implications in natural and engineered systems as well as in agricultural sites affected by anthropogenic activities, in which U and MPs coexist. A relevant result of our study is that HDPE modified the initial removal of aqueous U in agricultural soil depending on pH, particularly within the first hours of the experiments. Our study also confirms the weathering of HDPE surface after two weeks of agitation in water with the increase of surface cracks and oxygen content level as determined by SEM analysis and by XPS survey scan respectively. Given the persistent character of MPs in the environment, further research is needed to investigate the kinetics of surface adsorption and precipitation of U as well as the re-release conditions with the increase of the residence time, the content of organic matter, and the application of P fertilizers. Our study showed that the interaction of U and HDPE occurred at the natural pH range of soil rhizosphere, underlining the urgency to address the impact of U interaction with plastic polymer on their bioaccumulation and root-to-shoot translocation in crops and vegetation. Formation of U-plastic solids is a critical understudied exposure pathway that may enhance U bioavailability under the form of organic particulate. Future investigations should focus on a greater understanding of plastics on U toxicity and the associated health risks exposure.

CRediT authorship contribution statement

Casey Miller: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Andrew Neidhart:** Methodology, Investigation. **Kendra Hess:** Investigation. **Abdul-Mehdi S. Ali:** Resources. **Angelica Benavidez:** Resources, Investigation. **Michael Spilde:** Resources. **Eric Peterson:** Resources. **Adrian Brearley:** Resources, Funding acquisition. **Xuewen Wang:** Investigation. **B. Dulani Dhanapala:** Resources, Investigation. **José M. Cerrato:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Conceptualization. **Jorge Gonzalez-Estrella:** Writing – review & editing, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Eliane El Hayek:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Additional materials and methods, soil characterization (Table S1 and Fig. S4), experimental setup (Table S2), chemical equilibrium analysis for U speciation (Table S3), and CT scanning analysis details (Tables S4, S5 and Figs. S5, S6 and S8), FTIR, SEM images and size distribution of HDPE particles used in this study (Figs. S1-3), and SEM images and corresponding EDS spectra of U-bearing precipitates on HDPE surface (Fig. S7). Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.171834.

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