

Apportionment of Derivative Uranium from Phosphate Mineral Fertilizer in Agricultural Soil Components after Extended-Period uses

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Abstract

Long-term phosphorus (P) fertilizer application can lead to an accumulation of uranium (U) in the surface soil of agricultural fields. This study investigated the soil constituents that participate to the accumulation of U by using chemical extraction methods. Surface soil samples were obtained from paddy, wheat, barley and corn fields cultivated without any phosphate fertilizer (control site), with both NPK fertilizer and TSP for more than 18 years. In extension to the uranium total content in soil, the content of pyrophosphate and acid oxalate-extractable U were evaluation as a measurement of U related with soil total carbon and crystalline Fe/Al minerals in soil, respectively. The total, pyrophosphate-extractable, and acid oxalate-extractable U concentrations were higher in the soil obtained from fertilized sites than in that obtained from the control site. The difference in the U concentrations between phosphate fertilizer sites and the control sites corresponded with the increased U concentration observed after the application of phosphate fertilizer. In the paddy, wheat and barley field soil with different long application of phosphate fertilizers 57, 42 and 18 year, the increase in pyrophosphate-extractable U was 76.1, 72.9 and 85% of that in U_t respectively, but it was equivalent to the increase than in U_t at the corn fields soil with 34 years period added phosphate fertilizers. Moreover, the range of increase in acid oxalate-extractable U was 21.1–56.3% of that in U_t in the fields soil, In conclusion, most of the phosphate fertilizer-derived U was either incorporated into the soil organic matter or poorly crystalline Fe/Al minerals in the surface soil of agricultural fields. Therefore, soil total organic carbon was an aggregation of U in barley field and corn soil, whereas poorly crystalline Fe/Al minerals were important pools of U in wheat field and paddy soil experiencing alternating changes in redox conditions.

Keywords: Uranium; Pyrophosphate-Extractable; Fertilizer; Agricultural Fields.

1. Introduction

Radionuclides originate from uranium (U) exist in soils because of nuclear fuel production, nuclear weapons production, depleted uranium in ammunitions, coal combustion and chemical fertilizers (Morton et al., 2002; Gervais et al., 2006). The U in chemical fertilizers is linked to the high concentration in phosphate rock (Taylor, 2007, Sun et al., 2020a). Phosphorus (P)-based fertilizers have been widely used in Iraq as the application of plant nutrients. The application of phosphate fertilizers, has increased substantially due to the expanding cultivation of different crops (FAO, 1995) and also to the low phosphorus availability. Low phosphorus availability because almost the agriculture field soil in Iraq is calcareous soil (Andosols, Entisols) containing large amounts of amorphous minerals such as calcic or petrocalcic, gypsic within 50 cm of the surface soil (Muhameed et al., 2018).

On the basis of the U concentration in phosphate fertilizers, McBride and Spiers (2001) estimated that 50 years of the application of a specific phosphate fertilizer (100 kg ha⁻¹ year⁻¹ as P₂O₅) would lead to the addition of 2.4 kg of U per hectare to the topsoil, corresponding to an increase of about 1 mg kg⁻¹ in the soil U. In a paddy field in Japan, the increase in soil U due to the application of calcium superphosphate fertilizer (600 kg ha⁻¹ year⁻¹ as mixed fertilizer) for 10 years was estimated to be 5.3% of the total U in the soil (Tsumura, 1993). The results of the study conducted by Al-Saadie et al. (2018) indicated that the percentage of the cumulative increase in the isotope of uranium ²³⁸U resulted from repeated addition of phosphate fertilizers (NPK, TSP and SSP) amounted to 1.62% for 7-25 years of the application. The amount of U introduced into the soil due to the application of phosphate fertilizers during a single cultivation period is relatively low as compared to that naturally present in the soil. Many researchers have noted that although continuous application of phosphate fertilizers causes an

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increase in the U concentration in agricultural soil, the concentration rarely exceeds the naturally occurring U concentration (Wetterlind et al., 2012; Diederik et al., 2008). The study of Takeda et al. (2006) showed that the U concentrations in the soils of fields with continuous fertilizer applications and in contiguous uncultivated soils were determined that the surface soils in the experimental fields with fertilizer applications contained higher amounts of U compared with the uncultivated surface soils, the amount of high U in the soil was estimated by the vertical profile of U concentration, and a rise of about 200 mg m⁻² of U was found in the soils at 0–35 cm depth during a 61 year cultivation period. The evaluated value was nearly the same as the amount of U added through the fertilizers as calculated from U concentrations in the utilized fertilizers. Therefore, almost all the U from the fertilizers would yet remain in the upper part of the soils. Chemical extraction results suggested that organic carbon substances and non-crystalline clay minerals in the surface soil have played an essential part for accumulation of U derived from the fertilizers, despite the reality that the U concentration in the phosphate fertilizer used by some researcher was moderately lower than that in the fertilizer used by others, however, its accumulation in the soil was relatively more. So, it is apparent that soil properties are a substantial factor for define the accumulation rate of U in soil in addition to the amount of U anthropogenically added to soil. The main U species that exists under an oxidative environment is divalent uranyl ion (UO₂²⁺). This positively charged UO₂²⁺ is adsorbed on the negatively charged sites of soil components, and these sites increase with soil pH and clay content as well as clay mineral type. The U adsorption capacity of soil, thus, increases with soil pH (Ajay Kumar et al., 2013). However, when carbonate concentration increased with increase in pH, U became mobile in soil because of the formation of a soluble and negatively charged carbonate-U complex. UO₂²⁺ is also sorbed on Fe / Al sesquioxides (Aide, 2018). Fuller et al. (2020) demonstrated the relevance of U complexation with humic substances, which can influence U mobility and the solid-liquid distribution coefficient (K_d) values. Thus, K_d values are subject to uncertainties that may compromise disposal risk assessments if site-specific factors are not considered. This indicated that agricultural practices have significant effects on the mobility of U in soil. In Iraq, these agricultural practices affect the destiny of soil U added due to the intensive use of phosphate fertilizers. The amount of soil organic matter and the concentration of Fe oxide are possibly the key factors for controlling the accumulation of U in soil. The K_d of U is significantly correlated with organic matter and amorphous Fe contents of soil (Johansson, 2020). In order to identify the soil components that are the most important pools of phosphate fertilizer-derived U, comparison of U speciation in agricultural soil with and without phosphate fertilization is required by maintaining the same cultivation conditions except for phosphate fertilization. In this study, we compared the amounts of U

in different types of soil and operationally defined U speciation in the soil collected from paddy, wheat, barley and corn fields, cultivated with and without the application of phosphate fertilizers. The concentrations of U extracted by pyrophosphate and acid oxalate reagents were determined as a measure of U associated with soil organic carbon and Fe/Al sesquioxides, respectively. The aim of the study was to inspect the main soil components that act as pools of phosphate fertilizer-derived U and assessed the effects of various cultivation practices on the distribution of U in soil. The concentration and chemical speciation of U in agricultural soil will provide important information when considering the potential threat of agricultural food-derived U to human health.

2. Materials and methods

2.1. Soil samples

Ten to fifteen surface soil samples were taken from long-term (18 to 57 year) fertilized fields. The fields included 4 location; location1 was periodically submerged paddy fields; location2 and 3 were fields cultivated with wheat and barley; and location 4 was field cultivated with corn. The soil types, cultivation practices, cultivated crops, fertilization planned, and pertinent soil characteristics were listed in Table 1. To annotate the effects of the implementation of phosphate fertilizer on the accumulation of fertilizer-derived U in soil, fields under 3-4 different phosphate fertilizer regimes were selected and without phosphate fertilizer (control site). Different various types of commonly phosphate fertilizers were used on each site applied with superphosphate (TSP), single super phosphate (SSP), compound fertilizers of phosphate, nitrogen, and potassium (NPK) and ammonia phosphate fertilizers (DAP). Soil samples were selected from every plot and mixed to represent a representative sample. Each treatment had 4 replicates. Soil samples were carried out from each of these plots with a 20 cm extended cylindrical auger, with an internal diameter of 5 cm, all the soil from the experimental fields were air dried and passed through a 2-mm sieve and stored in plastic bottles until analyses.

2.2. Uranium analysis

Total pyrophosphate-extractable, and acid oxalate-extractable Uranium concentrations in the soil taken from unfertilized and fertilized fields were specified. Sequential chemical extraction has been applied by many researchers to assess the amounts of elements related with particular soil components. However, it has been pointed out that sequential extraction may cause redistribution of U in soil (Zhou and Baohua, 2005). For that, U associated with soil organic matter and Fe/Al oxides separately extracted to minimize the probable synthetic effects exerted on U distribution during the extraction processes. To evaluate the total concentration

of U (U_t) in soil, 0.5 g of air dried soil was digested with a mixture of HNO_3 , $HClO_4$, and HF in a Teflon beaker with heating at 393 K. Three analytical replicates were carried

out for each soil sample. After digestion, the acid-digested solution was evaporated to near dryness and then adjusted to 50 ml with 0.16 mol l^{-1} of HNO_3 .

Table 1. Properties of soil and field description

Location No. / Soil class	Experimental period and Cultivated crop	Fertilizer regime (kg ha ⁻¹ year ⁻¹)			Acid oxalate- extractable		Sodium pyrophosphate- extractable	
			TC	TN	Fe _o	Al _o	Al _p	U _t
			g kg ⁻¹					mg kg ⁻¹
1/Gleysols	57 Paddy rice	Unfertilized	4.21	0.19	19.7	2.83	0.39	1.56
		Fertilized (SSP, TSP, NPK, DAP) N:20+6 P2O5	2.06	0.3	16.9	2.27	0.77	3.41
2/ Gleysols	42 Wheat	Unfertilized	3.94	0.25	11.5	3.27	0.74	1.47
		Fertilized (TSP, NPK, DAP) N:17+10 P2O5	1.3	0.38	10	3.81	1.19	2.81
3/ Fluvisols	18 Barley	Unfertilized	4.01	0.2	8.41	2.55	0.66	1.38
		Fertilized (TSP, NPK, DAP) N:25+14 P2O5	7.65	0.47	8.8	2.19	0.75	1.9
4/ Andosols	34 Corn	Unfertilized	6.82	0.69	16.4	3.47	1.83	1.02
		Fertilized (TSP, NPK, DAP) N:28+20 P2O5	6.21	1.08	13.8	3.18	1.9	2.09

Pyrophosphate-extractable U (U_p) was extracted from 0.5 g of soil by adding 10 ml of 0.1 mol l^{-1} sodium pyrophosphate with the pH adjusted to 10. After the suspension was shaken for 24 h, it was centrifuged at 4000 g for 10 min, and the supernatant were filtered with paper filters (grade 14/N; Munktell & Filtrak GmbH, Bärenstein, GER).

Acid oxalate-extractable U (U_o) was extracted from 0.5 g of soil by adding 10 ml of a mixed solution containing 0.1 mol l^{-1} oxalic acid and 0.175 mol l^{-1} ammonium oxalate (pH 3.3) in a hot water bath at 353 K. After 1 h, the suspension was centrifuged at 4000 g, and the supernatant was filtered with paper filters. The uranium concentrations were determined using an Inductively Coupled Plasma Atomic Emission Spectrometer (ICPE-9000). External calibration standard was used and the accuracy of the analyzed data was confirmed by the standard addition method with U concentrations of 1, 3, 5 and 7 mg l^{-1} added to selected samples of (U_t , U_p and U_o extracts).

3. Results and discussion

3.1. Total U concentration (U_t)

The total U in soil taken from the control site (without phosphate fertilization) ranged from 1.02 to 1.56 mg kg^{-1} . The U_t in soil obtained from fields with long-term application of phosphate fertilization was clearly higher (1.90–3.41 mg kg^{-1}) than that in the control site soil (Table 1). The increase in U_t (ΔU_t) due to the application of the phosphate fertilizer was calculated by subtracting the U_t in the control site soil from that of the fertilized site soil. The ΔU_t was the lowest at location 3 with a value of 0.52 mg kg^{-1} and the highest at location 1 with a value of 1.85 mg kg^{-1} . Long-period phosphorus (P) fertilizer application can affect to an accumulation of (U) in agricultural soil, as reported previously (Eriksson et al.,

2015; Al-Saadie et al., 2016; Sun et al., 2020b). The results of this research indicated that the U as an impurity derived from the phosphate fertilizer remain in the surface soil of the fields of study. The U_t at all location, including phosphate fertilizers application, was remarkably high despite the fact that the U_t at the control site was the lowest among all the locations investigated in this study. On the other hand, the results of this study indicated that the long period of addition of phosphate chemical fertilizers of various kinds led to increase in the concentration of total uranium in fields of soil with extended use of fertilizers. It was found that the U_t concentration at location 1 when fertilizer was added for a period of 57 years, the concentration of uranium (3.41 mg kg^{-1}) was higher than the rest of the sites where the fertilizer was used for fewer years. The lowest U_t concentration was 1.90 mg kg^{-1} at location 3 when phosphate fertilizers added for 18 years. Depending on the periods of fertilizer application, ΔU_t at location 1 relative to location 2,3 and 4 reached 0.60, 1.51 and 1.32 mg U kg^{-1} when adding phosphate fertilizer for a period of 42, 18 and 34 years, respectively. respectively. Al-Mousawi et al. (2020) also observed ^{238}U , ^{40}K and ^{232}Th accumulation in soil cultivated with crops.

3.2. Pyrophosphate-extractable U concentration (U_p)

Uranium dissoluble by pyrophosphate (U_p) in soil without phosphate fertilization was minimal than that in soil with phosphate fertilization (Fig. 1). The U_p in the control site soil ranged from 0.482 to 0.864 mg kg^{-1} , while, it was ranged from 0.950–2.72 mg kg^{-1} in the soil agricultural fields fertilized with different long-term phosphate fertilizers application (18, 34, 42 and 57 years). ΔU_p calculated by subtracting the U_p in the control field soil from that in the fertilized field soil with different phosphate fertilizers. The ΔU_p was the lowest in the fertilized field soil at location 3, with 0.443 mg kg^{-1} (18

years), and the highest value was in the field soil at location 1 (57 years) which reached 1.86 mg kg⁻¹.

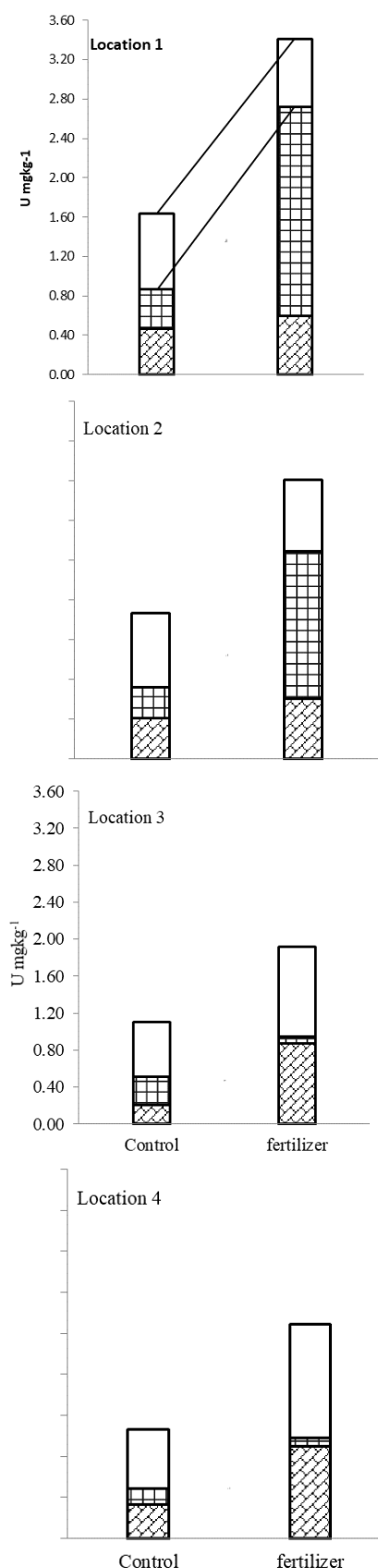


Figure 1. Total (U_t) pyrophosphate extractable (U_p) and oxalate extractable (U_o) concentration in the long-term fertility and unfertility experimental fields

The correlation between the U_t and U_p in the soil obtained from all fields investigated is shown in Figure 2. The U_t was linearly correlated with U_p in each location. Although the number of data sets was limited ($n=8$), linear relationships were also observed between the U_t and U_p at sites 1,2,3 and 4 ($R^2 = 0.971, 0.968, 0.959$ and 0.975 respectively). This suggested that U associated with soil components that were dissolved by the pyrophosphate solution and which it is a part of U associated with Fe/Al complexed with humic substances, actively contributes to an increase in U_t due to the implementation of the phosphate fertilizer.

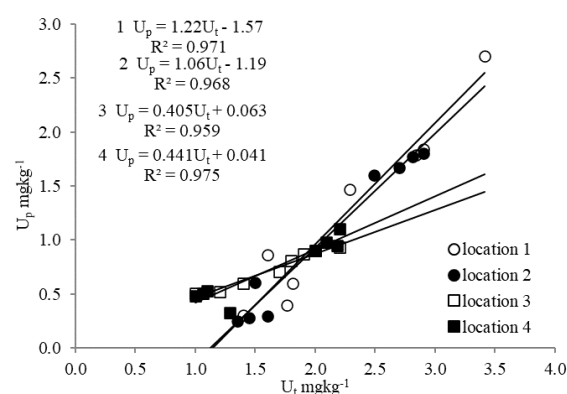


Figure 2. Correlation linear between U_t concentration and U_p influenced by application of phosphate fertilizer

The proportion of U_p to U_t in the control site soil (U_p/U_t) were .0527, 0.492, 0.461, and 0.455 at location 1,2,3 and 4 respectively. The (U_p/U_t) in the control site soil was significantly correlated with the TC content ($R^2 = 0.908, n=8$) and the concentration of pyrophosphate-extractable Al (Alp) ($R^2 = 0.920, n=8$) (Table 1). The significant relationship between the (U_p/U_t) and TC content in the control site soil showed that soil organic matter contributed to the retention of naturally occurring U in the soil. The binding nature of ^{238}U , ^{210}Po and ^{210}Pb was correlated for organic matter and Alp corresponds to the Al associated with soil humic substances has been experimentally proven by Al-Masri, et al. (2019). The (U_p/U_t) ratio in the fertilized site soil was higher than that in the control site soil and reached 0.271, 0.252, 0.034 and 0.014 at location 1,2,3 and 4 respectively. To evaluate the contribution of ΔU_p to ΔU_t , the ratio of ΔU_p to ΔU_t (R_p) was calculated using the following equation:

$$\frac{\Delta U_p}{\Delta U_t} = \frac{U_p(\text{fertilizer}) - U_p(\text{control})}{U_t(\text{fertilizer}) - U_t(\text{control})}$$

The R_p values are shown in Figure 2 by the numbers on the bar graph. R_p was nearly 1 for rice, wheat soils at site 1 and 2. This indicated that increasing in concentration of U_t was equivalent to increase in U_p , in other words, most of the fertilizer-derived U was stored in an pyrophosphate-extractable fraction in site soils. R_p where not exceeded 0.5 for the barley and corn fields (site 3 and 4 respectively), this indicated that the most of the fertilizer-derived U was stored in the acid oxalate-

extractable fraction and the acid oxalate-extractable soil components was a more important pool of U. Without the addition of U by the use of the phosphate fertilizer, U_p may be apparently decreased in the control site soils, showing that the application of addition phosphate fertilizer did always cause larger amounts of U accumulation than those observed in the fields without application fertilizers.

3.3. Acid oxalate-extractable U concentration (U_o)

The U_o in the control soil ranged from 0.205 to 0.464 at a rate of 0.353 mg kg⁻¹ and in the fertilized site soils ranged from 0.600 to 0.901 at a rate of 0.745 mg kg⁻¹. These results indicated that the U_o in the soil with phosphate fertilization were higher than that in the soil without phosphate fertilization by percentage increase amount 52.6%, while the percentage of U_p increase amount 61.9% (Fig. 1), This suggested that most of the uranium bound in the soil parts associated to pyrophosphate-extractable fraction. The increase in U_o (ΔU_o) in the fertilization field soil than in the control soil was calculated in the similar manner as for ΔU_p . The ΔU_o was the lowest in the rice and wheat fields which reached 0.136 and 0.199 mg kg⁻¹ respectively, and the highest values were in the barley and corn field which reached 0.664 and 0.570 mg kg⁻¹ respectively.

The correlation between the U_t and U_o in the soil obtained from all fields was less significant than U_p , the U_t was linearly correlated with U_o in each location, Although the number of data sets was limited (n=8), linear relationships were also observed between the U_t and U_o at location 1,2,3 and 4 ($R^2 = 0.954, 0.951, 0.952$ and 0.950 respectively; Figure 3). This suggested that the soil components dissolved by acid oxalate reagents, poorly crystalline Fe hydroxides minerals, influenced the U_t in the soil.

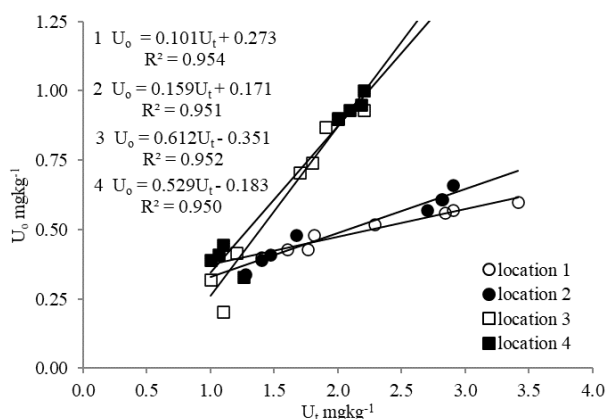


Figure 3. Correlation linear between U_t concentration and U_o influenced by application of phosphate fertilizer

Zhang et al.(2018) found that the U bind preferentially to FeO site than to AlO. So, it was suggested that poorly crystal Fe minerals played a more substantial role than Al minerals in retaining naturally occurring U in soil. The

ratio of ΔU_o to ΔU_t (R_o) was calculated in the same manner as R_p . The numbers on the graph bars in Fig. 1 represent the calculated values of R_o , were not exceeded 0.2 for the 1 and 2 sites, and were not exceeded 0.8 for 3 and 4 sites, this indicated that the most of the fertilizer-derived U was stored in the acid oxalate-extractable fraction and the acid oxalate-extractable soil components was a more important pool of U.

By comparing the soil samples collected from long and frequent use of phosphate fertilizers in fields soil with and without the application of the phosphate fertilizer, we indicated that most of the fertilizer-derived U in soil was stored in the soil components that were dissoluble by either pyrophosphate or acid oxalate reagents. A part of the accumulated U associated with Fe/Al complexed with humic substances which dissolved by both pyrophosphate and acid oxalate reagents, U_o was lower than U_p .

Conclusions

Using of mineral phosphate fertilizers frequently and for long periods increased the concentration of U in agricultural soils in Iraq. The uranium associated with phosphate fertilizers as impurities was either incorporated into soil organic matter, adsorbed, or precipitated with poorly crystal Fe/Al-bearing minerals in agricultural surface soils. Soil organic matter appeared to be a more important pool of U in the agriculture fields soil, whereas poorly crystalline Fe minerals were more important U pools in the paddy field soil that underwent alternating changes in redox conditions. Soil organic matter and Fe/Al minerals prevented fertilizer derived U from moving out to depth agricultural fields soil, thus causing the accumulation of U in the soil surface.

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