

Accumulation of uranium in soils from impurities in phosphate fertilisers¹

Matthew D. Taylor²

Abstract

Uranium (U) accumulation in four New Zealand soil types was assessed by comparing samples collected and archived in 1992 with soil samples collected and archived 36–43 years previously, from the same four sites. Mean levels of total U increased by $1.30 \pm 0.03 \mu\text{g g}^{-1}$ for soils sampled about 40 years ago compared with 1992 soil samples, an annual increase of $0.033 \pm 0.008 \mu\text{g g}^{-1} \text{yr}^{-1}$. These increases are probably associated with phosphate fertiliser application. All, or nearly all the U applied in fertiliser appeared to remain in the soil and so did not significantly leach to groundwater nor was taken up in significant amounts by plants. Levels of U in 1992 fertilisers in New Zealand are lower than those previously reported as manufacturers are sourcing raw phosphate rock containing low levels of U. It is calculated the present rate of U accumulation in soil has dropped to a quarter of that over the last 40 years.

Keywords: uranium accumulation in soil, phosphate fertiliser, lime, agricultural soils, volcanic soils, New Zealand

Zusammenfassung

Akkumulation von Uran in Böden durch Verunreinigungen aus Phosphatdüngern

In der Studie wurde die Veränderung der Gesamtgehalte an Uran (U) in vier Bodentypen Neuseelands über einen Zeitraum von 36 - 42 Jahren (Archivproben von 1944 - 1950 bis 1992 Neubeprobungen an den gleichen Punkten) untersucht. Die mittleren U Gehalte nahmen über einen mittleren Zeitraum von 40 Jahren hinweg um $1,30 \pm 0.03 \mu\text{g g}^{-1}$ zu, was einer jährlichen Zunahme von $0.033 \pm 0.008 \mu\text{g g}^{-1} \text{a}^{-1}$ entspricht. Dieser Anstieg wird auf U Verbindungen in Phosphatdüngern zurückgeführt. Alles, oder fast alles mit Düngemitteln zugeführte U scheint im Boden verblieben zu sein, so dass anzunehmen ist, dass nur unbedeutende Mengen ausgewaschen, oder von Pflanzen aufgenommen wurden. Die U Gehalte neuseeländischer Dünger lagen 1992 niedriger als 40 Jahre früher, was auf das Bemühen der Düngemittelhersteller Rohphosphate mit niedrigen U Gehalten zu verwenden zurückzuführen ist. Hierdurch ist die U-Akkumulationsrate im Vergleich zu den letzten 40 Jahre schätzungsweise auf ein Viertel zurückgegangen.

Schlüsselwörter: Urananreicherung im Boden, Phosphatdünger, Kalk, landwirtschaftliche Böden, vulkanische Böden, Neuseeland

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² Environment Waikato, PO Box 4010, Hamilton East, Hamilton, New Zealand; Email: matthew.taylor@ew.govt.nz

1 Introduction

Intensive pastoral farming in New Zealand requires large inputs of phosphate fertilisers and lime. These fertilisers contain U impurities (Syers et al., 1986) due to the substitution of U^{4+} for Ca^{2+} within the mineral structure of apatite (Burnett and Veeh 1977). Phosphate fertilisers sold in New Zealand from 1950 to 1980 were made from ancient guano deposits mined from Nauru and Christmas Island (Trueman 1965). The concentration of U in these deposits ranged from 31 - 56 mg U kg⁻¹ for Christmas Island and from 64 - 121 mg U kg⁻¹ for Nauru Island (Williams, 1974; Syers et al., 1986). U impurities may accumulate in the soil (Rothbaum et al., 1979) where they can be taken up by plants and transferred into the food chain, or be leached into ground and surface water (Jones, 1992). Barisic et al. (1992) suggest more than 20 % of fertiliser applied U annually deposited to the soil is leached to waterways as elevated U concentrations, up to 0.5 mg L⁻¹, in surface waters have been reported in Eastern Slovenia and Mexico (Barisic et al., 1992; Spalding and Sackett, 1972). In a study of U concentrations in the Morrow Plot soils over 85 years, Jones (1992) found U increased (presumably from lime or phosphate fertiliser) then declined, implying transfer to crops or leaching through soils to groundwater. In a separate study, Hamamo et al. (1995) compared a Morrow Plot fertilised and unfertilised soil and a farm soil with a cemetery soil. In agreement with Jones (1992), no differences, either accumulation or decline in soil U concentrations outside experimental error were observed, implying the U added in the fertiliser was lost into groundwater. On the other hand, Rothbaum et al. (1979) found most of the uranium applied in superphosphate to have accumulated in topsoils from Rothamsted Experimental Station, United Kingdom, and Papatoetoe, New Zealand.

Accumulation of U is of concern to regulatory authorities e.g. The United States Nuclear Regulatory Commission has established a residual contamination criterion for natural uranium in soil of 10 pCi g⁻¹, equivalent to 30 µg g⁻¹ (USNRC 1992).

Phosphate fertiliser associated cadmium (Cd), found in similar quantities to U, had increased over time in New Zealand soils (Taylor, 1997a), so it was expected U would show similar increases if it was accumulating. This study purposed to assess if there is an association between soil U levels and phosphate fertiliser, if U had accumulated in New Zealand soils and the rate of accumulation by comparing U concentrations in selected New Zealand soil samples collected and archived in 1992 with those collected and archived 36 - 43 years previously, from the same sites.

2 Materials and Methods

2.1 Collection of Samples

Previously, soils had been collected and analysed to assess the accumulation of cadmium in New Zealand soils (Taylor, 1997a). Four of these soils receiving different quantities of phosphate fertiliser were chosen to assess if U was accumulating from the addition of impurities in phosphate fertiliser (Table 1).

Table 1:
Soils and fertiliser regime

Series Name	Location	Sample Depth (cm)	Super-phosphate Fertiliser Regime (kg ha ⁻¹)	1992 soil pH
Mikimiki Stoney Silt Loam	Masterton	0 - 7.5	300 (moderate)	5.4
Eltham Peat	Eltham	0 - 15	600 (high)	5.4
Taupo Sandy Silt	Waikite Valley	0 - 7.5	100 (low)	5.4
Hinemaia Sand	Reporoa	0 - 7.5	500 (high)	5.9

The amount of fertiliser applied to each site was estimated after interviews with the farmer, although accurate records of fertiliser history were not kept. For the Taupo site, investigations showed the current farmer had owned the property less than a year and the farm had previously been „let go“ by the former owner (Mr Boolean [farmer] pers com). However, 1000 kg ha⁻¹ had been applied just before sampling. It is likely the amount of fertiliser applied to the Taupo soil is much less than that applied to the other three soils over the previous 40 years. Assuming this site had fertiliser applied at high rates for initial break in of the land (700 kg ha⁻¹) and fertiliser application at levels typical of the region for 4 years (2000 kg ha⁻¹) and taking into account the quantity of fertiliser applied by the current owner (1000 kg ha⁻¹, Mr Boolean personal com), an average value of 100 kg ha⁻¹ a⁻¹ was derived. Archived soils had been collected 36 - 43 years ago, either from pits or composites of core samples, and were from 0 - 15 or 0 - 7.5 cm depth. At the time of collection, the archived samples were air-dried, then hand ground to 2 mm, using an agate mortar and pestle. Ground soil was placed in glass jars and sealed with a screw-top cap. The jars were stored in a cool, dark room.

Soil samples collected and archived in 1992 consisted of 20 cores, 2.5 cm diameter, the same depth as the corresponding archived sample (0 - 7.5 cm or 0 - 15 cm), from an area of 400 m². Although it was possible to return to

the same field where the original soil sample was taken (as shown by Landcare Research records) it was impossible to identify the exact site. The soil samples were air dried, passed through a 2-mm plastic sieve, and then stored in glass jars in a cool, dark room until analysis.

Lime and fertiliser samples were bought from garden supply shops. The phosphate fertilisers consisted of 4 single superphosphates and 1 triple superphosphate. All were manufactured in New Zealand by local fertiliser companies. Various companies obtained their raw phosphate rock from different countries but were unable or unwilling to identify the source of the sample supplied. Samples were mixed thoroughly and a 50-g subsample was taken. This was fine ground in an agate mortar and pestle and stored in a 75-mL plastic specimen container until analysis.

2.2 Extraction of limes and easy to dissolve fertilisers

Lime samples and some superphosphate fertiliser (air dried soil < 2 mm) samples were totally dissolved in either HNO₃ or aqua regia (Taylor, 1997b).

2.3 Extraction of soil samples and difficult to dissolve fertilisers

Superphosphate fertiliser samples not dissolved in *aqua regia* and soil samples (air dried soil < 2 mm) required rigorous extraction with concentrated HNO₃/H₂O₂/HF/HClO₄. ²³²U in 1 mol L⁻¹ nitric acid solution was added as an internal standard to all samples (Taylor, 1997b). The extract was purified using solvent extraction and U concentration was determined by alpha spectrometry using a

Tennelec TC 256 alpha spectrometer interfaced with a 286 IBM compatible computer equipped with a PCA card and Nucleus Personal Computer Analyzer Software. Results were corrected for tailing, for background, for losses in the purification process (using ²³²U), and for soil moisture (Taylor, 1997b).

Total P and Cd had both been measured in a previous study (Taylor, 1997a).

3 Results and Discussion

3.1 U in Soil

Concentrations of total U ranged from 0.62 to 2.34 $\mu\text{g g}^{-1}$ for archived soil samples, and from 1.69 to 3.54 $\mu\text{g g}^{-1}$ for the present-day soil samples (Table 2.). Total U levels for both archived and soil samples collected and archived in 1992 are within the range reported previously in soils (0.79 - 11.0, Kabata-Pendias and Pendias 1984). The range of values in the archived soils in this study are due to differences in U concentrations in soil parent material, while those values for present-day samples are due to a combination of differences in U concentrations in soil parent material and differing inputs of U in phosphate fertiliser. Levels of total U in all samples were below the United States Nuclear Regulatory Commission residual contamination criteria of 30 $\mu\text{g g}^{-1}$ (USNRC 1992).

All soil samples in this study accumulated U over time (Table 2). The largest increase in total U concentration was in the Eltham Peat (from 0.79 to 2.48 $\mu\text{g g}^{-1}$, an annual increase of 0.046 $\mu\text{g g}^{-1} \text{ a}^{-1}$), reflecting the high inputs of superphosphate fertiliser (600 kg ha⁻¹ a⁻¹) and the low bulk

Table 2:

Comparison of archived and present-day soil samples

Sample ¹	Date Collected	Classification Soil Taxonomy ² (New Zealand Soil Classification ³)	Total P ($\mu\text{g g}^{-1}$)	Total Cd ($\mu\text{g g}^{-1}$)	Total U ($\mu\text{g g}^{-1}$)	²³⁴ U/ ²³⁸ U activity ratio
Mikimiki (SB 5369)	1949	Aquic Dystrudept (Mottled Orthic Brown Soil)	562	0.54	0.62	1.2
Mikimiki	1992	Aquic Dystrudept (Mottled Orthic Brown Soil)	859	0.74	1.69	1.0
Eltham (SB 6749)	1955	Typic Medihemist (Humic Organic Soil)	467	0.19	0.79	1.0
Eltham	1992	Typic Medihemist (Humic Organic Soil)	1378	4.26	2.48	0.9
Taupo (SB 6845)	1955	Typic Udivitrant (Typic Orthic Pumice Soil)	799	0.46	2.34	1.0
Taupo	1992	Typic Udivitrant (Typic Orthic Pumice Soil)	1722	2.18	2.89	0.8
Hinemaiaia (SB6826)	1956	Aquic Udivitrant (Mottled Orthic Pumice Soil)	308	0.10	1.85	1.0
Hinemaiaia	1992	Aquic Udivitrant (Mottled Orthic Pumice Soil)	2483	0.51	3.54	1.0

¹ Sample numbers prefixed by SB from Soils Database, Landcare Research; ² Soil Survey Staff (1999); ³ Hewitt (1992)

density of the peat. U is strongly sorbed by peat under reducing conditions (Edwards et al., 1995; Amrhein et al., 1993; Kabata-Pendias and Pendias, 1984) but even under drained conditions the peat appears to sorb the U applied in fertiliser.

The 1992 sample of the Hinemaia soil had the highest concentration of total U of the soil samples tested ($3.54 \mu\text{g g}^{-1}$) and the total U increase (1.85 to $3.54 \mu\text{g g}^{-1}$, an annual increase of $0.047 \mu\text{g g}^{-1} \text{a}^{-1}$) was similar to the Eltham Peat, reflecting the high inputs of superphosphate fertiliser ($500 \text{ kg ha}^{-1} \text{a}^{-1}$). The presence of mottles in this soil indicates imperfect drainage leading to intermittent reduction. U is considered immobile in reducing conditions (Edwards et al., 1995). The Hinemaia soil is also likely to receive inputs of rhyolitic ash from the Taupo Volcanic Zone, as well as phosphate fertiliser. However, the U contribution from ash is very small compared with that from phosphate fertiliser. Eruptions of Mt Ruapehu in 1995 and 1996 could supply 0.5 mm ash (Cronin et al., 1998) to the Hinemaia site since the initial sampling. Uranium in New Zealand rhyolitic rocks ranges from 1.5 to $5.3 \mu\text{g g}^{-1}$ (Briggs et al., 1993) and assuming the highest U content in the ash as $5.3 \mu\text{g g}^{-1}$, a total contribution to the soil of $0.04 \mu\text{g g}^{-1}$ could be expected (assuming bulk density = 0.6 , sample depth of $0 - 7.5 \text{ cm}$) compared with the total increase of $1.69 \mu\text{g g}^{-1}$ over 36 years. Therefore, ash may account for less than 3 % of the U accumulated.

The Taupo soil is also likely to have received similar inputs of rhyolitic ash as the site is only 15 km from that of the Hinemaia. However, it showed the least increase (from 2.34 to $2.89 \mu\text{g g}^{-1}$, an annual increase of $0.015 \mu\text{g g}^{-1} \text{a}^{-1}$) reflecting the low inputs of superphosphate fertiliser and the minor contribution from ash. In contrast, the Mikimiki soil showed an intermediate increase in total U content (0.62 to $1.69 \mu\text{g g}^{-1}$, an annual increase of $0.025 \mu\text{g g}^{-1} \text{a}^{-1}$), reflecting the moderate levels of superphosphate

fertiliser application. The concentration of total U increased on average by $1.30 \pm 0.03 \mu\text{g g}^{-1}$ for the 4 soils sampled, compared with 1992 soil samples, with an average annual increase of $0.033 \pm 0.008 \mu\text{g g}^{-1} \text{a}^{-1}$.

Accumulation rates of U were higher in this study (0.015 to $0.047 \mu\text{g g}^{-1} \text{a}^{-1}$) than those reported by Rothbaum et al. (1979), which ranged from 0.003 to $0.014 \mu\text{g g}^{-1} \text{a}^{-1}$ (Table 3). The differences in accumulation rate between this study and that of Rothbaum et al. (1979) possibly reflect the level of U contamination in the superphosphate used and differences in soil pH and organic carbon content. The superphosphate used in New Zealand (Nauru and Christmas Is.) has higher concentrations of U than the North African sourced superphosphate (Table 4) used by Rothbaum et al. (1979). Concentrations of U in superphosphate show considerable variance, depending on the source of the raw phosphate. Although the same quantity and type of fertiliser was applied to the three study sites at Rothamsted, the accumulation rates of the individual soils differ (Rothbaum et al., 1979). It appears that New Zealand's carbon, oxide and oxy-hydroxide rich, acidic soils are better able to bind and therefore accumulate more U compared with the neutral to slightly calcareous soils of the UK. This may be due to U solubility being limited by pH controlled adsorption on to organic matter (Crancon and van der Lee, 2003; Aubert et al., 2003; Zhou and Gu, 2005) iron oxides and oxy-hydroxides (Bostick et al., 2002; Barnett et al., 2002; Morrison et al., 1995) and clays (Pabalan and Turner, 1996; Edwards et al., 1995; Giblin et al., 1981) while uranium is readily leachable under alkaline conditions (Zielinski et al., 1997).

The time required to exceed the United States Nuclear Regulatory Commission residual contamination criteria of $30 \mu\text{g g}^{-1}$ (USNRC 1992) was calculated for the four soils in this study (Table 3). The soil with the highest concentration of U ($3.54 \mu\text{g g}^{-1}$) and the highest average annual

Table 3:
Comparison of uranium accumulation rate with pH for two studies

Site	Reference	pH	Average annual increase in U ($\mu\text{g g}^{-1} \text{a}^{-1}$)	Time to exceed USNRC contamination criteria (years)
Mikimiki, New Zealand	This study	5.6	0.025	1032
Eltham, New Zealand	This study	5.1	0.046	598
Taupo, New Zealand	This study	5.2	0.015	1807
Hinemaiaia, New Zealand	This study	5.5	0.047	563
Broadbalk, Rothamsted	Rothbaum et al., 1979	Neutral to slightly alkaline	0.004	-
Barnfield, Rothamsted	Rothbaum et al., 1979	Neutral to slightly alkaline	0.003	-
Park Grass, Rothamsted	Rothbaum et al., 1979	5.2	0.008	-
Papatoetoe, New Zealand	Rothbaum et al., 1979	5.9	0.014	-

increase ($0.047 \mu\text{g g}^{-1} \text{a}^{-1}$), the Hinemaiaia, would take 563 years to reach the contamination criteria if U application rates remained the same, while the Taupo soil would take the longest, 1807 years.

The rate of accumulation increased with increase in the fertiliser application rate for the 4 sites of this study ($R^2 = 0.909$) but little relationship was seen for the study by Rothbaum et al. (1979) (Figure 1). The superphosphate used in New Zealand (Nauru and Christmas Island) has higher concentrations of U and application rates have a wider range than the North African sourced superphosphate (Table 4) used by Rothbaum et al. (1979) enabling trends to be seen more distinctly.

The lack of relationship in the Rothbaum et al., study may be explained by soil pH. Echevarria et al. (2001) found soil pH had a significant effect on sorption ratio values. The sorption behaviour differences between UO_2^{2+} and UO_2^{2+} -carbonate complexes were so great that any other effect of soil properties on U sorption was hidden.

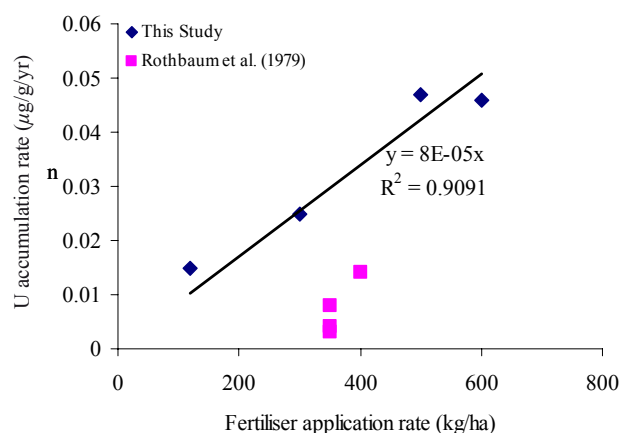


Figure 1:
Relationship between fertiliser rate and accumulation of U

3.2 U Concentrations in Phosphate Fertiliser and Lime

Levels of total U in phosphate fertiliser measured in this study were low (average $22.2 \mu\text{g g}^{-1}$) compared with historical data and calculated historical levels (Table 4 and Table 5). This is consistent with present-day fertiliser manufacturers preferentially obtaining raw phosphate rock from sources containing low levels of heavy metals, especially Cd and U, although these sources are also more expensive. (Hilton Furness, NZ Fertiliser Manufacturers Association, pers com). With the lower levels of U in present-day phosphate fertiliser, the rate of U accumulation in soil will slow.

Lime contained much lower concentrations (average $3.44 \mu\text{g g}^{-1}$) of U than phosphate fertiliser. These levels

are similar to those of the volcanic ash parent material of the Taupo and Hinemaia soils (Briggs et al., 1993). Therefore, U levels in these soils are unlikely to be significantly enhanced by lime additions.

Table 4:
Historic concentrations of U in phosphate rocks and fertiliser

Source (years sampled)	U ($\mu\text{g g}^{-1}$)	Reference
North Africa	34	Rothbaum et al., 1979
Nauru/Christmas superphosphate (1954 - 1975)	42	Rothbaum et al., 1979
Nauru Island (1925 - 1937)	65	Menzel 1968
Christmas Island (1925 - 1960)	27	Menzel 1968
North Carolina (1957 - 1964)	79	Menzel 1968
Florida (1946 - 1955)	148	Menzel 1968
Florida	189.7 ± 0.42	Hamamoto et al., 1995
Florida	119	Scholten and Timmermans 1996

Table 5:
Uranium in some New Zealand fertiliser and lime samples

Sample	^{234}U (ng g^{-1})	^{238}U ($\mu\text{g g}^{-1}$)
Superphosphate a	2.242	37.48
Superphosphate b	0.424	8.01
Superphosphate c	0.495	9.94
Superphosphate d	2.010	21.65
Triple Superphosphate	1.350	33.98
Dolomite Lime	0.197	4.06
Dolomite Lime	0.149	3.09
Lime	0.165	4.44
Lime	0.055	2.15

For three of the soils it was possible to estimate the average concentration of U in the superphosphate fertiliser applied between samplings using the formula:

$$c = \frac{iAdp}{f}$$

where c = concentration of U in fertiliser in $\mu\text{g g}^{-1}$
 i = average annual increase in U in soil in $\mu\text{g g}^{-1}$
 A = area of 1 ha in m^2
 d = sample depth in m
 ρ = bulk density in t m^3
 f = annual fertiliser application to 1 ha in kg

For the Mikimiki and Hinemaia soils the estimated U concentration in the superphosphate was 44 and 37 $\mu\text{g g}^{-1}$ respectively, while the Eltham soil gave a higher value of 63 $\mu\text{g g}^{-1}$. This higher value may be due to historical higher application of superphosphate than is currently applied, or application of superphosphate from a different and higher U content source from that applied to the other soils. However, all three of these estimated concentrations compare well with those in phosphate rocks and a superphosphate used in New Zealand (Table 4). It appears all or nearly all the U applied in fertiliser has remained in the soil and has not leached to ground water or been taken up by plants. It is evident that U is strongly sorbed onto acidic New Zealand soils.

The strong sorption onto soil indicates that plants will uptake little of the U. Shahandeh & Hossner (2002), studying U uptake in plants on 9 soils (pH range 4.7 - 8.1) found the lowest plant U occurred in clayey acid soils. They also found significant differences in U accumulation between plant species. Even if U uptake into some plants becomes a concern, there are alternative vegetables that could be grown with little likelihood of reaching levels that effect human health.

4 Conclusions

U has increased in all the soils measured in this study, and this increase may be attributed to U impurities in phosphate fertiliser. Mean levels of total U have increased by $1.30 \pm 0.030 \mu\text{g g}^{-1}$ for soils sampled 40 years ago compared with those in present-day soil samples. Accumulation rates ranged from 0.015 to 0.047, with an average increase of $0.033 \pm 0.008 \mu\text{g g}^{-1} \text{ a}^{-1}$. All or nearly all the U applied in phosphate fertiliser appears to have remained in the soil and has not leached to groundwater or been taken up by plants. The strong sorption onto soil indicates that plants will continue to uptake little of the U. Levels of total U in phosphate fertiliser measured in this study were low (average 22.2 $\mu\text{g g}^{-1}$) compared with historical data, due to present-day fertiliser manufacturers obtaining phosphate from sources containing low levels of U. With these lower levels of U in present-day phosphate fertiliser, the current rate of U accumulation in soil is likely to slow.

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