

# Monitoring of radioactivity in fertilizers in Austria

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## Abstract

This paper reports results of a monitoring project regarding natural radionuclides in fertilizers which are commonly used in Austria. Due to adaption of the Austrian Radiation protection rules to European legislative, fertilizers became part of products which have to be monitored by governmental authorities on their activity concentrations of natural radionuclides. In addition, the Austrian NORM ordinance lists processing of raw phosphate in chemical industry as well as in fertilizer industry as workplaces with possible elevated exposure to Uran and Thorium and their decay products and as industries, where residues could accumulate elevated concentrations of natural radionuclides. Therefore about 500 fertilizer samples were investigated by gamma spectroscopy. The fertilizers were provided by the governmental fertilizer inspection staff and preselected regarding usage in Austria and types of fertilizers with possible elevated activity concentrations (e.g. Tripelphosphate). A further distinction was made in order to compare fertilizers regarding their major components (organic-, nitrogen-, phosphate-, potassium-, multi plant nutrient- fertilizers ...).

The results are focused on U-238, Ra-226 and Pb-210. Using these results, input of Uranium in agricultural fields was calculated and three scenarios were discussed. These three scenarios include the total translocation of NORM nuclides into drinking water, the transfer of NORM nuclides into plants and the accumulation of Uranium in agricultural fields. The relation between Radium-226 and Uranium-238 is also discussed with focus on fertilizer processing and the probability to accumulate NORM residues in processing components.

**KEYWORDS:** *NORM, fertilizer*

## Introduction

The Radiation Protection Act § 37 [1] with the amendment to the Radiation Protection EU-Adaptation Law No. 137/2004, lists animal feed and fertilizers to the explicitly listed products and materials, which in terms of their content of radioactivity, have to be monitored by the authorities. The NORM ordinance [2] lists the processing of phosphates in the chemical industry and in the fertilizer industry, both as work areas with potentially elevated exposures to uranium and thorium and their decay products, and as a work area in which residues with increased content of uranium and thorium and its decay products may incur.

Next to nitrogen and potassium, phosphorus is one of three main components of fertilizer. The radioactive isotope  $^{40}\text{K}$  in potassium and the natural radionuclides of the uranium-radium decay series in phosphate rock are the radiological relevant radionuclides in fertilizers.

Since there is no routine yet for sampling scheme, an initial overview was first won by a broadly diversified selection of samples. The fertilizers were provided for investigation by the former Department of Fertilizers monitoring and microscopy within the former Institute for soil health and plant nutrition.

## Method of analysis and sample preparation

### Sample preparation

The received fertilizer were packed in conventional measurement geometry with a volume of 500 ml and sealed as airtight as possible. Then 40 samples stored for one month so that a secular equilibrium between the long-lived parent nuclide radium-226 and the short-lived daughter nuclide and noble gas radon-222 adjusted. The losses due to emanation of radon from the sample geometry were determined. It turned out to be the emanation of  $^{222}\text{Rn}$  strongly depends on the matrix of the fertilizer, so that in most fertilizers the losses due to radon emanation were 5% and in rare cases up to 20%. The average density of the samples was  $1.27 \pm 0.3 \text{ kg/dm}^3$ .

### **Method of analysis**

The samples were measured by high resolution gamma spectrometry. The measuring time on the HPGe (High Purity Germanium) detectors, with a relative efficiency of 30-40%, was 80,000 seconds. The analysis of the spectra and the decay-, random- and cascade-summing correction was performed by the GENIE2000 © software. A density correction was not applied. The uncertainty was calculated with a confidence interval of  $1.65 \sigma$ .

### **Investigation parameters**

During the evaluation of the measurement results, the activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were determined. The uranium-238 was determined via its secular equilibrium with protactinium-234m, radium-226 via  $^{214}\text{Pb}$  and thorium-232 via  $^{228}\text{Ac}$  and  $^{228}\text{Th}$ .

### **Phosphate extraction**

Since in the manufactory of phosphate fertilizer, phosphoric acid is used and sometimes already recovered Phosphatpentoxid is added, which is also used to produce phosphate acid, we will describe these processes briefly.

The production of phosphoric acid with the wet process is carried out by digestion of rock phosphate with inorganic acids. Sulfuric acid is worldwide most (90%) used for decomposition. At this digestion, 90%  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ , and about 95% to 80% of the nuclides of the thorium series will be transferred from rock phosphate raw material into the phosphogypsum. In contrast, remain about 90% uranium as uranyl phosphate, sulphate or fluoride complexes in the acid fraction.

In the thermal recovery of the phosphate from phosphate rock with silicic acid and coke in an electric resistance furnace at 1400-1500 °C is melted. The phosphorus is thereby transferred into gaseous form and is supplied with the hot furnace gases passed via electrostatic filter and condensed then with water. The condensed liquid is pumped into storage and covered with water. Subsequently, the phosphorus is atomized at the head of a combustion furnace and combusted under supply of air to phosphorus pentoxide. The main fraction of uranium and radium contains the sintered slag, over 90% of uranium and radium, but only about 2% of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  [3].

### **Results**

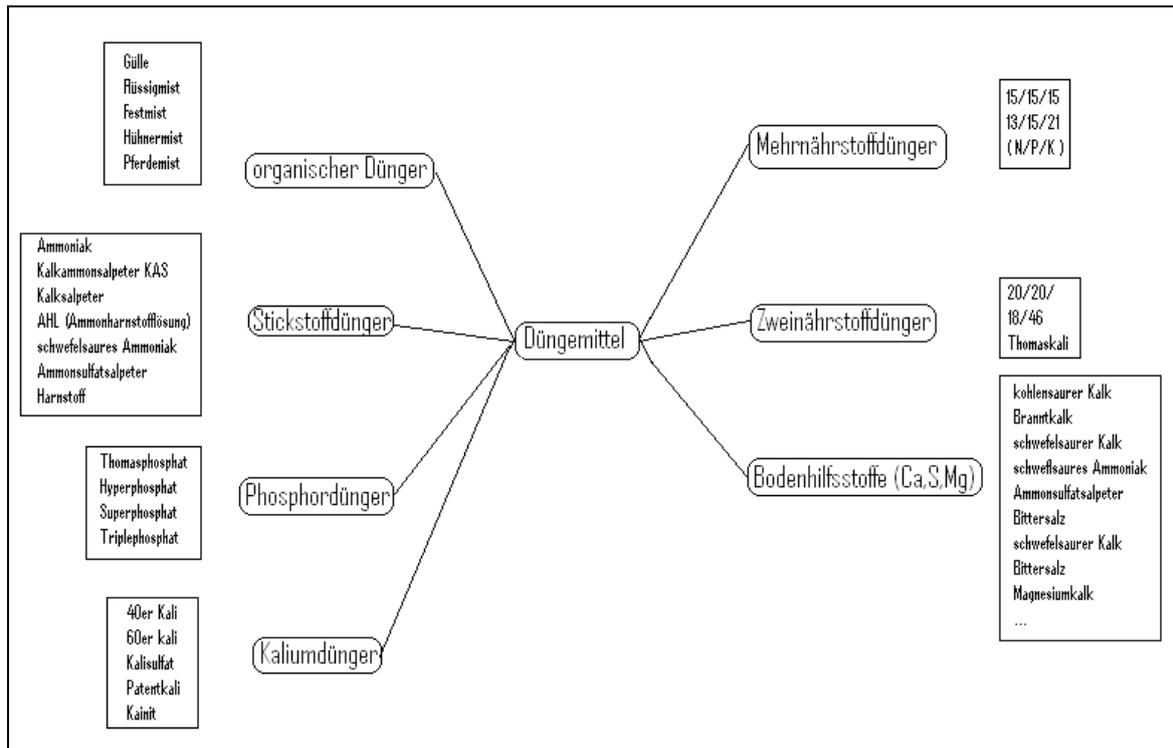
In the period from 2001-2007, about 500 fertilizers were studied. The samples were provided by the official control of fertilizers (AGES). A pre-selection of the available fertilizer was carried out by the Head of the Institute Dr. Katzlberger. The selection of the samples was taken to ensure that one part represents a cross section of Austrian fertilizers in use and fertilizer in which an increased activity concentration of natural radionuclides was suspected.

Another question regarding the subdivision or summary of measured fertilizer after the first interim analysis arises. The level of activity concentrations of uranium-radium series in the fertilizer depends on the activity concentration of natural radionuclides in phosphate rock, the preparation (partially or fully digested, ground) of the same, and the purity of the optionally used phosphoric acid. The manufacturer refers unfortunately only on the solubility of the major nutrients (N/P/K) and their percentage share.

Finally we could not always conclude from the solubility of phosphate fertilizers on the production. Hence the fertilizers were distinguished according to their main nutrients, as shown in figure 1. 53 of 500 fertilizers could not be assigned unambiguously to figure 1. The following activity concentrations of the different fertilizer types are arithmetically averaged.

### **Organic fertilizer**

Eleven fertilizers were assigned to the organic fertilizer. The activity concentrations of radionuclides of the natural decay series are all less than 50 Bq/kg. The mean activity concentration of  $^{40}\text{K}$  is 188Bq/kg. The maximum activity concentration of  $^{137}\text{Cs}$ , 40 Bq/kg, was detected. The origin of the Cs contamination is explained due to the accident of Chernobyl.



**Figure 1:** Classification by major fertilizer nutrients.

### Phosphate fertilizer

Phosphate fertilizers were distinguished according their availability of phosphate onto the plant and according to its solubility in water. The solubility of phosphates in turn depends on the production, phosphate fertilizer are thus divided into fully and partially digested and soft earthy rock phosphates. Because of the different names and the associated difficulties of an assignment regarding the digestion degree of phosphate fertilizers, they are classified and described according to Figure 1.

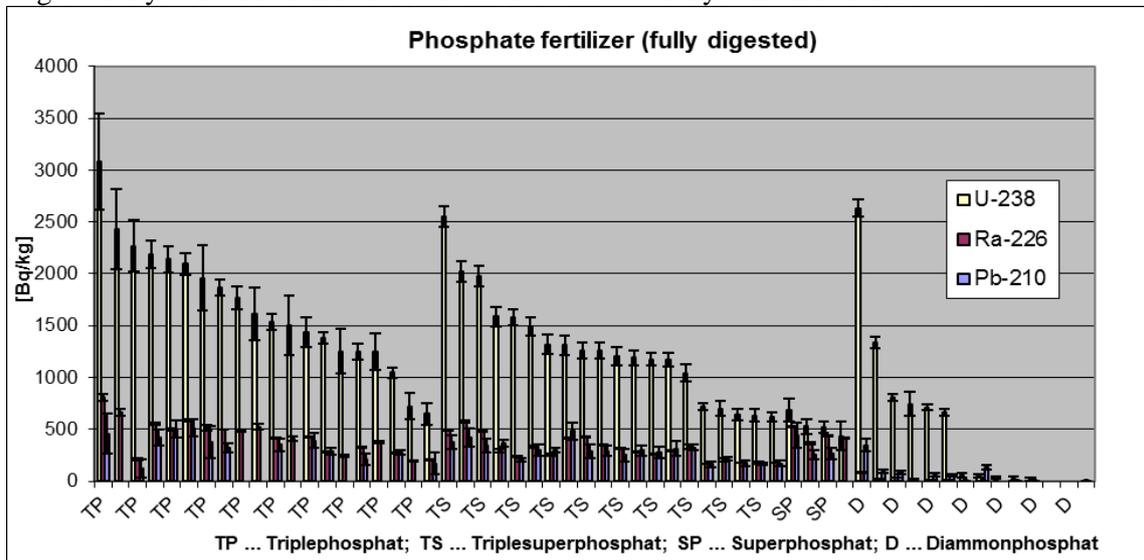
In Figure 2 different types of fully digested phosphate fertilizers (triple phosphate, triple super phosphate, di-ammonium phosphate and superphosphate, from left to right) are shown. A total of 58 fertilizers, 14 of them are assigned to di-ammonium phosphate, respectively 20 of them are assigned to triple phosphate and to triple superphosphate and 4 of them are assigned to superphosphate.

At triple phosphate fertilizers it can be seen that the rock phosphate was processed with phosphoric acid. The contribution of phosphoric acid can be identified because of the elevated  $^{238}\text{U}$  activity concentration, with an averaged mean of 1671 Bq/kg. The averaged activity concentrations of  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are in the range of normal rock phosphate with 424 and 339 Bq/kg. Nearly no nuclides of the Thorium decay chain could be detected. The averaged activity concentration ratio between  $^{226}\text{Ra}$  and  $^{238}\text{U}$  is 0.26.

Triple super phosphate fertilizers are similar except that in the preparation in addition to phosphoric acid also sulfuric acid is used. The average activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 1272 Bq/kg, 311 Bq/kg and 286 Bq/kg. Furthermore, the thorium series radionuclides could be measured, which, however, all with activity concentrations below 50 Bq/kg have no relevance from the viewpoint of radiation protection. The average ratio of  $^{226}\text{Ra}$  to  $^{238}\text{U}$  activity concentration is 0.25.

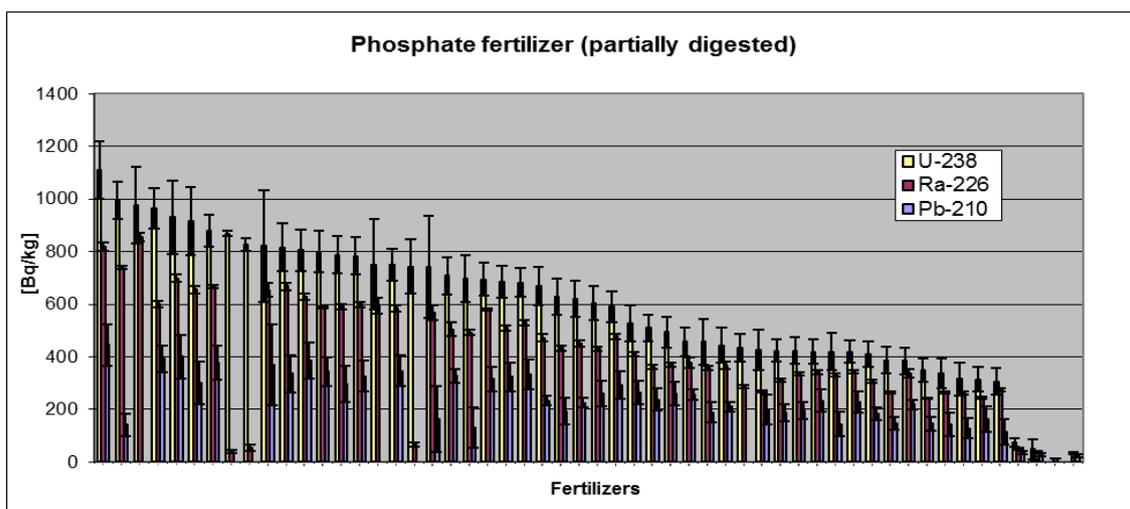
Super phosphate fertilizers are combined phosphate/sulphur fertilizer. During the processing of rock phosphate with sulphuric acid, no separation takes place between the phosphoric acid leached phosphate, uranium, thorium and the radium in the phosphogypsum. That's why the radium and uranium remains in the fertilizer [4]. The measured activity concentrations show that the radionuclides are still in equilibrium. The averaged activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 542 Bq/kg, 435 Bq/kg and 321 Bq/kg. The averaged activity concentration ratio between  $^{226}\text{Ra}$  and  $^{238}\text{U}$  is 0.81.

Di-ammonium phosphate or rather di-ammonium hydrogen phosphate is an ammonium salt of phosphoric acid. For the di-ammonium phosphate fertilizers a considerable variation in the  $^{238}\text{U}$  activity concentrations (18 -2632 Bq/kg) was observed. Causes of these fluctuations may be the initial activity concentration in used rock phosphate and secondly the quality of the phosphoric acid. The average activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 505 Bq/kg, 16 Bq/kg and 129 Bq/kg. The average activity concentration ratio of the  $^{226}\text{Ra}$  of  $^{238}\text{U}$  activity concentration is 0.03.



**Figure 2:**  $^{238}\text{U}$ -,  $^{210}\text{Pb}$ - and  $^{226}\text{Ra}$ - activity concentrations of different fully digested phosphate fertilizers.

Next to the fully digested phosphates are listed in Figure 1 also partially digested phosphates and fine ground earthy soft rock phosphates are present. A total of 54 of the partially digested fertilizer are assigned to the phosphate fertilizers. A more precise distinction as to the fully digested phosphate fertilizers is not possible. The average activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 609 Bq/kg, 414 Bq/kg and 238 Bq/kg. The mean activity concentration of  $^{40}\text{K}$  is 522 Bq/kg. The averaged activity concentration ratio between  $^{226}\text{Ra}$  and  $^{238}\text{U}$  is 0.72. Significant deviations from this value can be observed only in hyper phosphates and mineral compound fertilizers.



**Figure 3:**  $^{238}\text{U}$ -,  $^{210}\text{Pb}$ - and  $^{226}\text{Ra}$ - activity concentrations of different partially digested and earthy soft rock phosphates.

### Potassium fertilizer

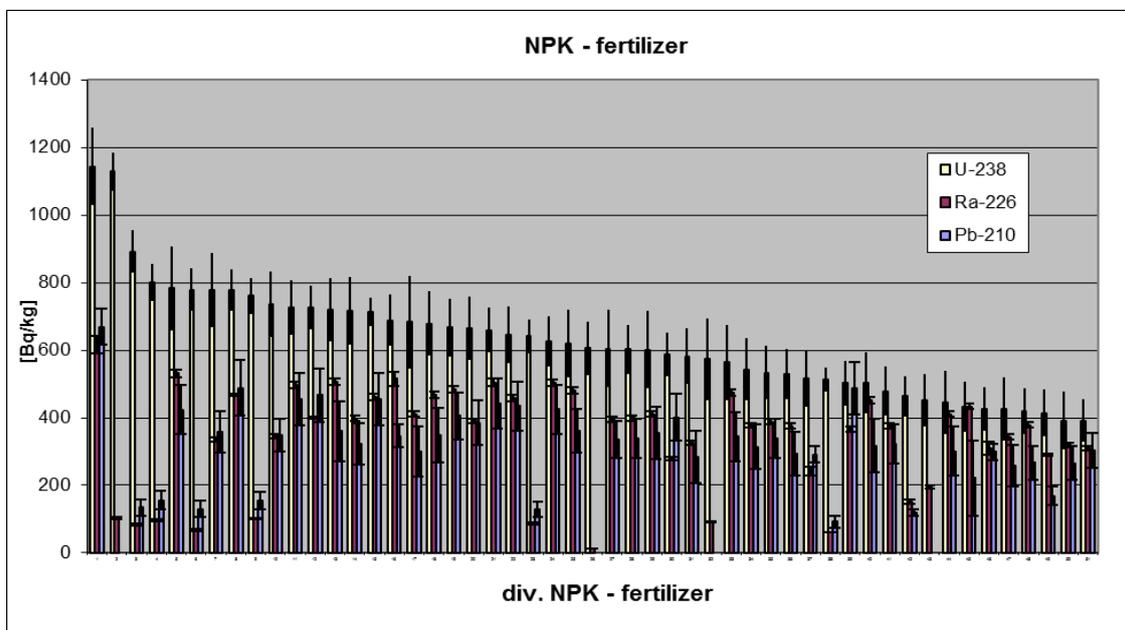
Potassium contains 0.012% of the radioactive isotope potassium-40 with a specific activity of 32.2 kBq/kg. The  $^{40}\text{K}$  activity concentrations vary greatly and span for potassium fertilizers within 6-16 Bq/g. The potassium content influences directly the activity concentration. Substituting the  $^{40}\text{K}$

activity concentration of the fertilizer at the specified concentration in  $K_2O$  ratio, shows that on average, the activity concentration is  $234 \pm 15$  Bq/kg for each per cent increase  $K_2O$ .

A total of 25 fertilizers are assigned to potassium fertilizers. Fertilizers designated Thomaskali, Donau Chemie 44-45 and Hyperkali are phosphor/potassium fertilizer and will be compared with the rest of PK fertilizers. Fourteen fertilizers exceeded the  $^{40}K$  activity concentrations of 10 Bq/g. Radionuclides of the natural decay series, with more than 10 Bq/kg, were not detected.

### Compound fertilizer

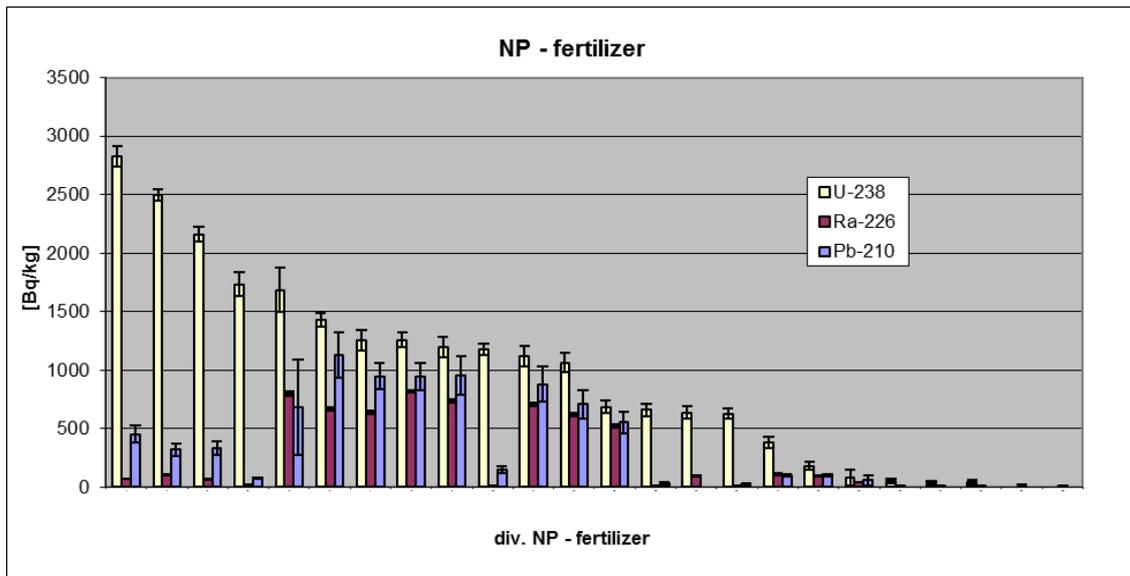
NPK fertilizers or compound fertilizer are a mixture of the three major fertilizer components (N is nitrogen, P for phosphorus and K for potassium). There are 184 fertilizers which were assigned to this group. These fertilizers are made by mixing of single-nutrient fertilizers. It is often unclear if the phosphorus is fully or partially digested. In 124 NPK-fertilizers uranium-238 could be measured. The activity concentration ratios between  $^{226}Ra$  and  $^{238}U$  show that for the manufacture of these fertilizers different sources of phosphate have been used. The average activity concentration of  $^{238}U$ ,  $^{226}Ra$  and  $^{210}Pb$  are 384 Bq/kg, 171 Bq/kg and 213 Bq/kg. Two fertilizers exceed the  $^{238}U$  activity concentration of 1 Bq/g. In figure 4 are 50 NPK-fertilizers shown, which represent the highest  $^{238}U$  activity concentrations.



**Figure 4:**  $^{238}U$ -,  $^{210}Pb$ - and  $^{226}Ra$ -activity concentrations of 50 compound fertilizers with the highest  $^{238}U$ -activity concentrations.

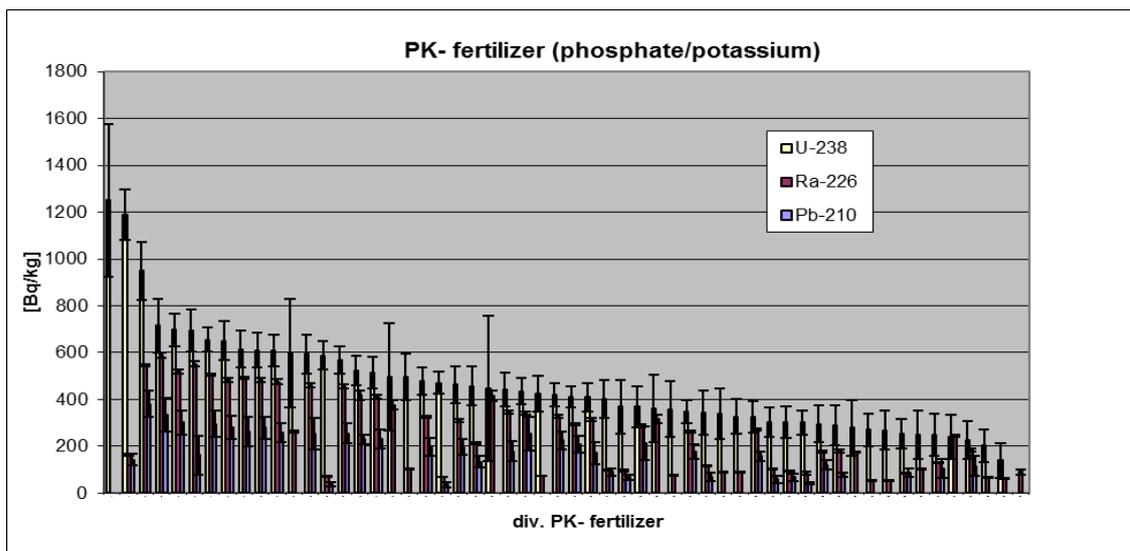
### Dual-nutrient fertilizer

Similar to the production of compound fertilizers, dual-nutrient fertilizers are made by mixing of single-nutrient fertilizers. A distinction between fully and partially digested phosphate fertilizers is not possible. Due to the large number of different dual-nutrient fertilizers, we focus only on NP (nitrogen and phosphate), PK (phosphate and potassium) and other phosphate-containing fertilizers. A total of 24 fertilizers are associated with NP-fertilizers (nitrogen, phosphorus). The average activity concentrations in NP-fertilizer of  $^{238}U$ ,  $^{226}Ra$  and  $^{210}Pb$  are 1038 Bq/kg, 259 Bq/kg and 471 Bq/kg. The mean activity concentration of  $^{40}K$  is 464 Bq/kg. At 12 NP-fertilizers exceed the  $^{238}U$  activity concentration of 1 Bq/g.



**Figure 5:**  $^{238}\text{U}$ -,  $^{210}\text{Pb}$ - and  $^{226}\text{Ra}$ - activity concentration of NP-fertilizers.

A total of 56 fertilizers are associated to PK-fertilizer (phosphorus, potassium). The average activity concentrations in PK-fertilizers of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 460 Bq/kg, 254 Bq/kg and 178 Bq/kg. Two PK-fertilizers exceeded the  $^{238}\text{U}$  activity concentration of 1 Bq/g.



**Figure 6:**  $^{238}\text{U}$ -,  $^{210}\text{Pb}$ - and  $^{226}\text{Ra}$ - activity concentration of PK-fertilizers.

### Other fertilizers

In this category lime-, nitrogen-, sulfur-fertilizers, soil conditioners and fertilizers which are not assignable, are summarized. A total of 93 fertilizers were assigned to the other-fertilizers. The average activity concentrations of  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  are 245 Bq/kg, 42 Bq/kg and 56 Bq/kg. Only in 11 fertilizers  $^{238}\text{U}$  were detected and one of them exceeded the activity concentration of 1Bq/g.

## Summary of the results

An overview of the minimal, maximal and the arithmetically averaged activity concentrations of radiological relevant nuclides of all assigned fertilizers is shown in table 1.

fertilizer / [Bq/g]	<sup>238</sup> U			<sup>226</sup> Ra			<sup>210</sup> Pb		
	Min.	Max.	MW	Min.	Max.	MW	Min.	Max.	MW
Org. fertilizer (11)	-	-	-	0,001	0,014	0,007	0,010	0,010	0,010
Di-ammon phosphate (14,fully digested)	0,018	2,632	0,642	0,001	0,082	0,016	0,056	0,346	0,129
Superphosphate (4, fully digested)	0,435	0,689	0,542	0,364	0,529	0,435	0,018	0,028	0,022
Triple superphosphate (20, fully digested)	0,622	2,549	<b>1,272</b>	0,164	0,570	0,311	0,164	0,483	0,286
Triple phosphate (20, fully digested)	0,647	3,080	<b>1,671</b>	0,196	0,803	0,424	0,119	0,513	0,339
Phosphate fertilizer (54,partly digested)	0,050	1,110	0,608	0,014	0,856	0,414	0,023	0,445	0,238
PK-fertilizer (56, f/p digested)	0,139	1,250	0,460	0,054	0,587	0,254	0,036	0,381	0,178
NP-fertilizer (24, f/p digested)	0,041	2,829	<b>1,038</b>	0,02	0,817	0,259	0,029	1,130	0,471
NPK-fertilizer (190, f/p digested)	0,039	1,143	0,382	0,001	0,617	0,162	0,002	0,668	0,210
Potassium fertilizer (25)	-	-	-	-	-	-	-	-	-
Other fertilizer (93)	0,019	1,294	0,246	0,001	0,761	0,042	0,002	0,355	0,056

**Table 1:** Overview of <sup>238</sup>U-, <sup>210</sup>Pb- and <sup>226</sup>Ra- activity concentrations in fertilizers

## Evaluation/Assessment

In the following chapter a conservative assessment were made with the measured values. This assessment contains the use of fertilizers and the following uptake of NORM nuclides into agricultural farmland. The activity concentration ratio between radium and uranium are investigated in the context of possible residues during processing of rock phosphate. An external ambient dose equivalent caused by <sup>40</sup>K for working in close proximity to large quantities of potassium fertilizers is estimated.

### Uranium uptake into agricultural farmland

Agricultural farmlands were fertilized up to 450 kg ha<sup>-1</sup> a<sup>-1</sup> P<sub>2</sub>O<sub>5</sub> in Austria. The overall mean is 59 kg ha<sup>-1</sup>a<sup>-1</sup> and contains mineral, organic and fully and partially digested phosphate fertilizers. For the following assessment a plough depth of 30 cm (homogenate) and a mean density of the soil of 1.5 kg/dm<sup>3</sup> is assumed. The assessed fertilizer is a triple phosphate fertilizer with a content of P<sub>2</sub>O<sub>5</sub> of 44% and an activity concentration from <sup>238</sup>U to <sup>230</sup>Th of 3 Bq/g and an activity concentration of <sup>226</sup>Ra and her daughter nuclides of 1 Bq/g. In table 2, an overview of the uranium and radium uptake into agricultural farmland for different phosphate fertilizer demand starting with the overall mean which is common in Austria to the maximum value. These uptakes are very conservative values. The real absorption of radionuclides cannot simply assessed because of the multitude of influencing parameters

such as pH-values, composition of the farmland, annual deposition, the cultivated fruits (maize, beet, wheat, ...), mineralization of the soil and the run off of radionuclides.

P <sub>2</sub> O <sub>5</sub> [kg/ha*year]	triple phosphate [kg/ha*year]	A <sup>U-238ges</sup> [kBq/ha*year]	A <sup>Ra-226ges</sup> [kBq/ha*year]	<sup>238</sup> U–uptake [Bq/kg*year]	<sup>226</sup> Ra–uptake [Bq/kg*year]
59	134	402	134	0,09	0,03
100	227	681	227	0,15	0,05
300	682	2046	682	0,45	0,15
450	1023	3069	1023	0,68	0,23

**Table 2:** The annual uptake of uranium and radium from triple phosphate fertilizer into agricultural farmland (44% P<sub>2</sub>O<sub>5</sub>).

After fertilizer application, the uranium passes through irrigation or deposition in the soil-solution. Under oxidized conditions, uranium is present in its 6-valent form, uranyl-ion, in the aqueous phase. From the soil-solution it can be absorbed by plants and accumulated in the plant tissue. Immobilization on clay minerals and organic matter takes place.

Under reduced conditions, uranium is present in its 4-valent form, which is less mobile as the 6-valent form. It forms sparingly soluble minerals and can therefore better be retained. A portion of the dissolved uranium remains in solution and can be washed into the groundwater. The discharge is favored because the uranyl forms with carbonate and phosphate zero-valued or negatively charged complexes like fulvic acids. These compounds can hardly be retained by the soil matrix [6].

In summary, three scenarios are discussed: the total leaching of uranium and its daughters in the groundwater, the availability of intake of uranium and its daughters in the cultivated fruit and enrichment of uranium in the fields. The dose coefficients for ingestion for members of the public were taken from ICRP Publication 67 [7]. The transfer factors for natural radionuclides from soil to the fruit are used in the NCRP Report No. 76 [8].

### The total leaching of uranium in the groundwater

The leaching of uranium and its daughters in the groundwater is a potential exposure scenario. This process is a continuous, since it is assumed that the field is fertilized each year with the same quantity of the same fertilizer. An average rainfall of 620 mm per year or 620 liters per square meter per year is assumed that represents the long-term average rainfall of the years 1971 to 2000 in Austria [9]. The effluent is continuously calculated over a year and thus diluted over one hectare with 6200000 liters. Thus, the maximum uptake of <sup>238</sup>U in the groundwater varies in the range between 0.065 and 0.495 Bq/l. For the dilution of seepage water with the aquifer, the IAEA proposed groundwater flow rate of 62500000 liters per year, is assumed [10]. We obtain the calculated activity concentration in groundwater and drinking water for <sup>238</sup>U between 0.006 and 0.045 Bq/l. The dose for a reference person of the population, who uses the groundwater without any further dilution as drinking water, is shown in Table 3. In this calculation it is assumed that <sup>238</sup>U to <sup>230</sup>Th as well as <sup>226</sup>Ra and its decay products are in radioactive equilibrium, and that the demand of drinking water is 25% or 180 liters per year, which is met by a water-well.

P <sub>2</sub> O <sub>5</sub> [kg/ha*Jahr]	A <sup>U-238+ges</sup> [Bq/l]	A <sup>Ra-226+ges</sup> [Bq/l]	Ingestionsdosis [μSv/a]
59	0,006	0,002	1,62
100	0,010	0,003	2,75
300	0,030	0,010	8,25
450	0,045	0,015	12,4

**Table 3:** Ingestion dose caused by a uptake of natural radionuclides from phosphate fertilizers into drinking water (well-water).

The ingestion doses in Table 3 are interpreted as the maximum doses, because only a fraction of the uranium and its decay products will be leached actually in the groundwater.

### Exposure through consumption of fertilized plants

Based on the previous assumptions regarding the total uptake of uranium and radium and their decay products into the plant is assumed. An expected maximum dose, which is caused by the food intake of natural radionuclides, which were taken by the plants, is calculated. The calculated doses for the ingestion of 89 kg of grain per year [11] are listed in Table 4. The available activity concentrations are calculated from the ratio of the annual uptake of radionuclides to the plants and the annual rainfall.

P <sub>2</sub> O <sub>5</sub> [kg/ha*Jahr]	A <sup>U-238+</sup> [Bq/l]	A <sup>Ra-226+</sup> [Bq/l]	Ingestionsdosis [μSv/a]
59	0,065	0,022	0,15
100	0,110	0,037	0,26
300	0,330	0,110	0,78
450	0,495	0,165	1,16

Table 4: Ingestion dose caused through the food intake of grain

### Enrichment of uranium in agricultural farmland

An increase of the uranium activity concentration through mineralized fertilizers (soft earthy rock phosphate) in agricultural farmland in the foreseeable future is unlikely. The rate of increase in the current good agricultural practice for farmland is moderate with maximum of 0.13% compared with the geogenic background of 3.3 Bq/kg or 40 Bq/kg <sup>238</sup>U. For this calculation, the highest measured activity concentrations were used, which usually occur only in fully digested phosphate fertilizers. These results agree very well with the results of the Federal Institute for Risk Assessment with a growth rate of 0.08% [12].

The increase in uranium contents in soil alone as a result of fertilization with mineral phosphates (e.g. uranium from limestone) is intrinsic, because the uptake in agro-ecosystems is considerably higher than the losses through leaching. Other studies on this topic support this assumption that on average about 0.5-1 mg/kg higher uranium concentrations in surface soils compared to soils under agricultural sites in northern Europe have been identified [13].

### <sup>226</sup>Ra/<sup>238</sup>U-ratio

The <sup>226</sup>Ra/<sup>238</sup>U-ratio indicates transfers of different species of NORM nuclides within rock phosphate processing. In figure 6 is shown that triple phosphate und triple super phosphate fertilizers have similar ratios between 0.25 and 0.26. A redistribution of NORM nuclides is to assume and that the additional processing with sulphuric acid at triple superphosphate fertilizers have no significant influence. Diammon phosphate fertilizers have an average ratio of 0.03 which matches with former investigations [14]. Super phosphate fertilizers have a comparable average ratio, like mineralized fertilizers, of 0.81 and 0.71. That's why we conclude that during processing there is no significant redistribution of NORM nuclides. The <sup>226</sup>Ra/<sup>238</sup>U ratio is relevant insofar as it can be concluded that in cases of small ratios a redistribution of NORM nuclides within the processing plants and elevated activity concentrations in the residues have to be taken into account. Furthermore, it allows an assignment of dual- and compound fertilizers with regard to the digestion of the phosphate.

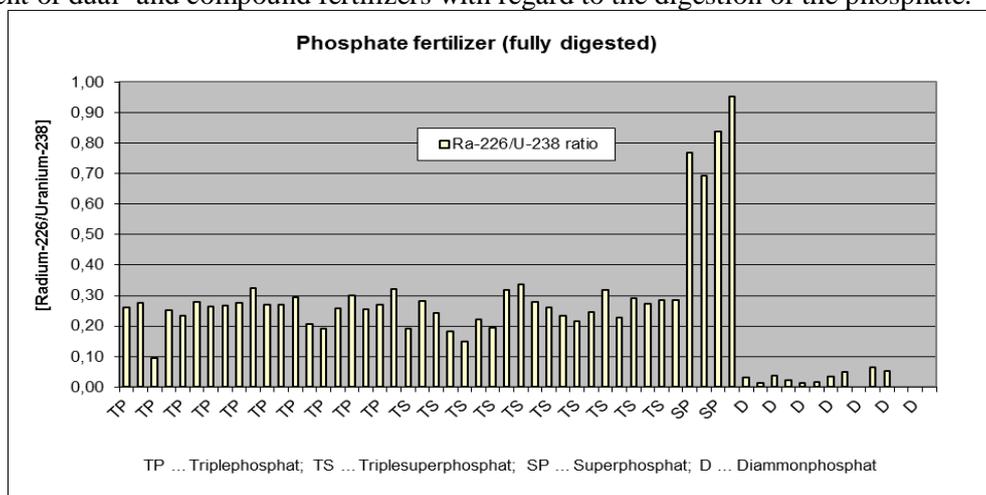


Figure 7: <sup>226</sup>Ra/<sup>238</sup>U ratios of different fully digested phosphate fertilizers.

## Exposure through potassium fertilizer

As mentioned in section 4.3, is the  $^{40}\text{K}$  activity concentration higher than 10 Bq/g in several fertilizers. The highest measured  $^{40}\text{K}$  activity concentration was 15.9 Bq/g. An exposure for a warehouse worker who works in the immediate near of large quantities of this fertilizer is estimated. With an average density of 1.4 kg/dm<sup>3</sup> and an assumed stack depth of 1 m, a surface activity concentration of 22260 kBq/m<sup>2</sup> can be calculated. The effective surface dose is estimated at 10 by 3 meters (30m<sup>2</sup>, very large as potassium-40 is a high-energy gamma emitter). The calculated dose, caused by  $^{40}\text{K}$  at 1 meter will be up to 14.7 µSv/h.

## Conclusion

At fully digested phosphate fertilizers an activity concentration with more than 1 Bq/g is possible. The average activity concentration is 1238 Bq/kg (99.5 mg/kg). At NP-fertilizer more than the half of them has activity concentrations higher than 1 Bq/g. The average activity concentration is 1035 Bq/kg (83.5 mg/kg). In all other investigated types of fertilizer, expect some outlier due to a possible false assignment, have significant lower activity concentrations of the uranium-radium decay chain of 1 Bq/g. Nuclides of the thorium decay chain does not matter with regard of the 1 Bq/g criteria. The assessment shows that even with conservative scenarios no real danger regarding to the NORM nuclides exists. Nevertheless we should keep in mind that phosphate fertilizing is the major large area distribution of uranium and its daughters into the environment in Europe. The potassium assessment scenario indicates that the storage of large amounts of potassium fertilizer (production, wholesale and distribution) can lead to doses above 1 mSv/year.

The  $^{226}\text{Ra}/^{238}\text{U}$  ratio is a tool to identify types of fertilizer and processes within the production which can accumulate NORM nuclides in processing system and residues.

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