

Dispersion of uranium in the environment by fertilization

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Abstract. It was the objective of this contribution to provide a comprehensive synopsis on the significance of the dispersion of uranium in the environment by common fertilizer practices. Recent studies revealed that uranium originating from fertilizers accumulates in soils over time and thereby increases uranium losses to water-bodies. Studies on the uranium content in soils and surface waters in relation to fertilizer practice substantiate such coherence. The most efficient and sustainable solution to the problem is the extraction of uranium from fertilizers.

Toxicological significance of uranium

Uranium (U) is a natural, chemo toxic and radiotoxic heavy metal. With view to the overall level of radioactivity in the environment U is certainly only a minor source of concern (Falck and Wymer 2006). The biochemical toxicity of the heavy metal U is estimated to be six orders of magnitude higher than the radiological toxicity (Milvy and Cothorn 1990; NRC 2005). Compared to other heavy metals, the chemical toxicity of U ranges between mercury and nickel, or christoballite and warfarin (Busby and Schnug 2008).

Uranium shows toxic effects on all forms of life: The most common and unspecific one is DNA damage followed by mutations (Envirhom 2005; Henner 2008; Lin et al. 1993; Thiebault et al. 2006). The effect of U on DNA is a sinister combination of the biochemical and radiological toxicology of U. Uranium builds up in living systems inter alia due to its high affinity to phosphorus containing components such as DNA (Busby and Hooper 2007). Once attached to the DNA U amplifies natural background radiation and causes through photoelectron enhancement effects damages to the DNA. This effect occurs to an excess that is obviously much stronger than that from α -radiation of U (Busby, 2005; Schmitz-Feuerhake and Bertell 2008). Bishop (2005) proposed also a signaling from

radiated to neighboring cells, which received no direct radiation so that cellular damages are multiplied.

The older an individual is, the higher will be the amount of U that is accumulated. This implies that the risk for contracting damages from U generally increases not only with the amount, but also with the time of exposure and thus with age (WHO 2004).

Mammals have a particularly high sensitivity against U (Fellows et al. 1998). Uranium tends to accumulate in the body, preferentially in kidneys, liver, spleen and bones. Uranium is a popular and long known nephrotoxin (Blantz 1975, Boshard et al. 1992; Flamenboum et al. 1976; Lin and Lin 1988; Lin et al. 1993; Zamora et al. 1998). The most remarkable damage of U coming along with low and medium contaminations is cancer (Linsalata 1994). The studies of Envirhom (2005) revealed that the brain is a target for U toxicity, too. Its sensitivity seems to be similar to that of kidneys (Envirhom 2005).

Uranium in food and uranium uptake by humans

Under non-exposed conditions the daily intake of U from air by humans amounts to about 1 ng U (WHO 2004). Uranium in soils enters the food chain indirectly through plant uptake or directly through consumption of U in drinking waters. The transfer of U from soil to plant is significantly higher for vegetative than for generative plant parts. Concentration factors for U are around 0.05 and similar to that determined for As, Co, Hg and Pb (Schick et al. 2008).

With view to food, lowest U concentrations were found in seeds, leaves and fruits, while approximately three times higher U contents were found in meat. For meat the ranking poultry < pork < beef reflects the animals lifespan and thus accumulation of U. The highest concentrations of U occur in offal and shellfish (Schnug et al. 2005).

A human has an average daily U intake of around 2.5 μg U when a simplified daily diet of 2000 kcal is assumed with 60% cereals and cereal products (1.5 $\mu\text{g}/\text{kg}$ U), 20% meat and meat products (5 $\mu\text{g}/\text{kg}$ U), 10% vegetables (2 $\mu\text{g}/\text{kg}$ U) and 10% fruits (1 $\mu\text{g}/\text{kg}$ U) (Schnug et al. 2005; Pais and Benton Jones jr. 1997; WHO 2004). Even a carnivore with a skewed affectation for offal or shellfish might increase this value only to at maximum 4 μg U, while a strict vegan cannot reduce the value below 1 μg U.

In contrast to solid food, the U concentration in drinking water has a distinctly stronger influence on the daily U intake by humans (Cothorn and Lappenbusch 1983; Schnug et al. 2005). Water is the most significant factor for the daily amount of U taken up by an individual. The intake of U by water can exceed the intake through solid foods in extreme cases by factor 10 and more if the daily water consumption is 2 L (40 mL/kg body mass according to Hesecker (2005)). Thus the evaluation of the U intake by humans requires comprehensive information about U concentrations in drinking waters.

Environmental loads of uranium originating from fertilization

The most likely largest non-point source emitters of U in Germany are agriculture, horticulture and forestry through the use of mineral phosphorus fertilizers. Even under conditions of good agricultural practice (Sharpley and Withers 2004) the annual amount of U unconsciously dispersed in the environment amounts on an average to 10 g/ha (Kratz et al. 2008). Depending on fertilizer type and intensity this U accumulates in soils with rates between 1- 46 $\mu\text{g}/\text{ha}\cdot\text{yr}$ (Rogasik et al. 2008; Taylor and Kim 2008). Utermann and Fuchs (2008) estimated that in Germany the mean U content in arable soils is 0.15 mg/kg higher than in soils under forestry. This difference can be completely explained by 45 years of cropping with phosphorus fertilization in an agricultural production system operating on a typical intensity level.

Fertilizer derived U in soils is prone to easy leaching, because U is comparatively mobile under pH and redox conditions of typical soils that underlie anthropogenic management (Jaques et al. 2005; Read et al. 2008). Transfer of U originating from fertilizers into water bodies is a fact and was proven in numerous research projects (Azuoazi et al. 2001; Barisic et al. 1992; Conceicao and Bonotto 2000; Hule et al. 2008; Kobal et al. 1990; Zielinski et al. 1995). Already in 1972 Spalding and Sackett attributed increased U concentrations in North American rivers of $\sim 0.7 - 0.9 \mu\text{g}/\text{L}$ U, compared to $\sim 0.1 - 0.2 \mu\text{g}/\text{L}$ U in South American rivers (Cothorn and Lappenbusch 1983), to the use of phosphate fertilizers in the region. Also Birke and Rauch (2008) found elevated U concentrations in river waters in some regions of Germany. The authors could not fully explain the data by geological factors and assumed that agricultural activities are accountable.

Most recently evidence was provided that, in full accordance with the prognosis of Jacques et al. (2005 & 2008), U originating from fertilizers starts to contaminate groundwater bodies and finally shows up in drinking waters:

Schäfer et al. (2007) found a close correlation between U and nitrate in drinking waters from the Rhine-Neckar region. Nitrate is like U easily mobile in soils, increases in ground and drinking water with fertilizer intensity and thus seems to be a suitable indicator for monitoring U transfer from fertilizers to waters.

Schulz et al. (2008) report twice as high U concentrations in drinking waters collected in former West Germany compared to former East Germany. A reasonable explanation is given by the facts that in the former East fertilization intensity was significantly lower and phosphate fertilizer products were employed, which had a lower U content than those used in the West. Consequently Rogasik et al. (2008) determined significantly higher amounts of U that accumulated in long-term phosphorus fertilization experiments in soils of the former East than in West Germany (Rogasik et al. 2008).

Knolle (2008) showed that the variability of boron concentrations explain a significant amount of the U concentrations in German tap waters. Boron is like U applied in significant amounts with phosphorus fertilizers: FAL-PB (2007) reports boron concentrations between 200 and 800 mg/kg B in various phosphorus

containing fertilizers, which makes the amount of boron applied with phosphorus fertilizers five times higher than the amount of U applied. Boron is like U easily mobile in soils, which makes it prone to leaching and hence a suitable indicator for monitoring the U transfers from fertilizers to waters.

Finally, leaching of fertilizer derived U from soils to water bodies is expected to increase in the Northern hemisphere in the course of global climate change because of increased rainfall during summer.

Controlling loads of uranium originating from fertilizers

With view to the negative impacts of U on humans and environment the ‘Precautionary Principle’¹ should be applied in order to protect water bodies from anthropogenic U contamination. Particularly soils deserve to be protected from U contamination through fertilization, as they are the most vulnerable interface between agriculture and adjacent ecosystems. In this context it is more than surprising that U is the only toxic heavy metal for which no critical or guideline values in soils exist, which address the protection of soils and water bodies, respectively (Ekardt and Schnug 2008). Only Canada released most recently a soil quality guideline value for the protection of both human and environmental health of 23 mg/kg U soil with the restriction that a lower content may need to be considered on sites where drinking water is sourced (CCME 2007).

The most effective measure to limit loads of fertilizer derived U to soils is to regulate U concentrations in fertilizers. Uranium is easy to separate during the manufacturing process of fertilizers (Kratz and Schnug 2006; Hu et al. 2008) and then no longer threat to health and environment, but a source of energy and a resource for chemical processes (Lindemann 2007; Hauser and Meyer 2007; Hu et al. 2008). Regulating U in fertilizers would be a significant contribution to prevent disease in humans through healthy environments (Prüss-Üstün and Corvalán 2006).

References

- Azouazi M, Ouahidi Y, Fakhi S, Andres Y, Abbe J C, Benmansour M (2001) Natural radioactivity in phosphates, phosphogypsum and natural waters in Morocco. *Journal of Environmental Radioactivity* 54: 231-242.
- Barisic D, Lulic S, Miletic P (1992) Radium and uranium in phosphate fertilizers and their impact on the radioactivity of waters. *Water Research* 26: 607-611.

¹ “When an activity raises threats of harm to human health or the environment, precautionary measures should be taken even if some cause and effect relationships are not fully established scientifically” (Montague 2008)

- Birke M, Rauch U (2008) Uranium in stream water of the Federal Republic of Germany. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Bishop D (ed.) (2005) *Compendium of Uranium and Depleted Uranium Research 1942 to 2004*. <http://www.idust.net/Compendium/Compendium.htm>
- Blantz R C (1975) The mechanism of acute renal failure after uranyl nitrate. *J. Clin. Invest.* 55: 621-635.
- Bosshard E, Zimmerli B, Schlatter C (1992) Uranium in the diet: risk assessment of its nephro- and radiotoxicity. *Chemosphere* 24: 309-321.
- Busby C (2005) Does uranium contamination amplify natural background radiation dose to the DNA? *Eur. J. Biol. Bioelectromagn.* 1: 120-131.
- Busby C, Hooper M (2007) Final Report of the UK Ministry of Defence Depleted Uranium Oversight Board (www.duob.org), pp. 51-74.
- Busby C, Schnug E (2008) Advanced biochemical and biophysical aspects of uranium contamination. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- CCME (2007) Canadian Council of Ministers of the Environment Canadian Soil Quality Guidelines for Uranium: Environmental and Human Health. Scientific Supporting Document PN 1371. ISBN 978-1-896997-64-3 PDF http://www.ccme.ca/assets/pdf/uranium_ssd_soil_1.0_e.pdf
- Conceicao F T, Bonotto D M (2000) Anthropogenic influences on the uranium concentration in waters of the Corumbatai river basin (SP), Brazil. *Revista Brasileira de Geosciencias* 30: 555-557.
- Cothern C R, Lappenbusch W L (1983) Occurrence of uranium in drinking water in the U.S.. *Health Physics* 45:89-99.
- Ekarde F, Schnug E (2008) Legal aspects of uranium in environmental compartments. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Envirhom (2005) Bioaccumulation of radionuclides in situations of chronic exposure of ecosystems and members of the public. Progress Report 2 covering the period June 2003 – September 2005. Report DRPH 2005-07 & DEI 2005-05; DIRECTION DE LA RADIOPROTECTION DE L'HOMME DIRECTION DE L'ENVIRONNEMENT ET DE L'INTERVENTION
- FAL-PB (2007) unpublished data of the institutes FAL-PB, deriving from the project: Kördel W, Herrchen M, Müller J, Kratz S, Fleckenstein J, Schnug E, Saring Dr, Thoma J, Haaman H, Reinhold J (2007) Begrenzung von Schadstoffeinträgen bei Bewirtschaftungsmaßnahmen in der Landwirtschaft bei Düngung und Abfallverwertung. Forschungsbericht 202 33 305 und 202 74 271, UBA-FB 001017. UBA-Texte 30/07.
- Falck W E, Wymer D (2006) Uranium in phosphate fertilizer production. In: Merkel, B. J. and Hasche-Berger, A. (Eds.) *Uranium in the environment*. Springer, Berlin Heidelberg 2006, pp. 857-866.
- Fellows R J, Ainsworth C C, Driver C J, Cataldo D A (1998) Dynamics and transformations of radionuclides in soils and ecosystem health. *Soil Chemistry and Ecosystem Health*. Soil Science Society of America. Special Publication No.52, 85-112.
- Flamenboum W, Hamburger R J, Huddleston J, Kaufman J S, McNeil J H, Schwartz J H, Nagle R (1976) The initiation phase of experimental acute renal failure: an evaluation of uranyl nitrate-induced renal failure in the rat. *Kidney Int.* 10: 115-122.

- Hauser C, Meyer K (2007) Uranchemie zwischen Phobie und Begeisterung. *Nachrichten aus der Chemie* 55: 1195-1194.
- Henner P (2008) Bioaccumulation of radionuclides and induced biological effects in situations of chronic exposure of ecosystems – a uranium case study. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Heseker H (2005) Untersuchungen zur ernährungsphysiologischen Bedeutung von Trinkwasser in Deutschland. DGE-aktuell 01/2005 vom 12.01.2005. http://www.forum-trinkwasser.de/studien/Studie1/Studie1_Inhalt.htm
- Hu Z H, Zhang H X, Wang F, Haneklaus S, Schnug E (2008) Combining energy and fertilizer production – vision for China's future. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Hule B, Kummer S, Stadler S, Merkel B (2008) Mobility of uranium from phosphate fertilizers in sandy soils. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Jacques D, Šimůnek J, Mallants D, van Genuchten M (2005) Modelling uranium leaching from agricultural soils to groundwater as a criterion for comparison with complementary safety indicators. In: *Symposium Proceedings. Scientific Basis for Nuclear Waste Management XXIX*, Gent, Belgium, 12-16 September 2005 / SCK•CEN, Warrendale, United States, Materials Research Society, 2006, pp. 1057-1064.
- Jacques D, Mallants M, Šimůnek J, van Genuchten M (2008) Modelling the fate of U from inorganic P-fertilizer applications in agriculture. In: De Kok, L.J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Knolle, F. 2008. Ein Beitrag zu Vorkommen und Herkunft von Uran in deutschen Mineral- und Leitungswässern. PhD thesis, TU Braunschweig, Germany (in press).
- Kobal I, Vaupotic J, Mitic D, Kriston J, Ancik M, Jerancic S, Skofljanec M (1990) Natural radioactivity of fresh waters in Slovenia, Yugoslavia. *Environmental International* 16: 141-154.
- Kratz S, Schnug E (2006) Rock phosphates and P fertilizers as sources of U contamination in agricultural soils. In: Merkel, B. J. and Hasche-Berger, A. (Eds.) *Uranium in the environment*, pp. 57-68. Springer, Berlin.
- Kratz S, Knappe F, Schnug E (2008) Uranium balances in agroecosystems. In: De Kok L. J. and Schnug, E. (Eds) *Loads and Fate of Fertilizer-derived Uranium*. Backhuys Publishers, Leiden, The Netherlands.
- Lin R H, Wu L J, Lee C H, Lin-Shiau S Y (1993) Cytogenetic toxicity of uranyl nitrate in Chinese hamster ovary cells. *Mutation Research* 319: 197-203.
- Lin J H, Lin T H (1988) Renal handling of drugs in renal failure I: Differential effects of uranyl nitrate- and glycerol-induced acute renal failure on renal excretion of TEAB and PAH in rats. *J Pharmacol. Exp. Ther.* 246: 896-901.
- Lindemann I (2007) Futter für Dimona. *Strahlentelex* 496-497, 6-19. http://www.strahlentelex.de/Stx_07_496_S06-10.pdf
- Linsalata P (1994) Uranium and thorium decay series radionuclides in human and animal food chains--A review. *J. Environ. Qual.* 23 : 633-642.
- Milvy P, Cothorn C R (1990) Scientific background for the development of regulations for radionuclides in drinking water. In: *Radon, Radium and Uranium in Drinking Water* (Eds. C. R. Cothorn and P. Rebers), Lewis Publishers, Chelsea, Michigan.

- Montague P (2008) The precautionary principle in the real world. Environmental Research Foundation. January 21, 2008. http://www.rachel.org/lib/pp_def.htm; Wingspread Statement 2002. Statement on the Precautionary Principle. In: Rampton, S. and Staubner, J. (Eds), Trust us we're experts! Jeremy Putnam Inc., New York.
- NRC (2005) The Nuclear Regulatory Commission (NRC) invites comment on petition for rulemaking concerning chemical toxicity of uranium. Docket No. PRM-20-26. Federal Register: June 15, 2005 (Volume 70).
- Pais I, Benton Jones jr. J (1997) Uranium. In: The handbook of trace elements. St. Lucie Press, Boca Raton, Florida, pp 143-144.
- Prüss-Üstün A, Corvalán A (2006) Preventing disease through healthy environments: Towards an estimate of the environmental burden of disease. ISBN 978 92 4 159420 2. http://www.who.int/quantifying_ehimpacts/publications/preventingdisease/en/index.html
- Read D, Black S, Trueman E, Arnold T, Baumann N (2008) The fate of depleted uranium in near surface environments. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- Rogasik J, Kratz S, Funder U, Panten K, Schnug E, Barkusky D, Baumecker M, Gutser R, Lausen P, Scherer H W, Schmidt L (2008) Uranium in soils of German long-term fertilizer trials. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- Schäf M, Daumann L, Erdinger L (2007) Uran in Trinkwasserproben im Rhein-Neckar Gebiet. Umweltmed Forsch Prax 12, 315.
- Schick J, Schroetter S, Lamas M C, Rivas M C, Kratz S, Schnug E (2008) Soil/plant interface of U. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- Schmitz-Feuerhake I, Bertell R (2008) Radiological aspects of uranium contamination. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- Schnug E, Steckel H, Haneklaus S (2005) Contribution of uranium in drinking waters to the daily uranium intake of humans - a case study from Northern Germany. Landbauforsch Völkenrode 55, 227-236. http://www.fal.de/nn_787874/SharedDocs/01__PB/DE/Downloads/LBF/2005/downloads-pb1900.templateId=raw.property=publicationFile.pdf/downloads-pb1900.pdf
- Schulz C, Rapp T, Conrad A, Hünken A, Seiffert L, Becker K, Seiwert M, Kolossa-Gehring M (2008) Elementgehalte im häuslichen Trinkwasser aus Haushalten mit Kindern in Deutschland. Forschungsbericht 202 62 219 UBA-FB 001026. WaBoLu Hefte 04/08. ISSN 1862-4340
- Sharpley A N, Withers J A (2004) The environmentally-sound management of agricultural phosphorus. Nutrient Cycling in Agroecosystems 39: 133-146
- Spalding RF, Sackett W M (1972) Uranium runoff from the Gulf of Mexico. Distribution province anomalous concentration. Science 175 : 629-631.
- Taylor M, Kim N (2008) The fate of uranium contaminants of phosphate fertilizers. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- Thiebault C, Carrière M, Milgram S, Gouget B (2006) Cytotoxicity and genotoxicity of natural uranium after acute or chronic exposures of normal rat kidney cells in favour of a cell transformation? Toxicology Letters 175: 14-23.

- Utermann J, Fuchs M (2008) Uranium contents in German soils. In: De Kok, L.J. and Schnug, E. (Eds) Loads and Fate of Fertilizer-derived Uranium. Backhuys Publishers, Leiden, The Netherlands.
- WHO (2004) Uranium in Drinking-water, Background document for development of WHO Guidelines for Drinking-water Quality, World Health Organization.
- Zamora M L, Tracy B L, Zielinski J M, Meyerhof D P, Moss M A (1998) Chronic ingestion in drinking water : a study of kidney bioeffects in humans. *Toxicol. Sci.* 43: 68-77.
- Zielinski R A, Asher-Bolinder S, Meier A L (1995) Uraniferous waters of the Arkansas River valleys, Colorado, USA: A function of geology and landuse. *Applied Geochemistry* 10: 133-144.