



Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpolUranium in agricultural soils and drinking water wells on the Swiss Plateau[☆]Moritz Bigalke^{a,*}, Lorenz Schwab^a, Agnes Rehmus^a, Patrick Tondo^b, Markus Flisch^b^a Institute of Geography, University of Bern, Hallerstr. 12, 3012 Bern, Switzerland^b Laboratory of the Canton of Bern, Muesmattstrasse 19, 3012 Bern, Switzerland

ARTICLE INFO

Article history:

Received 26 May 2017

Received in revised form

13 September 2017

Accepted 19 September 2017

Available online xxx

Keywords:

Uranium

Mineral fertilizer

Soil

Water

Activity ratio

ABSTRACT

Mineral phosphorus fertilizers are regularly applied to agricultural sites, but their uranium (U) content is potentially hazardous to humans and the environment. Fertilizer-derived U can accumulate in the soil, but might also leach to ground-, spring and surface waters. We sampled 19 mineral fertilizers from the canton of Bern and soils of three arable and one forest reference sites at each of four locations with elevated U concentrations (7–28 $\mu\text{g L}^{-1}$) in nearby drinking water wells. The total U concentrations of the fertilizers were measured. The soils were analysed at three depth intervals down to 1 m for general soil parameters, total Cd, P, U and NaHCO_3 -extractable U concentrations, and $^{234}\text{U}/^{238}\text{U}$ activity ratios (AR). The U concentrations and AR values of the drinking water samples were also measured. A theoretical assessment showed that fertilizer-derived U may cause high U concentrations in leaching waters (up to approx. 25 $\mu\text{g L}^{-1}$), but normally contributes only a small amount (approx. 0–3 $\mu\text{g L}^{-1}$). The arable soils investigated showed no significant U accumulation compared to the forest sites. The close positive correlation of AR with NaHCO_3 -extractable U ($R = 0.7$, $p < 0.001$) indicates that application of fertilizer can increase the extractable U pool. The lack of depth gradients in the soil U concentrations (1.5–2.7 mg kg^{-1}) and AR (0.90–1.06) ratios are inconsistent with the accumulation of U in the surface soil, and might indicate some leaching of fertilizer-derived U. The AR values in the water samples were close to 1, possibly suggesting an influence of fertilizer-derived U. However, based on findings from the literature and considering the heterogeneity of the catchment area, the agricultural practices, and the comparatively long distance to the groundwater, we conclude that fertilizer-derived U makes only a minor contribution to the elevated U concentrations in the water samples.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Mineral based phosphorous fertilizers are regularly applied to many agricultural sites, but often contain high concentrations of U. Depending on the soil conditions, fertilizer-derived U might be mobile and contaminate ground- and surface waters (Birke and Rauch, 2008; Huhle et al., 2008; Schnug and Lottermoser, 2013). Finally, the U may end up in drinking water, which is the major source for human uptake of toxic U (65%–95% of total U uptake; Schnug, 2012).

Mineral based phosphorous fertilizers are produced from phosphate rocks of igneous or sedimentary origin. Depending on

the source, sedimentary phosphorites are heavily enriched in U (up to 280 mg kg^{-1} ; McLaughlin et al., 1996). During the process of fertilizer production, U is enriched in the fertilizers by up to 150% (Sattouf, 2007). Thus, different types of fertilizers (e.g. triple superphosphate and diammoniumphosphate) may have very different U contents (Table S1) depending on the source and type of production. When mineral based fertilizers are applied to soil, they are rapidly solubilized and the mobilized U can be sorbed to soil components, precipitated, or leached. Uranium binds to organic matter, Al and Fe oxides, clay minerals and can bind to or being incorporated into soil organisms (Echevarria et al., 2001; Zheng et al., 2003; Zielinski et al., 2000). For soils with high clay and organic-matter content, U is anticipated not to migrate substantially (Rogasik et al., 2008). Accordingly, several studies have shown that long-term fertilization with mineral based P-fertilizers leads to substantial accumulation of U in soils, particularly topsoil, compared to unfertilized soils (Bigalke et al., 2017; Jones, 1992;

[☆] This paper has been recommended for acceptance by Prof. W. Wen-Xiong.

* Corresponding author.

E-mail address: moritz.bigalke@giub.unibe.ch (M. Bigalke).

Rogasik et al., 2008; Schipper et al., 2011; Takeda et al., 2005, 2006; Taylor, 2007; Wetterlind et al., 2012; Yamaguchi et al., 2009; Zielinski et al., 2006). In accordance with these findings, a number of authors have pointed to regional variations in concentrations of U in drinking water generally related to bedrock composition, even if the direct relationship might be complicated by groundwater flow through different rock formations (CCME, 2011; Stalder et al., 2012). However, a number of studies also found indications of U transfer from fertilizers to water bodies (Azouazi et al., 2001; Barisic et al., 1992; Conceicao and Bonotto, 2003; Huhle et al., 2008; Smidt et al., 2012; Zielinski et al., 2000). The influence of fertilizer-derived U inputs can be tested by comparing fertilized and unfertilized reference sites (Birke and Rauch, 2008; Huhle et al., 2008; Rogasik et al., 2008) or from correlations between U and other fertilizer-derived components such as nitrate, K and B, especially in shallow drinking waters at agricultural sites (Knolle, 2008; Popit et al., 2004; Schäf et al., 2007; Smidt et al., 2012).

One important reason for the different findings in terms of fertilizer derived U transfer to groundwaters is the U mobility in soils. The pH is one of the most important drivers of U mobility in soils (Echevarria et al., 2001; Tyler and Olsson, 2001). The lowest mobility of U is around pH 7 but mobility increases with decreasing pH because of decreasing number of binding sites and changes in U speciation. Depending on pH and the presence of organic and inorganic ligands such as phosphates (PO_4^{3-}), carbonates (CO_3^{2-}), sulphates (SO_4^{2-}), chlorides (Cl^-) or fluorides (F^-) U forms stable complexes of different mobilities (Bourdon et al., 2003; Vandenhove et al., 2007). Especially under slightly alkaline and oxic conditions – as they are common in many Swiss agricultural fields – U is therefore quite mobile (Jacques et al., 2006). The presence of carbonates particularly increases the mobility of U in alkaline soils because U–carbonate complexes are predominantly neutral or negatively charged and have a low affinity for soil minerals (Echevarria et al., 2001; Read et al., 2008; Zheng et al., 2003). Besides pH and complexing agents the amount of possible binding sites (organic material, Fe oxides and clay) controls the possible U sorption (Rogasik et al., 2008). Depending on soil properties, U does or does not significantly accumulate in soils but is readily transferred to aquifers (Hamamo et al., 1995; Mortvedt, 1994), especially if the ground water table is close to the soil surface (Huhle et al., 2008).

The $^{234}/^{238}\text{U}$ alpha activity ratio (AR) is a tool to relate U to its source directly. In an undisturbed closed system, as is assumed in a phosphate rock deposit, a secular equilibrium becomes established and AR levels approach 1 (Fig. 1; Bourdon et al., 2003; Keith et al., 2007). Conversely, recoil processes and the spontaneous oxidation of U^{4+} to U^{6+} during decay are reasons for preferential leaching of ^{234}U from soil, leaving it with $\text{AR} < 1$ and the leaching water with $\text{AR} > 1$ (Osmond and Cowart, 1976). The extent of $^{234}/^{238}\text{U}$ disequilibrium depends on the characteristics of the disturbance and values of 0.8–10 in waterbodies, 0.8–8 in precipitation and 0.5–1.2 in soil have been reported (Keith et al., 2007). The contribution of fertilizer-derived U to soils, surface and groundwater can therefore be recognized by a shift in AR towards 1, which has been successfully used in a number of studies to trace the sources of U in soil and water (Conceicao and Bonotto, 2003; Zielinski et al., 1997, 2000, 2006).

To study the influence of fertilizer-derived U on concentrations in soils and its possible leaching, we sampled soils from four agricultural catchments that showed elevated U concentrations in drinking water wells. These areas are under intensive agricultural usage and there is no obvious source of the high U concentrations in the water wells. The aims of this study were to determine:

1. Whether fertilizer-derived U has the potential to cause significantly elevated U concentrations in ground- and surface waters,
2. Whether there is any indication of fertilizer-derived U enrichment in the agricultural soils and,
3. Whether there is any indication of U leaching from the soils.

2. Material and methods

2.1. Study sites

Four study locations in the canton of Bern, Switzerland were chosen because of the occurrence of elevated U concentrations in ground- and spring waters (Table 1) nevertheless the geology is very similar to other sites with low U in waters. The locations Oberwil bei Büren (pumping station at Rossmatt), Oberwil bei Büren (well at Rüti), Ins (well) and Walliswil (well) were selected. All locations are in the Swiss molasse basin (Fig. S1). The agricultural fields and the nearby forest sites have similar underlying geology (www.geo.admin.ch). The two locations in Oberwil bei Büren (Rossmatt and Rüti) are close to each other and the area is underlain by Quaternary unconsolidated rock, mainly till from the last glaciation (Würm). The forest at Rüti covers the same formation, and the forest at Rossmatt lies on the same formation but is close to Tertiary molasse (Lower Freshwater Molasse) of the Zinshölzli Formation under a thin Quaternary cover. The pumping station at Rossmatt pumps water from 40 m depth. The water permeability is classified as normal for all fields, and the water-retention capacity is moderate to good. In Ins the agricultural fields and the forest site are situated on grey-green shell sandstones and marl of the Upper Marine Molasses. The springs are located on clayey marls, freshwater limestone and grey sandstones. The water permeability is characterized as normal with a good water-retention capacity. In Walliswil the area contains Quaternary unconsolidated rocks, probably from the last glaciation (Würm). The water permeability for the Walliswil sites is classified as excessive and the water-retention capacity as low. However, the forest site in Walliswil has a loose surface layer, the highest C_{org} concentrations, and a name indicating a historical peat land (Dängelmoos). In addition, local knowledge indicates that the forest site receives significant amounts of drainage water/surface runoff from other agricultural and forested sites. The water chemistry of all waters is dominated by dissolved Ca^{2+} (90–131 mg L^{-1}) and Mg^{2+} (5.9–26.9 mg L^{-1} ; total hardness 3.3–3.7 mmol L^{-1}), with less Na (3.3–16 and K (0.9–4.6) and has a neutral to slightly alkalic pH. The dominating soils are Cambisols, while at some sites also Luvisols might have formed (FAO, 2006). The depth of the ploughed surface horizon is between 25 and 30 cm, regular manure application is likely on most of the fields. Irrigation of the field sites is unlikely as mainly crops are grown which are not irrigated in this part of Switzerland (wheat, corn, grass etc.; Fuhrer, 2014).

2.2. Sampling

At each study site the soils of three agricultural fields and one forest were sampled. The arable sites were always located in the direct water protection area (inflow area) of the corresponding drinking water well. The information about the inflow areas are taken from the water protection maps (geoadmin.ch), and are mainly based on tests with fluorescent tracers in the past (personal communication Bruno Schlup, responsible person for the Rüti Well). One nearby forest site was chosen at every location as an unfertilized reference site. At each of the sites soil was sampled with a soil auger down to 1 m depth in ten replicates evenly distributed over the sites. The samples from all sites were separated

Download English Version:

<https://daneshyari.com/en/article/8857583>

Download Persian Version:

<https://daneshyari.com/article/8857583>

[Daneshyari.com](https://daneshyari.com)