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Indications for phosphorus fertilizer-derived uranium mobilization from arable soils to groundwater

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Introduction

The production of phosphorus (P) fertilizers requires raw phosphate rock material, which originates to about 87 % from marine sedimentary sources. Uranium (U) and many trace elements are enriched in these phosphates during deposition and diagenesis. Average U concentrations in P-fertilizers traded in Germany in 2007 of up to 283 mg kg^{-1} P_2O_5 indicate the potential of U contamination of the environment (Smidt et al., 2011a). The cumulative load of U from P fertilization to German arable soils was calculated to be 0.5 -1 kg ha^{-1} 54 $yr^{-1.}$ Several studies found indications of a higher U content in top soils compared to sub soils. The difference of the median values in forest and arable topsoils is 0.15 mg kg^{-1} U (Utermann and Fuchs, 2008). Under oxidizing conditions the hexavalent form of U is the most common, which occurs as the bivalent uranyl ion (UO_2^{2+}). It forms complexes with carbonate ($UO_2(CO_3)_2^{2-}$ or $UO_2(CO_3)_3^{4-}$), which are highly mobile in soil based on their low adsorption onto soil particles/colloids due to their negative charge. The anthropogenic loads of U to arable soils and its mobility might induce a contamination of groundwater and thus a hazard for the quality of drinking water.

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Material and Methods

Soil and drainage water samples were taken from wells and pipes of long-time soil-monitoring fields (Bodendauerbeobachtungsflächen - BDF) operated by the Agency for Mining, Energy and Geology in Lower Saxony (LBEG). The soil at Hornburg (BDF-H) site is a chernozem; it is used intensively for cropping production with high amounts of P-fertilizers. The Mariental site (BDF-M) is a stagnosol, which is also used for intense cropping production, but with a lower fertilization rate. The location of these sites close to the former German-German border allowed us to take soil samples from the still existing “Green Belt (GB)”, which was not fertilized for 65 years. This strip across Germany was kept free of trees during the period of two separated German states and provided us an optimal control site for comparison. Groundwater samples were collected from wells for the European Water Framework Directive by the Lower Saxon State Department for Waterway, Coastal and Nature Conservation (NLWKN) in an area of intense agricultural production known as the Hildesheimer Börde in Lower Saxony. The samples were analyzed according to DIN procedures for U, NO_3, NO_2, NH_3, ortho-P, F, Cl, SO_4, Ca, Mg, K, Na, Fe, Mn, Al, Pb, Cu, Cd, Zn, Ni, DOC, pH, EC and O_2.

Results and Discussion

The sites show different U concentration in drainage water samples (BDF-H: max. 4.9 and BDF-M: max. 0.3 µg kg^{-1}) as well as in the groundwater samples (BDF-H: 1.8 and BDF-M: 0.4 µg kg^{-1}). The total concentration of the BDF and GB topsoils ranges between 1.5 and 2.25 mg kg^{-1} U. The arable topsoils showed 0.15-0.30 mg kg^{-1} higher U concentration compared to the unfertilized GB soils. Batch experiments were performed to extract the easily mobilizable uranium fraction from the BDF and GB soils with 0.1M $NaHCO_3$ solution at a pH of 8.3 and liquid to solid ratio of 100ml/2g (Zielinski et al., 2006). The extracts from fertilized soils show higher U concentrations than the Green Belt soils. This result indicates that fertilizer-derived U is easier mobilized from top soils and hence might be leached faster to the deeper soil horizons and groundwater than the geogenic U in soils.

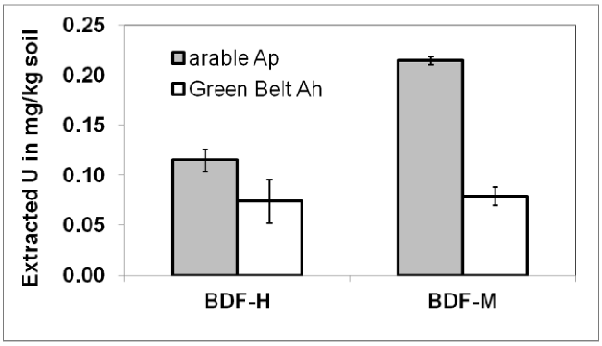

Fig. 1: Extractable (0.1M $NaHCO_3$) Uranium [mg kg^{-1} soil] from fertilized arable and unfertilized (Green Belt) top soils from long time soil monitoring fields in Lower Saxony

The groundwater samples (N=168) from the groundwater monitoring network wells (0-56 m sampling depths) in the Hildesheimer Börde show a wide concentration range of U (0.001 – 17.9 µg kg^{-1}). The large groundwater dataset was statistically analyzed and geochemical variables correlating with U were tested for interrelation using Principle Component Analysis (PCA). Two components were extracted; one is loading U together with the alkaline earth metals Ca and Mg, showing the geogenic U species in groundwater probably leached as carbonate-complexes. The second component is loading U with nitrate and oxygen. High nitrate concentrations in groundwater are the result of intensive agricultural fertilization. The relation between nitrate and U in groundwater is even more distinct in shallow wells (<15m) indicating the presence of anthropogenic fertilizer-derived U in the groundwater (Smidt et al., 2011b).

Conclusion

The results of soil, soil extract, drainage water and groundwater analyses show the high mobility of fertilizer-derived U and give indications for its leaching to groundwater aquifers. Fertilizer-derived U is a potential hazard for our drinking water resources.

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