

## A time-series phytoremediation experiment with sunflowers (*Helianthus annuus*) on a former uranium mining site

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**Abstract.** On a test field situated at a former uranium mining site near Ronneburg (Thuringia, Germany) a small scale time-series field experiment with sunflowers (*Helianthus annuus*) was carried out. This area has elevated contents for the heavy metals Cd, Co, Cr, Cu, Ni, Zn including the radionuclides U and Th. Over a time period of 24 weeks the sunflowers were cultivated on homogenized soil substrate and regularly harvested. The aim was to find the ideal moment to harvest the sunflowers, being defined as having the best balance between the extraction of the contaminants and a high biomass produced. The contents of the elements were determined in soil, roots and above-ground plant parts. The contents in the above-ground plant showed no clear increasing or decreasing trend over time, so they were not the appropriate values to determine the best moment to harvest. Instead the total extracted masses (content in  $\mu\text{g/g}$  x biomass in g) of the contaminants in the above-ground plant parts were calculated. According to this the best moment to harvest the sunflower plants was reached after 24 weeks of vegetation, because the highest extracted masses for all contaminants were calculated to this time. Additionally the biomass, which could be used e.g. for bio-fuel production, was highest at this time.

**Key words:** Heavy metals, time-series experiment, sunflower, phytoremediation

### Introduction

About 0.5 % of the area in Europe are contaminated by mining activities, whereby mining waste amounts to about 30% of waste produced in Europe (Piiro, 2010). Mined areas are often characterized by contamination with heavy metals, acidic soil pH, and low nutrient content. The classical methods used to remediate such areas are excavation and land-filling or washing with chemical solvents. So either huge amounts of soil have to be moved and suitable storage areas have to be found or much solvent has to be applied, which damages the soil and decreases its fertility (Singh and Prasad, 2011). Due to these negative impacts new in-situ decontamination methods should be investigated. Phytoremediation is a promising method to remove contaminants from soil in an eco-friendly and soft way. In addition it enhances the soil functionality and protects it from erosion. One further advantage is that these areas, which are not suitable for food crops can be used to grow plants for the bio-fuel production. In the area around Ronneburg (Thuringia, Germany) from 1946-1990 low grade

uranium ores were mined and leached. Altogether about 113,000 t of U were produced over the time. After 1990 the site was remediated by removing and filling the leached ores in open pit mines. New soil was applied and the landscape reconturized. The sealing of the former leaching heap was leaky, so heavy metal and radionuclide containing acid mine drainage could seep in the underlying basement. As a consequence the newly applied soil was again contaminated due to capillary rising. Nowadays the soil shows elevated contents for the heavy metals Cd, Co, Cr, Cu, Ni, and Zn and the radionuclides U and Th. On the area of the former leaching heap the test field "Gessenwiese" (2500 m<sup>2</sup>) was installed in 2004 (Grawunder et al., 2009). On a subplot (9 m<sup>2</sup>) at the test site filled with homogenized soil substrate, sunflowers were tested for their phytoextraction potential. Thereby one main question raised: When is the ideal moment to harvest the plants? Does it make sense to sow twice times a year, since over a vegetation period of e. g. three month the maximum extraction is reached and after that time only more biomass is formed? Is there only a "dilution" by biomass

production after an initial uptake? Or does it make sense to grow plants over 6 months since they form more biomass and in addition accumulate more contaminants? Although this problem is very obvious and important to answer, no hints can be found in literature. Often the harvesting time is selected randomly and no explanation why the harvest is done at a certain time can be found (Madejón et al., 2003; Marchiol et al., 2007; Sabudak et al., 2007)

## Materials and Methods

The study area is situated in middle-east of Germany near Ronneburg. As a result of former uranium mining activities the soil shows elevated contents for many heavy metals and radionuclides. The contents of the investigated elements Cd, Co, Cr, Cu, Ni, Zn, U, and Th in the homogenized soil substrate on which the sunflowers were planted can be found in Tab. 1. These total contents of the contaminants in the sandy loam soil were determined with ICP-MS after digestion with HF (suprapur, 40 %), HClO<sub>4</sub> (suprapur, 70 %), and HNO<sub>3</sub> (subboiled, 65 %) in a PicoTrace (DAS 30) apparatus. The soil has a pH about 4.6-5.0 (H<sub>2</sub>O) and an organic carbon content < 0.1%.

Tab. 1 Total element concentrations in soil ± confidence interval

Element	Concentration in µg/g
Cd	0.72 ± 0.04
Co	26.2 ± 0.1
Cr	45.8 ± 0.6
Cu	29.8 ± 0.3
Ni	54.8 ± 0.6
Zn	74.8 ± 0.4
Th	10.1 ± 0.1
U	4.68 ± 0.1

We selected sunflower (*Helianthus annuus*), based on preliminary experiments in this area which showed that they germinate and grow well on the contaminated, acidic soil substrate. We grew them over 24 weeks, which equals a normal vegetation period in agricultural use. During this time we took samples at least every five weeks samples. The plants were separated in above-ground parts and roots, three times washed with deionized water and dried at 40°C to weight constance. Following they were ground (RETSCH, MM400) and digested with HNO<sub>3</sub>, (subboiled, 65 %) in a microwave oven (CEM, MARSXPRESS). The concentrations of the elements in the digests were determined by ICP-MS (X-Series II, ThermoFisher Scientific).

## Results and Discussion

After 24 weeks of vegetation no toxicity symptoms were visible on the sunflowers. The experiment was finished after this time, because the sunflowers withered.

Focusing on the variation of the contents in the above-ground plant parts an ideal moment to harvest the sunflowers, that would be indicated by highest contents for all elements at a certain time, could not be determined. Over this time no clear decreasing or increasing trend for all elements is visible (Fig. 1). Cd, Cr and Zn, reach the highest content already after 5 weeks of vegetation, whereas the contents of Co, Cu, and Ni are highest after 20 week. U and Th have the highest values after 15 weeks of growth and are lower before and after this time. The biomass was about 0.7 g after five weeks, 2.1 g after 15 weeks increasing to 5 g dry weight per plant after 24 weeks of growth. Because of this contrasting behavior we calculated the total extracted mass of the elements (content in µg/g x biomass in g). The results (Fig. 2) indicate that the maximum extracted mass for all elements is reached at the 24<sup>th</sup> week of the vegetation period. At this time the extracted mass e.g. for Cu is about ten times higher than after five weeks and two times higher compared to 15 weeks of vegetation. An exception are Th and U, both reach their highest extraction already after 18 weeks. The sunflowers extracted the maximum of the contaminants after a vegetation period of 24 weeks and thereby also reached the highest biomass. So the sunflowers should be sown only once a year and cultured over 24 weeks, since with this approach the highest amount of contaminants can be removed. The exceptional extraction behavior of Th and U is not understood yet and will be investigated further.

## Conclusion

The time-series phytoremediation experiment showed that sunflowers have a time dependent extraction behavior for the investigated elements. The contents of the elements alone are not the appropriate parameters to determine the ideal moment to harvest the sunflowers. The total extracted masses of the elements have to be calculated instead. We found that after 24 weeks of growth the extracted amount of elements is highest, due to a higher biomass production, even if the content in this time decreases. Nevertheless this can only be stated for sunflower and should be investigated for other plants, which seem to be promising candidates for phytoremediation.

## Acknowledgements

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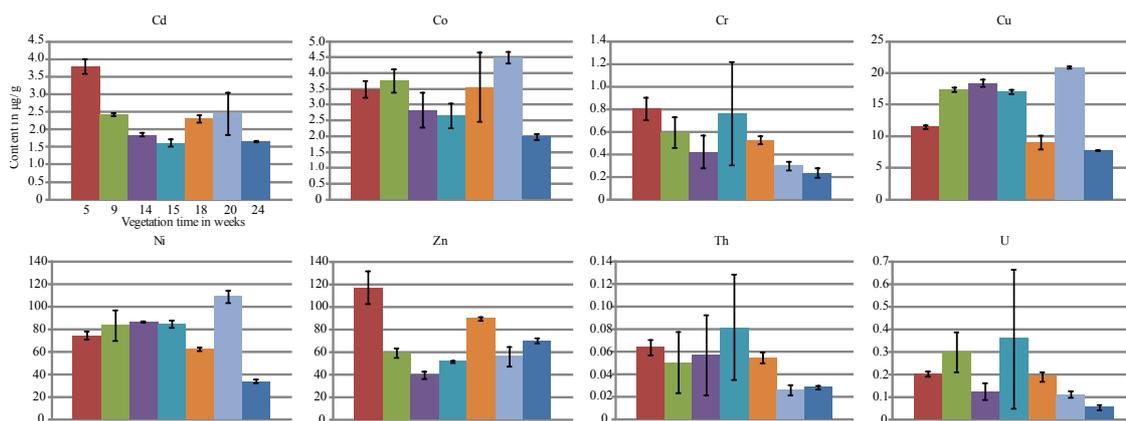


Fig 1. Concentration of the contaminants in the above-ground plant parts of sunflowers at different times of sampling (mean ± confidence interval; two replicates)

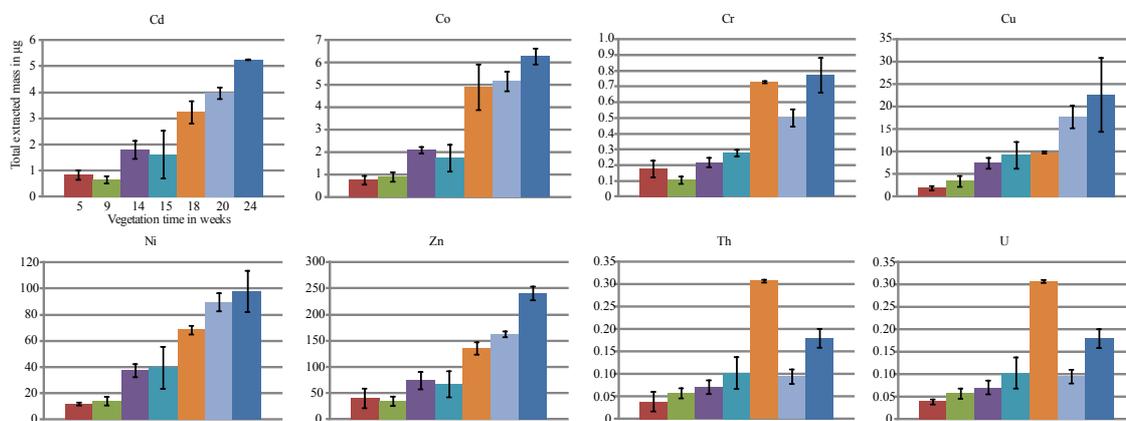


Fig. 2 Total extracted mass of the contaminants in the above-ground plant parts of sunflowers at different times of sampling (mean ± confidence interval; two replicates).

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