

# Geochemical Soil Atlas of Switzerland – Distribution of Toxic Elements

Jolanda E. Reusser\*, Maja B. Siegenthaler, Lenny H. E. Winkel, Daniel Wächter, Ruben Kretzschmar, and Reto G. Meuli

**Abstract:** Chemical elements such as copper and molybdenum are essential for animal and human health but may become toxic at elevated concentrations depending on the exposure and intake rate. Other elements such as mercury pose a threat to human health at already low concentrations. The soil acts as the main source of these elements for plant uptake and is thus driving accumulation along the food chain. However, in Switzerland, no nationwide information on elemental distributions in soils has existed up to now. The geochemical soil atlas of Switzerland will fill this gap by presenting the concentration ranges and the spatial distribution of 20 elements in the topsoil. In this summary, we present the methodological approaches and some main findings of the atlas with a focus on toxic elements as well as elements that can be or are toxic at higher concentrations.

**Keywords:** Geochemistry · Soil chemistry · Soil contamination · Soil mapping · Trace elements

**Jolanda E. Reusser** is an environmental engineer and received her PhD on the speciation of phosphorus compounds in soil using nuclear magnetic resonance spectroscopy at ETH Zurich. She worked as a postdoctoral researcher on the ‘Geochemical Soil Atlas of Switzerland’ project within the Swiss Soil Monitoring Network (NABO) at Agroscope, Zurich-Reckenholz. Currently, she is employed as a postdoctoral researcher in the group of Inorganic Environmental Geochemistry at ETH Zurich and in the group of Soil Quality and Soil Use at Agroscope.

**Maja B. Siegenthaler** is an agricultural scientist and conducted her PhD on phosphorus cycling in forest ecosystems at ETH Zurich. She worked as a postdoctoral researcher with a laboratory focus on the ‘Geochemical Soil Atlas of Switzerland’ at Agroscope, Zurich-Reckenholz, and is now investigating soil selenium speciation at Eawag, the Swiss Federal Institute of Aquatic Science and Technology.

**Lenny H. E. Winkel** is an Associate Professor of Inorganic Environmental Geochemistry at ETH Zurich and Eawag. She obtained her PhD in Geochemistry at ETH Zurich in 2006. After postdoctoral appointments in France, UK, and Greece, in the frame of an EU project, she was awarded a SNSF professorship in 2011 and promoted to Associate Professor at ETH Zurich in 2018. Her current research is aimed at understanding the processes controlling the biogeochemical cycling and environmental distribution of trace elements through a combination of field, lab, and modelling approaches.

**Daniel Wächter** holds an engineer (FH) degree in Environmental Engineering from the University of Applied Sciences (ZHAW). He recently transitioned to a role as a Data Management Specialist at the Swiss Competence Center for Soil (KOBO), part of the Bern University of Applied Sciences, starting in August 2023. Prior to this, he worked at Agroscope, Zurich-Reckenholz, where he was responsible for quality assurance within the Swiss Soil Monitoring Network (NABO).

**Ruben Kretzschmar** is a soil scientist specialized in environmental soil chemistry and mineralogy. He received his PhD in 1994 at North Carolina State University, NC, USA, and has been a Professor of Soil Chemistry in the Department of Environmental Systems Science at ETH Zurich since 1999. His recent research focused on the speciation and bioavailability of trace elements in redox-affected soils and their coupling to major elements such as iron, manganese, sulfur, and carbon.

**Reto G. Meuli** is a physical geographer specialized in soil science. After receiving his PhD from ETH Zurich in 1997 on ‘Geostatistical analysis of regional soil contamination by heavy metals’, he worked as a soil- and contaminated site specialist in Switzerland. From 2008 to 2023, he was the head of the research group Swiss Soil Monitoring Network (NABO) at Agroscope, Zurich-Reckenholz. He was also a board member of the Soil Science Society of Switzerland (2010–2022), of which he was President in 2012–2014.

## 1. Soil – Source of Essential and/or Toxic Elements

Elements such as calcium and magnesium are essential for organisms, others like mercury and cadmium can pose a health threat even at low concentrations.

The plant uptake from the topsoil (0–20 cm) is considered an important driver for the enrichment of essential and/or toxic elements in animal and human nutrition. In soils, elements originate from various sources, such as weathering of the bedrock or inputs from atmospheric deposition, and are influenced by different mobilisation and immobilisation processes<sup>[1]</sup> (Fig. 1).

The quantity of an element taken up by plants not only depends on its chemical form and bioavailability but also on its total concentration in soil. Total element concentrations exhibit great spatial variability due to the various sources and sinks. The spatial distribution of elements in soils can be assessed and visualised in geochemical soil atlases, for example, the geochemical soil atlas of England and Wales<sup>[2]</sup> or the geochemical soil atlas of Europe,<sup>[3]</sup> in which Switzerland is represented by 17 sampling sites. Based on the spatial distribution, regions with low concentrations of essential elements or toxic concentrations of elements can be defined. Therefore, geochemical soil atlases serve as a basis to develop measures for soil protection and protection of plant, animal, and human health in specific areas. Furthermore, important back-

\*Correspondence: Dr. J. E. Reusser, E-mail: jolanda.reusser@usys.ethz.ch  
Dept. Environmental Systems Science, ETH Zurich, CH-8092 Zurich

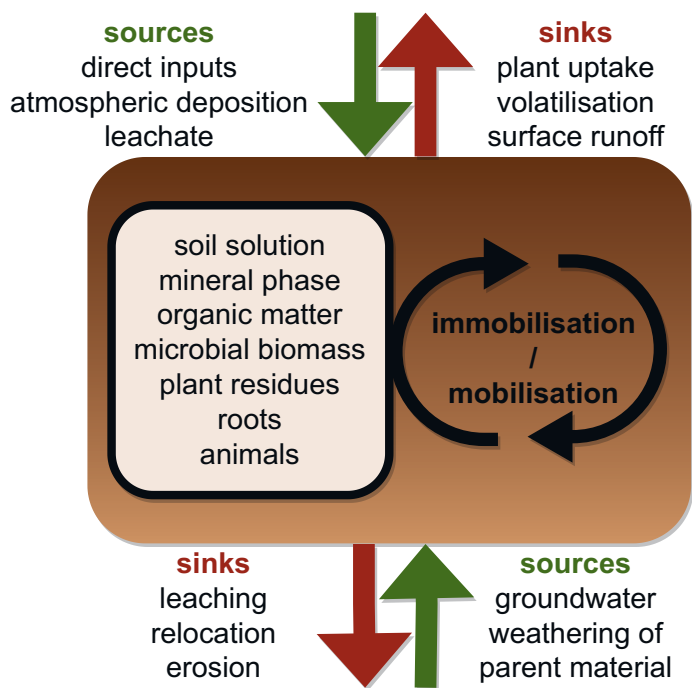


Fig. 1. Topsoil with examples of sources and sinks of elements as well as soil constituents influencing the mobility of elements in the topsoil through processes such as sorption/desorption, incorporation/mineralisation, chemical precipitation/solubilisation, etc. Sources are of geogenic (e.g. parent material), biogenic (e.g. manure) and anthropogenic (e.g. fertilisers) origin.

ground information can be derived to assess trace element cycling through soils as well as their potential effects on ecosystem health, forests, and water resources quality.

To date, no highly resolved geochemical soil atlas of Switzerland existed. Thus, the main aim of the geochemical soil atlas was to assess the concentrations of 20 elements in Swiss topsoils including both essential and/or elements that are toxic or can become toxic at higher concentrations (Fig. 2). The distribution of these elements among six different land use types was assessed (settlements, arable fields, meadows and pastures, forests, alpine pastures, unproductive alpine areas) and the spatial distribution based on a resolution of approx. one sampling site per 35 km<sup>2</sup> was visualised. The results and maps are published in the first geochemical soil atlas of Switzerland.<sup>[4]</sup> The sampling design, methods and main findings of the atlas as well as resulting future challenges are summarised in the following sections. Furthermore, additional information on the distribution of toxic elements

is provided for which threshold values were defined by the Swiss Ordinance on the Pollution of Soil (VBBo)<sup>[5]</sup> and by Eikmann and Kloke.<sup>[6]</sup>

## 2. Assessing Element Concentrations in Soils

To facilitate comparability, the methods used for the geochemical soil atlas of Europe<sup>[3]</sup> were also used for the geochemical soil atlas of Switzerland.

### 2.1 Soil Sampling

The geochemical soil atlas of Switzerland represents a total of 1'201 sampling sites. Most of these sites (1'082) were sampled between 2011 and 2015 within the framework of the Swiss Biodiversity monitoring BDM.<sup>[7]</sup> The sampling sites of the BDM are distributed on a regular 6 x 4 km grid across whole Switzerland. Four individual samples have been collected at each sampling site from a depth of 0–20 cm with an impact probe (diameter of 4.8 cm). The individual samples were evenly distributed on a circle of 3–3.5 m radius.<sup>[7]</sup>

The BDM sampling sites were complemented with 102 sampling sites of the Swiss Soil Monitoring Network NABO.<sup>[8]</sup> For the NABO sites, a composite soil sample was collected at each site (0–20 cm) consisting of 25 individual soil samples evenly distributed on a 10 x 10 m grid.

In addition, the 17 Swiss sampling sites of the European geochemical soil atlas GEMAS have been included in the dataset.<sup>[3]</sup> GEMAS sampling was based on a 50 x 50 km grid across Europe. At each sampling site, five subsamples were taken in a 10 x 10 m square and combined into a composite sample. The GEMAS total element concentrations in aqua regia digests were previously published in Reimann *et al.*<sup>[3]</sup>

### 2.2 Soil Preparation and Analyses

Equal to the soil analyses performed for the European geochemical soil atlas GEMAS<sup>[3]</sup> the collected soil samples were dried (40 °C, 48 h), sieved (< 2 mm) and milled prior to digestion with a modified aqua regia solution (HNO<sub>3</sub>:HCl:H<sub>2</sub>O, 1:1:1). The digestion and subsequent inductively coupled plasma mass spectrometry (ICP-MS) analyses were carried out in the laboratory 'Bureau Veritas Minerals' in Vancouver (Canada). In general, less than 0.5% of all measured values were below the limit of detection (LOD). The exclusion of values below the LOD could cause a bias towards high concentrations in the data distribution. Hence, the value half of the LOD was assigned to these samples, following Rawlins *et al.*<sup>[2]</sup> and Reimann *et al.*<sup>[3]</sup> An exception was sulfur with more than 18% of all measured data being below the LOD of 200 ppm. The re-digestion of these samples with subsequent analyses using ICP-MS/MS with a LOD of 3.6 ppm at

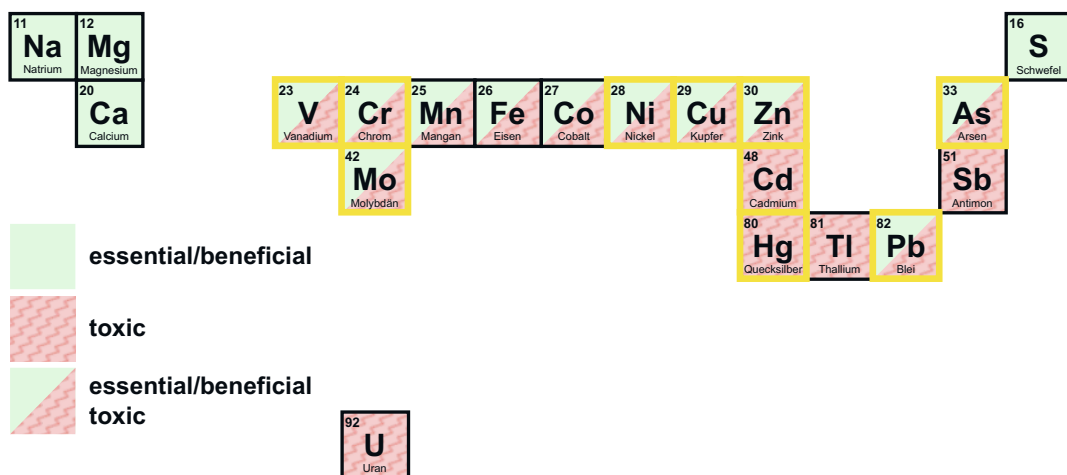


Fig. 2. Periodic table of elements that are assessed in the geochemical soil atlas of Switzerland. Yellow: the ten toxic elements discussed in this publication.

the Department Water Resources and Drinking Water (Eawag) in Dübendorf (Switzerland) substantially lowered the percentage of sulphur concentrations below LOD to less than 1%. In addition, soil samples from the BDM and NABO sites were analysed for other parameters such as pH, total organic C (TOC) and texture. Detailed information on the laboratory analyses of the soil samples can be found in Reusser *et al.*<sup>[4]</sup>

### 2.3 Exclusion of Anthropogenic Point Sources

The differentiation between geogenic sources and anthropogenic sources of elements in topsoils is challenging,<sup>[8]</sup> especially due to historical diffuse and direct inputs, *e.g.* atmospheric deposition as well as application of sewage sludge and slag in agriculture. Further data that could inform on sources, for example, element concentrations in subsoil and corresponding parent material, the isotopic composition, and the element speciation, is not available for most of the sites. The geochemical soil atlas of Switzerland represents the ‘ambient background’ concentrations, which are defined in the European Chemicals Agency REACH guidance on information requirements and chemical safety assessment<sup>[10]</sup> as the ‘sum of the natural background of an element with diffusive input in the past or present’. These ambient background concentrations do not include anthropogenic point sources such as (known) contaminated sites incl. shooting ranges, buffer zones along streets and railways, building backfill as well as other known significant anthropogenic overprint of the element concentrations or replacement of the initial soil layers. In addition to these exclusions, upper bends in the relative cumulative frequency functions of element concentrations could be used as an indicator for anthropogenically-influenced samples.<sup>[11]</sup> No such bends were apparent in the validated dataset.

### 2.4 Statistical Data Analyses

The median of all individual samples was calculated to represent the element concentrations and other parameter values at each sampling site. Most element concentrations followed a right-skewed, log-normal distribution which is typical for geochemical data.<sup>[12]</sup> For each element, the 5%, 10%, 25%, 50%, 75%, 90% and 95% percentiles of the data distribution were calculated to increase comparability between elements and assess low resp. high concentration ranges. Due to the non-normally distributed data, correlations were based on Spearman’s rank correlation coefficient and significant differences between groups were calculated using the pairwise Wilcoxon rank-sum-test as implemented in the package *stats* of R version 4.1.3.<sup>[13]</sup>

### 2.5 Interpolation and Visualisation

Several methods for spatial interpolation exist, some of them include covariates such as soil properties, others are solely based on differences in concentrations with increasing distances. The scope of the interpolation of the geochemical soil atlas was to better visualise regions of low resp. high concentration ranges, not to

model element concentrations at point locations.<sup>[9]</sup> The Ordinary Kriging spatial interpolation method was chosen for the geochemical soil atlas of Switzerland, the same method as for the geochemical atlas of Europe.<sup>[3]</sup> The semivariograms and interpolations on a 1 km x 1 km grid were computed using the *sp*<sup>[14,15]</sup> (version 1.5-1) and *geostat*<sup>[16,17]</sup> (version 2.1-0) R packages. Detailed information on the method is provided in Reusser *et al.*<sup>[4]</sup>

Instead of presenting individual concentrations, the sampling sites and interpolated concentrations were split into concentration ranges according to the percentiles of the data distribution.<sup>[12]</sup> Using this classification method, several elements could be aggregated into groups of exceptionally low (5%, 10%) or high (90%, 95%) concentration ranges with regards to the overall distribution.

### 2.6 Definition of Threshold Values

In this publication, we focus on ten elements that can be toxic at higher concentration levels. These elements were selected because they are either part of the VBBo (Cd, Cr, Cu, Hg, Mo, Ni, Pb, and Zn) or exceed threshold values defined by Eikmann and Kloke<sup>[6]</sup> at several sampling sites (As, V). For Switzerland, no binding thresholds exist for aqua regia digests. The defined threshold values in the VBBo are based on 2 M HNO<sub>3</sub> soil extracts. In these HNO<sub>3</sub> extracts, total element concentrations are underestimated.<sup>[18,19]</sup> Furthermore, element concentrations measured in 2 M HNO<sub>3</sub> extracts often correlate poorly with concentrations measured in aqua regia extracts,<sup>[18]</sup> invalidating a direct conversion between the two methods. Hence, the here presented element concentrations were compared with the threshold values defined by Eikmann and Kloke<sup>[6]</sup> for aqua regia soil extracts. In particular, the multifunctional BW I basis value was chosen, a threshold value indicating expected background concentrations of an element without ‘appreciable anthropological input’. According to Mailänder and Hammann,<sup>[20]</sup> the multifunctional BW I value is comparable to the so-called ‘Richtwert’ of VBBo. In contrast to BW I, the higher tolerance value BW II is use-related (*e.g.* agricultural land use) and comparable to the ‘Prüfwert’ (remediation target value) of VBBo,<sup>[20]</sup> no short or long-term negative effects are expected when concentrations are below this tolerance value.<sup>[6]</sup> However, these BW I and BW II threshold values are not binding in Switzerland and do not take into account the chemical speciation of an element, which influences its mobility and bio-availability. Therefore, we decided to present hotspots of elevated concentrations based on the data distribution rather than based on existing threshold values.

## 3. Element Concentrations in Swiss Topsoils – Distribution of Toxic Elements

The measured element concentrations in Swiss topsoils (Table 1) are in the range of typical soil concentrations in Europe when compared to the European geochemical soil atlas GEMAS.<sup>[3]</sup>

For most sites, concentrations are below existing threshold values. Some sites (from 0% for Hg to 21% for Ni) exceed the BW

Table 1. Percentiles (10% and 90%) and median of selected elements measured in aqua regia digests of soil samples (<2 mm) from the topsoil (0–20 cm) using ICP-MS. Number of total sampling sites = 1'201 (BDM, NABO, GEMAS). BW I and BW II: natural background value and tolerance value for agricultural land use as defined by Eikmann and Kloke,<sup>[6]</sup> percentage of sampling sites exceeding BW I = n.

	As	Cd	Cr	Cu	Hg	Mo	Ni	Pb	V	Zn
10%	3.6	0.10	13.8	7.7	0.033	0.47	10.2	15.0	16	35
Median	7.9	0.24	30.3	18.1	0.066	0.81	27.3	24.2	32	64
90%	21.9	0.62	52.1	35.0	0.140	1.78	49.8	41.3	61	106
BW I <sup>[6]</sup>	20	1	50	50	0.5	5	40	100	50	150
n > BW I (%)	11	5	12	2	0	1	21	1	17	2
BW II <sup>[6]</sup>	40	2	200	50	10	20	100	500	100	300

I threshold value defined by Eikmann and Kloke<sup>[6]</sup> but only few sites exceed the BW II tolerance value for agricultural land (from 0 sites for Hg to 19 sites for V). As anthropogenic point sources were excluded based on the available information, we assume that other reasons caused the high element concentrations resp. their accumulation in topsoils: (i) geogenic sources, (ii) diffusive inputs, (iii) missing sinks, (iv) soil processes such as preferential weathering and/or sorption, (v) unrecognized anthropogenic sources (e.g. slag input). These findings suggest that for Switzerland, the BW I value does not necessarily reflect the ‘actual value of natural soil subject to the upper, geological and pedological input’ as defined by Eikmann and Kloke<sup>[6]</sup> because in some regions, soil concentrations without presumed anthropogenic point sources are considerably higher. Furthermore, geochemical threshold values defined by Reimann *et al.*<sup>[9]</sup> for European soils are considerably higher depending on the calculation method.

For some elements (e.g. Hg, Zn, and Cu), significant differences were detected between land use types, especially arable fields, pastures/meadows, and forests. These results are discussed in more detail in Reusser *et al.*<sup>[4]</sup> The interpolated maps can be downloaded from the website of the Federal Office for the Environment FOEN.

### 3.1 Hotspots of Toxic Elements

On a spatial scale, large local and regional differences exist for all elements. In general, comparably low concentrations were detected in the Midlands, whereas elevated concentrations were prevalently observed in soils originating from the Northwestern part of Switzerland and regions in the Northern Prealps, the Eastern Alps and Southeastern Switzerland. Sites where half or more of the 10 selected elements exceed the 90% percentile of their concentration distributions are mostly located in the Jura (Fig. 3). Arsenic, for example, is known to be enriched in ferrous limestones and clays in the Jura.<sup>[21]</sup>

The presented hotspots indicate regions with high element concentrations in topsoils based on the data distribution on a national scale. However, the hotspots do not specify whether a threshold value was exceeded or not. In order to evaluate a possible contamination risk, further studies with a higher resolved sampling scheme in these regions including speciation and bio-availability assessments would be needed.

### 3.2 Assessment of Geogenic Impacts on Spatial Distribution

The lithology of the parent material may be an important factor in explaining elevated element concentrations in soils. However, nationwide information on the geochemistry and lithology of the bedrock resp. the parent material is scarce. The lithological map of Switzerland (1:500'000, Federal Office of Topography swisstopo) used in this study differentiates 25 main classes of the subsurface based on lithological and petrographic criteria. Where applicable, the measured element concentrations in the soil were compared to trace element contents in Swiss ‘soil-forming’ rock formations reported by Tuchs Schmid.<sup>[22]</sup>

When comparing all sampling sites in one lithological group as well as between groups, only in five lithological groups did half or more of the 10 selected elements exceed the 90% percentile of their respective concentration distributions (Fig. 4). With 20% of sites, this share is largest on carbonate rocks. Carbonate rocks are present as parent material in the Jura but also in the Swiss Alps. The correlations of the individual element concentrations with the lithological groups are discussed in more detail in Reusser *et al.*<sup>[4]</sup>

Several factors can contribute to the accumulation of (toxic) elements in soils developed on carbonate rocks: (i) high geogenic presence of the elements in carbonate rocks,<sup>[22]</sup> (ii) faster weathering of carbonates compared to silicates,<sup>[23]</sup> (iii) precipitation of the elements with carbonates at high pH,<sup>[3]</sup> (iv) enrichment of clay minerals with weathering and thus increasing sorption of ions.<sup>[24]</sup>

Furthermore, elevated concentrations of Cr and Ni were measured at sampling sites belonging to the group of basic rocks (Fig. 4). These two elements correlate with each other and with Co, Fe and V ( $R^2 > 0.7$ )<sup>[4]</sup>. The positive correlation could be due to the enrichment of these elements in iron-rich silicates, e.g. olivines, amphiboles and pyroxenes,<sup>[22,25]</sup> possibly indicating a common geogenic source (e.g. serpentinites<sup>[22]</sup>). In addition to the geogenic source, Cr and V are thought to be incorporated into iron oxides during their formation in the soil.<sup>[26]</sup>

The lithological group of basic rocks may serve as a potential indication for elevated Cr, Ni and Co concentrations, but more sampling sites belonging to this group need to be analysed in order to gain statistically significant results.

Number of elements exceeding 90 % percentile

- 0
- 1
- 2
- 3
- 4
- 5
- 6
- 7
- 8
- 9

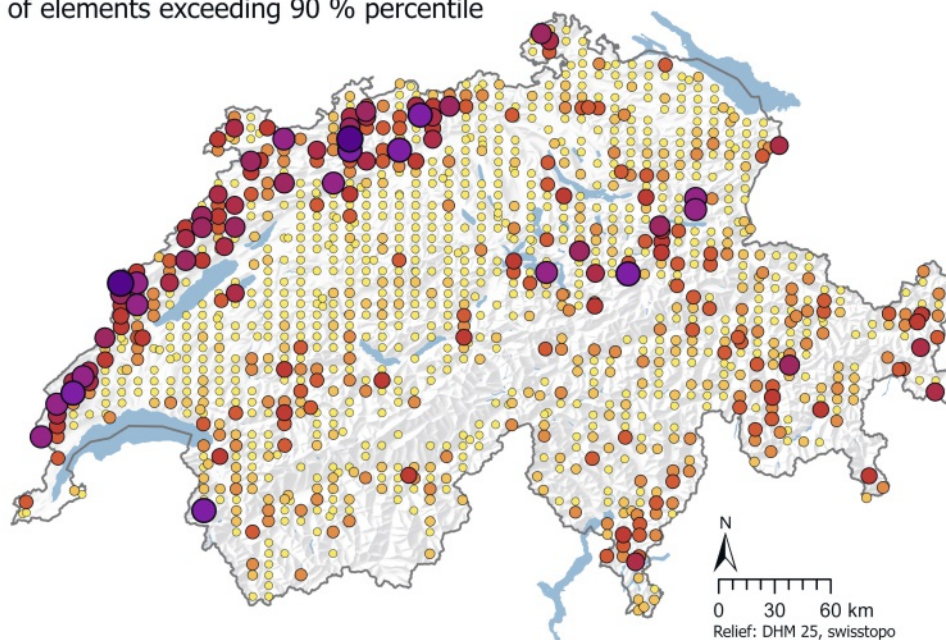


Fig. 3. Spatial distribution of hotspots with measured element concentrations exceeding 90% of the data distribution. The 10 elements As, Cd, Cr, Cu, Hg, Mo, Ni, Pb, V and Zn were considered. The number 0 indicates that none of the 10 elements exceeded 90% of the data distribution, whereas the number 9 indicates that in this region, 9 elements of 10 exceeded the 90% percentile of the respective concentration distribution. The indicated hotspots are solely based on the data distribution, the map does not provide information whether threshold values are exceeded or not.

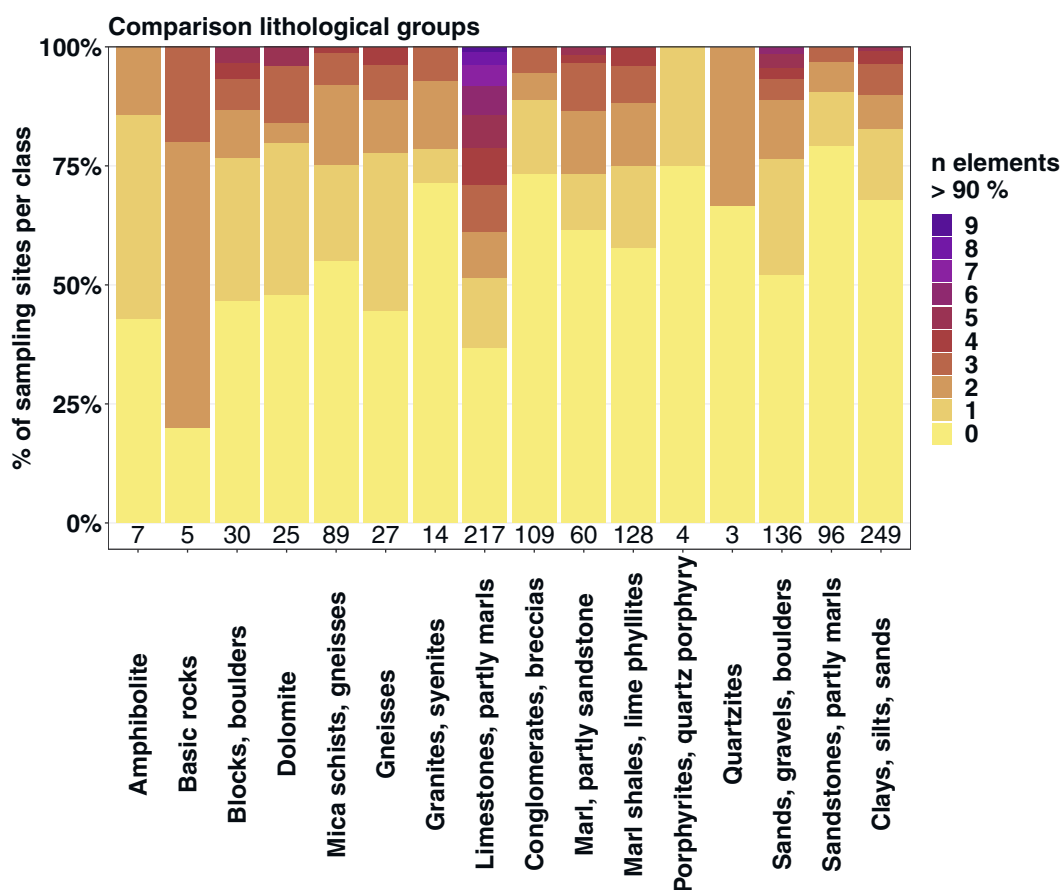


Fig. 4. Comparison of lithological groups between number of elements exceeding 90% of the data distribution. The 10 elements As, Cd, Cr, Cu, Hg, Mo, Ni, Pb, V and Zn were considered. The category with number 0 indicates that none of the 10 elements considered exceeded 90% of the data distribution, whereas the category with number 9 indicates that in this lithological group, 9 elements out of 10 exceeded the 90% percentile of the respective concentration distribution. On the y-axis, the percentage of sampling sites in each category compared to the total amount of sampling sites in the respective lithological group (number below bar) is presented. The sampling sites were grouped according to the lithological map of Switzerland, 1:500'000, Federal Office of Topography swisstopo.

#### 4. Conclusions

The geochemical soil atlas of Switzerland is the first nationwide assessment of total element concentrations in Swiss topsoils. The atlas provides the basis for future detailed studies of areas showing deficiencies or contaminations.

Hotspots of potential contamination, where several of the ten selected toxic elements exceed the 90% percentile of their respective concentration distributions, seem to be clustered in the Jura and at least partly caused by the prevalent geology. Some of these hotspots may be influenced by localized mineralizations (*e.g.* Zn and Pb mineralizations in carbonate and silicate rocks), others by elevated element concentrations throughout certain types of rocks (*e.g.* Cr and Ni in serpentinite). However, additional soil processes (*e.g.* weathering of carbonates), sources (*e.g.* pesticide and manure application in the case of Cu and Zn) and the lack of sinks influence element accumulation in soils. Hence, specific geology is not necessarily indicating hotspots of contamination.

#### 5. Outlook

The geochemical soil atlas is based on 1'201 sampling sites evenly distributed across Switzerland. Besides these sampling sites, much more data exists from analyses of element concentrations in the topsoil as well as lower soil layers carried out by a wide range of institutions.<sup>[18]</sup> However, extraction and analysis techniques vary markedly between studies, which hinders a direct combination of datasets. In addition, the purpose of most sampling campaigns was/is often the detection of hotspots, causing a bias towards high element concentrations. Nevertheless, knowledge of element concentrations in lower soil layers and the parent material combined with isotopic analyses is crucial for determining whether a contamination has been caused by geogenic sources or other processes and sources. The identification of the source may be important for predictions of element concentrations (over time as well as at other locations) and for the clarification of legal

responsibilities, but not necessarily for the evaluation of the actual toxicity of an element.

The digestion of soil samples using aqua regia with subsequent ICP-MS(/MS) analyses provides a reliable measure for the approximation of total element concentrations. However, to inform about the effective risks for human and animal health, not only the total element concentrations but mainly element bioavailability and the transfer into the food chain is decisive. Bioavailability could be estimated by models or by selective extraction procedures. Furthermore, element speciation can be crucial for their toxicity. For example, Cr(III) is essential, but Cr in the oxidation state VI is highly toxic. In the soil, Cr(VI) is much more mobile than Cr(III), which mostly precipitates or adsorbs onto surfaces of soil constituents at pH > 5.<sup>[27]</sup> Another example is mercury: it is assumed that less than 0.01% of elemental mercury is actually taken up by the human body, in contrast, 95% of Hg is taken up *via* the organic species methylmercury.<sup>[28]</sup>

In contrast to the Northwestern part of Switzerland with several hotspots, element concentrations in the Midlands are comparatively low. These regions are most important for agricultural production. Even though most of the trace elements examined can have toxic effects at elevated concentrations, they are often essential for plants, animals, and humans. Low soil concentrations of essential major elements (*e.g.* sulfur) as well as essential trace elements (*e.g.* molybdenum) could reduce the quality of food and fodder, possibly leading to deficiencies. Further studies would be needed to evaluate the status and supply of essential elements in these regions.

The geochemical soil atlas is based on one sampling campaign which took place between 2011–2015, hence representing the element concentrations in topsoils at a given time period. However, soil properties change over time as a result of *e.g.* land use or climate change. For example, pH is influenced by weathering of carbonates, redox conditions by compaction and drying/rewetting,

and sorption sites by the loss of soil organic matter. In addition to changing inputs (e.g. reduced atmospheric deposition of sulfur) and outputs (e.g. removal of crops), these changing soil properties affect the mobility of elements in the soil and thus potentially their accumulation or decrease. Where applicable, the concentrations of certain elements (e.g. Cd, Cu, Pb and Zn) were compared with the long-term monitoring findings of the Swiss Soil Monitoring Network (NABO) at selected sites.<sup>[4]</sup> A second sampling campaign would be needed to better understand the nationwide concentration development over time for all elements investigated. Knowledge of the dynamics in soil elemental concentrations combined with the information on soil properties, the geochemistry of the parent material, land use and related inputs and outputs as well as the chemical speciation of the elements could be used in models to predict element concentrations and their bioavailability in soils.

The geochemical soil atlas will be published in December 2023 and can be accessed via the Federal Office for the Environment FOEN and via the Swiss Centre of Excellence for Agricultural Research Agroscope.

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