Distribution and mobility of lead (Pb), copper (Cu), Zinc (Zn) and antimony (Sb) from ammunition residues on shooting ranges for small arms located on mires

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Abstract An environmental survey was performed on shooting ranges for small arms located on minerotrophic mires. The highest mean concentrations of Pb (13 g/kg), Cu (5.2 g/kg), Zn (1.1 g/kg) and Sb (0.83 g/kg) in the top soil were from a range located on a poor minerotrophic and acidic mire. This range had also the highest concentrations of Pb, Cu, Zn and Sb in discharge water (0.18 mg/L Pb, 0.42 mg/L Cu, 0.63 mg/L Zn and 65µg/L Sb) and subsurface soil water (2.5 mg/L Pb, 0.9 mg/L Cu, 1.6 mg/L Zn, and 0.15 mg/L Sb). No clear differences in the discharge of ammunition residues between the mires were observed based on the characteristics of the mires. In surface water with high pH (pH ~7) there was a trend with high concentrations of Sb and lower relative concentrations of Cu and Pb. The relatively low concentrations of ammunition residues both in the soil and soil water, 20 cm below the top soil, indicates limited vertical migration in the soil. Channels in the mires, made by plant roots or soil layer of less decomposed materials, may increase the rate of transport of contaminated surface water into deeper soil layers and ground water. A large portion of both Cu and Sb were associated to the oxidizable components in the peat, which may imply that these elements form inner-sphere complexes with organic matter. The largest portion of Pb and Zn were associated with the exchangeable and pH-sensitive components in the peat, which may imply that these elements form outer-sphere complexes with the peat.

Keywords: Shooting ranges; mires; peat; heavy metals; antimony; sequential extractions

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Introduction

In Norway it was common to establish shooting ranges for small arms on mires. Mires were regarded as less valuable and the ranges were relatively easy to maintain. Some of these ranges have been in use for decades and have accumulated tons of metal residues from spent ammunition. The bullets from small arms primarily contain an alloy of lead (Pb) and antimony (Sb) covered with an alloy of copper (Cu) and zinc (Zn) (Randich et al. 2002). The Norwegian army has now replaced leaded ammunition for small arms with bullets made of steel, which in the future will reduce the load of Pb at the shooting ranges. These bullets have a core of steel covered with an alloy of Cu and Zn. The deposition of ammunition residues at shooting ranges leads to significant discharge of metal residues into nearby water courses and may pose a threat to aquatic organisms living nearby the ranges (Haux et al. 1986; Mariussen et al. 2017). In addition, both wild animals and domestic animals pasture at contaminated areas are at risk of being exposed to toxic concentrations of metals. In particular, there are some concerns of domestic animals and birds that are shown vulnerable to Pb intoxication (Braun et al. 1997; Lewis et al. 2001; Peddicord and Lakind 2000).

Remediation of shooting ranges, which may cover areas of several square kilometers, can be challenging and costly, particularly at sites located on mires. Usually, metal contaminated soil at an abandoned shooting range is removed and deposited at an approved waste site. The removed soil is then replaced by clean soil and, depending on later use, subjected to revegetation for recreational use, or construction of infrastructure. At mires there are often large volumes of contaminated peat that presumably needs to be removed. In many countries, such as in the EU (Council Directive 1999/31/EC on the landfill of waste) and Norway (Directive FOR-2004-06-01-930 on the landfill of waste), there are restrictions on the levels of organic materials that are allowed to be deposited on regular waste sites, and there are few approved sites for disposal of such materials. Mires are, in addition, very vulnerable

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to intervention and it may take several decades to restore damaged mires into their original condition (Gorham and Rochefort 2003). Some peat mires may also contain endangered species of plants and animals.

Several studies have evaluated contamination by ammunition residues at shooting ranges. These surveys have primarily been performed on contaminated mineral soils and have shown limited movement of the contaminants vertically in the soil column (Jörgensen and Willems 1987; Knechtenhofer et al. 2003; Laporte-Saumure et al. 2011; Clausen and Korte 2009; Sanderson et al. 2012; Cao et al. 2003; Murray et al. 1997; Ash et al. 2013), but a significant spread of contaminants by surface runoff (Martin et al. 2013; Labare et al. 2004; Craig et al. 1999; Strømseng et al. 2009; Mariussen et al. 2012). Less is known about the fate of metal contamination at shooting ranges at sites with soil rich in organic matter. Mires are wet terrains dominated by peat-forming plants. Peat accumulated at mires is remnants of primarily dead plants accumulated for centuries. Humic and fulvic acids, which are decomposition products of the peat, are complex organic components of aromatics and sugars with carboxylic and phenolic substituents and form most of the organic components in natural waters (Klavins and Purmalis 2013). Dissolved metals from the ammunition residues, particularly cations such as Pb and Cu, have high affinity to humic and fulvic acids (e.g. Christl et al. 2005) which may increase their mobility in the environment.

To evaluate the contamination pattern at shooting ranges located on mires, an environmental survey was performed at six different ranges located at sites with different characteristics. The main goal of this survey was to investigate the spread of ammunition residues at shooting ranges located on mires in order to provide a better basis to evaluate the more appropriate clean up measures to be performed at such sites. In addition, the mobility of metals and metalloids in mires was compared with their mobility in mineral soils, and the major runoff pathways of the metal residues were identified. The result of the survey has been presented

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for the Norwegian army in a technical report (Mariussen et al., 2016) in Norwegian. Peat and soil, rich in organic matter, and soil water samples were taken vertically in the soil column from the shooting ranges and analyzed for different elements. Water samples of discharge water were analyzed to get an overview of the spread of contaminants by surface runoff and to investigate fluctuations in the concentrations of contaminants in the runoff water during the summer season. In addition, the binding characteristics of Pb, Cu, Zn and Sb to peat and organic rich soil were evaluated by sequential extractions.

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Materials and Methods

Area description

Avgrunnsdalen shooting range (**Av**) is an abandoned range located in the south-eastern part of Norway (59.58042°N, 10.51218°E), just outside Oslo near the coast in the Buskerud County (Fig 1 and Fig 1S, supplementary). The range was established in 1917 and was used for approximately 80 years. One of the shooting ranges in the area is located at a mire, which covers an area of approximately 10000 m^2. A small drainage creek is running through the contaminated range. The drainage basin of the stream is approximately 0.5 km^2 with an approximate annual mean discharge of 8 L/s (Heier et al. 2009). The shooting range was abandoned in 1997 and in 2013 subjected to remediation measures. Steinsjøen (**St**) shooting range is a military training area located in the south-eastern part of Norway (60.54248°N, 11.09554°E), just north of Oslo in the Oppland County (Fig 1 and Fig 2S) The area has been used for military shooting training for approximately 50 years and is still active. A small erosion creek runs through the mire, which during heavy rainfall represents an input of metal contaminated water to nearby fish bearing waters. Small lakes in this area have elevated levels of heavy metals from the shooting activity (Mariussen et al. 2009). Mauken (**Ma**) shooting range is a military training area used for approximately 50 years and is still active (Fig 1 and Fig 3S). The shooting range is located in the northern part of Norway in the Troms County (69.04592°N, 19.24131°E). A small drainage creek runs just below the berm and represent an input of metal contaminated water to a nearby river. Tittelsnes (**Ti**) military shooting range was closed in 2005 and had been used for approximately 55 years (Fig 1 and Fig 4S). The range, which was subjected to remediation measures in 2014, is located on the west-coast of Norway (59.72306°N, 5.51481°E). At this range there were two small drainage brooks. One of the brooks ran along the firing point and was acidic (pH 4.6). The other one ran across the range, and had a higher pH (pH 6.4). A small fish bearing lake downstream the range had

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elevated levels of heavy metals (~9µg/L Cu, ~5µg/L Pb, ~13µg/L Zn) due to discharge of contaminated water from the shooting range (Forsvarsbygg 2012). Kjoselvdalen (**Kj**) shooting range for small arms has been used since 1950 and is located in the northern part of Norway (Fig 1 and Fig 5S), near the city of Tromsø in the Troms County (69.73210°N, 19.14590°E). Hengsvann (**He**) shooting range covers an area of approximately 37 km^2 near the small town of Kongsberg in Buskerud County (59.64462313°N, 9.44673313°E), in the south–eastern part of Norway (Fig 1 and Fig 6S). The area has been used for training with small arms and heavily armed weapons for more than 60 years. Large parts of the area are covered by woodland and mires. The survey was performed at a range for small arms at a fairly shallow mire (~0.5 m deep).

Sample collection of soil and soil water

A network of sampling points, distributed approximately 5 meters apart, were established at the six sites (Table 1S). In each sampling point, soil and soil water samples were collected in three vertical layers. One sample was taken on the surface, one sample was taken approximately at 15-30 cm and one at 30-45 cm below the surface, respectively. The soil water at **Av** was collected by squeezing the water from the soil by hand. The squeezed water samples were later filtered through 0.45 µm filters at the laboratory. Close to the same sampling points, for the superficial soil at **Av,** soil water was also collected with Rhizon soil water samplers (Rhizosphere Research Products, Wageningen, the Netherlands). At **St**, **Ma**, **Ti**, **He** and **Kj** soil water samples were collected only with Rhizon soil water samplers. Rhizon samplers extract soil water under vacuum. The vacuum was set with a 50 ml syringe. According to the manufacturer, the Rhizon sampler has a pore size of 0.1 µm. All water samples were preserved with ultra-pure concentrated nitric acid (HNO_3) to a final

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concentration of 0.65 %. The soil samples were collected in polyethylene zip-lock bags and placed in a refrigerator at 4°C before further processing.

Sample collection of surface water

Water samples were taken from the discharge creeks running through the investigated sites. These samples give a snapshot of the concentrations of metal contaminants from the ranges in the discharge stream at the day the soil samples were collected. The water samples were analyzed for both total- and dissolved (0.45µm filtrate) metals. The metal concentrations obtained from a 0.45 µm filtrate correspond to elements dissolved as free ions and colloidal bound elements (Gustaffson and Geschwend 1997). In 2013 **Av** was subjected to some remediation measures. In order to monitor fluctuations in the concentrations of contaminants in the runoff water due to construction works on the range, two water samples were collected daily by an ISCO automatic water sampler (ISCO 6700) from 13th of April to 13th of October. The ISCO water sampler was placed approximately 200 m downstream of the mire (Fig 1S). pH in the runoff water was measured weekly when the water samples were collected from the ISCO. All water samples were preserved with concentrated HNO_3 (final concentration 0.65 % HNO_3).

Sample preparation of soil and chemical analyses

A subset of soil samples was subjected to chemical analyses. Aliquots of the soil were dried at 105°C for approximately 24 hours. The dried soil, which consisted of organic rich soil and peat, was then crushed by hand into polyethylene zip-lock bags. Aliquots of approximately 0.3-0.4 g of the crushed and powdered soil were subjected to microwave assisted digestion with Aqua Regia (1:3 mixtures of concentrated HNO_3 and HCl, respectively). The samples were digested in a microwave oven (Mars$_x$, CEM corporation NC) with the following

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temperature program: 9°C/min to 180°C, followed by 5 min at 180°C, then 2°C/min to 190°C followed by 10 min at 190°C. After digestion, the samples were allowed to cool down to room temperature into their vessels before being transferred to 50 ml polyethylene vials (Sarstedt) with deionized water. A certified soil reference material (GBW07406, Institute of Geophysical and Geochemical Exploration, China) was used to assess the correct quantification of Pb, Cu, Zn and Sb. Mean recoveries of these elements in the reference material in our lab between 2011 and 2014 were 95% (± 4 SD), 101% (± 10), 97% (± 15) and 85% (± 13). Evaluations of recovery of Sb, Pb and Cu in soil with use of Aqua Regia have previously been described (Nash et al. 2000; Hjortenkrans et al. 2009, Mariussen 2012). The water and soil samples were analyzed for different elements by ICP-MS (Thermo X-series II). An internal standard was added in each sample, and the samples were quantified using a four point standard curve (0.1 – 1000 µg/L). Representative detection limits for the analytes, estimated as 10 times the standard deviation of the blanks from 5 analytical runs, were 20 µg/L, 40 µg/L, 0.70 µg/L, 1.1 µg/L, 5.0 µg/L, 0.03 µg/L, 1.6 µg/L. 0.10 µg/L, 0.20 µg/L, 0.05 µg/L and 0.03 for Na, K, Mg, Al, Ca, Mn, Fe, Cu, Zn, Sb and Pb, respectively. To ensure the correct quantification of the metals, reference solutions of known element concentrations (TM 23.4, TMDA 61.2, Rain-97, Battle-02 Analytical reference material, Environment Canada, Canada) were analyzed in addition to in-house prepared standards. A deviation of 5% from the given concentration in the reference solution was accepted. Blanks were regularly analyzed to control for background contamination.

Measurement of soil pH and loss on ignition

An aliquot of 5 ml dried and crushed soil were mixed with 50 ml deionized water and subjected to continuous mixing for 24 hours. After settling down for 15 minutes, the soil pH was determined by introducing the pH electrode into the soil water. Loss on ignition (L.O.I.)

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was used as an index of the amount of organic materials in the soil and was measured after burning an aliquot of approximately 3-4 g dried soil at 550°C in a furnace (Heraeus K750) for 2 hours (Chambers et al. 2011).

Sequential extractions of peat soil

Moist peat from **Av** and a moist pooled organic rich top soil sample from **Ti** were subjected to sequential extractions by using the method described by Tessier et al. (1979) and modified by Oughten et al. (1992). Aliquots of soils (~2-3 g wet weight) were extracted sequentially into 6 fractions in a liquid to solid ratio of 15 or 20 (LS15/20) as described in Table 2S. Fractions 1, 2 and 3 were mixed with a Reax overhead mixer (10 rpm), whereas fractions 4 and 5 were put in a water bath and regularly mixed by hand. The residual fraction (fraction 6) of the soil was washed twice with deionized water and dried (105°C) overnight in pre-weighed vials followed by digestion with aqua regia as described above. In addition, one part of the soil was used to determine the dry-weight.

Data analysis

Descriptive statistics, correlation analysis and mathematical calculations were computed by GraphPad Prism 6 or Excel 2010. Correlation analyses (Pearson correlation analysis) were performed between each analyzed element in the soil and soil water from the six shooting ranges sampled at three vertical layers in the soil profile, a total of 36 groups or datasets. Significant correlation ($p<0.05$) was given a score of one and all the scores were then added together. Elements that correlated in more than half of the group were highlighted and considered as a general trend in the dataset. The data from each of the correlation analyses (Pearson's correlation coefficient and p values) are shown in the Appendix of the Supplementary materials. Normal distribution of the variables was assessed using the

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D´Augostino & Person omnibus normality test. In general the data did not pass the normality test, and log_e-transformation was applied to obtain normality.

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Results and Discussion

Characteristics of the mires

Mires are classified from both the water and soil chemistry and the biodiversity of the flora (Rydin and Jeglum 2006). Several of the investigated sites were heavily disturbed by the training activities and data on vegetation were not collected. A crude classification was done based on the chemistry of the soil (Table 3S), soil water (Table 4S) and discharge water (Table 1). Ombrotrophic mires or peatlands, which primarily are influenced by precipitation and not the underlying bedrock, are acidic (pH 3.5-4.2) with low surface water concentrations of Ca (< 1 mg/L) and other ions, such as Mg, Na and K. Minerotrophic mires can be classified into poor (pH 4-5.5) and rich mires or fens (pH > 5) and are influence by both precipitation and the underlying bedrock. The concentration of Ca in the surface water from minerotrophic mires varies between less than 1 mg/L in poor mires to more than 50 mg/L in extremely rich mires. All the studied mires were in close vicinity of mineral edges or relatively shallow and some of the collected samples contained mineral soil from stop-butts or from the underlying bedrock. This was reflected in high variations in the content of organic matter in the soil samples from some of the sites (L.O.I., Table 2). The range located at **Ti** could not be classified as a mire per se, but had mixed soil characteristics, which included soil profiles with a predominance of peat with more than 90% content of organic matter and soil profiles which can be classified as organic rich mineral soil. According to the classification described in Rydin and Jeglund (2008) none of the sites could be classified as ombrotrophic peatlands. The mire at **Av** could be regarded as a poor and minerotrophic mire, with bedrock primarily of granite (Norwegian Geological Survey). Sequential extractions of peat from **Av** showed a low portion of Ca in the residual fraction compared to the richer organic soil from **Ti** (Table 5S). A slightly higher Na concentration (3.6 mg/L) in the discharge water from **Av** compared to some of the other sites indicated some influence by costal climate and sea salt inputs

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(Skjelkvåle et al. 2007). The peat, soil water and discharge water from **Av** had relatively low concentrations of Ca (~2-4 mg/L in soil water) and Mg (~0.5 mg/L in soil water). The mires at **St** and **He** could be regarded as poor and minerotrophic. The peat and soil water had low concentrations of Ca (< 4.0 mg/L in soil water), Mg (< 1.0 mg/L in soil water) and Na (< 1.2 mg/L in soil water). Both areas have acidic bedrock primarily of granite and gneiss (Norwegian Geological Survey). The mires at **Ma** and **Kj** could be regarded as rich minerotrophic fens. The discharge water from these mires had a high pH and Ca concentrations (8.8 and 22 mg/L, respectively). The range at **Kj** has bedrock consisting of gneiss, muscovite and garnet (Norwegian Geological Survey). The mire itself is localized on top of a moraine. At **Ma** the bedrock consists of amphibolite and metagabbro (Norwegian Geological Survey). The organic rich soil from **Ti** had high concentrations of Fe (> 13 g/kg), Al (> 15 g/kg), Ca (> 9g/kg) and Mg (>2.5 g/kg). This was not reflected in the soil water or drainage water, which, compared to the other sites, primarily had high concentrations of Na (6.7 and 8.9 mg/L). Sequential extractions of a pooled sample of the organic rich top soil from **Ti** showed that Ca, Mg, Fe and Al primarily were associated with the residual fraction indicating high contribution from the mineral fraction (Table 5S). The brook, which ran through the range, had a pH of 6.4. The area has bedrock of gneiss and schist (Norwegian Geological Survey). The soil water and discharge water from **Kj** and **Ti** had high concentrations of Na (> 6.5 m/L) indicating an influence from coastal sea sprays (Skjelkvåle et al. 2007). The mires in our investigation had a soil water pH between 4 and 5, indicating less difference between the mires than suggested by our crude classification. According to Sjörs and Gunnarsson (2002) the pH in the soil water is usually lower than in the surface water. Peat and organic rich soil have an acidic nature and contain different types of degradation products of plant remnants, which include carbohydrates, proteins, humic and fulvic acids, lignins and different lipid containing compounds (Klavins and Purmalis, 2013).

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The largest part of peat is made of humic compounds that maintain the peat environment with low pH.

Run-off patterns of ammunition residues

Particularly high concentrations of Pb, Cu, Zn and Sb were found in the run-off water from **St** (Table 1). This was probably attributed to near stagnant run-off water on highly contaminated acidic mire. At **Ti** there were two brooks running through the range with different pH. The acidic creek, with a pH of 4.6, had a much higher concentration of Cu (86µg/L vs 19 µg/L) and Zn (53µg/L vs 12 µg/L) than the less acidic brook which had a pH of 6.2. The less acidic brook had, in addition, a higher relative concentration of Pb and Sb in proportion to Cu (Table 1). The acidic drainage brook at **Ti** ran along the firing point and the high Cu and Zn concentration may be attributed both to the acidity of the water and the finding of a substantial amount of cartridges near and in the creek, which primarily are made of brass. The brooks with a low pH appeared to have a higher proportional level of Pb compared to Sb (Table 1). It is known that the solubility of Sb increases with increasing pH and that the mobility of cations, such as Pb and Cu, decreases with decreasing pH (e.g. Santillan-Medrano and Jurinak 1975; Johnson et al. 2005; Gundersen and Steinnes 2003; Sanderson et al. 2012). The concentrations of Pb, Cu, Zn and Sb in the water in filtered and non-filtered samples were similar indicating that the contaminants primarily are dissolved as free ions or bound to organic or inorganic colloids. Several studies have monitored discharge of Pb, Cu and Sb in surface waters from shooting ranges showing similar concentration ranges as in our study (Martin et al. 2013; Craig et al. 1999; Mariussen et al. 2012, 2017; Strømseng et al. 2009). In addition, the concentrations of Pb and Cu were comparable to the concentrations in rivers influenced by lead mining in the U.S. (Schmitt and Finger 1982; Czarnezki 1987). Bearing in mind the numerous shooting ranges that exist, they represent a considerable source of

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contamination that may spread into local watersheds leading to potential harmful effects on aquatic organisms (Haux et al. 1986; Mariussen et al. 2017).

The run-off patterns of the discharge water from **Av** were studied in more detail. Two discharge water samples were taken daily from 13th of April to 13th of October 2013, showing considerable day to day variations in the concentrations of Pb, Cu, Sb and Zn in the creek (Fig. 2). Fig 7S shows the day to day variations in the concentrations of other elements (Mg, Ca, Fe, Al and Mn) in the runoff creek. During the observation period, some remediation measures were performed for few days at the shooting range area (e.g., in August some non-vegetated hot spots at the contaminated mire were covered with clean peat). These measures had apparently only minor or no effects on the concentrations of the ammunition residues in the runoff water. Mean total concentrations of Pb, Cu, Sb and Zn in the discharge creek during the period were 20 ± 13 µg/L, 14 ± 6.5 µg/L, 2.0 ± 0.85 µg/L and 20 ± 10 µg/L, respectively. The 95% percentiles were 30, 25, 3.1 and 32 µg/L, whereas the 5% percentiles were 9.4, 5.5, 0.8 and 13 µg/L for Pb, Cu, Sb and Zn, respectively. During heavy rain there was an increase in the concentrations of Pb, Cu and Sb in the creek. Using the 5% and 95% percentiles to avoid outliers, the concentrations of Pb, Cu, Sb and Zn increased by a factor of 3.1, 4.5, 3.8 and 2.6 respectively during precipitation. Similar episodic increase in runoff concentration of contaminants during stormflow or high flow has been observed from contaminated peatland (Rothwell et al. 2007; Broder and Biester 2015), from urban runoff (Lieb and Carline 2000; Roberts et al. 2007), in rivers draining abandoned mines (Byrne et al. 2013; Czarnezki 1987) as well as at shooting ranges (Strømseng et al. 2009; Mariussen et al. 2012). The increase in the trace metal concentration in the creek following precipitation is accompanied by an increased flow in the river. Previously we estimated that the temporarily increased flow in a creek running through a shooting range due to heavy precipitation contributed approximately 20-25% of the total annual discharge of ammunition residues from

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the creek. Snow melting contributed approximately 25-30% to the total annual discharge of Pb, Cu and Sb (Strømseng et al. 2009). At a shooting range most ammunition residues are left on the upper surface layer and, during rainfall, surface water submerges the ammunition residues in the top soil layer. The increase in the dissolved metal concentration observed after rainfall is therefore probably due to larger transport of water through the upper soil horizons and soil surface (Strømseng et al. 2009; Broder and Biester 2015). In order to get a satisfactory picture of the contamination in a creek it is therefore important to do repeated analysis for a sufficiently long period of time, which includes both wet and dry periods.

Contamination of ammunition residues in soil and soil water

Subsets of the collected soil samples from the six areas were analyzed. With the exception of the site at **He,** the soils contained very high concentrations of ammunition residues (Table 2). Particularly, the concentrations of Pb were high (13 g/kg at **St**) reflecting the use of Pb containing ammunition for years. Correlation analysis of the analyzed elements in the soil from the ranges showed in general that Pb, Cu and Sb were correlated with each other (Table 6S), which is reasonable since these elements are the main constituents of small arms ammunition. None of these elements correlated with other elements analyzed in the soils. We could not observe any prominent spatial difference in the distribution of Pb, Cu and Sb at the investigated sites. Higher concentrations of ammunitions residues are expected to be found near the target areas. With the exception of **He,** the ranges studied in this investigation have been used for decades and for different training purposes which probably has led to a less heterogeneous distribution of the ammunition residues.

The concentration of Pb, Cu and Sb rapidly decreased vertically in the peat soil. Between 90 and 99 % of the contamination was located in the surface soil. The figure is probably even higher since the chemical analyses of the soil only consider the smallest

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fragments or dissolved ammunition residues. Even at **Av**, which has been used for almost 100 years as a shooting range for small arms, more than 95 % of the Pb residues were located at the top 0-15 cm of the soil, indicating limited migration of the contaminants vertically in the soil column. Previous reports have shown that atmospheric deposited Cu and Pb, and even Sb, have little mobility and are well preserved in ombrotrophic peatlands (Nieminen et al., 2002; Cloy et al., 2009; Novak et al. 2011). Similar contamination patterns have been previously shown also in mineral soils contaminated with ammunition from small arms (Jörgensen and Willems 1987; Knechtenhofer et al. 2003; Laporte-Saumure et al. 2011; Clausen and Korte 2009; Sanderson et al. 2012; Cao et al. 2003; Murray et al. 1997). The concentrations of Pb and Sb at 30-45 cm were nevertheless higher than background levels analyzed in ombrotrophic peatlands measured in Finland (Nieminen et al. 2002), Switzerland and Scotland (Shotyk et al. 2004; Cloy et al. 2009), and peat cores from a bog in Estonia (Syrovetnik et al. 2007). The expected background concentrations of Pb, Cu, Zn and Sb in mires depend on the bedrock and anthropogenic inputs. Analyses of peat cores from different sites in Europe show background concentrations of Pb, Cu, Zn and Sb of less than approximately 20 mg/kg, 10 mg/kg, 50 mg/kg and 0.2 mg/kg respectively (Nieminen et al. 2002; Shotyk et al. 2004; Silamikele et al. 2010), indicating some vertical transport of contaminants in the soil profiles in our study. Elevated concentrations of contaminants from ammunition residues in the deepest soil layer may be attributed to vertical transport by soil water. With the exception of the soil water samples from **Kj** and **He**, similar contamination patterns as for soils were observed for the soil water samples (Table 3). Correlation analysis of the analyzed elements in the soil water from the ranges showed that Pb and Cu were correlated in 17 of 18 groups (Table 7S). Sb, which is used in alloy in Pb-bullets, correlated with Pb in only 9 out of 18 groups, which may be due to their very different chemical properties leading to different rates of dissolution from the bullets. Sb in soil water was in

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addition correlated with Al (10 out of 18 groups). Only few have studied the interaction between Al and Sb in water, but it has been shown that Sb, particularly the trivalent Sb(III), has affinity to Al-oxides in soil (Nakamuru and Altansuvd 2014). Aluminum may therefore have a role in the mobility of Sb in soil water. Other elements with a prominent correlation in soil water were Pb and Mn, Cu and Zn and Mn, Ca and Mg, Fe and Mn, Mg and Mn.

At four of the sites, the concentrations of Pb, Cu, Sb and Zn in the soil water rapidly decreased vertically in the soil column. Irrespectively of mire type, more than 90 % of Pb, Cu and Sb in soil water were located in the top soil column. Although a decrease in the concentration of Pb, Cu, Sb in the soil column was observed, the concentrations of Pb and Cu in the deepest layer were higher than observed in pore water collected at 40 cm depth from bogs near industrial sites in Czech Republic and Finland (Novak and Pacherova 2008; Shotyk et al. 2016). When comparing the soil and soil water concentrations of Pb, Sb and Cu some differences appeared between the ranges. The shooting ranges at **Av** and **St** had the highest soil concentrations of Pb and Cu. The ranges at **St** and **Ma** had the highest soil concentrations of Sb, and highest soil water concentrations of Pb, Cu and Sb (based on the soil water concentrations sampled with Rhizon samplers). **St** and **Ma** are both shooting ranges that are still active and probably have more freshly deposited residues of ammunition. It is reasonable to believe that freshly deposited bullets leak more metal due to a more exposed metal surface. During weathering, a coating of oxides develops on the surface of the bullet residue that may act as a protective layer reducing the leaching of soluble metals. On a Pb bullet a cover of litharge (PbO), hydrocerrussite ($Pb(CO_3)_2(OH)_2$) and cerussite ($PbCO_3$) may develop (Vantelon et al. 2005). The surface of bullets covered with brass and brass containing cartridges are usually covered by cuprous oxide (Cu_2O) and zinc oxide (ZnO) (Qiu and Leygraf 2011). Hardison Jr et al. (2004) observed a higher decomposition rate of the smaller Pb bullet fragments than the larger ones. They also claimed that the smallest fragments are

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too small to develop a protective layer of oxides and decompose relatively fast, thus releasing Pb into the water. An acidic environment is likely to slow the oxidation process and facilitate leakage.

The soil water concentration of Pb, Cu, Zn, and Sb at the range at **Kj** and **He** were more evenly spread in the soil column (Table 3). The mire at **He** was the most acidic mire and had less ammunition residues deposited. This mire was relatively shallow (~0.5 m deep) and it is possible that the high Cu concentration in the soil water may have a natural origin from the bedrock beneath. The soil water at **Kj** had relatively low concentrations of both Pb (3.0 µg/L) and Cu (3.6 µg/L) near the surface. The mire at the **Kj** range had similar characteristics as the organic soil at the **Ti** range. Below the 15 cm layer the soil water concentrations of Pb, Cu and Sb between the two sites were similar (based on the median concentrations). The low concentrations of Pb and Cu in the soil water near the surface at **Kj** may be attributed to more alkaline conditions.

The sampled soil water at **Av** shooting range was squeezed from the soil by manual power and represent crude soil water fractions, which probably also include metal species that are less mobile in the soil water. Even though the water samples were filtered (0.45 µm filtered) before analysis they contained considerable amount of dissolved organic matter seen as precipitated matter after storage. The element concentrations in the soil water were, therefore, probably overestimated. The median soil water concentrations at the top soil showed concentrations of 378 µg/L, 189 µg/L, 166 µg/L and 70 µg/L of Pb, Cu, Zn and Sb respectively (Table 3). For comparison, soil water was sampled with designated samplers (Rhizon samplers) at similar sampling points. These had median soil water concentrations of 43 µg/L, 12 µg/L, 64 µg/L and 13 µg/L of Pb, Cu, Zn and Sb respectively. These concentrations are a factor of 9, 16, 3 and 5 lower than the concentrations in the water achieved by squeezing the water out of the soil. For other elements, such as Fe, Mn, Al, Ca,

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Mg and K, the difference was much less between the sampling methods (Table 4S) indicating that these elements are less associated with particulate matter compared to Pb, Cu, Sb and Zn.

At three of the sites, some of the holes that were dug for soil sampling were quickly filled with water, apparently from cracks in the soil columns. The mean concentrations of Pb, Cu, Zn and Sb in this water collected from **St** and **He** resembled the concentrations in the discharge creeks running through the mires (Table 4), indicating an origin from the surface water. At the third site, **Ti**, there was a much larger variation in the element concentration in this water, which made it more difficult to assess its origin. This may partly be due to the mixed soil characteristics at this site. Nevertheless, the concentrations of Pb, Cu and Sb in this water from **Ti** were much higher than in the soil water collected from the deeper soil layer (30-45 cm) indicating an origin from the soil surface (Table 4). Typically, the hydraulic conductivity of water in mires is very low. In moderately decomposed peat, a flow rate of 10^{-6} to 10^{-8} m/s has been reported (Rydin and Jeglum 2008). Zones in the mire with high hydraulic conductivity are, however, reported (Rydin and Jeglum 2008). The observed cracks in the soil column are probably channels in the mires from remnants of roots and plants, or layers in the mire with less decomposed materials. Such layers in the soil column probably contribute to an increased transport rate of trace metal contaminated water into deeper soil layers. Similar observations have been previously reported by Knechtenhofer et al. (2003) who showed that plant roots facilitated vertical transport of Pb, Cu and Sb in an acidic mineral soil with some accumulation of organic matter. Cracks in clay-rich soil as a result of desiccation are also reported to facilitate downward transport of contaminated water in the soil profile (Murray et al. 1997).

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Sequential extractions of peat soil

At shooting ranges located on mire, the deposited ammunition is typically submerged into more or less water saturated conditions in soil rich in organic matter. To assess the adsorption characteristic of different elements, sequential extractions of peat from **Av** and the organic rich soil from **Ti** were performed (Fig 3, Table 5S and 8S). Pb and Zn had similar adsorption characteristics. A high proportion of Pb and Zn were found in fractions 2 and 3. Elements in these fractions usually occur in ion exchangeable forms, sensitive to pH-fluctuation, and are probably adsorbed to the surface of iron (hydr)oxides and organic matter as outher-sphere complexes (Stumm 1995; Violante 2013). In addition, Pb-carbonates are presumed to be dissolved during the extraction of fraction 3. This observation is similar to what observed by Syrovetnik et al. (2007), who determined binding mechanisms of different elements in peat from an Estonian bog. The binding characteristics of Pb depend on the soil type and probably the degree of contamination. In Pb contaminated acidic forest soil with high content of organic matter (30-50%), approximately 50% of the Pb was associated with the salt-exchangeable fraction, resembling fraction 2 in this study (Ettler et al. 2005). In addition a substantial amount of anglesite ($PbSO_4$) was identified (Ettler et al. 2005). In mineral soils from shooting ranges, Pb was shown to be primarily associated with the carbonate fraction resembling fraction 3 in this study (Cao et al. 2003; Duggan and Dhawan 2007). This was attributed to the identification of a substantial amount of hydrocerussite ($Pb_3(CO_3)_2(OH)_2$), which, in addition to cerrusite ($PbCO_3$), is one of the main transformation products of Pb bullets in shooting range soils (Jörgensen and Willems 1987). In a study by Hartikainen and Kerko (2009) a larger portion of Pb was found in the residual fraction (35-88%) in both mineral soil and in soil with high content of organic matter (49-76%). The authors suggested that this was due to naturally occurring reserves in the mineral matrix, or solely to an artifact produced by the extraction procedure. Also Cao et al. (2003) found

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substantial amounts of Pb in the residual fraction. They attributed this finding to the formation of less soluble Pb species, such as phosphate containing hydroxypyromorphite ($Pb_5(PO_4)_3OH$). Sekaly et al. (1999) observed that the proportion of inert complexes of Pb with fulvic acids increased as the metal/fulvic acid ratio decreased. This implies that with a high input of Pb the strong binding sites becomes saturated and the proportion of labile complexes increases.

In contrast to Pb and Zn, a large portion of Cu was associated to the oxidizable components (fraction 5), which may imply that a significant portion of Cu form inner-sphere complexes with the peat and organic soil. Similar results were observed in peat from an Estonian bog (Syrovetnik et al. 2007). Peat and bottom sediments contain a large portion of humic and fulvic acids, which are complex organic components made of aromatics and sugars with carboxylic and phenolic moieties. The nature of humic and fulvic acids favor the adsorption of cationic substances, such as Cu, Pb and Zn. Several studies have evaluated metal binding to humic and fulvic acids and showed that Cu has a higher affinity to peat and humic substances than Pb (e.g. Manunza et al. 1995; Logan et al. 1997; Christl et al. 2001). Christ et al. (2001) showed that Cu was more strongly bound to humic substances than Pb, particularly at low environmentally relevant metal concentrations. It was suggested that Cu formed both monodentate and bidentate complexes with humic substances, whereas Pb predominantly was bound as monodentate complexes. In addition, it has been shown that Cu has a higher affinity to phenolic containing sites than Pb (Manunza et al. 1995; Christl et al. 2001).

A large portion of Sb (> 50%) was associated to the oxidizable components. Sb in natural water can be found in two oxidation states, as Sb(III) and Sb(V) respectively (Filella et al. 2002). Only few studies have performed speciation analysis of Sb in soil water, sampled from shooting ranges, and found that the major Sb species is Sb(V), probably as the

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negatively charged $Sb(OH)_6^-$ (Johnson et al. 2005; Okkenhaug et al. 2013, 2016). Similarly, most investigations have primarily found Sb(V) in contaminated soils (Ettler et al. 2007; Scheinost et al. 2006; Ackermann et al. 2009). The large portion of Sb associated with the oxidizable components in the peat soil may therefore appear unexpected since the organic substances in peat should not favor adsorption of the negatively charged $Sb(OH)_6^-$. Metallic antimony is used as an alloy in lead bullets and on impact in berms the metals in the bullets may be subjected to oxidation processes. In a study by Ilgen et al. (2014) it was shown that Sb^0 is relatively fast oxidized to Sb(III) and Sb(V). The rate of oxidation of Sb(III) to Sb(V) was shown to increase in the presence inorganic salts, such as Ca and Na, and decrease with the presence of the organic ligand EDTA. Under anaerobic conditions it has also been postulated that Sb(V) in soil water can be reduced to Sb(III) and in shooting range soils under anaerobic conditions it has been observed an increased level of Sb(III) (Hockmann et al. 2015). Mires with water saturation conditions and low oxygen pressure may therefore favor increased formation of Sb(III). Ettler er al. (2007) found considerable amounts of Sb(III) in organic rich acidic soils, which indicates that Sb(III), once formed, may be associated with organic matter. Some studies have also indicated that Sb(III) form complexes with organic ligands and may adsorb to humic substances (Pilarski et al. 1995; Buschmann and Sigg 2004; Johnson et al. 2005). In a study by Tighe et al. 2005 it was observed that both Sb(III) and Sb(V) can adsorb to humic substances, particularly at pH lower than 6.5. The mechanisms of Sb(V) adsorption were linked to the adsorption to amino functional groups or complexation with Fe and Al impurities of the humic acid. In a later study by Tella and Pokrovski (2012), it was shown that Sb(V) could form stable bidentate complexes with carboxylic and phenolic functional groups of organic ligands, particularly in acidic organic-rich waters with pH $\leq$ 4. These findings may therefore imply that Sb may form inner-sphere complexes with organic matter in the peat.

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In several studies, Sb has been shown to have high affinity for amorphous iron (hydr)oxides (e.g. Ambe 1987; Tighe al. 2005; Johnson et al. 2005; Leverett et al. 2012) and aluminum (hydr)oxides (Nakamaru and Altansuvd, 2014). Although most of the Fe and Al were found in fraction 4 and in the residual fraction (Table 5S), a relatively large portion of both were associated with the oxidizable fraction (fraction 5) indicating that a substantial amount of Fe and Al was associated to organic materials. Depending on the surface charge, iron oxide surfaces (containing FeO^-, $FeOH^0$, $FeOH_2^+$ moieties) are able to sorb both cationic and anionic species on hydroxyl groups. The surface charge of iron (hydr)oxides depends on the pH and higher pHs favor cation adsorption. The iso-electrical point of iron (hydr)oxides varies between 7 and 10 (Kosmulski 2016). It is therefore plausible that the acidic environment in the mire favor adsorption of $Sb(OH)_6^-$ to iron (hydr)oxide particles associated with organic components in the soil. Similar sorption mechanisms of Sb as for iron (hydr)oxides may be applicable for Al. Few studies have, however, evaluated the interaction between Al and Sb, but it has been shown that Sb, particularly Sb(III) has affinity for Al-oxides in soil (Nakamuru and Altansuvd 2014). Aluminum has high affinity for organic materials (e.g. Lee 1985; Weng et al. 2002) and the major Al species in acidic conditions in natural waters are Al^{3+}, $AlOH^{2+}$, and $Al(OH)^+$ (Lee 1985). In addition Al may form several polynuclear complexes (Lee 1985). Al may therefore have a role in the mobility of Sb in soil and water.

Conclusions

Substantial amounts of ammunition residues have been deposited during the years at the investigated shooting ranges. The relatively low concentrations of Pb, Zn, Sb and Cu both in the soil and soil water already 20 cm below the top soil compared to the top soil, indicate limited vertical migration of these elements in the soil. The concentrations of Pb and Sb at 30-

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45 cm in soil and soil water were nevertheless in general higher than the expected background levels, indicating that some vertical transport is occurring. The spread of metals from the deposited small arms ammunition to the nearby environment most likely occurs primarily by surface runoff. However, channels in the mires, made by plant roots or soil layer of less decomposed materials, may increase the rate of transport of contaminated surface water into deeper soil layers and ground water. The highest concentrations of Pb, Cu and Sb were found in soil water from ranges that are still in use, namely **St,** which is an acidic mire, poor in ions such as Ca and Mg, and **Ma**, which is a less acidic mire with higher content of Ca. No clear differences in the discharge of ammunition residues between the mires were observed based on the characteristics of the mires. A more frequent sampling of surface water and soil water under different meteorological conditions may have revealed larger differences between the sites. The relatively high concentrations of Pb and Cu in the soil water and discharge water from **He** as compared to the soil concentration levels, indicates, however, that acidic mires poor on ions, such as Ca and Mg, probably have a higher potential for spreading the contaminants in the environment. In the surface waters with higher pH (pH ~7) a trend was observed with high concentrations of Sb and lower relative concentrations of Cu and Pb. A large portion of both Cu and Sb appeared to be associated to the oxidizable components in the peat, which may imply that these elements form inner-sphere complexes with organic matter in the peat. The largest fractions of Pb and Zn were associated with the exchangeable and pH-sensitive components in the peat, which may imply that these elements primarily form outer-sphere complexes with the peat.

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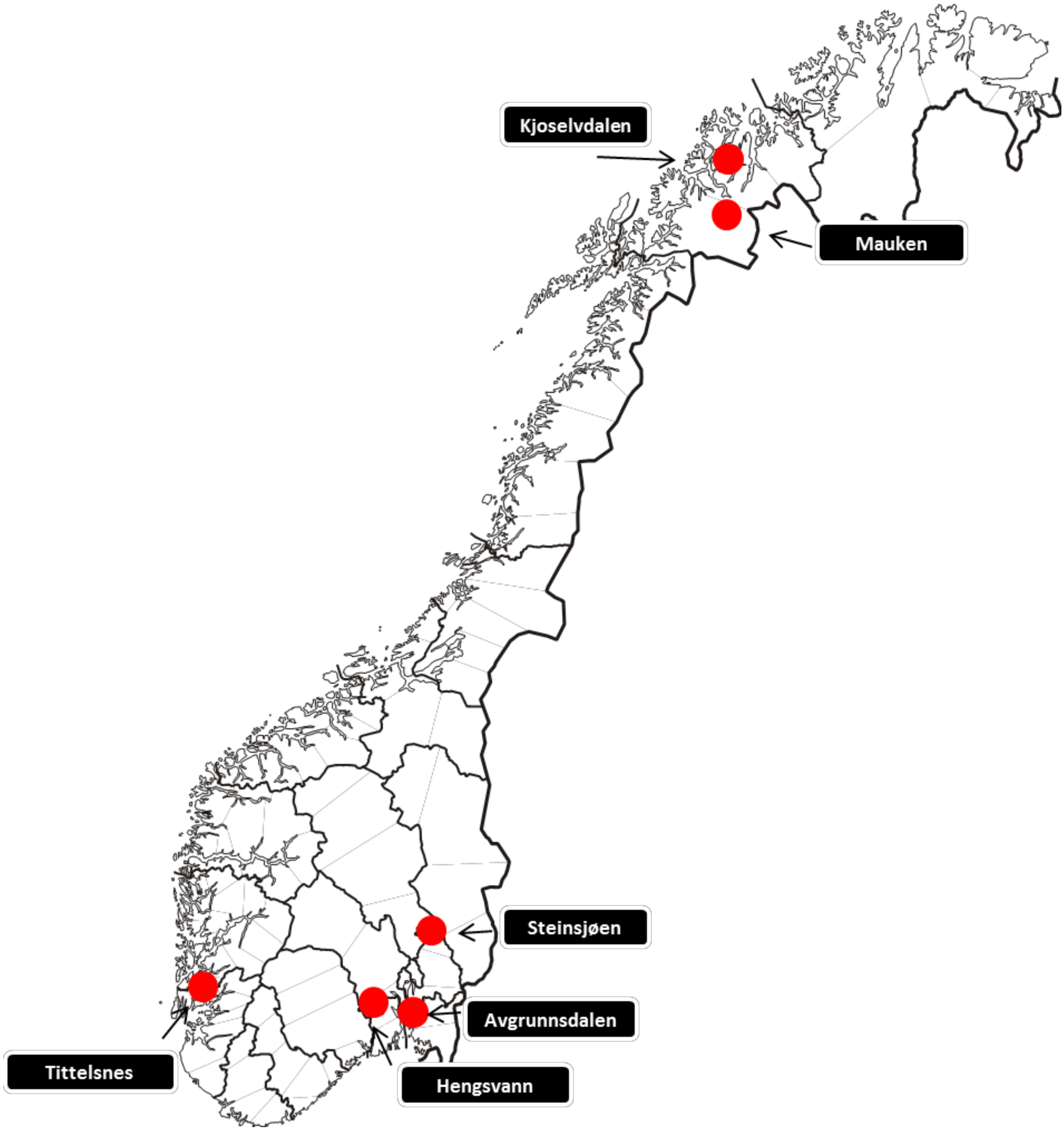

Fig 1 Map of Norway showing the locations of the investigated shooting ranges.

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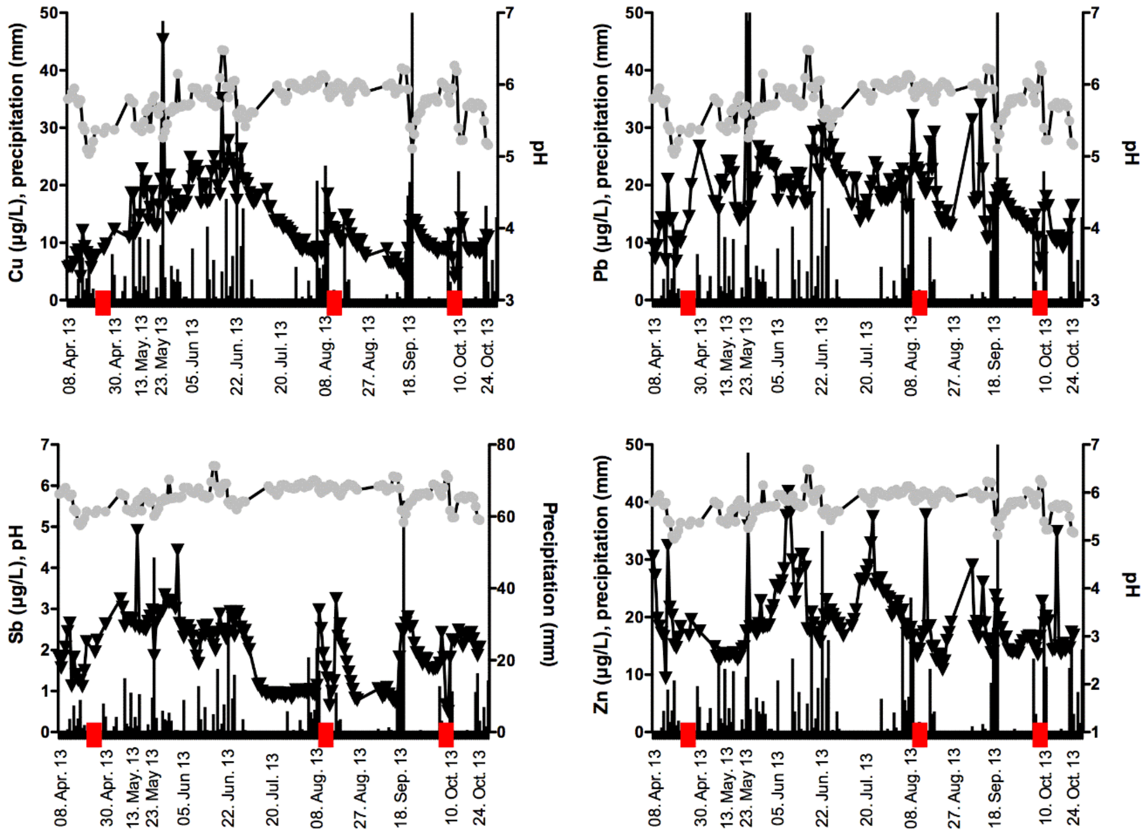

Fig 2 Daily concentrations of Cu, Pb, Sb and Zn in the runoff water at Avgrunnsdalen shooting range for small arms as a function of precipitation. pH was measured in the creek weekly. The pH fluctuations of the water are shown in higher resolution in Fig 7S. The boxes in red indicate periods where some remediation measures were performed at the shooting range area. —▼— -data for total concentration (µg/L) of the elements. ━●━ pH of the inlet water. —— Precipitation (mm) data from Sande in the Vestfold County near Avgrunnsdalen shooting range (Norwegian Meteorological Institute).

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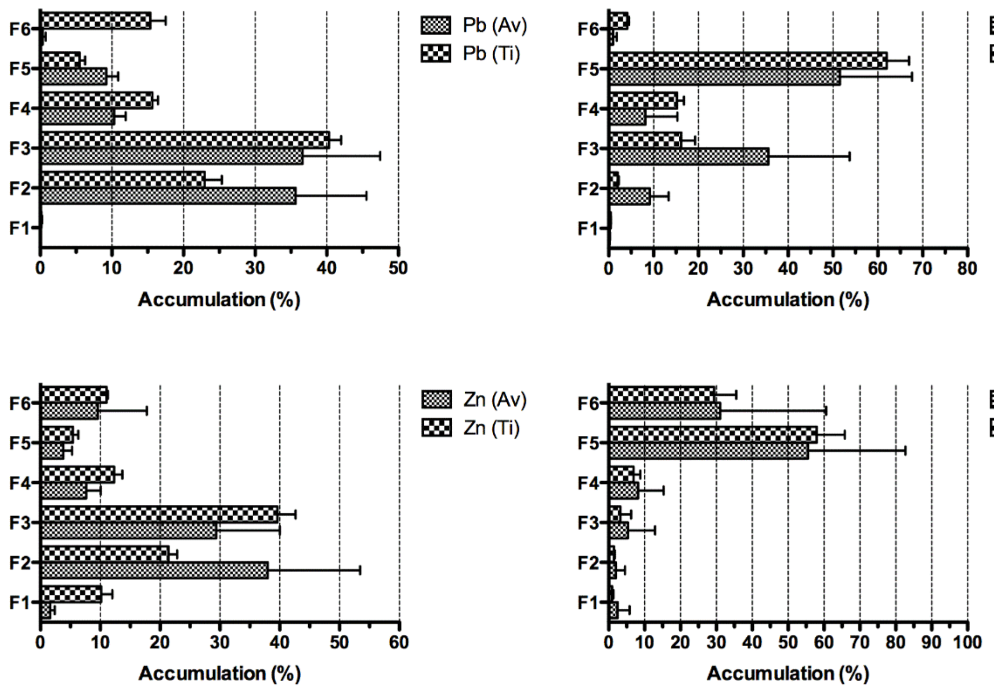

Fig 3 Sequential extractions of peat and organic rich soil from **Av** and **Ti.** The figure shows, for each extracted fraction, the proportion of the total concentration of Pb, Cu, Zn and Sb, respectively. Fractions F1-F3 are the easy dissolvable, more bioavailable fractions. Fractions F4 and F5 contain less available elements, associated with, for example, Fe- and Mn-oxides and organic materials, respectively. Fraction F6 is the residual fraction.

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Table 1 Concentrations of 10 elements in non-filtered and filtered discharge water from the six shooting ranges for small arms. The results are from one sample, which was taken at the day the field work was performed and represents a snapshot of the element concentration in the streams. The numbers in brackets are the proportion between Pb and Cu, and Pb and Sb. At **Ti** there were two small streams running through the range.

Range	Pb (µg/L)	Cu (µg/L)	Sb (µg/L)	Zn (µg/L)	Na (mg/L)	K (mg/L)	Mg (mg/L)	Ca (mg/L)	Al (mg/L)	Fe (mg/L)	Mn (µg/L)	pH
Av	28/24	14 (2)/13 (1.8)	3.1 (9.0)/3.1 (7.7)	18/18	3.6/3.6	0.42/0.40	0.53/0.54	1.9/1.9	0.35/0.33	0.77/0.64	65/66	5.85
St	176/174	415 (0.4)/413 (0.4)	65 (2.7)/66 (2.6)	629/586	0.83/0.86	0.50/0.52	0.42/0.41	1.4/1.4	0.44/0.42	0.53/0.49	109/109	4.77
Ma	7.3/6.2	11/ (0.7)/10 (0.6)	8.3 (0.9)/8.2 (0.8)	5.7/5.6	1.9/1.9	0.65/0.64	1.2/1.2	8.8/8.8	0.06/0.06	0.36/0.30	36/35	7.14
Ti-1	18/15	19 (0.9)/17 (0.9)	1.8 (10)/1.8 (8.3)	12/12	8.9/8.9	0.62/0.59	1.5/1.5	1.9/1.9	0.19/0.21	0.49/0.48	4.6/4.7	6.20
Ti-2	7.5/7.7	86 (0.09)/78 (0.1)	0.34 (22)/0.34 (23)	53/49	6.7/6.7	0.13/0.10	0.84/0.84	1.0/1.0	0.44/0.44	0.67/0.68	6.9/6.8	4.64
Kj[a]	1.9	36 (0.05)	3.9 (0.50)	273	6.5	1.5	3.6	22	0.03	0.03	4.2	7.00
He	3.9/3.6	12 (0.33)/11 (0.32)	0.39 (10)/0.39 (9.2)	9.1/11	0.62/0.61	0.06/0.03	0.10/0.10	0.42/0.41	0.40/0.39	0.47/0.44	11/11	4.46

[a]The results from **Kj** is for filtered samples only (0.45 µm).

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Table 2 Mean (± SD) concentration of Pb, Cu, Sb and Zn, and pH in peat soil collected from three vertical layers from six shooting ranges for small arms, and the average soil-water pH. The median soil concentrations are shown in brackets.

Soil origin	Pb (g/kg)	Cu (g/kg)	Sb (mg/kg)	Zn (g/kg)	pH	L.O.I.[b] (%)	*N*[a]
Av 0-15 cm	6.3 ± 4.9 (4.7)	0.80 ± 0.48 (0.71)	170± 11 (140)	0.43 ± 0.36 (0.30)	4.32 ± 0.29 (4.34)	76 ± 17 (77)	13/13
Av 15-30 cm	0.57 ± 1.2 (0.13)	0.25 ± 0.54 (0.03)	30± 50 (7.0)	0.20± 0.45 (0.04)	4.10 ± 0.14 (4.24)	87 ± 15 (95)	12/13
Av 30-45 cm	0.15 ± 0.29 (0.04)	0.02 ± 0.02 (0.01)	6.0± 10 (2.0)	0.03± 0.02 (0.02)	3.98 ± 0.16 (4.12)	90 ± 16 (97)	11/12
St 0-15 cm	13 ± 10 (11)	5.2 ± 2.9 (4.3)	830 ± 1100 (520)	1.1 ± 0.65 (1.0)	4.06 ± 0.25 (4.10)	84 ± 17 (87)	30/20
St 15-30 cm	0.36 ± 0.82 (0.11)	0.19 ± 0.33 (0.06)	40 ± 50 (20)	0.18 ± 0.19 (0.15)	3.79 ± 0.40 (3.78)	93 ± 7.6 (96)	23/19
St 30-45 cm	0.06 ± 0.06 (0.04)	0.04 ± 0.05 (0.03)	4.0 ± 5.0 (2.0)	0.03 ± 0.06 (0.01)	3.73 ± 0.35 (3.78)	71 ± 40 (94)	19/18
Ti 0-15 cm	2.3 ± 1.5 (2.6)	0.62 ± 0.60 (0.41)	50 ± 50 (30)	0.14 ± 0.19 (0.08)	4.70 ± 0.12 (4.86)	32 ± 1.1 (33)	12/5[bc]
Ti 15-30 cm	0.39 ± 0.77 (0.11)	0.58 ± 1.5 (0.09)	30 ± 70 (4.0)	0.11 ± 0.14 (0.06)	4.78 ± 0.24 (4.88)	39 ± 31 (38)	12/13
Ti 30-45 cm	0.11 ± 0.15 (0.04)	0.03 ± 0.02 (0.02)	2.0 ± 2.0 (2.0)	0.05 ± 0.03 (0.03)	4.91 ± 0.39 (4.85)	19 ± 24 (10)	12/10
Ma 0-15	1.4 ± 1.8 (0.52)	0.78 ± 1.1 (0.34)	175 ± 180 (140)	0.22 ± 0.20 (0.15)	4.68 ± 0.31 (4.69)	89 ± 7.5 (93)	11/10
Ma 15-30	0.13 ± 0.17 (0.05)	0.05 ± 0.05 (0.03)	10 ± 10 (7.0)	0.01 ± 0.01 (0.01)	4.61 ± 0.50 (4.63)	94 ± 0.9 (94)	12/11
Ma 30-45	0.05 ± 0.03 (0.05)	0.03 ± 0.02 (0.02)	4.0 ± 4 (3.0)	0.01 ± 0.01 (0.01)	4.68 ± 0.59 (4.70)	94 ± 1.1 (94)	10/9
Kj 0-15	2.5 ± 1.9 (2.1)	1.1 ± 1.1 (0.57)	80 ± 10 (3.0)	0.22 ± 0.16 (0.17)	4.91 ± 0.27 (4.94)	62 ± 21 (61)	12/13
Kj 15-30	0.03 ± 0.05 (0.01)	0.09 ± 0.06 (0.07)	1.0 ± 2.0 (1.0)	0.03 ± 0.03 (0.02)	4.67 ± 0.37 (4.72)	69 ± 32 (90)	12/13
Kj 30-45	0.02 ± 0.02 (0.01)	0.06 ± 0.03 (0.06)	< 0.001	0.03 ± 0.03 (0.03)	4.74 ± 0.41 (4.78)	64 ± 37 (83)	12/13
He 0-15	0.26 ± 0.26 (0.13)	0.05 ± 0.07 (0.04)	0.02 ± 0.03 (0.005)	0.03 ± 0.03 (0.005)	4.11 ± 0.36 (4.14)	70 ± 15 (69)	13/13
He 15-30	0.03 ± 0.01 (0.03)	0.01 ± 0.01 (0.01)	0.001 ± 0.001 (0.001)	0.001 ± 0.001 (0.001)	4.09 ± 0.47 (4.08)	55 ± 30 (65)	13/13
He 30-45	0.03 ± 0.02 (0.03)	0.01 ± 0.01 (0.01)	0.001 ± 0.001 (0.001)	0.001 ± 0.001 (0.001)	4.17 ± 0.23 (4.20)	53 ± 37 (59)	13/13

[a] Number of samples analyzed for elements and pH and L.O.I. respectively

[b] L.O.I.: loss on ignition

[c] L.O.I. was analyzed on five subsamples of a pooled sample

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Table 3 Mean (± SD) soil water concentrations of Pb, Cu, Sb and Zn collected from three vertical layers from three small arms shooting ranges. The median soil water concentrations are shown in brackets. The soil water was sampled with Rhizon soil water samplers.

Water origin	Pb (µg/L)	Cu (µg/L)	Sb (µg/L)	Zn (µg/L)	N
Av 0-15 cm	75 ± 81 (43)	16 ± 19 (12)	24 ± 28 (13)	77 ± 52 (65)	20
Av 0-15 cm[a]	552 ± 389 (378)	860 ± 2610 (189)	108 ± 128 (70)	200 ± 139 (166)	55
Av 15-30 cm[a]	113 ± 327 (36)	93 ± 431 (18)	65 ± 336 (9.1)	183 ± 111 (156)	49
Av 30-45 cm[a]	63 ± 165 (27)	61 ± 277 (16)	12 ± 23 (5.4)	265 ± 228 (207)	46
St 0-15 cm	2500 ± 4100 (1100)	900 ± 1200 (600)	150 ± 93 (150)	1600 ± 1300 (1300)	54
St 15-30 cm	130 ± 420 (8.3)	25 ± 18 (21)	15 ± 16 (9.1)	190 ± 230 (100)	40
St 30-45 cm	14 ± 36 (4.5)	18 ± 18 (21)	5.5 ± 6.0 (4.3)	62 ± 38 (57)	38
Ti 0-15	65 ± 44 (49)	57 ± 28 (46)	8.9 ± 9.0 (6.5)	82 ± 54 (63)	24
Ti 15-30	22 ± 47 (2.2)	11 ± 17 (5.4)	1.7 ± 2.0 (0.74)	68 ± 40 (46)	24
Ti 30-45	7.1 ± 12 (3.4)	4.6 ± 5.6 (2.3)	0.42 ± 0.36 (0.27)	40 ± 27 (33)	19
Ma 0-15 cm	470 ± 1200 (110)	260 ± 900 (76)	120 ± 120 (66)	170 ± 160 (130)	35
Ma 15-30 cm	82 ± 150 (34)	33 ± 37 (22)	14 ± 22 (8.6)	62 ± 53 (43)	30
Ma 30-45 cm	28 ± 33 (15)	19 ± 34 (6.3)	3.6 ± 3.6 (2.5)	36 ± 13 (31)	30
Kj 0-15	3.0 ± 2.7 (2.4)	3.6 ± 2.8 (3.0)	2.9 ± 3.2 (1.3)	44 ± 32 (32)	20
Kj 15-30	3.0 ± 5.4 (2.0)	1.3 ± 0.88 (1.2)	1.0 ± 1.3 (0.57)	41 ± 35 (36)	20
Kj 30-45	4.4 ± 6.6 (2.2)	1.8 ± 1.8 (1.2)	0.40 ± 0.33 (0.30)	37 ± 35 (29)	20
He 0-15	5.5 ± 4.5 (4.8)	32 ± 26 (25)	3.5 ± 4.9 (1.7)	72 ± 24 (62)	24
He 15-30	4.9 ± 5.9 (3.5)	24 ± 20 (17)	0.92 ± 0.94 (0.53)	81 ± 40 (74)	24
He 30-45	3.1 ± 2.7 (2.1)	25 ± 27 (10)	0.59 ± 0.35 (0.52)	77 ± 36 (73)	25

[a]The soil water was sampled by squeezing manually the soil followed by filtration of the water sample through a 0.45 µm filter.

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Table 4 Mean (± SD) concentrations of different elements in filtered (0.45 µm) water from channels in the soil column. The element concentrations are compared with the element concentrations in the soil water sampled 30-45 cm below the soil surface and the element concentrations in filtered water from the creeks running through the ranges. The median soil water concentrations are shown in brackets.

Elements	**St** (water channels)	**St** (creek)	**St** (soil water)	**Ti** (water channels)	**Ti**[a] (creek)	**Ti** (soil water)	**He** (water channels)	**He** (creek)	**He** (soil water)
Pb (µg/L)	340 ± 220 (340)	174	14 ± 36 (4.5)	75 ± 57 (60)	15/7.7	7.1 ± 12 (3.4)	3.6 ± 3.3 (2.2)	3.6	3.1 ± 2.7 (2.1)
Cu (µg/L)	500 ± 380 (390)	413	18 ± 18 (21)	43 ± 41 (34)	17/78	4.6 ± 5.6 (2.3)	7.4 ± 7.7 (5.4)	11	25 ± 27 (10)
Sb (µg/L)	88 ± 74 (50)	66	5.5 ± 6.0 (4.3)	6.8 ± 8.9 (2.8)	1.8/0.34	0.42 ± 0.36 (0.27)	1.5 ± 0.90 (1.2)	0.39	0.59 ± 0.35 (0.52)
Zn (µg/L)	390 ± 260 (340)	586	62 ± 38 (57)	19 ± 14 (15)	12/49	40 ± 27 (33)	23 ± 8.2 (20)	11	77 ± 36 (73)
Fe (mg/L)	0.69 ± 0.43 (0.53)	0.49	1.9± 2.2 (1.0)	0.80 ± 0.67 (0.57)	0.48/0.68	2.50 ± 2.5 (1.2)	0.43 ± 0.40 (0.21)	0.44	0.21 ± 0.41 (0.10)
Mn (µg/L)	100 ± 32 (100)	109	87 ± 136 (44)	23 ± 15 (16)	4.7/6.8	36 ± 22 (34)	6.8 ± 5.7 (4.4)	11	5.4 ± 2.7 (4.8)
Al (mg/L)	0.80 ± 0.37 (0.82)	0.42	0.67 ± 0.48 (0.59)	0.46 ± 0.34 (0.40)	0.21/0.44	0.31 ± 0.25 (0.18)	0.82 ± 0.26 (0.82)	0.39	0.58 ± 0.22 (0.56)
Ca (mg/L)	1.9 ± 0.42 (1.8)	1.4	1.6 ± 1.3 (1.4)	1.5 ± 0.520 (1.4)	1.9/1.0	3.2 ± 2.3 (2.2)	0.75 ± 0.45 (0.55)	0.41	1.7 ± 0.86 (1.5)
Mg (mg/L)	0.48 ± 0.12 (0.40)	0.41	0.41 ± 0.31 (0.34)	1.0 ± 0.36 (0.95)	1.5/0.84	1.4 ± 0.38 (1.3)	0.09 ± 0.05 (0.08)	0.10	0.15 ± 0.06 (0.14)
Na (mg/L)	0.74 ±0.07 (0.73)	0.83	1.2 ± 0.33 (1.2)	7.1 ± 2.1 (7.0)	8.9/6.7	11 ±2.1 (11)	0.68 ± 0.27 (0.56)	0.61	0.83 ± 0.46 (0.69)
K (mg/L)	0.31 ± 0.14 (0.31)	0.52	0.47 ± 0.18 (0.47)	0.64 ± 0.35 (0.64)	0.59/0.10	0.52 ± 0.34 (0.46)	0.32 ± 0.20 (0.28)	0.03	0.58 ± 0.62 (0.34)

[a]At **Ti** there were two small drainage creeks running through the range (ref. Table 1).

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Supplementary materials,

Distribution and mobility of lead (Pb), copper (Cu), Zinc (Zn) and antimony (Sb) from ammunition residues on shooting ranges for small arms located on mires

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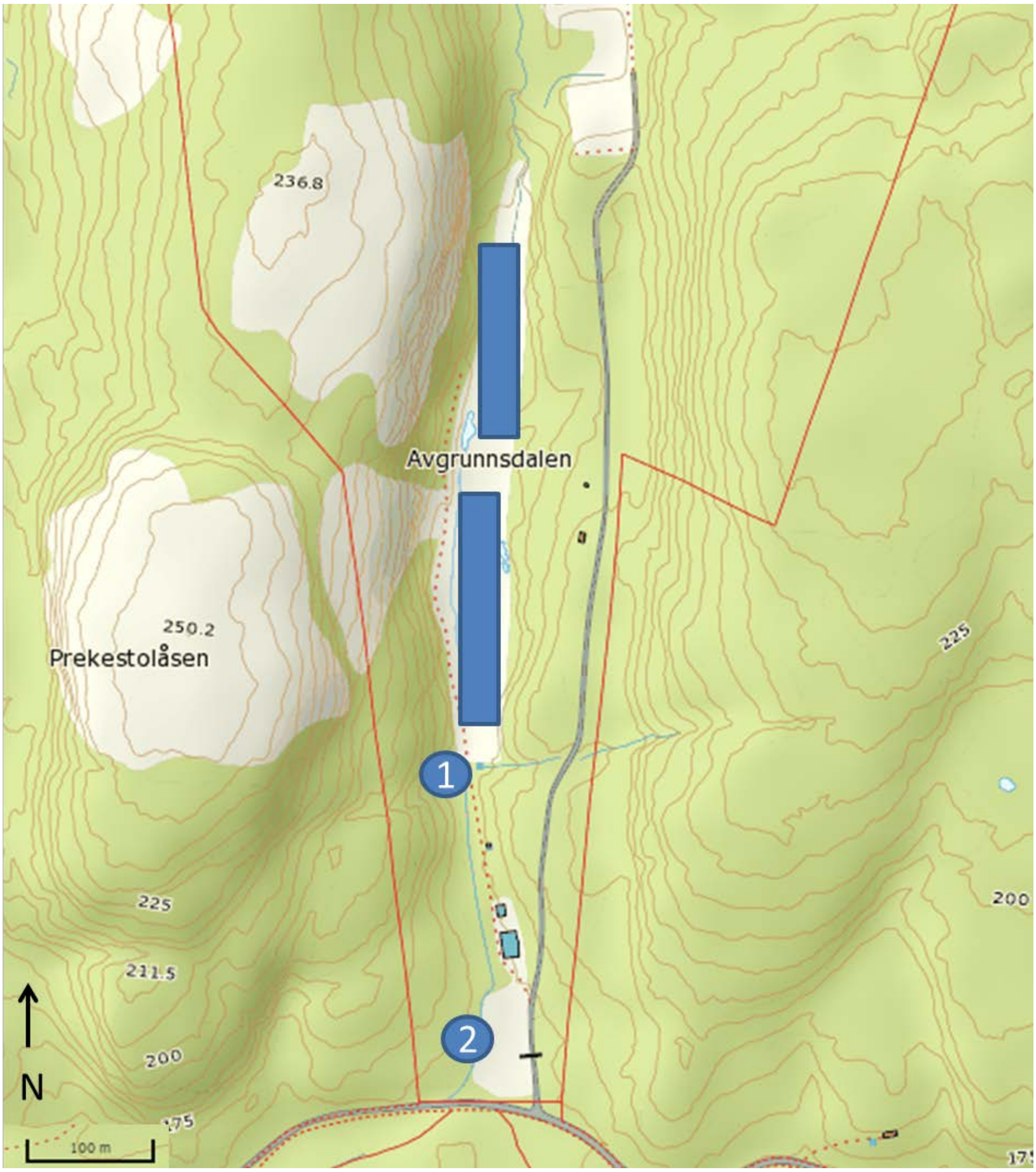

Fig 1S Map of Avgrunnsdalen (**Av**) shooting range for small arms (59.58042°N, 10.51218°E). A network of sampling points was distributed approximately 5 meters apart within the two rectangular blue boxes. Water samples were taken at a small creek at the site marked with the blue circle no. 1. In 2013, an ISCO water sampler was placed approximately 200 m downstream of the mire at the site marked with the blue circle no. 2 (Map: The Norwegian Mapping Authority).

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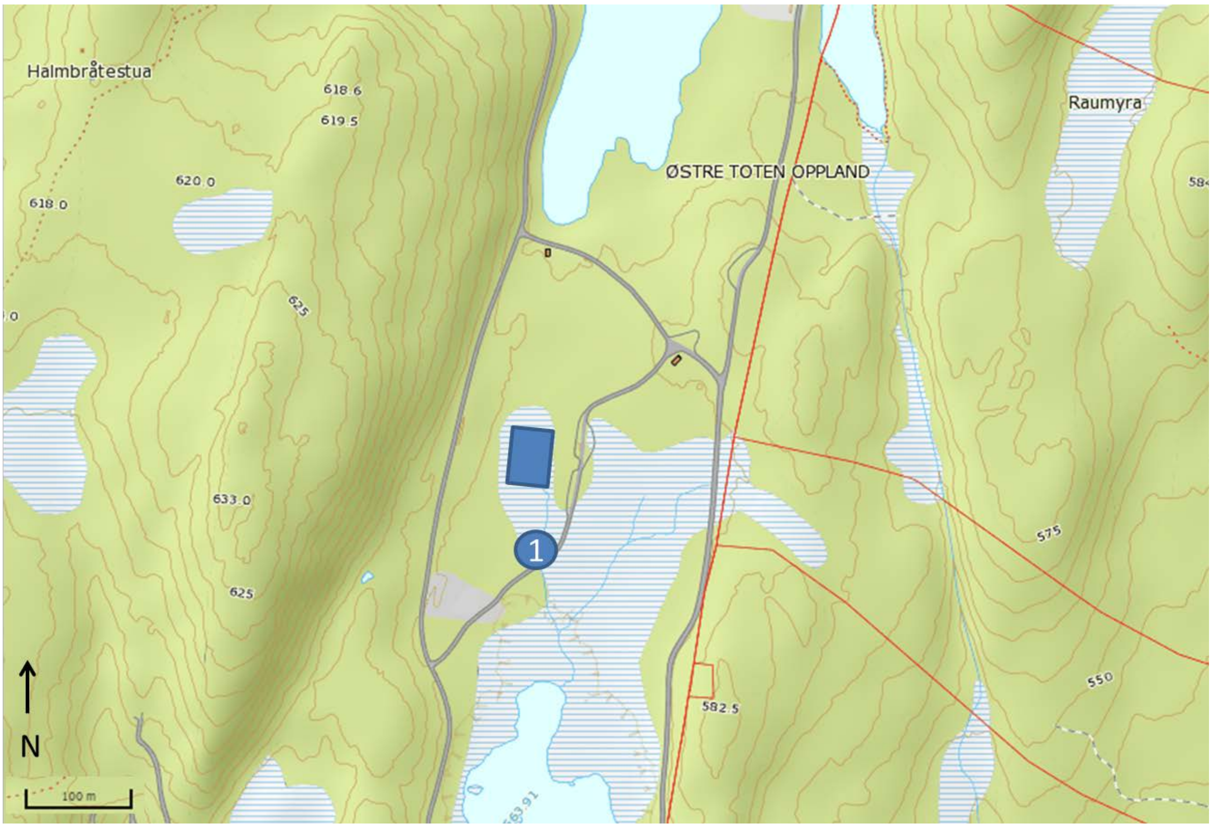

Fig 2S Map of Steinsjøen (**St**) shooting range for small arms (60.54248°N, 11.09554°E). A network of sampling points was distributed approximately 5 meters apart within the rectangular blue box. Water samples were taken at a small creek at the site marked with a blue circle (Map: The Norwegian Mapping Authority).

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Supplementary materials,

Fig 3S Map of Mauken (**Ma**) shooting range for small arms (69.04592°N, 19.24131°E). A network of sampling points was distributed approximately 5 meters apart within the red box. Water samples were taken at a small creek at the site marked with a blue circle (Map: The Norwegian Mapping Authority).

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Supplementary materials,

Fig 4S Map of Tittelsnes (**Ti**) shooting range for small arms (59.72306°N, 5.51481°E). A network of sampling points was distributed approximately 5 meters apart within the rectangular blue box. Water samples were taken at a small creek at the site marked with a blue circle (Map: The Norwegian Mapping Authority).

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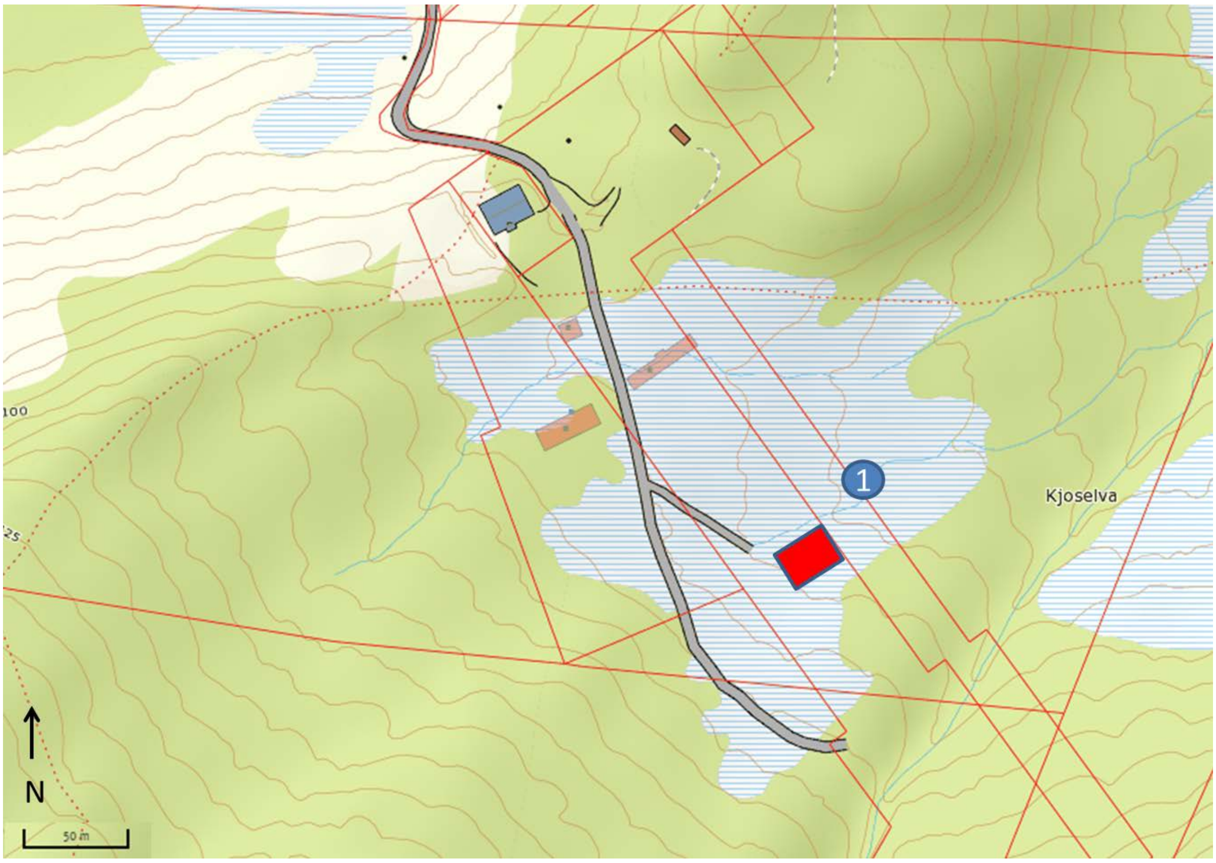

Fig 5S Map of Kjoselvdale (**Kj**) shooting range for small arms (69.73210°N, 19.14590°E). A network of sampling points was distributed approximately 5 meters apart within the red box. Water samples were taken at a small creek at the site marked with a blue circle (Map: The Norwegian Mapping Authority).

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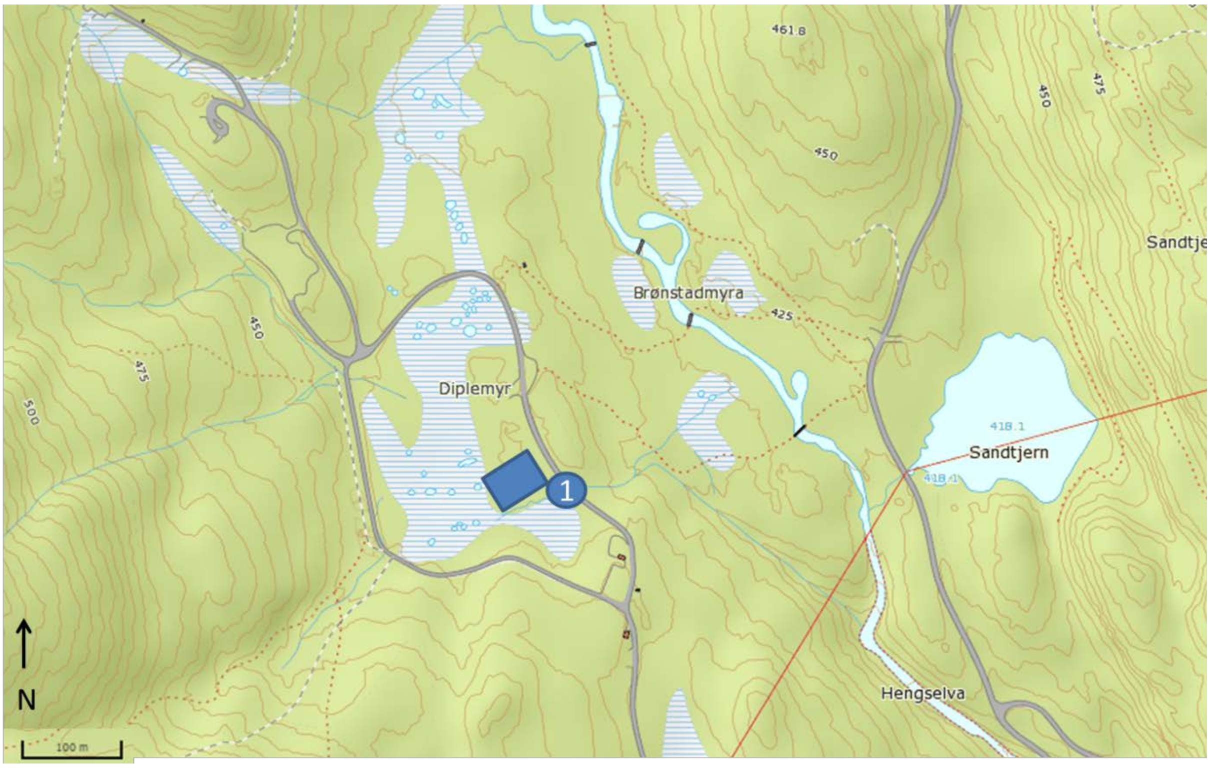

Fig 6S Map of Hengsvann (**He**) shooting range for small arms (59.64462313°N, 9.44673313°E). A network of sampling points was distributed approximately 5 meters apart within the rectangular blue box. Water samples were taken at a small creek at the site marked with a blue circle (Map: The Norwegian Mapping Authority).

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Supplementary materials,

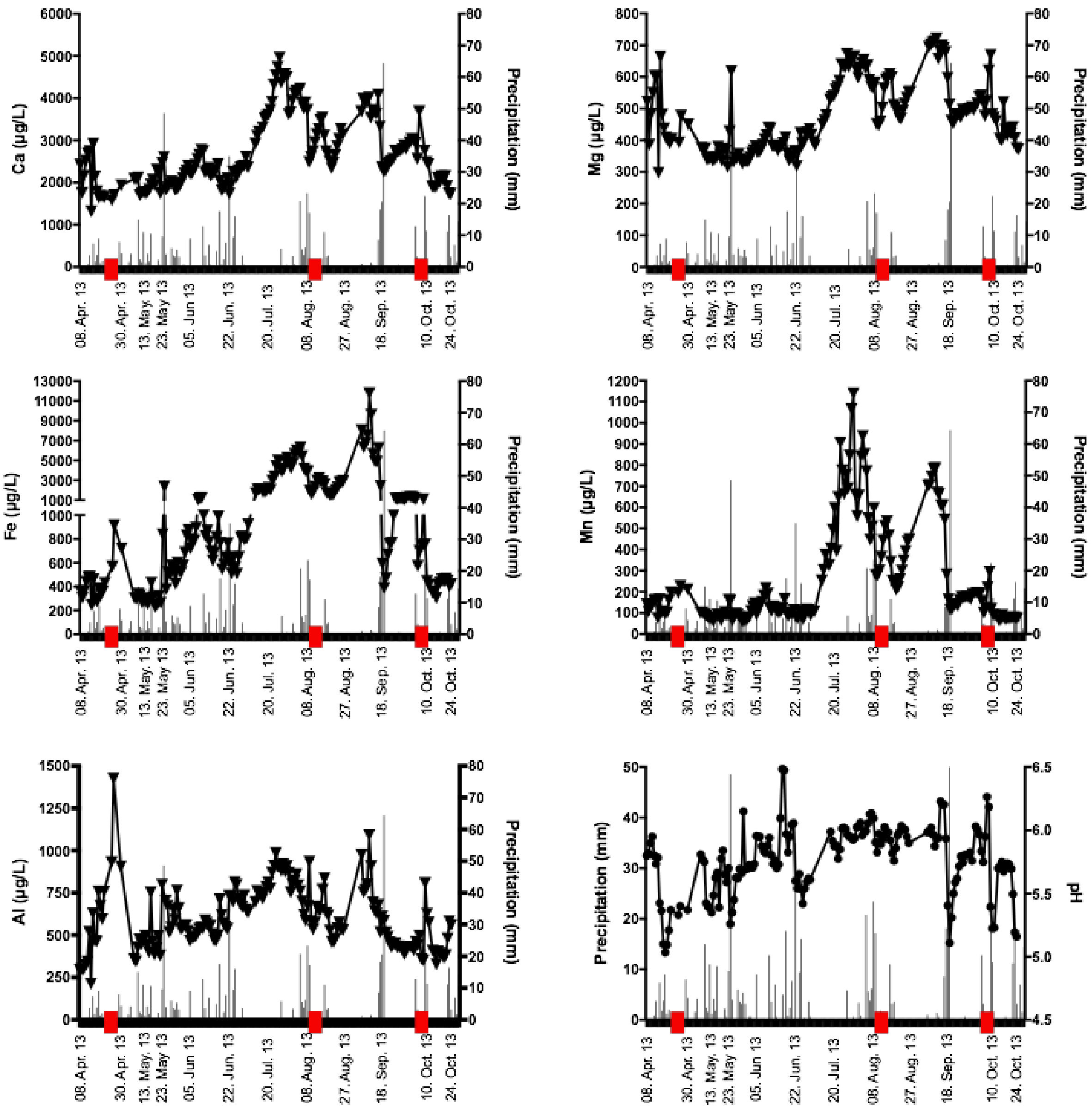

Fig 7S Daily concentrations of Ca, Mg, Fe, Mn and Al, and the pH fluctuation in the runoff water at Avgrunnsdalen shooting range for small arms as a function of precipitation. pH was measured in the creek weekly. The boxes in red indicate periods where some remediation measures were performed at the shooting range area. ─▼─ -data for total concentration (µg/L) of the elements. ——— Precipitation (mm) data from Sande in the Vestfold County near Avgrunnsdalen shooting range (Norwegian Meteorological Institute).

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Supplementary materials,

Table 1S Summary of positions, date of the surveys, the number of sampling points at each site, and the approximate area of which the survey covered.

Site	Position	Area (m^2)	Sampling points	Date
Avgrunnsdalen (Av)	59.58042°N, 10.51218°E	5000	52	3-4th of May 2011
Steinsjøen (St)	60.54248°N, 11.09554°E	1000	56	30th of May 2011
Mauken (Ma)	69.04592°N, 19.24131°E	600	35	3rd of July 2012
Kjoselvdalen (Kj)	69.73210°N, 19.14590°E	300	20	15th of October 2012
Tittelsnes (Ti)	59.72306°N, 5.51481°E	400	24	25th of June 2013
Hengsvann (He)	59.64462313°N, 9.44673313°E	400	25	3rd of July 2012

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Table 2S Procedure for the sequential extraction of soil samples.

Reaction step	Fraction	Extraction reagent (LS 15/20)[a]	pH	Contact time (h)	Temp. (°C)
F1	Water soluble	H_2O		1	20
F2	Reversibly bound	1 M NH_4Ac	7.0	2	20
F3	Reversibly bound, associated with carbonates	1 M NH_4Ac (pH adjusted with HAc)	5.0	2	20
F4	Easily reduced compounds (Fe/Mn hydroxides)	0.04 M NH_2OH•HCl in 25% HAc		6	80
F5	Oxidized compounds, soil organic material	a) 3 parts of total volume 30% H_2O_2 b) 1 part of total volume 3.2 M NH_4Ac in 20% HNO_3		a) 5.5 b) 0.5	a) 80 b) 20
F6[b]	Acid dissolvable	Aqua Regia (1 part conc. HNO_3, 3 parts conc. HCl)		6	200

[a] Peat from **Av** and **Ti** were extracted with LS15 and LS20 respectively

[b]Fraction 6 was extracted by Aqua Regia in an Ultra Wave micro wave oven as described in the method section.

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Supplementary materials,

Table 3S The mean (± SD) concentration of Fe, Mn, Al, Ca and Mg in peat soil collected from three vertical layers from six shooting ranges for small arms, and the median soil-water pH measured as described in materials and methods. The median soil concentrations are shown in brackets.

Soil origin	**Fe (g/kg)**	**Mn (mg/kg)**	**Al (g/kg)**	**Ca (g/kg)**	**Mg (g/kg)**	**Ignition loss (%)**	**pH**	***N***[a]
Av 0-15 cm	12 ± 9.3 (8.4)	190 ± 110 (160)	10 ± 7.0 (6.6)	5.2 ± 2.8 (5.0)	1.4 ± 1.6 (0.87)	76 ± 17 (77)	4.34	13/13
Av 15-30 cm	2.7 ± 1.8 (2.4)	130 ± 130 (110)	7.9 ± 8.8 (3.7)	4.8 ± 2.8 (4.8)	0.89 ± 0.45 (0.94)	87 ± 15 (95)	4.24	12/13
Av 30-45 cm	1.9 ± 1.7 (1.0)	70 ± 80 (40)	8.1 ± 10 (3.4)	4.1 ± 2.3 (4.7)	0.89 ± 0.54 (0.82)	90 ± 16 (97)	4.12	11/12
St 0-15 cm	7.4 ± 4.4 (6.5)	200 ± 130 (170)	6.4 ± 3.3 (5.6)	3.3 ± 1.7 (3.1)	0.79 ± 0.81 (0.60)	84 ± 17 (87)	4.10	30/20
St 15-30 cm	3.0 ± 2.3 (2.8)	120 ± 80 (90)	8.1 ± 5.8 (6.0)	2.5 ± 1.2 (2.3)	0.55 ± 0.44 (0.40)	93 ± 7.6 (96)	3.80	23/19
St 30-45 cm	3.3 ± 2.9 (2.9)	90 ± 120 (70)	11 ± 7.6 (8.5)	1.5 ± 0.54 (1.6)	0.73 ± 0.90 (0.29)	71 ± 40 (94)	3.80	19/18
Ti 0-15 cm	16 ± 6.9 (17)	340 ± 250 (200)	15 ± 6.9 (13)	9.4 ± 4.0 (8.3)	2.5 ± 1.4 (2.1)	32 ± 1.1 (33)	4.86	12/5[b]
Ti 15-30 cm	13 ± 6.7 (12)	270 ± 180 (240)	20 ± 11 (19)	9.8 ± 3.4 (9.3)	3.1 ± 2.6 (1.8)	39 ± 31 (38)	4.88	12/13
Ti 30-45 cm	15 ± 6.3 (16)	390 ± 190 (370)	28 ± 12 (24)	13 ± 3.4 (13)	3.8 ± 2.8 (2.9)	19 ± 24 (10)	4.84	12/10
Ma 0-15 cm	7.5 ± 3.1 (7.1)	50 ± 60 (40)	2.4 ± 0.95 (2.2)	14 ± 4.4 (14)	1.0 ± 0.49 (0.84)	89 ± 7.5 (93)	4.69	11/10
Ma 15-30 cm	4.3 ± 1.4 (4.2)	20 ± 9 (20)	2.1 ± 0.82 (1.9)	14 ± 5.1 (14)	0.80 ± 0.19 (0.76)	94 ± 0.9 (94)	4.63	12/11
Ma 30-45 cm	3.5 ± 1.2 (3.2)	20 ± 5 (20)	1.9 ± 0.37 (1.8)	13 ± 5.1 (15)	0.80 ± 0.13 (0.78)	94 ± 1.1 (94)	4.70	10/9
Kj 0-15	27 ± 16 (27)	370 ± 360 (310)	15 ± 6.1 (15)	12 ± 2.9 (13)	4.6 ± 2.5 (4.7)	62 ± 21 (61)	4.94	12/13
Kj 15-30	13 ± 15 (5.7)	370 ± 600 (130)	15 ± 11 (11)	9.9 ± 4.5 (8.1)	4.0 ± 4.6 (1.7)	69 ± 32 (90)	4.72	12/13
Kj 30-45	14 ± 11 (12)	470 ± 380 (420)	20 ± 10 (18)	14 ± 6.5 (13)	6.3 ± 5.1 (5.1)	64 ± 37 (83)	4.78	12/13
He 0-15	12 ± 8.3 (9.0)	50 ± 20 (50)	10 ± 2.3 (9.8)	1.3 ± 0.59 (1.3)	0.41 ± 0.16 (0.46)	70 ± 15 (69)	4.14	13/13
He 15-30	5.0 ± 2.1 (4.2)	70 ± 40 (60)	12 ± 4.7 (13)	2.0 ± 0.99 (1.7)	0.65 ± 0.39 (0.61)	55 ± 30 (65)	4.08	13/13
He 30-45	5.1 ± 2.1 (5.0)	70 ± 50 (70)	13 ± 8.4 (11)	2.2 ± 1.2 (2.0)	0.65 ± 0.43 (0.63)	53 ± 37 (59)	4.20	13/13

[a] The number of samples analyzed for elements and pH and ignition loss respectively

[b] Ignition loss were analyzed on five subsamples of a pooled sample **Ti**

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Table 4S The mean (± SD) concentration of Fe, Mn, Al, Ca, Mg, Na and K in soil water collected from three vertical layers from six shooting ranges for small arms. The median soil concentrations are shown in brackets.

Water	**Fe (mg/L)**	**Mn (µg/L)**	**Al (mg/L)**	**Ca (mg/L)**	**Mg (mg/L)**	**Na (mg/L)**	**K (mg/L)**	***N***
Av 0-15 cm[a]	0.11 ± 1.2 (0.52)	100 ± 180 (63)	0.34 ± 0.17 (0.33)	2.2 ± 0.91 (1.6)	0.57 ± 0.50 (0.44)	2.5 ± 0.55 (2.4)	0.48 ± 0.24 (0.44)	55
Av 0-15 cm[b]	0.80 ± 1.5 (0.35)	94 ± 200 (47)	0.70 ± 1.2 (0.41)	3.5 ± 3.7 (2.5)	0.50 ± 0.47 (0.34)	14 ± 6.6 (14)	1.2 ± 2.6 (0.59)	20
Av 15-30 cm[b]	0.19 ± 0.14 (0.14)	43 ± 39 (30)	0.47 ± 0.34 (0.37)	3.4 ± 1.8 (3.5)	0.47 ± 0.28 (0.40)	15 ± 4.1 (14)	0.78 ± 1.1 (0.38)	19
Av 30-45 cm[b]	0.14 ± 0.12 (0.09)	39 ± 45 (26)	0.47 ± 0.36 (0.31)	4.6 ± 3.3 (4.1)	0.61 ± 0.42 (0.57)	17 ± 7.3 (15)	2.2 ± 3.9 (1.1)	20
St 0-15 cm	0.70 ± 2.4 (0.12)	240 ± 140 (230)	0.52 ± 0.33 (0.46)	4.0 ± 1.9 (3.4)	1.0 ± 0.51 (0.93)	1.1 ± 0.32 (1.1)	1.1 ± 0.82 (1.0)	54
St 15-30 cm	1.4 ± 2.2 (0.69)	115 ± 117 (92	0.57 ± 0.27 (0.55)	2.0 ± 1.2 (1.6)	0.58 ± 0.35 (0.52)	1.2 ± 0.33 (1.1)	0.56 ± 0.22 (0.55)	40
St 30-45 cm	1.9 ± 2.2 (1.0)	87 ± 136 (44)	0.67 ± 0.48 (0.59)	1.5 ± 0.71 (1.6)	0.41 ± 0.31 (0.34)	1.2 ± 0.33 (1.2)	0.47 ± 0.18 (0.47)	38
Ti 0-15 cm	0.69 ± 0.89 (0.43)	22 ± 12 (21)	0.63 ± 1.0 (0.39)	1.8 ± 0.86 (1.5)	0.93 ± 0.36 (0.80)	6.8 ± 1.9 (6.7)	1.7 ± 1.5 (1.2)	24
Ti 15-30 cm	1.6 ± 1.8 (0.76)	28 ± 19 (27)	0.52 ± 0.64 (0.31)	2.4 ± 1.4 (2.1)	1.1 ± 0.48 (1.2)	8.8 ± 2.6 (9.6)	0.79 ± 0.42 (0.73)	24
Ti 30-45 cm	2.5 ± 2.5 (1.2)	36 ± 22 (34)	0.31 ± 0.25 (0.18)	3.2 ± 2.3 (2.2)	1.4 ± 0.38 (1.3)	11 ±2.1 (11)	0.52 ± 0.34 (0.46)	19
Ma 0-15 cm	0.38 ± 0.49 (0.21)	16 ± 16 (11)	0.09 ± 0.07 (0.07)	6.3 ± 2.8 (6.8)	0.87 ± 0.41 (0.83).	1.8 ± 0.29 (1.8)	1.8 ± 1.3 (1.5)	35
Ma 15-30 cm	1.1 ± 0.75 (0.93)	10 ± 7.7 (8.2)	0.08 ±0.05 (0.08)	7.3 ± 4.0 (6.3)	0.78 ± 0.29 (0.70)	1.9 ± 0.22 (1.9)	1.0 ± 0.61 (1.0)	30
Ma 30-45 cm	1.2 ± 0.81 (1.2)	11 ± 5.3 (10)	0.06 ± 0.03 (0.04)	8.1 ± 3.9 (7.7)	0.96 ± 0.28 (0.92)	2.0 ± 0.21 (2.0)	0.73 ± 0.51 (0.63)	30
Kj 0-15 cm	0.53 ± 0.65 (0.27)	17 ± 15 (14)	0.09 ± 0.03 (0.09)	2.9 ± 1.2 (2.8)	0.52 ± 0.27 (0.80)	3.9 ± 0.78 (4.0)	0.11 ± 0.10 (0.09)	20
Kj 15-30 cm	1.5 ± 1.3 (0.95)	38 ± 28 (32)	0.10 ± 0.03 (0.10)	4.9 ± 1.9 (4.6)	1.3 ± 0.33 (1.3)	5.0 ± 0.51 (4.9)	0.14 ± 0.14 (0.10)	20
Kj 30-45 cm	2.4 ± 1.7 (2.1)	62 ± 47 (48)	0.12 ± 0.06 (0.11)	6.6 ± 2.7 (6.5)	1.6 ± 0.48 (1.5)	5.7 ± 0.59 (5.7)	0.18 ± 0.24 (0.11)	20
He 0-15	0.41 ± 0.51 (0.19)	4.8 ± 2.1 (4.5)	0.46 ± 0.22 (0.45)	1.5 ± 0.89 (1.2)	0.18 ± 0.06 (0.18)	0.61 ± 0.23 (0.66)	1.4 ± 0.64 (1.5)	24
He 15-30	0.35 ± 0.52 (0.14)	4.6 ± 2.2 (4.1)	0.63 ± 0.29 (0.88)	1.5 ± 0.71 (1.6)	0.16 ± 0.06 (0.15)	0.80 ± 0.44 (0.79)	0.86 ±0.64 (0.59)	24
He 30-45	0.25 ± 0.41 (0.10)	5.4 ± 2.7 (4.8)	0.58 ± 0.22 (0.56)	1.7 ± 0.86 (1.5)	0.15 ± 0.06 (0.14)	0.83 ± 0.46 (0.69)	0.58 ± 0.62 (0.34)	25

[a] The soil water was sampled with Rhizon soil water sampler.

[b] The soil water was sampled by squeezing manually the soil followed by filtration of the water sample through a 0.45 µm filter.

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Supplementary materials,

Table 5S Sequential extraction of peat from **Av** and **Ti** small arms shooting ranges. The results are shown as mean distribution (percent distribution ± SD) between the 6 fractions of different elements. The median values are shown in brackets.

	K (%) Av (n = 9)	**K (%) Ti (n = 4)**	**Mg (%) Av**	**Mg (%) Ti**	**Ca (%) Av**	**Ca (%) Ti**	**Al (%) Av**	**Al (%) Ti**	**Mn (%) Av**	**Mn (%) Ti**	**Fe (%) Av**	**Fe (%) Ti**
Fraction 1	0.20 ± 0.09 (0.17)	7.9 ± 0.85 (7.9)	1.7 ± 0.92 (1.9)	5.8 ± 0.98 (6.0)	2.5 ± 0.52 (2.2)	1.6 ± 0.17 (1.6)	0.08 ± 0.03 (0.08)	0.05 ± 0.004 (0.05)	2.0 ± 0.86 (1.8)	4.4 ± 0.85 (4.3)	0.04 ± 0.03 (0.04)	0.06 ± 0.003 (0.06)
Fraction 2	0.60 ± 0.34 (0.50)	2.8 ± 0.44 (2.6)	20 ± 12 (18)	8.2 ± 1.3 (7.8)	52 ± 9.45 (56)	4.6 ± 0.47 (4.6)	0.27 ± 0.13 (0.27)	0.05 ± 0.006 (0.05)	35 ± 13 (35)	9.5 ± 1.1 (9.4)	0.11 ± 0.06 (0.09)	0.04 ± 0.003 (0.04)
Fraction 3	1.7 ± 0.59 (1.5)	1.2 ± 0.33 (1.3)	4.5 ± 3.8 (3.1)	1.7 ± 0.35 (1.8)	18 ± 5.6 (16)	1.8 ± 0.30 (1.9)	16 ± 6.9 (19)	2.9 ± 0.24 (2.9)	13 ± 7.1 (11)	5.7 ± 1.2 (5.7)	10 ± 8.4 (7.7)	0.96 ± 0.09 (0.96)
Fraction 4	1.4 ± 0.31 (1.4)	1.0 ± 0.26 (1.2)	2.4 ± 1.0 (2.1)	1.2 ± 0.20 (1.2)	6.4 ± 3.6 (5.1)	0.92 ± 0.17 (0.86)	15 ± 5.7 (17)	3.9 ± 0.59 (3.7)	9.7 ± 6.7 (6.3)	6.3 ± 2.1 (5.5)	32 ± 15 (32)	16 ± 1.1 (16)
Fraction 5	0.43 ± 0.13 (0.37)	0.32 ± 0.04 (0.31)	4.4 ± 4.5 (3.0)	2.0 ± 0.44 (1.9)	5.9 ± 2.7 (4.8)	0.55 ± 0.12 (0.55)	25 ± 12 (24)	13 ± 2.5 (13)	3.8 ± 2.3 (2.5)	1.9 ± 0.43 (2.0)	22 ± 13 (21)	7.8 ± 1.9 (7.6)
Fraction 6	96 ± 0.92 (96)	87 ± 1.8 (87)	68 ± 20 (70)	81 ± 2.4 (82)	16 ± 12 (13)	91 ± 0.87 (91)	46 ± 19 (43)	80 ± 3.0 (80)	38 ± 22 (32)	72 ± 4.7 (73)	37 ± 23 (32)	75 ± 2.6 (76)

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Supplementary materials,

Table 6S Summary of the correlation analysis (Pearson correlation analysis) of the element concentrations in the soil from the six shooting ranges sampled at three vertical layers in the soil profile, a total of 18 groups. Significant correlation ($p<0.05$) was given a score of one and eachs score was then added together (e.g. the soil concentration of Pb and Cu correlated in 15 of 18 groups). Elements that correlated in more than half of the groups are highlighted in the table. The results from each analysis with Pearson's correlation coefficient and degree of significance are shown in the Appendix, Tables 1A-6A.

Elements	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K
Pb										
Cu	15/18									
Sb	16/18	10/18								
Ca	5/18	3/18	3/18							
Zn	4/18	8/18	5/18	3/18						
Fe	2/18	1/18	2/18	4/18	3/18					
Mn	3/18	1/18	0/18	8/18	3/18	11/18				
Mg	4/18	5/18	5/18	9/18	6/18	9/18	11/18			
Al	3/18	5/18	2/18	5/18	3/18	11/18	6/18	12/18		
K	1/18	2/18	1/18	6/18	3/18	6/18	5/6	7/18	9/18	
pH	2/18	3/18	2/18	2/18	1/18	6/18	6/18	7/18	4/18	6/18

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Supplementary materials,

Table 7S Summary of the correlation analysis (Pearson correlation analysis) of the element concentrations in the soil water from the six shooting ranges sampled at three vertical layers in the soil profile, a total of 18 groups. Significant correlation ($p<0.05$) was given a score of one and each score was then added together (e.g. the soil concentration of Pb and Cu correlated in 15 of 18 groups). Elements that correlated in more than half of the groups are highlighted in the table. The results from each analysis with Pearson's correlation coefficient and degree of significance are shown in the Appendix, Tables 7A-12A.

Elements	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na
Pb										
Cu	17/18									
Sb	9/18	10/18								
Ca	6/18	6/18	1/18							
Zn	7/18	10/18	5/18	9/18						
Fe	6/18	6/18	5/18	8/18	8/18					
Mn	11/18	10/18	2/18	11/18	6/18	14/18				
Mg	5/18	5/18	4/18	18/18	7/18	8/18	12/18			
Al	9/18	9/18	10/18	6/18	5/18	9/18	4/18	5/18		
Na	4/18	4/18	6/18	9/18	5/18	5/18	6/18	8/18	4/18	
K	1/18	4/18	1/18	5/18	4/18	3/18	1/18	5/18	5/18'	5/18

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Table 8S Sequential extraction of soil from **Av** and **Ti** small arms shooting ranges. The results are shown as mean distribution (percent distribution ± SD) between the 6 fractions of Pb, Cu. Sb and Zn. The median values are shown in brackets.

	Pb (%) Av (n = 9)	**Pb(%) Ti (n = 4)**	**Cu (%) Av (n = 9)**	**Cu (%) Ti**	**Sb (%) Av**	**Sb (%) Ti**	**Zn (%) Av**	**Zn (%) Ti**
Fraction 1	0.06 ± 0.02 (0.07)	0.19 ± 0.03 (0.20)	0.23 ± 0.06 (0.24)	0.46 ± 0.04 (0.46)	1.1 ± 0.62 (1.2)	0.96 ± 0.33 (1.1)	1.6 ± 0.77 (1.8)	10 ± 1.8 (10)
Fraction 2	36 ± 9.9 (39)	23 ± 2.4 (24)	9.2 ± 4.2 (10)	2.0 ± 0.21 (2.1)	0.93 ± 0.55 (0.99)	1.5 ± 0.20 (1.5)	38 ± 15 (42)	21 ± 1.4 (21)
Fraction 3	34 ± 26 (37)	40 ± 1.7 (41)	34 ± 27 (34)	16 ± 3.1 (16)	3.3 ± 27 (2.8)	3.3 ± 3.0 (1.8)	29 ± 26 (30)	40 ± 3.1 (38)
Fraction 4	10 ± 1.6 (10)	16 ± 0.77 (15)	6.8 ± 5.3 (5.9)	15 ± 1.6 (16)	8.2 ± 7.1 (6.9)	6.9 ± 1.8 (6.9)	7.6 ± 2.4 (8.0)	12 ± 1.3 (12)
Fraction 5	9.2 ± 1.7 (9.4)	5.5 ± 0.77 (5.3)	51 ± 16 (46)	62 ± 5.0 (61)	56 ± 27 (66)	58 ± 7.8 (61)	3.8 ± 1.5 (3.5)	5.4 ± 0.90 (5.5)
Fraction 6	0.35 ± 0.43 (0.13)	15 ± 2.1 (15)	0.96 ± 0.82 (0.51)	4.1 ± 0.42 (4.2)	25 ± 23 (18)	29 ± 6.2 (28)	9.6 ± 8.2 (8.4)	11 ± 0.22 (11)

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Supplementary materials,

Appendix

Table 1A Correlation analysis between the element concentrations and pH in the soil from **Kj** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations (p<0.05) are marked in yellow.

Kj, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.685762	0.689245	0.277208	0.298788	0.413299	0.462774	0.358402	0.294078	0.125533	-0.06811
Cu	0.685762		0.611342	0.08101	0.53312	-0.06924	-0.00365	0.024362	-0.11166	-0.14824	-0.17886
Sb	0.689245	0.611342		-0.2004	0.341385	0.081495	-0.02253	-0.16801	-0.16332	-0.18961	-0.37407
Ca	0.277208	0.08101	-0.2004		-0.09716	0.682787	0.808186	0.936748	0.867103	0.179087	0.447562
Zn	0.298788	0.53312	0.341385	-0.09716		0.025983	-0.09984	-0.14075	-0.1867	0.121709	-0.05847
Fe	0.413299	-0.06924	0.081495	0.682787	0.025983		0.896175	0.795537	0.923084	0.528209	0.229518
Mn	0.462774	-0.00365	-0.02253	0.808186	-0.09984	0.896175		0.921737	0.934763	0.452133	0.255689
Mg	0.358402	0.024362	-0.16801	0.936748	-0.14075	0.795537	0.921737		0.930111	0.267706	0.314783
Al	0.294078	-0.11166	-0.16332	0.867103	-0.1867	0.923084	0.934763	0.930111		0.465189	0.393403
K	0.125533	-0.14824	-0.18961	0.179087	0.121709	0.528209	0.452133	0.267706	0.465189		0.584133
pH	-0.06811	-0.17886	-0.37407	0.447562	-0.05847	0.229518	0.255689	0.314783	0.393403	0.584133	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.009664	0.009158	0.359178	0.321371	0.160398	0.111292	0.229171	0.329425	0.682809	0.833423
Cu	0.009664		0.026418	0.792484	0.060648	0.822169	0.990565	0.937034	0.716482	0.628878	0.57807
Sb	0.009158	0.026418		0.511537	0.25363	0.791265	0.941752	0.583246	0.593955	0.534975	0.230962
Ca	0.359178	0.792484	0.511537		0.752191	0.010113	0.000828	2.40E-06	1.24E-04	0.558271	0.144577
Zn	0.321371	0.060648	0.25363	0.752191		0.932852	0.745535	0.64649	0.541366	0.692038	0.856771
Fe	0.160398	0.822169	0.791265	0.010113	0.932852		3.38E-05	0.001146	6.85E-06	0.063517	0.473008
Mn	0.111292	0.990565	0.941752	0.000828	0.745535	3.38E-05		7.51E-06	2.83E-06	0.120862	0.422487
Mg	0.229171	0.937034	0.583246	2.40E-06	0.64649	0.001146	7.51E-06		4.10E-06	0.37655	0.318981
Al	0.329425	0.716482	0.593955	1.24E-04	0.541366	6.85E-06	2.83E-06	4.10E-06		0.109193	0.205803
K	0.682809	0.628878	0.534975	0.558271	0.692038	0.063517	0.120862	0.37655	0.109193		0.046112
pH	0.833423	0.57807	0.230962	0.144577	0.856771	0.473008	0.422487	0.318981	0.205803	0.046112	

Kj, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.484043	0.562302	0.081527	0.501167	0.113447	-0.04348	0.154034	0.111053	-0.04045	0.337226
Cu	0.484043		0.448734	-0.31028	0.252115	-0.39075	-0.42062	-0.2903	-0.11525	-0.14528	0.301739
Sb	0.562302	0.448734		-0.409	0.23138	-0.414	-0.53688	-0.34192	-0.39984	-0.58732	0.12901
Ca	0.081527	-0.31028	-0.409		0.689827	0.972279	0.93432	0.98215	0.938662	0.519643	-0.18907
Zn	0.501167	0.252115	0.23138	0.689827		0.648134	0.543333	0.721001	0.682436	0.158706	0.123161
Fe	0.113447	-0.39075	-0.414	0.972279	0.648134		0.963068	0.97212	0.879713	0.542433	-0.08168
Mn	-0.04348	-0.42062	-0.53688	0.93432	0.543333	0.963068		0.929387	0.846719	0.624333	-0.03116
Mg	0.154034	-0.2903	-0.34192	0.98215	0.721001	0.97212	0.929387		0.948106	0.559496	-0.04807
Al	0.111053	-0.11525	-0.39984	0.938662	0.682436	0.879713	0.846719	0.948106		0.574179	-0.1193
K	-0.04045	-0.14528	-0.58732	0.519643	0.158706	0.542433	0.624333	0.559496	0.574179		0.367498
pH	0.337226	0.301739	0.12901	-0.18907	0.123161	-0.08168	-0.03116	-0.04807	-0.1193	0.367498	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.093724	0.045469	0.791187	0.08104	0.712115	0.887835	0.615363	0.717962	0.895604	0.283746
Cu	0.093724		0.124032	0.302179	0.405991	0.186782	0.152381	0.335968	0.707724	0.635819	0.34051
Sb	0.045469	0.124032		0.16523	0.446893	0.159614	0.058513	0.252838	0.175836	0.034813	0.689454
Ca	0.791187	0.302179	0.16523		0.009076	2.75E-08	2.94E-06	2.49E-09	2.03E-06	0.068746	0.556176
Zn	0.08104	0.405991	0.446893	0.009076		0.016585	0.05498	0.005419	0.010167	0.604551	0.702955
Fe	0.712115	0.186782	0.159614	2.75E-08	0.016585		1.31E-07	2.84E-08	7.35E-05	0.055464	0.800769
Mn	0.887835	0.152381	0.058513	2.94E-06	0.05498	1.31E-07		4.33E-06	2.61E-04	0.022555	0.923413
Mg	0.615363	0.335968	0.252838	2.49E-09	0.005419	2.84E-08	4.33E-06		8.26E-07	0.046795	0.882066
Al	0.717962	0.707724	0.175836	2.03E-06	0.010167	7.35E-05	2.61E-04	8.26E-07		0.040154	0.711907
K	0.895604	0.635819	0.034813	0.068746	0.604551	0.055464	0.022555	0.046795	0.040154		0.239911
pH	0.283746	0.34051	0.689454	0.556176	0.702955	0.800769	0.923413	0.882066	0.711907	0.239911	

Kj, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.277831	0.818725	0.261043	0.454978	0.260723	0.295033	0.323683	0.306743	0.028622	0.27439
Cu	0.277831		0.137183	-0.15234	0.005574	-0.42766	-0.26835	-0.0969	0.045042	-0.05351	-0.07927
Sb	0.818725	0.137183		0.116004	0.439606	0.182174	0.14863	0.237498	0.114771	-0.03155	0.068255
Ca	0.261043	-0.15234	0.116004		0.874938	0.830259	0.935194	0.969268	0.940222	0.640349	0.697674
Zn	0.454978	0.005574	0.439606	0.874938		0.756298	0.829734	0.928446	0.836	0.490532	0.635491
Fe	0.260723	-0.42766	0.182174	0.830259	0.756298		0.924914	0.854677	0.764158	0.54183	0.501582
Mn	0.295033	-0.26835	0.14863	0.935194	0.829734	0.924914		0.945073	0.895188	0.494801	0.648716
Mg	0.323683	-0.0969	0.237498	0.969268	0.928446	0.854677	0.945073		0.948557	0.61026	0.687725
Al	0.306743	0.045042	0.114771	0.940222	0.836	0.764158	0.895188	0.948557		0.610207	0.749006
K	0.028622	-0.05351	-0.03155	0.640349	0.490532	0.54183	0.494801	0.61026	0.610207		0.4825
pH	0.27439	-0.07927	0.068255	0.697674	0.635491	0.501582	0.648716	0.687725	0.749006	0.4825	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.358054	0.00062	0.38899	0.118251	0.389593	0.327784	0.280656	0.308018	0.926048	0.388099
Cu	0.358054		0.654943	0.6193	0.985582	0.144917	0.37535	0.752833	0.883835	0.862179	0.806535
Sb	0.00062	0.654943		0.705882	0.132816	0.551393	0.627959	0.434621	0.708885	0.918517	0.833069
Ca	0.38899	0.6193	0.705882		9.02E-05	4.42E-04	2.73E-06	4.82E-08	1.77E-06	0.018389	0.01165
Zn	0.118251	0.985582	0.132816	9.02E-05		0.002775	4.49E-04	4.65E-06	3.70E-04	0.088768	0.026367
Fe	0.389593	0.144917	0.551393	4.42E-04	0.002775		6.02E-06	1.98E-04	0.002355	0.055789	0.096628
Mn	0.327784	0.37535	0.627959	2.73E-06	4.49E-04	6.02E-06		1.12E-06	3.56E-05	0.085607	0.022483
Mg	0.280656	0.752833	0.434621	4.82E-08	4.65E-06	1.98E-04	1.12E-06		7.88E-07	0.02676	0.013443
Al	0.308018	0.883835	0.708885	1.77E-06	3.70E-04	0.002355	3.56E-05	7.88E-07		0.026777	0.005055
K	0.926048	0.862179	0.918517	0.018389	0.088768	0.055789	0.085607	0.02676	0.026777		0.112125
pH	0.388099	0.806535	0.833069	0.01165	0.026367	0.096628	0.022483	0.013443	0.005055	0.112125	

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Supplementary materials,

Table 2A Correlation analysis between the element concentrations and pH in the soil from **Ma** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations (p<0.05) are marked in yellow

Ma, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.923035	0.653206	-0.4789	0.745658	0.36883	0.708206	0.805735	0.59787	0.443775	0.596367
Cu	0.923035		0.720563	-0.31647	0.71489	0.324785	0.582063	0.7862	0.632107	0.288788	0.693316
Sb	0.653206	0.720563		0.163688	0.606535	-0.11771	0.307933	0.471201	0.169244	-0.03615	0.210669
Ca	-0.4789	-0.31647	0.163688		-0.13542	-0.57672	-0.47734	-0.55243	-0.58247	-0.71036	-0.22239
Zn	0.745658	0.71489	0.606535	-0.13542		0.173873	0.783998	0.784773	0.464492	0.323949	0.605313
Fe	0.36883	0.324785	-0.11771	-0.57672	0.173873		0.459065	0.35135	0.391331	0.844856	0.558556
Mn	0.708206	0.582063	0.307933	-0.47734	0.783998	0.459065		0.642077	0.488433	0.706671	0.39811
Mg	0.805735	0.7862	0.471201	-0.55243	0.784773	0.35135	0.642077		0.835665	0.459876	0.632235
Al	0.59787	0.632107	0.169244	-0.58247	0.464492	0.391331	0.488433	0.835665		0.38692	0.513678
K	0.443775	0.288788	-0.03615	-0.71036	0.323949	0.844856	0.706671	0.459876	0.38692		0.30115
pH	0.596367	0.693316	0.210669	-0.22239	0.605313	0.558556	0.39811	0.632235	0.513678	0.30115	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.00014	0.040562	0.161415	0.013291	0.294272	0.021905	0.004893	0.067917	0.198889	0.068793
Cu	0.00014		0.01873	0.372989	0.020145	0.359852	0.077499	0.006999	0.049901	0.418378	0.0262
Sb	0.040562	0.01873		0.651374	0.063009	0.746053	0.386717	0.16923	0.640202	0.921018	0.559076
Ca	0.161415	0.372989	0.651374		0.70914	0.080922	0.162977	0.097732	0.077241	0.021325	0.536885
Zn	0.013291	0.020145	0.063009	0.70914		0.630944	0.007271	0.007174	0.176224	0.361163	0.063687
Fe	0.294272	0.359852	0.746053	0.080922	0.630944		0.182006	0.31948	0.26345	0.002093	0.093299
Mn	0.021905	0.077499	0.386717	0.162977	0.007271	0.182006		0.045327	0.152036	0.022323	0.25453
Mg	0.004893	0.006999	0.16923	0.097732	0.007174	0.31948	0.045327		0.002604	0.181135	0.04984
Al	0.067917	0.049901	0.640202	0.077241	0.176224	0.26345	0.152036	0.002604		0.269347	0.128835
K	0.198889	0.418378	0.921018	0.021325	0.361163	0.002093	0.022323	0.181135	0.269347		0.397798
pH	0.068793	0.0262	0.559076	0.536885	0.063687	0.093299	0.25453	0.04984	0.128835	0.397798	

Ma, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.827179	0.809371	0.012181	0.875848	-0.18701	0.553603	0.527695	0.226356	0.376783	0.398174
Cu	0.827179		0.515501	-0.06007	0.867913	-0.20275	0.4418	0.479101	0.294679	0.354033	0.442049
Sb	0.809371	0.515501		0.361071	0.612256	-0.03797	0.487243	0.653866	0.488796	0.452632	0.443246
Ca	0.012181	-0.06007	0.361071		-0.08203	0.206314	0.246027	0.166685	0.477775	0.468378	-0.00648
Zn	0.875848	0.867913	0.612256	-0.08203		-0.47488	0.248636	0.639121	0.197838	0.30856	0.490007
Fe	-0.18701	-0.20275	-0.03797	0.206314	-0.47488		0.307066	-0.13676	0.18254	0.346418	-0.13201
Mn	0.553603	0.4418	0.487243	0.246027	0.248636	0.307066		0.121152	0.384447	0.543674	-0.07504
Mg	0.527695	0.479101	0.653866	0.166685	0.639121	-0.13676	0.121152		0.569433	0.384659	0.693677
Al	0.226356	0.294679	0.488796	0.477775	0.197838	0.18254	0.384447	0.569433		0.63349	0.374285
K	0.376783	0.354033	0.452632	0.468378	0.30856	0.346418	0.543674	0.384659	0.63349		0.418578
pH	0.398174	0.442049	0.443246	-0.00648	0.490007	-0.13201	-0.07504	0.693677	0.374285	0.418578	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.000901	0.001426	0.97003	0.000188	0.560566	0.061851	0.077852	0.479294	0.227324	0.225185
Cu	0.000901		0.086278	0.852876	0.000253	0.527393	0.150451	0.115047	0.352481	0.258882	0.173413
Sb	0.001426	0.086278		0.248861	0.034332	0.906732	0.108129	0.02109	0.106842	0.139532	0.172114
Ca	0.97003	0.852876	0.248861		0.799935	0.520008	0.44082	0.60462	0.1162	0.124595	0.984905
Zn	0.000188	0.000253	0.034332	0.799935		0.118745	0.435833	0.025256	0.537662	0.329157	0.126007
Fe	0.560566	0.527393	0.906732	0.520008	0.118745		0.331625	0.671686	0.570148	0.269985	0.698829
Mn	0.061851	0.150451	0.108129	0.44082	0.435833	0.331625		0.70761	0.217239	0.067685	0.826429
Mg	0.077852	0.115047	0.02109	0.60462	0.025256	0.671686	0.70761		0.053287	0.216964	0.017908
Al	0.479294	0.352481	0.106842	0.1162	0.537662	0.570148	0.217239	0.053287		0.026994	0.256782
K	0.227324	0.258882	0.139532	0.124595	0.329157	0.269985	0.067685	0.216964	0.026994		0.2001
pH	0.225185	0.173413	0.172114	0.984905	0.126007	0.698829	0.826429	0.017908	0.256782	0.2001	

Ma, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.808581	0.774762	-0.52638	0.624432	-0.39552	0.076673	0.078388	0.538727	0.346672	-0.46059
Cu	0.808581		0.790776	-0.24168	0.896919	-0.6988	-0.34909	0.060427	0.067853	-0.04591	-0.4473
Sb	0.774762	0.790776		-0.05667	0.685705	-0.68201	-0.37543	0.366593	0.190197	0.350816	-0.3757
Ca	-0.52638	-0.24168	-0.05667		-0.04139	-0.07157	-0.53736	0.167601	-0.48127	0.008093	0.43219
Zn	0.624432	0.896919	0.685705	-0.04139		-0.53744	-0.376	0.059262	-0.03229	0.024586	-0.36907
Fe	-0.39552	-0.6988	-0.68201	-0.07157	-0.53744		0.772578	-0.23151	0.264771	0.072255	-0.05806
Mn	0.076673	-0.34909	-0.37543	-0.53736	-0.376	0.772578		-0.1233	0.547347	0.16433	-0.23497
Mg	0.078388	0.060427	0.366593	0.167601	0.059262	-0.23151	-0.1233		-0.40898	0.169479	0.243666
Al	0.538727	0.067853	0.190197	-0.48127	-0.03229	0.264771	0.547347	-0.40898		0.637818	-0.22231
K	0.346672	-0.04591	0.350816	0.008093	0.024586	0.072255	0.16433	0.169479	0.637818		0.220483
pH	-0.46059	-0.4473	-0.3757	0.43219	-0.36907	-0.05806	-0.23497	0.243666	-0.22231	0.220483	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.004629	0.008493	0.118045	0.053624	0.257919	0.83326	0.829577	0.108114	0.326411	0.212143
Cu	0.004629		0.006457	0.501141	4.35E-04	0.024557	0.32282	0.868297	0.852252	0.899791	0.227357
Sb	0.008493	0.006457		0.876425	0.028603	0.029824	0.285044	0.297437	0.598672	0.320267	0.319032
Ca	0.118045	0.501141	0.876425		0.909621	0.844245	0.10919	0.6435	0.159046	0.982297	0.245339
Zn	0.053624	4.35E-04	0.028603	0.909621		0.109127	0.284252	0.870819	0.929432	0.946251	0.328336
Fe	0.257919	0.024557	0.029824	0.844245	0.109127		0.008802	0.519851	0.459738	0.842765	0.882044
Mn	0.83326	0.32282	0.285044	0.10919	0.284252	0.008802		0.734351	0.101504	0.650079	0.542806
Mg	0.829577	0.868297	0.297437	0.6435	0.870819	0.519851	0.734351		0.240583	0.639731	0.527516
Al	0.108114	0.852252	0.598672	0.159046	0.929432	0.459738	0.101504	0.240583		0.047244	0.565354
K	0.326411	0.899791	0.320267	0.982297	0.946251	0.842765	0.650079	0.639731	0.047244		0.568636
pH	0.212143	0.227357	0.319032	0.245339	0.328336	0.882044	0.542806	0.527516	0.565354	0.568636	

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Supplementary materials,

Table 3A Correlation analysis between the element concentrations and pH in the soil from **St** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

St, 0-15 cm.

Pears co	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**K**	**pH**
Pb		0.690234	0.791425	-0.456	-0.07514	0.309668	0.078068	-0.17647	-0.14197	-0.04938	-0.17955
Cu	0.690234		0.326844	-0.11606	0.491445	0.031603	-0.22521	-0.51975	-0.42593	-0.46278	-0.26382
Sb	0.791425	0.326844		-0.36791	-0.19142	0.250734	0.203373	0.09278	0.047737	0.215657	-0.0804
Ca	-0.456	-0.11606	-0.36791		0.331452	-0.35444	-0.00588	-0.01203	-0.02152	-0.40535	0.215301
Zn	-0.07514	0.491445	-0.19142	0.331452		-0.42084	-0.65631	-0.68226	-0.64443	-0.73381	-0.43369
Fe	0.309668	0.031603	0.250734	-0.35444	-0.42084		0.565356	0.432179	0.6036	0.444498	0.451014
Mn	0.078068	-0.22521	0.203373	-0.00588	-0.65631	0.565356		0.503032	0.564912	0.409116	0.51076
Mg	-0.17647	-0.51975	0.09278	-0.01203	-0.68226	0.432179	0.503032		0.758414	0.765806	0.624507
Al	-0.14197	-0.42593	0.047737	-0.02152	-0.64443	0.6036	0.564912	0.758414		0.651842	0.624131
K	-0.04938	-0.46278	0.215657	-0.40535	-0.73381	0.444498	0.409116	0.765806	0.651842		0.277371
pH	-0.17955	-0.26382	-0.0804	0.215301	-0.43369	0.451014	0.51076	0.624507	0.624131	0.277371	

P	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**K**	**pH**
Pb		2.43E-05	1.92E-07	0.011322	0.693126	0.095859	0.69294	0.350899	0.454248	0.822948	0.342415
Cu	2.43E-05		0.077916	0.541376	0.005816	0.868327	0.249219	0.003243	0.018933	0.026175	0.158917
Sb	1.92E-07	0.077916		0.045472	0.310903	0.181407	0.299277	0.625806	0.802203	0.323021	0.67276
Ca	0.011322	0.541376	0.045472		0.073573	0.054633	0.976305	0.94968	0.910145	0.054998	0.253203
Zn	0.693126	0.005816	0.310903	0.073573		0.020569	1.49E-04	3.29E-05	1.21E-04	6.75E-05	0.016652
Fe	0.095859	0.868327	0.181407	0.054633	0.020569		0.001718	0.017078	4.13E-04	0.033582	0.012369
Mn	0.69294	0.249219	0.299277	0.976305	1.49E-04	0.001718		0.006363	0.001736	0.058675	0.00548
Mg	0.350899	0.003243	0.625806	0.94968	3.29E-05	0.017078	0.006363		1.20E-06	2.05E-05	2.25E-04
Al	0.454248	0.018933	0.802203	0.910145	1.21E-04	4.13E-04	0.001736	1.20E-06		0.000752	2.28E-04
K	0.822948	0.026175	0.323021	0.054998	6.75E-05	0.033582	0.058675	2.05E-05	0.000752		0.200066
pH	0.342415	0.158917	0.67276	0.253203	0.016652	0.012369	0.00548	2.25E-04	2.28E-04	0.200066	

St, 15-30 cm.

Pears co	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**K**	**pH**
Pb		0.937084	0.695952	0.448751	0.561763	0.414261	0.193784	0.569927	0.219873	0.311165	0.355759
Cu	0.937084		0.816757	0.621727	0.666928	0.387222	0.189674	0.519817	0.182607	0.225842	0.387382
Sb	0.695952	0.816757		0.801988	0.788521	0.434687	0.383466	0.561062	0.189326	0.237646	0.474426
Ca	0.448751	0.621727	0.801988		0.591592	0.383179	0.316065	0.489526	0.20708	0.194722	0.594835
Zn	0.561763	0.666928	0.788521	0.591592		0.001828	0.101926	0.245814	-0.10773	-0.04592	0.261823
Fe	0.414261	0.387222	0.434687	0.383179	0.001828		0.754145	0.664915	0.643646	0.648945	0.48734
Mn	0.193784	0.189674	0.383466	0.316065	0.101926	0.754145		0.353714	0.378373	0.327973	0.256693
Mg	0.569927	0.519817	0.561062	0.489526	0.245814	0.664915	0.353714		0.749277	0.801601	0.798917
Al	0.219873	0.182607	0.189326	0.20708	-0.10773	0.643646	0.378373	0.749277		0.886858	0.77879
K	0.311165	0.225842	0.237646	0.194722	-0.04592	0.648945	0.327973	0.801601	0.886858		0.622437
pH	0.355759	0.387382	0.474426	0.594835	0.261823	0.48734	0.256693	0.798917	0.77879	0.622437	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		4.61E-11	0.000226	0.031726	0.005278	0.049384	0.387531	0.004524	0.313418	0.181743	0.095707
Cu	4.61E-11		1.99E-06	0.00154	0.00051	0.067932	0.397868	0.011011	0.404302	0.338359	0.067809
Sb	0.000226	1.99E-06		4.19E-06	7.84E-06	0.038193	0.078122	0.005348	0.386926	0.313022	0.022179
Ca	0.031726	0.00154	4.19E-06		0.002946	0.071108	0.151863	0.017747	0.343094	0.410695	0.002755
Zn	0.005278	0.00051	7.84E-06	0.002946		0.993397	0.651739	0.258225	0.624655	0.84756	0.2275
Fe	0.049384	0.067932	0.038193	0.071108	0.993397		5.04E-05	0.000537	0.000921	0.001964	0.01834
Mn	0.387531	0.397868	0.078122	0.151863	0.651739	5.04E-05		0.106327	0.0825	0.170431	0.248841
Mg	0.004524	0.011011	0.005348	0.017747	0.258225	0.000537	0.106327		3.88E-05	2.15E-05	4.85E-06
Al	0.313418	0.404302	0.386926	0.343094	0.624655	0.000921	0.0825	3.88E-05		1.90E-07	1.20E-05
K	0.181743	0.338359	0.313022	0.410695	0.84756	0.001964	0.170431	2.15E-05	1.90E-07		0.00338
pH	0.095707	0.067809	0.022179	0.002755	0.2275	0.01834	0.248841	4.85E-06	1.20E-05	0.00338	

St, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.750123	0.840738	0.700345	0.503357	0.181137	0.091917	0.127088	0.058589	0.150142	0.261291
Cu	0.750123		0.662189	0.745497	0.520439	-0.22259	-0.27319	-0.23378	-0.24426	-0.21223	0.089432
Sb	0.840738	0.662189		0.732512	0.721838	0.12135	0.067853	0.091242	-0.07058	0.091048	0.220216
Ca	0.700345	0.745497	0.732512		0.383988	-0.07292	-0.06302	-0.36329	-0.36111	-0.37116	-0.07262
Zn	0.503357	0.520439	0.721838	0.383988		0.25973	0.272939	0.312624	0.206034	0.272072	0.39751
Fe	0.181137	-0.22259	0.12135	-0.07292	0.25973		0.943958	0.753122	0.857925	0.766462	0.510164
Mn	0.091917	-0.27319	0.067853	-0.06302	0.272939	0.943958		0.568028	0.737957	0.600431	0.326935
Mg	0.127088	-0.23378	0.091242	-0.36329	0.312624	0.753122	0.568028		0.894023	0.97647	0.771328
Al	0.058589	-0.24426	-0.07058	-0.36111	0.206034	0.857925	0.737957	0.894023		0.947983	0.737312
K	0.150142	-0.21223	0.091048	-0.37116	0.272072	0.766462	0.600431	0.97647	0.947983		0.855095
pH	0.261291	0.089432	0.220216	-0.07262	0.39751	0.510164	0.326935	0.771328	0.737312	0.855095	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.000217	6.56E-06	0.00084	0.028021	0.458009	0.716797	0.604132	0.811688	0.552074	0.279906
Cu	0.000217		0.002009	0.000249	0.022346	0.359698	0.272689	0.335413	0.313546	0.397851	0.71579
Sb	6.56E-06	0.002009		0.000362	4.84E-04	0.620687	0.789072	0.710272	0.774032	0.719371	0.364976
Ca	0.00084	0.000249	0.000362		0.104576	0.766713	0.8038	0.126309	0.128768	0.129409	0.767668
Zn	0.028021	0.022346	0.000484	0.104576		0.282894	0.273156	0.192527	0.397416	0.274744	0.091921
Fe	0.458009	0.359698	0.620687	0.766713	0.282894		4.10E-09	1.98E-04	2.64E-06	2.07E-04	0.025638
Mn	0.716797	0.272689	0.789072	0.8038	0.273156	4.10E-09		0.013923	4.72E-04	0.010818	0.185427
Mg	0.604132	0.335413	0.710272	0.126309	0.192527	1.98E-04	0.013923		2.49E-07	4.39E-12	1.10E-04
Al	0.811688	0.313546	0.774032	0.128768	0.397416	2.64E-06	4.72E-04	2.49E-07		2.29E-09	3.16E-04
K	0.552074	0.397851	0.719371	0.129409	0.274744	2.07E-04	0.010818	4.39E-12	2.29E-09		6.14E-06
pH	0.279906	0.71579	0.364976	0.767668	0.091921	0.025638	0.185427	1.10E-04	3.16E-04	6.14E-06	

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Supplementary materials,

Table 4A Correlation analysis between the element concentrations and pH in the soil from **Av** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations (p<0.05) are marked in yellow.

Av, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.401276	0.617189	-0.04719	0.25674	-0.16728	-0.35074	-0.30365	-0.4007	-0.38447	-0.3788
Cu	0.401276		0.013554	-0.65962	0.188343	0.255784	-0.30016	-0.38742	0.253398	-0.20793	-0.30102
Sb	0.617189	0.013554		0.1561	0.36019	-0.46747	-0.51546	-0.29362	-0.62493	-0.47444	-0.37773
Ca	-0.04719	-0.65962	0.1561		0.427146	-0.5441	0.118846	0.467234	-0.70176	-0.08533	0.27831
Zn	0.25674	0.188343	0.36019	0.427146		-0.49937	-0.41957	-0.13417	-0.34162	-0.6479	-0.02664
Fe	-0.16728	0.255784	-0.46747	-0.5441	-0.49937		0.365329	0.155042	0.696427	0.287736	0.196554
Mn	-0.35074	-0.30016	-0.51546	0.118846	-0.41957	0.365329		0.792822	0.235011	0.770866	8.62E-01
Mg	-0.30365	-0.38742	-0.29362	0.467234	-0.13417	0.155042	0.792822		-0.03413	0.715589	0.824803
Al	-0.4007	0.253398	-0.62493	-0.70176	-0.34162	0.696427	0.235011	-0.03413		0.21096	0.207255
K	-0.38447	-0.20793	-0.47444	-0.08533	-0.6479	0.287736	0.770866	0.715589	0.21096		0.592842
pH	-0.3788	-0.30102	-0.37773	0.27831	-0.02664	0.196554	8.62E-01	0.824803	0.207255	0.592842	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.174149	0.024623	0.884209	0.397137	0.584913	0.240006	0.313178	0.17483	0.1946	0.201823
Cu	0.174149		0.964948	0.019608	0.537755	0.398959	0.319052	0.190899	0.403525	0.495444	0.317587
Sb	0.024623	0.964948		0.628059	0.226688	0.107236	0.071409	0.330211	0.022389	0.101405	0.203203
Ca	0.884209	0.019608	0.628059		0.16607	0.067427	0.712966	0.125644	0.010967	0.79204	0.381078
Zn	0.397137	0.537755	0.226688	0.16607		0.082315	0.153515	0.662123	0.253276	0.016638	0.931158
Fe	0.584913	0.398959	0.107236	0.067427	0.082315		0.219642	0.613025	0.008179	0.340446	0.519832
Mn	0.240006	0.319052	0.071409	0.712966	0.153515	0.219642		0.001226	0.43959	0.002038	1.51E-04
Mg	0.313178	0.190899	0.330211	0.125644	0.662123	0.613025	0.001226		0.911862	0.005954	0.000521
Al	0.17483	0.403525	0.022389	0.010967	0.253276	0.008179	0.43959	0.911862		0.489041	0.496878
K	0.1946	0.495444	0.101405	0.79204	0.016638	0.340446	0.002038	0.005954	0.489041		0.032732
pH	0.201823	0.317587	0.203203	0.381078	0.931158	0.519832	1.51E-04	0.000521	0.496878	0.032732	

Av, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.87619	0.801961	0.327409	0.557379	0.056932	0.069022	0.381477	-0.16372	0.37461	-0.20171
Cu	0.87619		0.854056	0.35126	0.791519	0.199698	0.230362	0.162619	0.125802	0.549527	0.232108
Sb	0.801961	0.854056		0.532445	0.673097	0.036011	0.392984	0.546208	-0.21879	0.285306	0.242551
Ca	0.327409	0.35126	0.532445		0.316644	-0.09891	0.525191	0.690574	-0.47827	-0.0962	0.433613
Zn	0.557379	0.791519	0.673097	0.316644		0.214107	0.355745	0.205688	0.093718	0.454193	0.303718
Fe	0.056932	0.199698	0.036011	-0.09891	0.214107		0.538207	-0.49051	0.753597	0.33665	0.672464
Mn	0.069022	0.230362	0.392984	0.525191	0.355745	0.538207		0.311712	0.033346	-0.04784	0.712635
Mg	0.381477	0.162619	0.546208	0.690574	0.205688	-0.49051	0.311712		-0.85741	-0.18384	-0.108
Al	-0.16372	0.125802	-0.21879	-0.47827	0.093718	0.753597	0.033346	-0.85741		0.506592	0.390332
K	0.37461	0.549527	0.285306	-0.0962	0.454193	0.33665	-0.04784	-0.18384	0.506592		0.139737
pH	-0.20171	0.232108	0.242551	0.433613	0.303718	0.672464	0.712635	-0.108	0.390332	0.139737	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.000185	0.001702	0.298874	0.059728	0.860498	0.831216	0.221115	0.61115	0.256337	0.529568
Cu	0.000185		2.02E-04	0.239255	0.001265	0.51304	0.448952	0.595552	0.68216	0.064202	0.445424
Sb	0.001702	0.000202		0.061037	0.011685	0.907025	0.184059	0.053454	0.472658	0.368714	0.424612
Ca	0.298874	0.239255	0.061037		0.291849	0.747843	0.065326	0.008971	0.098293	0.766155	0.138801
Zn	0.059728	0.001265	0.011685	0.291849		0.482429	0.232893	0.500211	0.760725	0.138002	0.313058
Fe	0.860498	0.51304	0.907025	0.747843	0.482429		0.057776	0.088786	0.002932	0.284622	0.011794
Mn	0.831216	0.448952	0.184059	0.065326	0.232893	0.057776		0.299841	0.913881	0.882623	0.006262
Mg	0.221115	0.595552	0.053454	0.008971	0.500211	0.088786	0.299841		1.79E-04	0.567355	0.725456
Al	0.61115	0.68216	0.472658	0.098293	0.760725	0.002932	0.913881	1.79E-04		0.092811	0.187301
K	0.256337	0.064202	0.368714	0.766155	0.138002	0.284622	0.882623	0.567355	0.092811		0.664914
pH	0.529568	0.445424	0.424612	0.138801	0.313058	0.011794	0.006262	0.725456	0.187301	0.664914	

Av, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.836828	0.818462	0.389674	0.403532	0.228474	0.13046	0.500146	-0.18336	0.103687	0.354532
Cu	0.836828		0.789511	0.470973	0.35965	0.395513	0.281396	0.348953	0.037059	0.173051	0.60711
Sb	0.818462	0.789511		0.346495	0.569292	0.055135	0.22073	0.650639	-0.31154	0.169547	0.280714
Ca	0.389674	0.470973	0.346495		0.208549	-0.16878	-0.02871	0.636852	-0.4345	-0.34012	0.328105
Zn	0.403532	0.35965	0.569292	0.208549		0.169039	0.532024	0.535854	-0.27758	0.131275	0.05486
Fe	0.228474	0.395513	0.055135	-0.16878	0.169039		0.793497	-0.45442	0.824548	0.53179	0.793833
Mn	0.13046	0.281396	0.22073	-0.02871	0.532024	0.793497		-0.14411	0.516482	0.325755	0.683707
Mg	0.500146	0.348953	0.650639	0.636852	0.535854	-0.45442	-0.14411		-0.83262	-0.08043	-0.13558
Al	-0.18336	0.037059	-0.31154	-0.4345	-0.27758	0.824548	0.516482	-0.83262		0.454099	0.601876
K	0.103687	0.173051	0.169547	-0.34012	0.131275	0.53179	0.325755	-0.08043	0.454099		0.362658
pH	0.354532	0.60711	0.280714	0.328105	0.05486	0.793833	0.683707	-0.13558	0.601876	0.362658	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.001317	0.00207	0.236152	0.218428	0.499216	0.702216	0.117183	0.589421	0.775608	0.284713
Cu	0.001317		0.003852	0.14369	0.277321	0.228585	0.401881	0.292894	0.913857	0.632586	0.047612
Sb	0.00207	0.003852		0.296539	0.067553	0.872088	0.514258	0.030169	0.351033	0.639595	0.403071
Ca	0.236152	0.14369	0.296539		0.538301	0.619829	0.933219	0.035103	0.181748	0.336242	0.324593
Zn	0.218428	0.277321	0.067553	0.538301		0.619283	0.092071	0.089316	0.408572	0.717735	0.872721
Fe	0.499216	0.228585	0.872088	0.619829	0.619283		0.003556	0.160278	0.001792	0.113626	0.003532
Mn	0.702216	0.401881	0.514258	0.933219	0.092071	0.003556		0.672483	0.103822	0.358335	0.020354
Mg	0.117183	0.292894	0.030169	0.035103	0.089316	0.160278	0.672483		0.001468	0.825195	0.691026
Al	0.589421	0.913857	0.351033	0.181748	0.408572	0.001792	0.103822	0.001468		0.187393	0.050093
K	0.775608	0.632586	0.639595	0.336242	0.717735	0.113626	0.358335	0.825195	0.187393		0.303048
pH	0.284713	0.047612	0.403071	0.324593	0.872721	0.003532	0.020354	0.691026	0.050093	0.303048	

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Supplementary materials,

Table 5A Correlation analysis between the element concentrations and pH in the soil from **Ti** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations (p<0.05) are marked in yellow.

Ti, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.900121	0.814451	-0.21849	0.297168	-0.35856	0.094547	-0.8042	-0.74329	-0.19023	0.26848
Cu	0.900121		0.908385	-0.43978	0.187246	-0.24919	-0.08489	-0.73661	-0.82796	-0.3995	0.367419
Sb	0.814451	0.908385		-0.50723	0.104333	-0.07054	-0.25726	-0.60385	-0.6946	-0.44359	0.299601
Ca	-0.21849	-0.43978	-0.50723		0.326224	0.345014	0.70023	0.282236	0.495479	0.843805	-0.08037
Zn	0.297168	0.187246	0.104333	0.326224		-0.1206	0.254228	-0.20123	-0.37729	0.556463	0.646089
Fe	-0.35856	-0.24919	-0.07054	0.345014	-0.1206		0.069492	0.420435	0.347684	0.204047	0.072901
Mn	0.094547	-0.08489	-0.25726	0.70023	0.254228	0.069492		0.120293	0.026493	0.430198	0.017306
Mg	-0.8042	-0.73661	-0.60385	0.282236	-0.20123	0.420435	0.120293		0.642065	0.09928	-0.0592
Al	-0.74329	-0.82796	-0.6946	0.495479	-0.37729	0.347684	0.026493	0.642065		0.435955	-0.52873
K	-0.19023	-0.3995	-0.44359	0.843805	0.556463	0.204047	0.430198	0.09928	0.435955		0.061027
pH	0.26848	0.367419	0.299601	-0.08037	0.646089	0.072901	0.017306	-0.0592	-0.52873	0.061027	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		6.61E-05	0.001257	0.495106	0.348236	0.252408	0.770077	0.001615	0.005598	0.553712	0.398808
Cu	6.61E-05		4.35E-05	0.152546	0.560072	0.434785	0.793091	0.006287	0.000882	0.198229	0.240021
Sb	0.001257	4.35E-05		0.092335	0.746934	0.827545	0.41955	0.037595	0.012183	0.148607	0.344112
Ca	0.495106	0.152546	0.092335		0.30073	0.272062	0.011219	0.374113	0.101417	0.00056	0.803918
Zn	0.348236	0.560072	0.746934	0.30073		0.70889	0.425235	0.530571	0.226648	0.060238	0.023219
Fe	0.252408	0.434785	0.827545	0.272062	0.70889		0.830081	0.173551	0.26812	0.524706	0.82186
Mn	0.770077	0.793091	0.41955	0.011219	0.425235	0.830081		0.709604	0.934864	0.162736	0.957427
Mg	0.001615	0.006287	0.037595	0.374113	0.530571	0.173551	0.709604		0.02438	0.75886	0.854986
Al	0.005598	0.000882	0.012183	0.101417	0.226648	0.26812	0.934864	0.02438		0.156563	0.077167
K	0.553712	0.198229	0.148607	0.00056	0.060238	0.524706	0.162736	0.75886	0.156563		0.85056
pH	0.398808	0.240021	0.344112	0.803918	0.023219	0.82186	0.957427	0.854986	0.077167	0.85056	

Ti, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.82231	0.68509	-0.14484	0.469211	-0.18464	0.05544	-0.13828	-0.34711	-0.06305	0.245498
Cu	0.82231		0.729586	-0.16753	0.819362	-0.39078	-0.18643	-0.13009	-0.56663	-0.30122	0.500887
Sb	0.68509	0.729586		-0.37685	0.562368	-0.45356	-0.36962	-0.23491	-0.46859	-0.31617	0.401471
Ca	-0.14484	-0.16753	-0.37685		0.169864	0.644787	0.83558	0.44162	0.57073	0.742332	-0.22111
Zn	0.469211	0.819362	0.562368	0.169864		-0.20201	-0.054	0.186846	-0.27982	-0.09309	0.535436
Fe	-0.18464	-0.39078	-0.45356	0.644787	-0.20201		0.836693	0.690891	8.85E-01	0.801383	-0.55044
Mn	0.05544	-0.18643	-0.36962	0.83558	-0.054	0.836693		0.395884	0.677654	9.05E-01	-0.52441
Mg	-0.13828	-0.13009	-0.23491	0.44162	0.186846	0.690891	0.395884		0.741343	0.365894	0.012303
Al	-0.34711	-0.56663	-0.46859	0.57073	-0.27982	8.85E-01	0.677654	0.741343		0.786691	-0.50406
K	-0.06305	-0.30122	-0.31617	0.742332	-0.09309	0.801383	9.05E-01	0.365894	0.786691		-0.59353
pH	0.245498	0.500887	0.401471	-0.22111	0.535436	-0.55044	-0.52441	0.012303	-0.50406	-0.59353	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.001027	0.01395	0.653343	0.123836	0.565644	0.864123	0.668225	0.26896	0.845647	0.441836
Cu	0.001027		0.007079	0.602754	0.001109	0.209115	0.561823	0.686971	0.054741	0.341379	0.097166
Sb	0.01395	0.007079		0.227231	0.057	0.138625	0.237003	0.462375	0.124406	0.31674	0.19582
Ca	0.653343	0.602754	0.227231		0.597645	0.023591	0.000713	0.150637	0.052625	0.005693	0.48981
Zn	0.123836	0.001109	0.057	0.597645		0.528933	0.867627	0.560926	0.378387	0.773527	0.072804
Fe	0.565644	0.209115	0.138625	0.023591	0.528933		0.000691	0.012851	1.31E-04	0.001725	0.063671
Mn	0.864123	0.561823	0.237003	0.000713	0.867627	0.000691		0.2027	0.015457	5.19E-05	0.080063
Mg	0.668225	0.686971	0.462375	0.150637	0.560926	0.012851	0.2027		0.005793	0.242126	0.96973
Al	0.26896	0.054741	0.124406	0.052625	0.378387	1.31E-04	0.015457	0.005793		0.002401	0.094725
K	0.845647	0.341379	0.31674	0.005693	0.773527	0.001725	5.19E-05	0.242126	0.002401		0.041898
pH	0.441836	0.097166	0.19582	0.48981	0.072804	0.063671	0.080063	0.96973	0.094725	0.041898	

Ti, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.618013	0.59812	-0.6919	-0.01656	-0.72903	-0.59244	-0.53925	-0.57361	-0.8344	0.56085
Cu	0.618013		0.354961	-0.28326	0.262981	-0.29598	-0.11994	-0.39833	-0.43014	-0.62244	0.349792
Sb	0.59812	0.354961		-0.19796	-0.33978	-0.5101	-0.32051	-0.6217	-0.42789	-0.62245	0.659159
Ca	-0.6919	-0.28326	-0.19796		0.049821	0.459024	0.420133	0.308297	0.581592	0.513376	-0.02534
Zn	-0.01656	0.262981	-0.33978	0.049821		0.278463	-0.05519	0.619904	0.334043	0.355479	-0.03444
Fe	-0.72903	-0.29598	-0.5101	0.459024	0.278463		0.776122	0.683324	0.584249	0.805287	-0.69147
Mn	-0.59244	-0.11994	-0.32051	0.420133	-0.05519	0.776122		0.39381	0.323241	0.459389	-0.5407
Mg	-0.53925	-0.39833	-0.6217	0.308297	0.619904	0.683324	0.39381		0.840271	0.855418	-0.38126
Al	-0.57361	-0.43014	-0.42789	0.581592	0.334043	0.584249	0.323241	0.840271		0.798671	-0.15785
K	-0.8344	-0.62244	-0.62245	0.513376	0.355479	0.805287	0.459389	0.855418	0.798671		-0.59606
pH	0.56085	0.349792	0.659159	-0.02534	-0.03444	-0.69147	-0.5407	-0.38126	-0.15785	-0.59606	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.032216	0.039945	0.012668	0.959266	0.007144	0.042373	0.070403	0.051176	0.000738	0.05782
Cu	0.032216		0.257547	0.372309	0.408905	0.350261	0.710424	0.199666	0.162799	0.030653	0.265033
Sb	0.039945	0.257547		0.537404	0.27988	0.090198	0.309765	0.030911	0.165254	0.030652	0.019724
Ca	0.012668	0.372309	0.537404		0.877799	0.133336	0.173894	0.329591	0.0473	0.087808	0.937698
Zn	0.959266	0.408905	0.27988	0.877799		0.380807	0.864734	0.031542	0.288602	0.256804	0.915385
Fe	0.007144	0.350261	0.090198	0.133336	0.380807		0.002999	0.014298	0.046058	0.001573	0.012746
Mn	0.042373	0.710424	0.309765	0.173894	0.864734	0.002999		0.205291	0.305432	0.132987	0.069501
Mg	0.070403	0.199666	0.030911	0.329591	0.031542	0.014298	0.205291		0.000622	0.000388	0.221403
Al	0.051176	0.162799	0.165254	0.0473	0.288602	0.046058	0.305432	0.000622		0.001837	0.624151
K	0.000738	0.030653	0.030652	0.087808	0.256804	0.001573	0.132987	3.88E-04	0.001837		0.040815
pH	0.05782	0.265033	0.019724	0.937698	0.915385	0.012746	0.069501	0.221403	0.624151	0.040815	

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Supplementary materials,

Table 6A Correlation analysis between the element concentrations and pH in the soil from **He** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

He, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.820269	0.702173	0.295488	0.326708	0.310501	0.265117	0.309027	-0.22528	0.374897	-0.23003
Cu	0.820269		0.697166	0.024336	0.459512	0.338285	0.099217	0.143572	0.09633	0.223119	0.134726
Sb	0.702173	0.697166		0.179968	0.237636	0.438138	0.162855	0.106396	-0.04404	0.085683	-0.20282
Ca	0.295488	0.024336	0.179968		0.088977	0.317786	0.963487	0.937683	-0.33909	0.410166	-0.08409
Zn	0.326708	0.459512	0.237636	0.088977		-0.22511	0.013102	0.079866	0.218116	0.698991	-0.28441
Fe	0.310501	0.338285	0.438138	0.317786	-0.22511		0.405466	0.41501	-0.15968	-0.02967	0.381459
Mn	0.265117	0.099217	0.162855	0.963487	0.013102	0.405466		0.974912	-0.26108	0.306047	0.130782
Mg	0.309027	0.143572	0.106396	0.937683	0.079866	0.41501	0.974912		-0.16641	0.450647	0.131481
Al	-0.22528	0.09633	-0.04404	-0.33909	0.218116	-0.15968	-0.26108	-0.16641		0.212256	0.213661
K	0.374897	0.223119	0.085683	0.410166	0.698991	-0.02967	0.306047	0.450647	0.212256		-0.46112
pH	-0.23003	0.134726	-0.20282	-0.08409	-0.28441	0.381459	0.130782	0.131481	0.213661	-0.46112	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.000594	0.007454	0.327002	0.275925	0.301824	0.381359	0.304245	0.459298	0.206888	0.44963
Cu	0.000594		0.008083	0.937102	0.11417	0.258246	0.747087	0.639833	0.75424	0.463726	0.660788
Sb	0.007454	0.008083		0.556304	0.434347	0.134265	0.59501	0.729382	0.886407	0.78077	0.506332
Ca	0.327002	0.937102	0.556304		0.772537	0.290016	1.23E-07	2.21E-06	0.257041	0.163912	0.784747
Zn	0.275925	0.11417	0.434347	0.772537		0.459641	0.966115	0.795358	0.474066	0.007849	0.3463
Fe	0.301824	0.258246	0.134265	0.290016	0.459641		0.169275	0.158501	0.6023	0.923339	0.198411
Mn	0.381359	0.747087	0.59501	1.23E-07	0.966115	0.169275		1.60E-08	0.388912	0.309174	0.670206
Mg	0.304245	0.639833	0.729382	2.21E-06	0.795358	0.158501	1.60E-08		0.586875	0.122241	0.668534
Al	0.459298	0.75424	0.886407	0.257041	0.474066	0.6023	0.388912	0.586875		0.486313	0.483364
K	0.206888	0.463726	0.78077	0.163912	0.007849	0.923339	0.309174	0.122241	0.486313		0.112749
pH	0.44963	0.660788	0.506332	0.784747	0.3463	0.198411	0.670206	0.668534	0.483364	0.112749	

He, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.905513	0.240568	-0.58519	0.295799	-0.34323	-0.66933	-0.6345	0.563099	-0.15711	-0.64421
Cu	0.905513		0.330811	-0.50506	0.425957	-0.12918	-0.52578	-0.43239	0.596361	-0.01882	-0.549
Sb	0.240568	0.330811		-0.03237	0.492569	-0.25852	-0.08402	-0.16622	-0.16572	0.139883	-0.47019
Ca	-0.58519	-0.50506	-0.03237		0.309453	0.382371	0.907662	0.791447	-0.48736	0.608353	0.513251
Zn	0.295799	0.425957	0.492569	0.309453		-0.09771	0.116059	0.143999	-0.19206	0.25313	0.008003
Fe	-0.34323	-0.12918	-0.25852	0.382371	-0.09771		0.615922	0.682966	0.195439	0.360837	0.249951
Mn	-0.66933	-0.52578	-0.08402	0.907662	0.116059	0.615922		0.940245	-0.32399	0.580867	0.596327
Mg	-0.6345	-0.43239	-0.16622	0.791447	0.143999	0.682966	0.940245		-0.17848	0.545539	0.684324
Al	0.563099	0.596361	-0.16572	-0.48736	-0.19206	0.195439	-0.32399	-0.17848		0.221497	-0.22729
K	-0.15711	-0.01882	0.139883	0.608353	0.25313	0.360837	0.580867	0.545539	0.221497		0.16125
pH	-0.64421	-0.549	-0.47019	0.513251	0.008003	0.249951	0.596327	0.684324	-0.22729	0.16125	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		2.05E-05	0.428527	0.035643	0.326469	0.250904	0.012344	0.019837	0.045097	0.60824	0.017477
Cu	2.05E-05		0.26958	0.078332	0.146702	0.674046	0.064971	0.140045	0.031454	0.951343	0.052
Sb	0.428527	0.26958		0.916405	0.08725	0.393752	0.784933	0.587321	0.58845	0.648543	0.104935
Ca	0.035643	0.078332	0.916405		0.303544	0.197251	1.82E-05	0.001268	0.091165	0.027371	0.07284
Zn	0.326469	0.146702	0.08725	0.303544		0.750807	0.705747	0.638826	0.5296	0.404038	0.979298
Fe	0.250904	0.674046	0.393752	0.197251	0.750807		0.025004	0.010086	0.522251	0.225794	0.410167
Mn	0.012344	0.064971	0.784933	1.82E-05	0.705747	0.025004		1.77E-06	0.280168	0.037367	0.031466
Mg	0.019837	0.140045	0.587321	0.001268	0.638826	0.010086	1.77E-06		0.559631	0.053807	0.009879
Al	0.045097	0.031454	0.58845	0.091165	0.5296	0.522251	0.280168	0.559631		0.467067	0.45519
K	0.60824	0.951343	0.648543	0.027371	0.404038	0.225794	0.037367	0.053807	0.467067		0.598694
pH	0.017477	0.052	0.104935	0.07284	0.979298	0.410167	0.031466	0.009879	0.45519	0.598694	

He, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		0.874165	0.392582	-0.06719	0.227099	-0.09382	-0.3105	-0.37423	0.604388	0.268349	-0.61201
Cu	0.874165		0.353237	-0.37642	0.363176	-0.21608	-0.55266	-0.57541	0.784882	-0.05706	-0.5806
Sb	0.392582	0.353237		-0.0662	0.129272	-0.16401	0.008359	0.011245	0.234242	-0.00799	-0.14005
Ca	-0.06719	-0.37642	-0.0662		-0.22805	0.247758	0.815316	0.773037	-0.33781	0.675258	0.35021
Zn	0.227099	0.363176	0.129272	-0.22805		-0.67339	-0.39771	-0.28605	0.272364	-0.03668	-0.13848
Fe	-0.09382	-0.21608	-0.16401	0.247758	-0.67339		0.4025	0.380526	0.120233	0.153989	0.331762
Mn	-0.3105	-0.55266	0.008359	0.815316	-0.39771	0.4025		0.973226	-0.40396	0.537247	0.308774
Mg	-0.37423	-0.57541	0.011245	0.773037	-0.28605	0.380526	0.973226		-0.35731	0.53114	0.430636
Al	0.604388	0.784882	0.234242	-0.33781	0.272364	0.120233	-0.40396	-0.35731		0.138291	-0.21983
K	0.268349	-0.05706	-0.00799	0.675258	-0.03668	0.153989	0.537247	0.53114	0.138291		0.030697
pH	-0.61201	-0.5806	-0.14005	0.35021	-0.13848	0.331762	0.308774	0.430636	-0.21983	0.030697	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	K	pH
Pb		9.32E-05	0.184548	0.82737	0.45558	0.760478	0.301818	0.207761	0.028675	0.375362	0.026208
Cu	9.32E-05		0.236438	0.204897	0.222577	0.478309	0.050143	0.039628	0.001483	0.853122	0.037476
Sb	0.184548	0.236438		0.829862	0.673822	0.59237	0.978378	0.970916	0.441131	0.979325	0.648145
Ca	0.82737	0.204897	0.829862		0.453635	0.414421	0.000682	0.001943	0.258961	0.01132	0.240759
Zn	0.45558	0.222577	0.673822	0.453635		0.011635	0.178372	0.343414	0.367979	0.905296	0.651863
Fe	0.760478	0.478309	0.59237	0.414421	0.011635		0.172717	0.199603	0.695613	0.615467	0.268123
Mn	0.301818	0.050143	0.978378	0.000682	0.178372	0.172717		2.28E-08	0.171017	0.05831	0.304662
Mg	0.207761	0.039628	0.970916	0.001943	0.343414	0.199603	2.28E-08		0.23069	0.061793	0.141839
Al	0.028675	0.001483	0.441131	0.258961	0.367979	0.695613	0.171017	0.23069		0.652314	0.470519
K	0.375362	0.853122	0.979325	0.01132	0.905296	0.615467	0.05831	0.061793	0.652314		0.920702
pH	0.026208	0.037476	0.648145	0.240759	0.651863	0.268123	0.304662	0.141839	0.470519	0.920702	

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Supplementary materials,

Table 7A Correlation analysis between the element concentrations and pH in the soil water from **Kj** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

Kj, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.5727	0.248988	-0.19438	0.299653	-0.33621	-0.44407	-0.11515	0.135628	0.155832	0.155789
Cu	0.572701		0.694554	-0.42295	0.47589	-0.53532	-0.48415	-0.37966	0.004871	-0.08013	0.052156
Sb	0.248988	0.69455		-0.25218	0.114037	-0.40617	-0.28802	-0.23059	0.026555	-0.29857	-0.03793
Ca	-0.19438	-0.42295	-0.25218		-0.71303	0.881884	0.848774	0.974139	0.105521	0.582568	-0.36548
Zn	0.299653	0.47589	0.114037	-0.71303		-0.66233	-0.64425	-0.61917	0.043937	-0.07461	0.341245
Fe	-0.33621	-0.53532	-0.40617	0.881884	-0.66233		0.943101	0.831399	0.189413	0.420411	-0.1339
Mn	-0.44407	-0.48415	-0.28802	0.848774	-0.64425	0.943101		0.799926	0.041178	0.425288	-0.24692
Mg	-0.11515	-0.37966	-0.23059	0.974139	-0.61917	0.831399	0.799926		0.137857	0.680516	-0.32261
Al	0.135628	0.00487	0.026555	0.105521	0.043937	0.189413	0.041178	0.137857		0.130094	0.627854
Na	0.155832	-0.08013	-0.29857	0.582568	-0.07461	0.420411	0.425288	0.680516	0.130094		-0.341
K	0.155789	0.05216	-0.03793	-0.36548	0.341245	-0.1339	-0.24692	-0.32261	0.627854	-0.341	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.00831	0.289783	0.411531	0.199303	0.147235	0.049822	0.628809	0.568584	0.511788	0.511906
Cu	0.008311		0.000678	0.063168	0.033926	0.015001	0.030531	0.098726	0.983741	0.737007	0.827135
Sb	0.289783	0.00068		0.283436	0.632141	0.075566	0.21817	0.328023	0.911515	0.201016	0.873855
Ca	0.411531	0.06317	0.283436		0.000417	2.75E-07	2.24E-06	4.47E-13	0.65794	0.00703	0.113049
Zn	0.199303	0.03393	0.632141	0.000417		0.001464	0.00217	0.003602	0.85407	0.754571	0.1409
Fe	0.147235	0.015	0.075566	2.75E-07	0.001464		4.82E-10	5.57E-06	0.423817	0.064939	0.573554
Mn	0.049822	0.03053	0.21817	2.24E-06	0.00217	4.82E-10		2.3E-05	0.863149	0.061571	0.293935
Mg	0.628809	0.09873	0.328023	4.47E-13	0.003602	5.57E-06	2.3E-05		0.562186	0.000959	0.16535
Al	0.568584	0.98374	0.911515	0.65794	0.85407	0.423817	0.863149	0.562186		0.584606	0.003037
Na	0.511788	0.73701	0.201016	0.00703	0.754571	0.064939	0.061571	0.000959	0.584606		0.141209
K	0.511906	0.82713	0.873855	0.113049	0.1409	0.573554	0.293935	0.16535	0.003037	0.141209	

Kj, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.626084	0.489934	-0.47755	0.300346	-0.52524	-0.55101	-0.44315	-0.05678	-0.13516	0.115418
Cu	0.626084		0.470134	-0.57926	0.517066	-0.65091	-0.65612	-0.49021	0.146491	0.268066	0.267295
Sb	0.489934	0.470134		-0.35417	0.132361	-0.2626	-0.54729	-0.22824	0.05943	-0.16019	0.185712
Ca	-0.47755	-0.57926	-0.35417		-0.57039	0.85079	0.852972	0.9008	0.058297	0.213623	0.010758
Zn	0.300346	0.517066	0.132361	-0.57039		-0.51398	-0.54808	-0.51867	0.092016	0.297884	0.203096
Fe	-0.52524	-0.65091	-0.2626	0.85079	-0.51398		0.835736	0.722663	0.274938	0.008762	0.121073
Mn	-0.55101	-0.65612	-0.54729	0.852972	-0.54808	0.835736		0.628723	-0.02648	0.147164	-0.14789
Mg	-0.44315	-0.49021	-0.22824	0.9008	-0.51867	0.722663	0.628723		0.126476	0.16274	0.12169
Al	-0.05678	0.146491	0.05943	0.058297	0.092016	0.274938	-0.02648	0.126476		-0.04867	0.795072
Na	-0.13516	0.268066	-0.16019	0.213623	0.297884	0.008762	0.147164	0.16274	-0.04867		0.116914
K	0.115418	0.267295	0.185712	0.010758	0.203096	0.121073	-0.14789	0.12169	0.795072	0.116914	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.003146	0.028319	0.033223	0.198216	0.0174	0.011802	0.050357	0.812066	0.569933	0.627992
Cu	0.003146		0.03646	0.00744	0.019563	0.001882	0.00168	0.028216	0.537709	0.253161	0.254581
Sb	0.028319	0.03646		0.125504	0.578021	0.263346	0.012505	0.333109	0.80345	0.49991	0.433093
Ca	0.033223	0.00744	0.125504		0.008636	2E-06	1.77E-06	6.13E-08	0.807128	0.365816	0.964097
Zn	0.198216	0.019563	0.578021	0.008636		0.020434	0.012354	0.019122	0.699622	0.202099	0.390456
Fe	0.0174	0.001882	0.263346	2E-06	0.020434		4.48E-06	0.000319	0.240727	0.970755	0.611126
Mn	0.011802	0.00168	0.012505	1.77E-06	0.012354	4.48E-06		0.002985	0.911778	0.535819	0.533788
Mg	0.050357	0.028216	0.333109	6.13E-08	0.019122	0.000319	0.002985		0.595183	0.493011	0.609297
Al	0.812066	0.537709	0.80345	0.807128	0.699622	0.240727	0.911778	0.595183		0.838537	2.8E-05
Na	0.569933	0.253161	0.49991	0.365816	0.202099	0.970755	0.535819	0.493011	0.838537		0.623514
K	0.627992	0.254581	0.433093	0.964097	0.390456	0.611126	0.533788	0.609297	2.8E-05	0.623514	

Kj, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.737412	0.429597	0.006178	0.138778	0.010898	-0.01168	-0.06951	0.678846	0.030945	0.4101143
Cu	0.737412		0.405882	-0.33615	0.645964	-0.41528	-0.35722	-0.41561	0.47735	0.417195	0.5097591
Sb	0.429597	0.405882		-0.17304	0.125577	-0.01667	-0.31706	-0.18797	0.357437	0.125231	0.3213553
Ca	0.006178	-0.33615	-0.17304		-0.74318	0.907284	0.893203	0.934829	0.126778	0.133177	-0.000163
Zn	0.138778	0.645964	0.125577	-0.74318		-0.76843	-0.69309	-0.71975	-0.19295	0.203692	0.0288868
Fe	0.010898	-0.41528	-0.01667	0.907284	-0.76843		0.854263	0.822244	0.135169	-0.0281	0.0404773
Mn	-0.01168	-0.35722	-0.31706	0.893203	-0.69309	0.854263		0.75473	0.045848	0.047051	-0.188991
Mg	-0.06951	-0.41561	-0.18797	0.934829	-0.71975	0.822244	0.75473		-0.0188	0.025636	-0.05269
Al	0.678846	0.47735	0.357437	0.126778	-0.19295	0.135169	0.045848	-0.0188		0.052407	0.6368491
Na	0.030945	0.417195	0.125231	0.133177	0.203692	-0.0281	0.047051	0.025636	0.052407		0.1976577
K	0.410114	0.509759	0.321355	-0.00016	0.028887	0.040477	-0.18899	-0.05269	0.636849	0.197658	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.000479	0.058707	0.979376	0.559551	0.963628	0.961024	0.770898	0.000998	0.896955	0.0725019
Cu	0.000479		0.094678	0.172609	0.003779	0.086564	0.14559	0.086295	0.045149	0.084979	0.0306893
Sb	0.058707	0.094678		0.46566	0.597823	0.944389	0.173173	0.427421	0.121812	0.598842	0.1671006
Ca	0.979376	0.172609	0.46566		0.000174	3.42E-08	1.16E-07	1.59E-09	0.594298	0.575658	0.999455
Zn	0.559551	0.003779	0.597823	0.000174		7.57E-05	0.000704	0.000347	0.41505	0.389037	0.9037766
Fe	0.963628	0.086564	0.944389	3.42E-08	7.57E-05		1.64E-06	8.65E-06	0.569908	0.906377	0.8654562
Mn	0.961024	0.14559	0.173173	1.16E-07	0.000704	1.64E-06		0.00012	0.847791	0.843844	0.4248683
Mg	0.770898	0.086295	0.427421	1.59E-09	0.000347	8.65E-06	0.00012		0.937309	0.914564	0.8253897
Al	0.000998	0.045149	0.121812	0.594298	0.41505	0.569908	0.847791	0.937309		0.826316	0.0025314
Na	0.896955	0.084979	0.598842	0.575658	0.389037	0.906377	0.843844	0.914564	0.826316		0.403536
K	0.072502	0.030689	0.167101	0.999455	0.903777	0.865456	0.424868	0.82539	0.002531	0.403536	

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Supplementary materials,

Table 8A Correlation analysis between the element concentrations and pH in the soil water from **Ma** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

Ma, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.82429	0.415999	0.56307	0.696727	-0.07284	0.623489	0.642212	0.374568	0.30263	0.283398
Cu	0.824293		0.350912	0.464199	0.677751	0.011118	0.419206	0.429779	0.376023	0.226787	0.221789
Sb	0.415999	0.35091		0.079222	0.205542	-0.6381	0.186277	0.335279	0.51786	0.24153	0.230364
Ca	0.56307	0.4642	0.079222		0.79391	0.037767	0.509615	0.871716	-0.06983	0.410638	0.416754
Zn	0.696727	0.67775	0.205542	0.79391		0.000273	0.45781	0.696737	0.028292	0.177557	0.164085
Fe	-0.07284	0.01112	-0.6381	0.037767	0.000273		-0.10133	-0.27294	-0.00673	-0.28643	-0.13781
Mn	0.623489	0.41921	0.186277	0.509615	0.45781	-0.10133		0.670367	0.155478	0.389032	0.247655
Mg	0.642212	0.42978	0.335279	0.871716	0.696737	-0.27294	0.670367		0.05005	0.545931	0.458949
Al	0.374568	0.37602	0.51786	-0.06983	0.028292	-0.00673	0.155478	0.05005		-0.04666	0.263456
Na	0.30263	0.22679	0.24153	0.410638	0.177557	-0.28643	0.389032	0.545931	-0.04666		0.544867
K	0.283398	0.22179	0.230364	0.416754	0.164085	-0.13781	0.247655	0.458949	0.263456	0.544867	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		1.2E-09	0.012936	0.000429	3.33E-06	0.67754	6.3E-05	3.2E-05	0.02663	0.077218	0.098997
Cu	1.16E-09		0.038747	0.004969	7.72E-06	0.94946	0.012188	0.009975	0.026	0.190158	0.200374
Sb	0.012936	0.03875		0.650997	0.2362	3.73E-05	0.28398	0.048963	0.001441	0.162171	0.183076
Ca	0.000429	0.00497	0.650997		1.27E-08	0.829459	0.001766	9.38E-12	0.69017	0.014274	0.012757
Zn	3.33E-06	7.7E-06	0.2362	1.27E-08		0.99876	0.005686	3.33E-06	0.871834	0.307525	0.34625
Fe	0.67754	0.94946	3.73E-05	0.829459	0.99876		0.562462	0.112647	0.969411	0.095285	0.429832
Mn	6.3E-05	0.01219	0.28398	0.001766	0.005686	0.562462		1.05E-05	0.372472	0.0209	0.15146
Mg	3.2E-05	0.00997	0.048963	9.38E-12	3.33E-06	0.112647	1.05E-05		0.775243	0.000693	0.005552
Al	0.02663	0.026	0.001441	0.69017	0.871834	0.969411	0.372472	0.775243		0.790108	0.12622
Na	0.077218	0.19016	0.162171	0.014274	0.307525	0.095285	0.0209	0.000693	0.790108		0.000713
K	0.098997	0.20037	0.183076	0.012757	0.34625	0.429832	0.15146	0.005552	0.12622	0.000713	

Ma, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.940165	0.469144	-0.02082	0.576	0.090678	0.343933	-0.11464	0.826264	-0.4661	-0.31244
Cu	0.940165		0.471425	-0.04879	0.540794	0.067875	0.323782	-0.15085	0.799794	-0.51341	-0.32043
Sb	0.469144	0.471425		-0.16247	0.476196	-0.38872	0.074396	-0.22899	0.494694	-0.6995	0.093802
Ca	-0.02082	-0.04879	-0.16247		0.396311	0.645802	0.039146	0.945347	0.058385	0.290109	0.302338
Zn	0.576	0.540794	0.476196	0.396311		0.239661	0.552447	0.265613	0.423414	-0.32037	-0.08355
Fe	0.090678	0.067875	-0.38872	0.645802	0.239661		0.281242	0.550323	0.055423	0.335291	0.134057
Mn	0.343933	0.323782	0.074396	0.039146	0.552447	0.281242		-0.02547	0.178004	-0.10887	-0.26328
Mg	-0.11464	-0.15085	-0.22899	0.945347	0.265613	0.550323	-0.02547		0.041647	0.3401	0.270897
Al	0.826264	0.799794	0.494694	0.058385	0.423414	0.055423	0.178004	0.041647		-0.46382	-0.2468
Na	-0.4661	-0.51341	-0.6995	0.290109	-0.32037	0.335291	-0.10887	0.3401	-0.46382		0.285002
K	-0.31244	-0.32043	0.093802	0.302338	-0.08355	0.134057	-0.26328	0.270897	-0.2468	0.285002	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		1.28E-14	0.008915	0.913044	0.000866	0.633687	0.062746	0.54637	1.87E-08	0.00943	0.092775
Cu	1.28E-14		0.008545	0.797939	0.002032	0.721556	0.080909	0.426203	1.14E-07	0.003713	0.084281
Sb	0.008915	0.008545		0.391003	0.007812	0.03376	0.696011	0.223537	0.005452	1.7E-05	0.62199
Ca	0.913044	0.797939	0.391003		0.03015	0.000116	0.837275	3.71E-15	0.759252	0.119914	0.104403
Zn	0.000866	0.002032	0.007812	0.03015		0.202102	0.001548	0.156005	0.019728	0.084348	0.66069
Fe	0.633687	0.721556	0.03376	0.000116	0.202102		0.132184	0.001628	0.771136	0.070099	0.480029
Mn	0.062746	0.080909	0.696011	0.837275	0.001548	0.132184		0.893701	0.346662	0.566864	0.159806
Mg	0.54637	0.426203	0.223537	3.71E-15	0.156005	0.001628	0.893701		0.827031	0.06593	0.147631
Al	1.87E-08	1.14E-07	0.005452	0.759252	0.019728	0.771136	0.346662	0.827031		0.009833	0.188582
Na	0.00943	0.003713	1.7E-05	0.119914	0.084348	0.070099	0.566864	0.06593	0.009833		0.126874
K	0.092775	0.084281	0.62199	0.104403	0.66069	0.480029	0.159806	0.147631	0.188582	0.126874	

Ma, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.885938	0.189296	-0.03305	0.269806	-0.07205	0.001083	-0.02956	0.479255	-0.03869	-0.074811
Cu	0.885938		0.220929	-0.14559	0.16444	-0.29046	-0.07666	-0.1518	0.531065	-0.09752	0.0606739
Sb	0.189296	0.220929		-0.25495	0.022865	-0.36471	-0.00805	-0.39625	0.547574	-0.52921	-0.139539
Ca	-0.03305	-0.14559	-0.25495		0.157087	0.792253	0.089359	0.875499	-0.13992	0.537513	0.4882793
Zn	0.269806	0.16444	0.022865	0.157087		0.017632	0.131717	0.351272	0.171561	0.292942	-0.080405
Fe	-0.07205	-0.29046	-0.36471	0.792253	0.017632		0.301157	0.727669	-0.25384	0.393758	0.2130292
Mn	0.001083	-0.07666	-0.00805	0.089359	0.131717	0.301157		0.205661	0.036882	-0.03698	-0.211967
Mg	-0.02956	-0.1518	-0.39625	0.875499	0.351272	0.727669	0.205661		-0.22977	0.687158	0.2495174
Al	0.479255	0.531065	0.547574	-0.13992	0.171561	-0.25384	0.036882	-0.22977		-0.36969	-0.123197
Na	-0.03869	-0.09752	-0.52921	0.537513	0.292942	0.393758	-0.03698	0.687158	-0.36969		0.3556441
K	-0.07481	0.060674	-0.13954	0.488279	-0.08041	0.213029	-0.21197	0.249517	-0.1232	0.355644	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		7.6E-11	0.316414	0.862334	0.149332	0.705179	0.995467	0.876779	0.007371	0.83915	0.6943949
Cu	7.6E-11		0.240705	0.442695	0.385211	0.119445	0.687226	0.423275	0.002532	0.608166	0.7501078
Sb	0.316414	0.240705		0.173926	0.904538	0.047527	0.966313	0.03018	0.001737	0.002638	0.4620838
Ca	0.862334	0.442695	0.173926		0.407101	1.82E-07	0.638653	2.42E-10	0.460845	0.00219	0.0061901
Zn	0.149332	0.385211	0.904538	0.407101		0.926319	0.487793	0.056988	0.364672	0.116178	0.6727545
Fe	0.705179	0.119445	0.047527	1.82E-07	0.926319		0.105832	5.2E-06	0.175885	0.031327	0.2583653
Mn	0.995467	0.687226	0.966313	0.638653	0.487793	0.105832		0.275583	0.846574	0.846154	0.2608027
Mg	0.876779	0.423275	0.03018	2.42E-10	0.056988	5.2E-06	0.275583		0.22192	2.74E-05	0.1836046
Al	0.007371	0.002532	0.001737	0.460845	0.364672	0.175885	0.846574	0.22192		0.044359	0.5166088
Na	0.83915	0.608166	0.002638	0.00219	0.116178	0.031327	0.846154	2.74E-05	0.044359		0.0537602
K	0.694395	0.750108	0.462084	0.00619	0.672755	0.258365	0.260803	0.183605	0.516609	0.05376	

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Supplementary materials,

Table 9A Correlation analysis between the element concentrations and pH in the soil water from **St** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

St, 0-15 cm.

Pears co	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**Na**	**K**
Pb		0.76512	0.421367	0.623307	0.521312	-0.02304	0.409971	0.347557	0.248998	0.024039	0.151918
Cu	0.765124		0.196587	0.700687	0.781497	0.062214	0.628785	0.606836	0.53445	-0.20955	-0.07198
Sb	0.421367	0.19659		-0.05507	-0.01466	-0.29789	-0.21105	-0.33982	-0.398	-0.17639	0.050144
Ca	0.623307	0.70069	-0.05507		0.676355	0.084642	0.651292	0.842571	0.591001	0.078977	0.172315
Zn	0.521312	0.7815	-0.01466	0.676355		-0.20812	0.614957	0.760675	0.446874	-0.03095	0.120241
Fe	-0.02304	0.06221	-0.29789	0.084642	-0.20812		0.33237	0.022946	0.370323	-0.21505	-0.42589
Mn	0.409971	0.62879	-0.21105	0.651292	0.614957	0.33237		0.678912	0.629611	-0.2165	-0.0425
Mg	0.347557	0.60684	-0.33982	0.842571	0.760675	0.022946	0.678912		0.661995	0.031002	0.087778
Al	0.248998	0.53445	-0.398	0.591001	0.446874	0.370323	0.629611	0.661995		-0.06327	-0.07482
Na	0.024039	-0.20955	-0.17639	0.078977	-0.03095	-0.21505	-0.2165	0.031002	-0.06327		0.534148
K	0.151918	-0.07198	0.050144	0.172315	0.120241	-0.42589	-0.0425	0.087778	-0.07482	0.534148	

P	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**Na**	**K**
Pb		1.6E-11	0.001508	4.8E-07	5.3E-05	0.868657	0.002079	0.010022	0.069424	0.863012	0.272806
Cu	1.62E-11		0.154229	3.66E-09	3.09E-12	0.654939	3.55E-07	1.15E-06	3.14E-05	0.128315	0.604988
Sb	0.001508	0.15423		0.692483	0.916229	0.028686	0.125537	0.011935	0.002878	0.201982	0.71878
Ca	4.8E-07	3.7E-09	0.692483		1.99E-08	0.542838	9.66E-08	1.37E-15	2.54E-06	0.570261	0.212777
Zn	5.3E-05	3.1E-12	0.916229	1.99E-08		0.130999	7.52E-07	2.49E-11	0.000705	0.824178	0.386456
Fe	0.868657	0.65494	0.028686	0.542838	0.130999		0.014068	0.869186	0.005845	0.118369	0.001324
Mn	0.002079	3.6E-07	0.125537	9.66E-08	7.52E-07	0.014068		1.68E-08	3.39E-07	0.115851	0.760253
Mg	0.010022	1.1E-06	0.011935	1.37E-15	2.49E-11	0.869186	1.68E-08		5.01E-08	0.823896	0.527934
Al	0.069424	3.1E-05	0.002878	2.54E-06	0.000705	0.005845	3.39E-07	5.01E-08		0.649461	0.590789
Na	0.863012	0.12832	0.201982	0.570261	0.824178	0.118369	0.115851	0.823896	0.649461		3.18E-05
K	0.272806	0.60499	0.71878	0.212777	0.386456	0.001324	0.760253	0.527934	0.590789	3.18E-05	

St, 15-30 cm.

Pears co	**Pb**	**Cu**	**Sb**	**Ca**	**Zn**	**Fe**	**Mn**	**Mg**	**Al**	**Na**	**K**
Pb		0.647202	0.123805	0.554904	0.622174	-0.29861	0.049326	0.474865	0.302819	-0.17628	0.379607
Cu	0.647202		0.201897	0.40023	0.296198	0.035416	0.412788	0.484115	0.207907	0.180832	0.268244
Sb	0.123805	0.201897		0.008971	0.319545	-0.30052	0.071868	0.102005	-0.33481	-0.44757	0.165829
Ca	0.554904	0.40023	0.008971		0.609846	0.012641	0.586337	0.931261	0.549922	0.246089	0.388369
Zn	0.622174	0.296198	0.319545	0.609846		-0.48498	0.177537	0.508398	0.240311	-0.40156	0.455655
Fe	-0.29861	0.035416	-0.30052	0.012641	-0.48498		0.460094	0.128495	0.112388	0.696495	-0.32882
Mn	**0.049326**	**0.412788**	**0.071868**	**0.586337**	**0.177537**	0.460094		0.727232	0.257176	0.506276	0.036819
Mg	0.474865	0.484115	0.102005	0.931261	0.508398	0.128495	0.727232		0.444689	0.318621	0.292188
Al	0.302819	0.207907	-0.33481	0.549922	0.240311	0.112388	0.257176	0.444689		0.182603	0.195869
Na	-0.17628	0.180832	-0.44757	0.246089	-0.40156	0.696495	0.506276	0.318621	0.182603		-0.00381
K	0.379607	0.268244	0.165829	0.388369	0.455655	-0.32882	0.036819	0.292188	0.195869	-0.00381	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		6.39E-06	0.446588	0.000202	1.82E-05	0.061252	0.762455	0.001961	0.057521	0.276566	0.015696
Cu	6.39E-06		0.211543	0.010498	0.063481	0.828241	0.008118	0.00155	0.197976	0.264142	0.094219
Sb	0.446588	0.211543		0.956188	0.044438	0.059541	0.659438	0.531109	0.034708	0.003782	0.306478
Ca	0.000202	0.010498	0.956188		2.95E-05	0.938293	7.02E-05	2.99E-18	0.000237	0.125844	0.01327
Zn	1.82E-05	0.063481	0.044438	2.95E-05		0.001516	0.27309	0.00081	0.135267	0.010222	0.00313
Fe	0.061252	0.828241	0.059541	0.938293	0.001516		0.002816	0.429417	0.489907	6E-07	0.038291
Mn	0.762455	0.008118	0.659438	7.02E-05	0.27309	0.002816		1.07E-07	0.10915	0.000859	0.821549
Mg	0.001961	0.00155	0.531109	2.99E-18	0.00081	0.429417	1.07E-07		0.004041	0.045092	0.067323
Al	0.057521	0.197976	0.034708	0.000237	0.135267	0.489907	0.10915	0.004041		0.259415	0.225788
Na	0.276566	0.264142	0.003782	0.125844	0.010222	6E-07	0.000859	0.045092	0.259415		0.981405
K	0.015696	0.094219	0.306478	0.01327	0.00313	0.038291	0.821549	0.067323	0.225788	0.981405	

St, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.748067	-0.13807	0.540518	0.44585	-0.04985	0.011715	0.323243	0.472777	-0.22023	0.2824774
Cu	0.748067		0.170313	0.386189	0.0594	0.302601	0.38165	0.447838	0.411639	0.301147	0.2720513
Sb	-0.13807	0.170313		-0.11293	-0.0789	0.13713	0.105315	0.118532	-0.1921	0.230648	-0.00244
Ca	0.540518	0.386189	-0.11293		0.381704	0.133348	0.218912	0.808215	0.746048	-0.04182	0.2219265
Zn	0.44585	0.0594	-0.0789	0.381704		-0.45631	-0.32019	0.231378	0.173348	-0.54456	0.2074967
Fe	-0.04985	0.302601	0.13713	0.133348	-0.45631		0.807889	0.444834	0.348145	0.71734	-0.203039
Mn	0.011715	0.38165	0.105315	0.218912	-0.32019	0.807889		0.574399	0.186637	0.643765	-0.189794
Mg	0.323243	0.447838	0.118532	0.808215	0.231378	0.444834	0.574399		0.598182	0.345769	0.1645543
Al	0.472777	0.411639	-0.1921	0.746048	0.173348	0.348145	0.186637	0.598182		0.111825	0.2179272
Na	-0.22023	0.301147	0.230648	-0.04182	-0.54456	0.71734	0.643765	0.345769	0.111825		0.0169083
K	0.282477	0.272051	-0.00244	0.221927	0.207497	-0.20304	-0.18979	0.164554	0.217927	0.016908	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		6.72E-08	0.408444	0.00046	0.005025	0.766299	0.944348	0.047755	0.002723	0.183973	0.0857363
Cu	6.72E-08		0.306639	0.016627	0.723152	0.064805	0.018062	0.004811	0.010237	0.066167	0.0984638
Sb	0.408444	0.306639		0.499628	0.637724	0.411658	0.52917	0.478464	0.247916	0.163565	0.9883998
Ca	0.00046	0.016627	0.499628		0.018045	0.424796	0.186679	8.48E-10	7.62E-08	0.803142	0.1805328
Zn	0.005025	0.723152	0.637724	0.018045		0.003984	0.050019	0.1622	0.297968	0.000408	0.2112902
Fe	0.766299	0.064805	0.411658	0.424796	0.003984		8.72E-10	0.005138	0.032199	4.03E-07	0.2214841
Mn	0.944348	0.018062	0.52917	0.186679	0.050019	8.72E-10		0.000162	0.261875	1.3E-05	0.2537455
Mg	0.047755	0.004811	0.478464	8.48E-10	0.1622	0.005138	0.000162		7.3E-05	0.033475	0.3235217
Al	0.002723	0.010237	0.247916	7.62E-08	0.297968	0.032199	0.261875	7.3E-05		0.503872	0.1887184
Na	0.183973	0.066167	0.163565	0.803142	0.000408	4.03E-07	1.3E-05	0.033475	0.503872		0.9197451
K	0.085736	0.098464	0.9884	0.180533	0.21129	0.221484	0.253746	0.323522	0.188718	0.919745	

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Table 10A Correlation analysis between the element concentrations and pH in the soil water from **Av** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

Av, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.32755	0.328746	-0.0394	0.088524	0.575925	0.395893	-0.04727	0.379985	0.040198	0.231945
Cu	0.327555		0.016557	0.304764	0.524825	0.504019	0.311456	0.065293	0.752187	0.683268	0.482341
Sb	0.328746	0.01656		-0.28087	-0.12554	-0.19551	0.138108	-0.0157	-0.24787	-0.40026	-0.3135
Ca	-0.0394	0.30476	-0.28087		0.290776	0.383554	0.712749	0.810438	0.375982	0.626155	0.796556
Zn	0.088524	0.52482	-0.12554	0.290776		0.407854	0.206964	-0.01391	0.603438	0.557342	0.34609
Fe	0.575925	0.50402	-0.19551	0.383554	0.407854		0.604397	0.110406	0.760645	0.46597	0.631819
Mn	0.395893	0.31146	0.138108	0.712749	0.206964	0.604397		0.550804	0.788372	0.643041	0.819877
Mg	-0.04727	0.06529	-0.0157	0.810438	-0.01391	0.110406	0.550804		0.049473	0.36283	0.669698
Al	0.379985	0.75219	-0.24787	0.375982	0.603438	0.760645	0.788372	0.049473		0.566098	0.596365
Na	0.040198	0.68327	-0.40026	0.626155	0.557342	0.46597	0.643041	0.36283	0.566098		0.693648
K	0.231945	0.48234	-0.3135	0.796556	0.34609	0.631819	0.819877	0.669698	0.596365	0.693648	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.01896	0.018503	0.869011	0.536753	9.78E-06	0.004032	0.843117	0.098415	0.866378	0.325113
Cu	0.018958		0.908194	0.191372	7.7E-05	0.000163	0.026097	0.784477	0.000131	0.000898	0.031252
Sb	0.018503	0.90819		0.230315	0.380062	0.169153	0.333803	0.947627	0.29202	0.08034	0.178313
Ca	0.869011	0.19137	0.230315		0.213593	0.095035	0.000421	1.47E-05	0.102308	0.003141	2.64E-05
Zn	0.536753	7.7E-05	0.380062	0.213593		0.00297	0.14506	0.953598	0.004848	0.010679	0.134981
Fe	9.78E-06	0.00016	0.169153	0.095035	0.00297		2.65E-06	0.643093	9.88E-05	0.038382	0.002805
Mn	0.004032	0.0261	0.333803	0.000421	0.14506	2.65E-06		0.01184	3.64E-05	0.002226	9.66E-06
Mg	0.843117	0.78448	0.947627	1.47E-05	0.953598	0.643093	0.01184		0.835908	0.115887	0.001238
Al	0.098415	0.00013	0.29202	0.102308	0.004848	9.88E-05	3.64E-05	0.835908		0.00927	0.005514
Na	0.866378	0.0009	0.08034	0.003141	0.010679	0.038382	0.002226	0.115887	0.00927		0.000694
K	0.325113	0.03125	0.178313	2.64E-05	0.134981	0.002805	9.66E-06	0.001238	0.005514	0.000694	

Av, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.600097	0.857015	-0.60423	-0.19042	0.360022	0.309353	-0.46986	-0.49403	-0.64872	-0.09297
Cu	0.600097		0.535636	-0.11602	0.268374	0.494497	0.238427	-0.08332	0.122061	0.008746	-0.19
Sb	0.857015	0.535636		-0.6902	-0.2864	0.204158	0.308983	-0.57896	-0.60724	-0.78168	-0.19557
Ca	-0.60423	-0.11602	-0.6902		0.525015	0.355995	0.267322	0.844534	0.605319	0.85848	0.453733
Zn	-0.19042	0.268374	-0.2864	0.525015		0.371442	0.080976	0.388782	0.746129	0.640167	0.077449
Fe	0.360022	0.494497	0.204158	0.355995	0.371442		0.691105	0.089031	0.630476	0.286845	0.178474
Mn	**0.309353**	**0.238427**	**0.308983**	**0.267322**	**0.080976**	0.691105		0.128536	0.315464	0.114757	0.197444
Mg	-0.46986	-0.08332	-0.57896	0.844534	0.388782	0.089031	0.128536		0.348692	0.782343	0.313998
Al	-0.49403	0.122061	-0.60724	0.605319	0.746129	0.630476	0.315464	0.348692		0.748213	-0.06842
Na	-0.64872	0.008746	-0.78168	0.85848	0.640167	0.286845	0.114757	0.782343	0.748213		0.386563
K	-0.09297	-0.19	-0.19557	0.453733	0.077449	0.178474	0.197444	0.313998	-0.06842	0.386563	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		5.18E-06	3.93E-15	0.004778	0.190011	0.011056	0.030546	0.036585	0.026831	0.001973	0.705017
Cu	5.18E-06		7.31E-05	0.626183	0.062253	0.000304	0.098986	0.726927	0.608197	0.970809	0.435931
Sb	3.93E-15	7.31E-05		0.000757	0.046039	0.159403	0.030756	0.007478	0.004519	4.69E-05	0.422328
Ca	0.004778	0.626183	0.000757		0.017457	0.123433	0.254531	2.82E-06	0.004683	1.28E-06	0.051026
Zn	0.190011	0.062253	0.046039	0.017457		0.008596	0.580188	0.09024	0.000158	0.002364	0.752651
Fe	0.011056	0.000304	0.159403	0.123433	0.008596		3.87E-08	0.708953	0.002882	0.220131	0.464756
Mn	0.030546	0.098986	0.030756	0.254531	0.580188	3.87E-08		0.589153	0.175463	0.629976	0.417806
Mg	0.036585	0.726927	0.007478	2.82E-06	0.09024	0.708953	0.589153		0.131876	4.58E-05	0.190475
Al	0.026831	0.608197	0.004519	0.004683	0.000158	0.002882	0.175463	0.131876		0.000148	0.780777
Na	0.001973	0.970809	4.69E-05	1.28E-06	0.002364	0.220131	0.629976	4.58E-05	0.000148		0.102074
K	0.705017	0.435931	0.422328	0.051026	0.752651	0.464756	0.417806	0.190475	0.780777	0.102074	

Av, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.759185	0.817595	-0.32475	-0.18692	0.159755	0.064095	0.324138	-0.50198	-0.38494	0.1698992
Cu	0.759185		0.715674	-0.3303	0.069264	0.150636	0.052255	0.196256	-0.34503	-0.31675	-0.105563
Sb	0.817595	0.715674		-0.3536	-0.22065	0.266619	0.265241	0.201552	-0.34339	-0.48838	-0.047028
Ca	-0.32475	-0.3303	-0.3536		0.400097	0.407266	0.437268	0.524343	0.565789	0.656662	0.6410379
Zn	-0.18692	0.069264	-0.22065	0.400097		0.20471	0.113025	0.22462	0.378706	0.339757	0.2180081
Fe	0.159755	0.150636	0.266619	0.407266	0.20471		0.749191	0.139484	0.659182	0.005275	-0.01649
Mn	0.064095	0.052255	0.265241	0.437268	0.113025	0.749191		0.325358	0.5045	0.09204	0.2187912
Mg	0.324138	0.196256	0.201552	0.524343	0.22462	0.139484	0.325358		-0.16413	0.09554	0.5933703
Al	-0.50198	-0.34503	-0.34339	0.565789	0.378706	0.659182	0.5045	-0.16413		0.368158	0.068685
Na	-0.38494	-0.31675	-0.48838	0.656662	0.339757	0.005275	0.09204	0.09554	0.368158		0.6873831
K	0.169899	-0.10556	-0.04703	0.641038	0.218008	-0.01649	0.218791	0.59337	0.068685	0.687383	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		9.65E-10	4.1E-12	0.162414	0.213539	0.288908	0.672162	0.163248	0.024113	0.093743	0.4739168
Cu	9.65E-10		2.27E-08	0.154936	0.647389	0.317669	0.730176	0.406945	0.136257	0.173619	0.6578117
Sb	4.1E-12	2.27E-08		0.126155	0.140596	0.073276	0.07483	0.394145	0.138259	0.028901	0.8439216
Ca	0.162414	0.154936	0.126155		0.080473	0.074706	0.053861	0.017628	0.009317	0.001661	0.0023211
Zn	0.213539	0.647389	0.140596	0.080473		0.172347	0.454535	0.34105	0.099647	0.142753	0.35582
Fe	0.288908	0.317669	0.073276	0.074706	0.172347		2.11E-09	0.557536	0.001571	0.982392	0.9449885
Mn	0.672162	0.730176	0.07483	0.053861	0.454535	2.11E-09		0.161578	0.023301	0.699545	0.3540518
Mg	0.163248	0.406945	0.394145	0.017628	0.34105	0.557536	0.161578		0.489274	0.68866	0.0058177
Al	0.024113	0.136257	0.138259	0.009317	0.099647	0.001571	0.023301	0.489274		0.110238	0.7735534
Na	0.093743	0.173619	0.028901	0.001661	0.142753	0.982392	0.699545	0.68866	0.110238		0.0008116
K	0.473917	0.657812	0.843922	0.002321	0.35582	0.944989	0.354052	0.005818	0.773553	0.000812	

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Table 11A Correlation analysis between the element concentrations and pH in the soil water from **Ti** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations (p<0.05) are marked in yellow.

Ti, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.16076	0.054802	-0.29778	0.247381	0.211447	-0.54798	-0.19265	-0.06036	-0.28483	0.050116
Cu	0.160759		0.635339	0.248207	0.441279	-0.40795	-0.41799	-0.26881	0.407356	-0.56314	0.674941
Sb	0.054802	0.63534		0.21531	0.170249	-0.14656	-0.33304	-0.43509	0.427097	-0.47426	0.36108
Ca	-0.29778	0.24821	0.21531		0.039315	-0.066	0.490362	0.433205	0.346258	-0.25737	0.364599
Zn	0.247381	0.44128	0.170249	0.039315		0.088591	0.024983	-0.2313	0.390996	-0.53582	0.737567
Fe	0.211447	-0.40795	-0.14656	-0.066	0.088591		0.080897	0.136409	0.179998	0.213914	0.035881
Mn	-0.54798	-0.41799	-0.33304	0.490362	0.024983	0.080897		0.511899	0.001782	0.225713	0.003914
Mg	-0.19265	-0.26881	-0.43509	0.433205	-0.2313	0.136409	0.511899		-0.09968	0.536862	-0.15733
Al	-0.06036	0.40736	0.427097	0.346258	0.390996	0.179998	0.001782	-0.09968		-0.35157	0.457477
Na	-0.28483	-0.56314	-0.47426	-0.25737	-0.53582	0.213914	0.225713	0.536862	-0.35157		-0.48324
K	0.050116	0.67494	0.36108	0.364599	0.737567	0.035881	0.003914	-0.15733	0.457477	-0.48324	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.45301	0.79924	0.157587	0.243839	0.32127	0.00557	0.367118	0.784401	0.177328	0.816105
Cu	0.453006		0.00085	0.242217	0.030881	0.047826	0.042098	0.204037	0.053694	0.004167	0.000297
Sb	0.79924	0.00085		0.312301	0.426409	0.494367	0.111786	0.033598	0.042091	0.019212	0.083002
Ca	0.157587	0.24222	0.312301		0.855276	0.759289	0.014987	0.034461	0.105553	0.224696	0.079828
Zn	0.243839	0.03088	0.426409	0.855276		0.680599	0.90775	0.276835	0.065066	0.006962	3.91E-05
Fe	0.32127	0.04783	0.494367	0.759289	0.680599		0.707086	0.525056	0.411165	0.315526	0.867804
Mn	0.00557	0.0421	0.111786	0.014987	0.90775	0.707086		0.010555	0.993563	0.28892	0.985519
Mg	0.367118	0.20404	0.033598	0.034461	0.276835	0.525056	0.010555		0.650892	0.006832	0.462815
Al	0.784401	0.05369	0.042091	0.105553	0.065066	0.411165	0.993563	0.650892		0.099963	0.028172
Na	0.177328	0.00417	0.019212	0.224696	0.006962	0.315526	0.28892	0.006832	0.099963		0.016751
K	0.816105	0.0003	0.083002	0.079828	3.91E-05	0.867804	0.985519	0.462815	0.028172	0.016751	

Ti, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.787821	0.74187	-0.00382	0.636042	0.238178	0.152922	0.062603	0.236108	-0.0318	0.110041
Cu	0.787821		0.780457	-0.33184	0.513304	0.009582	-0.19207	-0.29417	0.261439	-0.29784	0.144521
Sb	0.74187	0.780457		-0.37097	0.500875	0.092547	-0.22993	-0.35275	0.41829	-0.39439	0.321526
Ca	-0.00382	-0.33184	-0.37097		-0.04758	0.537239	0.66045	0.718924	-0.12839	0.675637	-0.3617
Zn	0.636042	0.513304	0.500875	-0.04758		0.19631	0.011712	0.06748	0.203356	0.133232	-0.01203
Fe	0.238178	0.009582	0.092547	0.537239	0.19631		0.546416	0.498336	0.456842	0.39424	-0.59963
Mn	0.152922	-0.19207	-0.22993	0.66045	0.011712	0.546416		0.83326	-0.04467	0.808454	-0.39357
Mg	0.062603	-0.29417	-0.35275	0.718924	0.06748	0.498336	0.83326		0.07927	0.909959	-0.33913
Al	0.236108	0.261439	0.41829	-0.12839	0.203356	0.456842	-0.04467	0.07927		-0.07454	-0.00414
Na	-0.0318	-0.29784	-0.39439	0.675637	0.133232	0.39424	0.808454	0.909959	-0.07454		-0.35449
K	0.110041	0.144521	0.321526	-0.3617	-0.01203	-0.59963	-0.39357	-0.33913	-0.00414	-0.35449	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		4.88E-06	3.33E-05	0.985855	0.000836	0.262389	0.475611	0.771355	0.266682	0.882725	0.608743
Cu	4.88E-06		6.84E-06	0.113152	0.010308	0.964555	0.368581	0.16293	0.217198	0.157493	0.500465
Sb	3.33E-05	6.84E-06		0.074314	0.012664	0.667117	0.279769	0.090896	0.041936	0.056507	0.125504
Ca	0.985855	0.113152	0.074314		0.825256	0.006786	0.000444	7.56E-05	0.549925	0.000291	0.082436
Zn	0.000836	0.010308	0.012664	0.825256		0.357904	0.956685	0.75406	0.340559	0.534842	0.955527
Fe	0.262389	0.964555	0.667117	0.006786	0.357904		0.005734	0.013196	0.024819	0.056611	0.001955
Mn	0.475611	0.368581	0.279769	0.000444	0.956685	0.005734		4.32E-07	0.835792	1.76E-06	0.057068
Mg	0.771355	0.16293	0.090896	7.56E-05	0.75406	0.013196	4.32E-07		0.712732	7.13E-10	0.104981
Al	0.266682	0.217198	0.041936	0.549925	0.340559	0.024819	0.835792	0.712732		0.729221	0.984696
Na	0.882725	0.157493	0.056507	0.000291	0.534842	0.056611	1.76E-06	7.13E-10	0.729221		0.089199
K	0.608743	0.500465	0.125504	0.082436	0.955527	0.001955	0.057068	0.104981	0.984696	0.089199	

Ti, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.861809	0.586396	-0.03599	0.685567	0.457013	0.477795	0.235565	0.52135	0.068097	0.0241892
Cu	0.861809		0.464582	-0.05375	0.58251	0.158121	0.342712	0.045477	0.263036	0.119541	0.2699206
Sb	0.586396	0.464582		-0.25937	0.736026	0.369836	0.138695	-0.00412	0.790855	-0.13165	0.1627253
Ca	-0.03599	-0.05375	-0.25937		-0.09152	0.20917	0.476707	0.659482	-0.22907	0.492012	-0.329564
Zn	0.685567	0.58251	0.736026	-0.09152		0.56366	0.279863	0.064515	0.739127	-0.02765	-0.117824
Fe	0.457013	0.158121	0.369836	0.20917	0.56366		0.709779	0.653172	0.725669	0.251295	-0.437218
Mn	0.477795	0.342712	0.138695	0.476707	0.279863	0.709779		0.814057	0.243704	0.403218	-0.102156
Mg	0.235565	0.045477	-0.00412	0.659482	0.064515	0.653172	0.814057		0.173551	0.387303	-0.284723
Al	0.52135	0.263036	0.790855	-0.22907	0.739127	0.725669	0.243704	0.173551		-0.08609	-0.204516
Na	0.068097	0.119541	-0.13165	0.492012	-0.02765	0.251295	0.403218	0.387303	-0.08609		0.0642403
K	0.024189	0.269921	0.162725	-0.32956	-0.11782	-0.43722	-0.10216	-0.28472	-0.20452	0.06424	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		2.12E-06	0.00832	0.883718	0.001196	0.049165	0.038549	0.331623	0.022071	0.781781	0.9216987
Cu	2.12E-06		0.045069	0.827	0.008868	0.517937	0.1509	0.853333	0.27659	0.625943	0.2637443
Sb	0.00832	0.045069		0.283586	0.000328	0.119116	0.571216	0.986629	5.55E-05	0.591116	0.5056618
Ca	0.883718	0.827	0.283586		0.709434	0.390109	0.039057	0.002128	0.345504	0.032373	0.1682438
Zn	0.001196	0.008868	0.000328	0.709434		0.011959	0.245865	0.793015	0.0003	0.910532	0.6309469
Fe	0.049165	0.517937	0.119116	0.390109	0.011959		0.000664	0.002427	0.000437	0.299373	0.0612208
Mn	0.038549	0.1509	0.571216	0.039057	0.245865	0.000664		2.22E-05	0.31469	0.08693	0.6773002
Mg	0.331623	0.853333	0.986629	0.002128	0.793015	0.002427	2.22E-05		0.47736	0.101363	0.2374148
Al	0.022071	0.27659	5.55E-05	0.345504	0.0003	0.000437	0.31469	0.47736		0.726019	0.4009791
Na	0.781781	0.625943	0.591116	0.032373	0.910532	0.299373	0.08693	0.101363	0.726019		0.7938793
K	0.921699	0.263744	0.505662	0.168244	0.630947	0.061221	0.6773	0.237415	0.400979	0.793879	

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Supplementary materials,

Table 12A Correlation analysis between the element concentrations and pH in the soil water from **He** collected at 0-15 cm, 15-30 cm and 30-45 cm respectively. The tables show Pearson correlation coefficients and the *p* values of the analysis respectively. Statistical significant correlations ($p<0.05$) are marked in yellow.

He, 0-15 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.68664	0.171587	0.340605	0.306339	0.3753	0.475379	0.258896	0.189752	0.589361	-0.03127
Cu	0.68664		0.141573	0.51561	0.173063	0.132746	0.421429	0.353831	0.090337	0.38784	0.1711
Sb	0.171587	0.14157		0.158175	0.322747	-0.39858	-0.11374	0.131119	-0.32079	-0.20152	0.373151
Ca	0.340605	0.51561	0.158175		0.175973	-0.36953	-0.04835	0.809936	-0.52956	0.51968	0.312356
Zn	0.306339	0.17306	0.322747	0.175973		-0.20154	0.005209	0.199622	-0.39268	0.193861	0.216925
Fe	0.3753	0.13275	-0.39858	-0.36953	-0.20154		0.623901	-0.19526	0.686056	0.282694	-0.49815
Mn	0.475379	0.42143	-0.11374	-0.04835	0.005209	0.623901		0.132478	0.455319	0.33025	-0.18145
Mg	0.258896	0.35383	0.131119	0.809936	0.199622	-0.19526	0.132478		-0.49044	0.451386	0.35501
Al	0.189752	0.09034	-0.32079	-0.52956	-0.39268	0.686056	0.455319	-0.49044		-0.1081	-0.35949
Na	0.589361	0.38784	-0.20152	0.51968	0.193861	0.282694	0.33025	0.451386	-0.1081		0.169956
K	-0.03127	0.1711	0.373151	0.312356	0.216925	-0.49815	-0.18145	0.35501	-0.35949	0.169956	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.00021	0.422728	0.103384	0.145418	0.070739	0.018889	0.221867	0.3745	0.002441	0.884659
Cu	0.000211		0.509335	0.009914	0.418687	0.536346	0.040266	0.089838	0.674637	0.061116	0.424065
Sb	0.422728	0.50933		0.460397	0.123994	0.053708	0.59667	0.541395	0.126419	0.345031	0.072498
Ca	0.103384	0.00991	0.460397		0.410785	0.07554	0.822467	1.62E-06	0.007785	0.009248	0.137277
Zn	0.145418	0.41869	0.123994	0.410785		0.344983	0.980729	0.349687	0.057687	0.364051	0.308598
Fe	0.070739	0.53635	0.053708	0.07554	0.344983		0.001122	0.360527	0.000215	0.180742	0.013236
Mn	0.018889	0.04027	0.59667	0.822467	0.980729	0.001122		0.537174	0.025366	0.115	0.396149
Mg	0.221867	0.08984	0.541395	1.62E-06	0.349687	0.360527	0.537174		0.014968	0.026825	0.088698
Al	0.3745	0.67464	0.126419	0.007785	0.057687	0.000215	0.025366	0.014968		0.615113	0.084464
Na	0.002441	0.06112	0.345031	0.009248	0.364051	0.180742	0.115	0.026825	0.615113		0.427217
K	0.884659	0.42406	0.072498	0.137277	0.308598	0.013236	0.396149	0.088698	0.084464	0.427217	

He, 15-30 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.594432	0.065801	0.351258	0.040735	0.595293	0.497073	0.240548	0.231445	0.541376	0.30567
Cu	0.594432		0.378785	0.351299	-0.06235	0.363016	0.235565	0.073123	-0.00491	0.341536	0.264695
Sb	0.065801	0.378785		0.001503	0.373526	-0.14635	0.015498	0.261445	-0.44605	0.048197	0.486288
Ca	0.351258	0.351299	0.001503		0.201568	0.017927	0.271018	0.655546	-0.24905	0.276661	0.242176
Zn	0.040735	-0.06235	0.373526	0.201568		-0.13577	0.131604	0.402878	-0.17717	0.284682	0.526751
Fe	0.595293	0.363016	-0.14635	0.017927	-0.13577		0.596884	-0.09643	0.573789	0.411578	0.069966
Mn	0.497073	0.235565	0.015498	0.271018	0.131604	0.596884		0.468283	0.138912	0.500883	0.037939
Mg	0.240548	0.073123	0.261445	0.655546	0.402878	-0.09643	0.468283		-0.53645	0.379814	0.498288
Al	0.231445	-0.00491	-0.44605	-0.24905	-0.17717	0.573789	0.138912	-0.53645		0.086682	-0.42276
Na	0.541376	0.341536	0.048197	0.276661	0.284682	0.411578	0.500883	0.379814	0.086682		0.526502
K	0.30567	0.264695	0.486288	0.242176	0.526751	0.069966	0.037939	0.498288	-0.42276	0.526502	

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Supplementary materials,

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.00219	0.759999	0.092364	0.850105	0.002149	0.013467	0.257528	0.276518	0.006294	0.146346
Cu	0.00219		0.067956	0.092323	0.77227	0.081244	0.267817	0.734188	0.981819	0.102384	0.211318
Sb	0.759999	0.067956		0.994439	0.072188	0.495012	0.9427	0.217187	0.028909	0.823036	0.015976
Ca	0.092364	0.092323	0.994439		0.344911	0.933737	0.200205	0.000506	0.240564	0.190628	0.254222
Zn	0.850105	0.77227	0.072188	0.344911		0.527015	0.539889	0.050943	0.407549	0.177564	0.00818
Fe	0.002149	0.081244	0.495012	0.933737	0.527015		0.002076	0.653975	0.003372	0.04569	0.745284
Mn	0.013467	0.267817	0.9427	0.200205	0.539889	0.002076		0.021006	0.517408	0.012663	0.86029
Mg	0.257528	0.734188	0.217187	0.000506	0.050943	0.653975	0.021006		0.006883	0.06715	0.013206
Al	0.276518	0.981819	0.028909	0.240564	0.407549	0.003372	0.517408	0.006883		0.687137	0.039576
Na	0.006294	0.102384	0.823036	0.190628	0.177564	0.04569	0.012663	0.06715	0.687137		0.008216
K	0.146346	0.211318	0.015976	0.254222	0.00818	0.745284	0.86029	0.013206	0.039576	0.008216	

He, 30-45 cm.

Pears co	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.656163	-0.04837	-0.03362	0.367093	0.679688	0.540419	-0.05375	0.433668	0.200986	0.0950467
Cu	0.656163		-0.08996	0.238213	0.289827	0.520854	0.529187	0.069453	0.492172	0.526066	0.4146252
Sb	-0.04837	-0.08996		-0.1732	0.298767	-0.39637	-0.38409	0.047528	-0.32995	0.050243	0.3844117
Ca	-0.03362	0.238213	-0.1732		-0.07689	0.142877	0.332109	0.710649	-0.14785	0.176099	0.126423
Zn	0.367093	0.289827	0.298767	-0.07689		0.198662	0.053952	-0.05522	0.139321	0.23942	0.4204018
Fe	0.679688	0.520854	-0.39637	0.142877	0.198662		0.771538	0.128521	0.279456	0.219735	0.038267
Mn	0.540419	0.529187	-0.38409	0.332109	0.053952	0.771538		0.389598	0.298375	0.247481	0.0795476
Mg	-0.05375	0.069453	0.047528	0.710649	-0.05522	0.128521	0.389598		-0.34436	0.199306	0.3044102
Al	0.433668	0.492172	-0.32995	-0.14785	0.139321	0.279456	0.298375	-0.34436		0.171884	-0.238753
Na	0.200986	0.526066	0.050243	0.176099	0.23942	0.219735	0.247481	0.199306	0.171884		0.7305105
K	0.095047	0.414625	0.384412	0.126423	0.420402	0.038267	0.079548	0.30441	-0.23875	0.730511	

P	Pb	Cu	Sb	Ca	Zn	Fe	Mn	Mg	Al	Na	K
Pb		0.000498	0.822429	0.876076	0.077635	0.000259	0.006405	0.803026	0.034248	0.346335	0.6586505
Cu	0.000498		0.66893	0.251511	0.159926	0.007595	0.006527	0.741487	0.012449	0.006911	0.0393223
Sb	0.822429	0.66893		0.407696	0.146854	0.049808	0.058019	0.821513	0.107232	0.81149	0.0577887
Ca	0.876076	0.251511	0.407696		0.71487	0.49567	0.104814	6.87E-05	0.480615	0.399773	0.5470561
Zn	0.077635	0.159926	0.146854	0.71487		0.341101	0.797845	0.793185	0.506568	0.249045	0.0363958
Fe	0.000259	0.007595	0.049808	0.49567	0.341101		6.31E-06	0.54037	0.176093	0.291246	0.8558923
Mn	0.006405	0.006527	0.058019	0.104814	0.797845	6.31E-06		0.054216	0.147412	0.23298	0.7054491
Mg	0.803026	0.741487	0.821513	6.87E-05	0.793185	0.54037	0.054216		0.091862	0.339507	0.139006
Al	0.034248	0.012449	0.107232	0.480615	0.506568	0.176093	0.147412	0.091862		0.411326	0.2504051
Na	0.346335	0.006911	0.81149	0.399773	0.249045	0.291246	0.23298	0.339507	0.411326		3.379E-05
K	0.658651	0.039322	0.057789	0.547056	0.036396	0.855892	0.705449	0.139006	0.250405	3.38E-05	

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