



# Downwash of atmospherically deposited trace metals in peat and the influence of rainfall intensity: An experimental test



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## HIGHLIGHTS

- 4 rainfall treatments, including pollutant metals, were added to 13 peat cores.
- The depth at which added metals was detected increased in the order  $Pb < Cu < Zn \leq Ni$ .
- Higher precipitation leads to deeper downwashing of metals.
- Downwashing exerts a strong influence on the vertical distribution of metals.

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## ABSTRACT

Accumulation records of pollutant metals in peat have been frequently used to reconstruct past atmospheric deposition rates. While there is good support for peat as a record of relative changes in metal deposition over time, questions remain whether peat archives represent a quantitative or a qualitative record. Several processes can potentially influence the quantitative record of which downwashing is particularly pertinent as it would have a direct influence on how and where atmospherically deposited metals are accumulated in peat. The aim of our study was two-fold: first, to compare and contrast the retention of dissolved Pb, Cu, Zn and Ni in peat cores; and second, to test the influence of different precipitation intensities on the potential downwashing of metals. We applied four 'rainfall' treatments to 13 peat cores over a 3-week period, including both daily (2 or 5.3 mm day<sup>-1</sup>) and event-based additions (37 mm day<sup>-1</sup>, added over 1 h or over a 10 h rain event). Two main trends were apparent: 1) there was a difference in retention of the added dissolved metals in the surface layer (0–2 cm): 21–85% for Pb, 18–63% for Cu, 10–25% for Zn and 10–20% for Ni. 2) For all metals and both peat types (sphagnum lawn and fen), the addition treatments resulted in different downwashing depths, i.e., as the precipitation-addition increased so did the depth at which added metals could be detected. Although the largest fraction of Pb and Cu was retained in the surface layer and the remainder effectively immobilized in the upper peat ( $\leq 10$  cm), there was a smearing effect on the overall retention, where precipitation intensity exerts an influence on the vertical distribution of added trace metals. These results indicate that the relative position of a deposition signal in peat records would be preserved, but it would be quantitatively attenuated.

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## 1. Introduction

Accumulation records of trace metals in peat cores have been used in numerous studies to reconstruct past atmospheric metal deposition rates, not only for lead (Pb) – the most studied pollutant metal – but also for a suite of other pollutant metals, e.g., copper (Cu), nickel (Ni), antimony (Sb) and zinc (Zn) (Shotyk, 1996; Cloy et al., 2005; Pontevedra-Pombal et al., 2013). Based on the general temporal coherence between peat and other natural archives, especially for the record of Pb accumulation (Renberg et al., 2001), there is good

evidence that peat cores are reliable archives for studying relative changes in atmospheric metal deposition over centennial to millennial timescales (Shotyk, 1998; Martinez-Cortizas et al., 1999). Comparisons of the stableisotopic composition of Pb accumulated in peat cores against herbaria samples with known dates of collection (spanning the past 100–150 years) also demonstrate that peat tracks these general changes in deposition over shorter (decadal) timescales (Weiss et al., 1999; Farmer et al., 2002). For other pollutant metals the utility of peat has been justified largely on the co-variation with Pb over time (e.g., Krachler et al., 2003; Cloy et al., 2005). While such studies provide strong support for peat as a record of relative changes in metal deposition over time, questions remain regarding the temporal resolution possible with peat and the extent to which the accumulation records of different metals in peat represent absolute, quantitative

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records rather than only a qualitative record of past deposition (Bindler et al., 2004; Biester et al., 2007).

Some of the key processes that have been discussed as potentially influencing the quantitative record of metal accumulation rates include nutrient cycling (Malmer, 1988), decomposition (Biester et al., 2012) and post-depositional remobilization (Urban et al., 1990), spatial variability of deposition (Bindler et al., 2004), and downwashing (Damman, 1978; Urban et al., 1990; Oldfield et al., 1995; Biester et al., 2007; Hansson et al., 2014b). Downwashing, our main interest here, is particularly pertinent because it would have a direct influence on how and where atmospherically deposited metals are accumulated in the peat record. We define downwashing here as the rapid percolation of elements, deposited on the mire surface via rainfall, through the upper layers of the peat.

Based on a recent study of the natural, short-lived radioisotope beryllium-7 ( $^7\text{Be}$ ;  $t_{1/2} = 53.4$  day) in 5 peat cores from two sites in Sweden (Hansson et al., 2014a), we found that 25–80% of the total inventory was located in the upper 2-cm layer, but that the atmospherically supplied  $^7\text{Be}$  could be detected from 4 cm to as much as 20 cm depth. Given the short-half life for  $^7\text{Be}$ , this required that the  $^7\text{Be}$  was transported recently to the sub-surface peat. Our objective in this study was to further test the importance of downwashing for trace metals in peat, beyond that of  $^7\text{Be}$ , based on a controlled experimental setup. We used a set of 13 peat cores collected from Röd mossamyran, a site we have studied both for radiometric tracers ( $^7\text{Be}$ ,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{241}\text{Am}$ ) (Hansson et al., 2014a,b) and Hg (Rydberg et al., 2010).

Our first aim was to compare and contrast the retention or downwashing of dissolved Pb, Cu, Zn and Ni in peat cores. As noted above, the immobility or mobility of trace metals in peat records is often argued on the basis of similarities or dissimilarities to the distribution of Pb. Previous experimental studies of, e.g., Pb and Be suggest different potentials for downwashing, where 50% of the added Be (Wieder et al., 2010) and 30% of added Pb (Vile et al., 1999) were retained in the top 2-cm layer, with the remainder of the additions distributed downwards towards (Be) or just below (Pb) the water table at 15 cm. However, because these experimental studies were conducted at different times using cores from different sites, it is possible that differences in downwashing between those elements could be related to differences in more basic peat properties (e.g., bulk density, decomposition and hydraulic connectivity). Using a similar experimental design and established approach, we directly compare the retention behavior of four commonly studied pollutant trace metals, Pb, Cu, Ni and Zn within the same cores simultaneously, and use peat cores collected from two contrasting environments on a nutrient-poor mire (Sphagnum lawn and dominant fen section). The four different metals were also chosen to represent a range in known retention from the relatively immobile Pb and Cu to the generally considered more mobile metals Ni and Zn (Bunzl et al., 1976; Kerndorff and Schnitzer, 1980; Berg and Steinnes, 1997; Ceburnis and Valiulis, 1999; Steinnes et al., 2003). We also included the conservative element Ti in our analyses to serve as an immobile checkpoint between cores.

Our second, novel aim was to test the influence of different simulated precipitation intensities on the potential downwashing of metals in peat cores. Rainfall intensities are quite variable over time, and we hypothesized that the amount and intensity of rainfall would influence the extent of downwashing of atmospherically deposited trace metals. We would predict that a low, steady addition would facilitate retention at shallower depths whereas more rapid flushing of rainwater through a peat column during more intense events would reduce the contact time possible between metal cations and the peat substrate, thus increasing the depth at which the deposited metals can be immobilized and retained. Over seasonal and especially annual timescales both precipitation extremes – slow, steady events and extreme rainfall – will occur. In our study a total of 4 ‘rainfall’ treatments were applied to 13 peat cores over a 3-week period, including both daily and event-based applications.

## 2. Material & method

### 2.1. Site description and rainfall history

The peat cores were collected from Röd mossamyran, a small (~7 ha) oligotrophic mire located in Umeå, northern Sweden ( $63^\circ 47' \text{N}$ ,  $20^\circ 20' \text{E}$ ; 40 m a.s.l.). Field and laboratory measured pH values of the peat pore water have been in the range 3.5–4.0. For a more detailed description of the mire, including peat stratigraphy, see Rydberg et al. (2010). Based on climate data from the Swedish Meteorological and Hydrological Institute (SMHI.se) the mean (defined as the period 1961–1990) annual temperature for Umeå is  $3^\circ \text{C}$  and the mean annual precipitation is 600 mm, but annual rainfall has varied from 400 to 1020 mm year $^{-1}$ . The average 24-hr rainfall during 2012 in Umeå was 2.2 mm day $^{-1}$ , but with 187 days of recorded precipitation, the 24-hr average per rainfall day would be 4.2 mm day $^{-1}$ . 10% of the 24-hr rainfall days in 2012 exceeded 10.3 mm day $^{-1}$ , and the highest values were 25 and 28 mm day $^{-1}$  (5 and 2 October 2012, respectively); however, only within the past several years 24-hr rainfall as high as 67 and 119 mm day $^{-1}$  were recorded (7 July 2009 and 27 August 2007, respectively). Rainfall intensities also vary within events; for example, at SMHI's Holmön station 26 km from Umeå, where hourly data are available, the maximum hourly rainfall was 12.7 mm h $^{-1}$  within a 24-hr rainfall event with 63 mm of rain in total (27 August 2007, 12 p.m. to 28 August 2007, 12 p.m.).

### 2.2. Sampling and experimental setup

We collected thirteen surface peat cores from the mire in late June 2013, of which 8 cores were retrieved on the open sphagnum lawn (dominated by *Sphagnum centrale* and *Sphagnum subsecundum*) and 5 cores on the dominant fen area. The main fen area consists of a mixture of *Sphagnum* spp. (*S. centrale*, *S. subsecundum* and *Sphagnum palustre*), shrubs (e.g., *Calluna vulgaris* and *Ledum palustre* L.). Small, scattered pine (*Pinus sylvestris*) also occurs on the fen area but not in close proximity to our coring location. The two sampling sites were located approximately 5 m apart. Using a handsaw sharpened to a knife blade we removed  $10 \times 10 \times 30$  cm deep monoliths, to fit the dimensions of our plexiglas core boxes. Each core was placed into its own core box directly in the field, and care was taken to minimize compaction of the peat during coring and transferring to the core boxes. The cores were then transported back to the laboratory, after which the pre-conditioning commenced immediately, and all cores were stored in the laboratory overnight to ensure that no leakage occurred from the boxes. The cores were then placed outside in a sheltered courtyard and covered by a tent built of see-through plastic with the short ends open to reduce any greenhouse effect. The cores were placed outside to ensure natural conditions of daylight and temperature, and the tent prevented any addition to the core surface other than our additions.

**Table 1**

Recipe per liter stock artificial rain water as described by Wieder et al. (2010). In total 11 L stock synthetic rain was prepared and the pH of the rainwater was adjusted to 4.7 using NaOH and H<sub>2</sub>SO<sub>4</sub> to approximate the precipitation chemistry of the Umeå-region.

Compound	Quantity (1 L)
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	0.5965 g
NH <sub>4</sub> HCO <sub>3</sub>	0.5453 g
Ca(NO <sub>3</sub> ) <sub>2</sub>	0.6973 g
Mg(NO <sub>3</sub> ) <sub>2</sub> · 6H <sub>2</sub> O	0.4105 g
NaNO <sub>3</sub>	0.0764 g
KNO <sub>3</sub>	0.0818 g
NH <sub>4</sub> NO <sub>3</sub>	0.1520 g
NH <sub>4</sub> Cl	0.0588 g
NaOH	0.8586 g
H <sub>2</sub> SO <sub>4</sub>	221 µL

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