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Incorporation of radiometric tracers in peat and implications for estimating accumulation rates

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HIGHLIGHTS

- ^{210}Pb, ^{137}Cs, ^{241}Am and ^{7}Be, and tot-Pb and tot Hg were measured in 5 peat cores.
- Two age–depth models were applied resulting in different accumulation rates.
- The CRS-model overestimated peat mass accumulation.
- The new IP-CRS-model improved mass accumulation rates.
- A downwash component gave accumulation rates in better agreement with monitoring.

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ABSTRACT

Accurate dating of peat accumulation is essential for quantitatively reconstructing past changes in atmospheric metal deposition and carbon burial. By analyzing fallout radionuclides ^{210}Pb, ^{137}Cs, ^{241}Am, and ^{7}Be, and total Pb and Hg in 5 cores from two Swedish peatlands we addressed the consequence of estimating accumulation rates due to downwashing of atmospherically supplied elements within peat. The detection of ^{7}Be down to 18–20 cm for some cores, and the broad vertical distribution of ^{241}Am without a well-defined peak, suggest some downward transport by percolating rainwater and smearing of atmospherically deposited elements in the uppermost peat layers. Application of the CRS age–depth model leads to unrealistic peat mass accumulation rates (400–600 g m^{-2} yr^{-1}), and inaccurate estimates of past Pb and Hg deposition rates and trends, based on comparisons to deposition monitoring data (forest moss biomonitoring and wet deposition). After applying a newly proposed IP-CRS model that assumes a potential downward transport of ^{210}Pb through the uppermost peat layers, recent peat accumulation rates (200–300 g m^{-2} yr^{-1}) comparable to published values were obtained. Furthermore, the rates and temporal trends in Pb and Hg accumulation correspond more closely to monitoring data, although some off-set is still evident. We suggest that downwashing can be successfully traced using ^{7}Be, and if this information is incorporated into age–depth models, better calibration of peat records with monitoring data and better quantitative estimates of peat accumulation and past deposition are possible, although more work is needed to characterize how downwashing may vary between seasons or years.

1. Introduction

For more than a century peatlands have been a valuable archive for studying past environmental changes, not only for the history of bog development (von Post, 1913), but also for the changes in landscape vegetation and climate (Granlund, 1931), carbon cycling (Gorham, 1991), and the atmospheric deposition of trace metals and soil dust (Shotyk, 1998; Kylander et al., 2007). Advancements in peatland research require that we continue to develop our understanding of important processes that influence how environmental signals are incorporated and preserved in this natural archive, and those that influence the precise dating of these records, both on long-term (centennial to millennial) and short-term (annual to decadal) scales.

Effort has been put into improving the tools used to date the long-term record; e.g., determining which organic fraction to radiocarbon date (Nilsson et al., 2001), improving radiocarbon age–depth modeling (Blaauw, 2010), and establishing supporting independent chronological markers such as with lead (Pb) pollution (Renberg et al., 2001). For the more recent record (years to decades) radiometric dating using atmospherically deposited radionuclides, i.e., lead-210 (^{210}Pb), cesium-137 (^{137}Cs), and americium-241 (^{241}Am), has been the main technique (Oldfield et al., 1995). While many studies have validated the general temporal patterns of pollutant metal accumulation based on historical

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usage (Shotyk et al., 1997; Novak et al., 2003) or by the comparability between peat and other archives (Weiss et al., 1999; Farmer et al., 2002), there has been limited critical examination of accumulation records in quantitative terms (Le Roux et al., 2005). To be certain that we extract not only a qualitative record from peat, we must establish a quantitative link between the archive and the few to several decades of data that are now available from contemporary monitoring and research.

Age–depth modeling using ^{210}Pb has been the main tool since the late 1970s for dating recent stratigraphic deposits such as sediments (Appleby and Oldfield, 1978; Appleby, 2008), peat (Aaby et al., 1979; Oldfield et al., 1995), and soil organic horizons (Kaste et al., 2011). Given its half-life of 22.3 years, ^{210}Pb age–depth models provide dating on a 100–150 yr time scale, where the Constant Rate of Supply (CRS) model (Appleby and Oldfield, 1978) is the most frequently applied. While the general applicability of ^{210}Pb dating for peat is well established (Turetsky et al., 2004), refinements in age–depth modeling continue to be developed (Olid et al., 2008; van der Plicht et al., 2013). Critical questions have been raised regarding processes that might affect the accuracy of the modeling such as the mobility of ^{210}Pb in the peat, especially in the uppermost layers (Urban et al., 1990; Mitchell et al., 1992; Oldfield et al., 1995; Lamborg et al., 2002; Biester et al., 2007; Olid et al., 2008).

The short-lived radioisotope beryllium-7 (^{7}Be, $T_{1/2}$ = 53.3 days), which forms high in the atmosphere, is an ideal tracer of rapid downward movement through peat, i.e. downwashing of precipitation, because ^{7}Be, like ^{210}Pb, is delivered to the surface primarily via wet deposition (Wallbrink and Murray, 1996; Whiting et al., 2005). ^{7}Be is rapidly bound to soil and organic particles once deposited (You et al., 1989; Taylor et al., 2012), and thus any ^{7}Be found at depth in the peat must have been rapidly transported to these underlying levels. In the five peat cores from two peatlands studied here, we found that ^{7}Be could be measured to depths of 16 cm in a *Sphagnum* lawn core and 4 cm in an adjacent fen core from an oligotrophic fen, and to depths of 8 to as much as 20 cm in hummock cores (n = 3) from an ombrotrophic bog (Hansson et al., 2014). Experimental studies in the laboratory have indicated similar downwashing profiles based on additions of Pb (Vile et al., 1999), Be (Wieder et al., 2010) and Pb, Cu, Ni, and Zn (Hansson et al., under revision). The distributions of added metals and Be with depth in these three experimental studies were similar to our ^{7}Be data (Hansson et al., 2014 and here), where only 30% (Pb) and 45% (Be) were retained in the top 2 cm with the remaining additions distributed exponentially down to the approximate height of the water table. Combined, these field and experimental studies provide compelling evidence that the retention of atmospherically deposited elements, which should include ^{210}Pb, is not always limited to the top-surface layer in the peat, but that a significant fraction can be downwashed to and immobilized in layers below the surface. This is contrary to a main assumption in age–depth models that ^{210}Pb is immobile.

Further discussion on the downwash process itself can be found in Hansson et al. (2014), but for clarity a brief description of the definition follows. Downwash is the process that allows atmospherically deposited elements to penetrate the peat surface following percolating rainwater thereby reaching further into the peat column before attaching to the organic substrate and being deposited. It is thus not a question of post-deposition mobility but an extension of the actual deposition itself. Downwash should not, however, be equated with 'smearing' that can be caused by both downwash, or post-deposition mobility and hydrology (fluctuating groundwater, evaporation and capillary activity). The ^{7}Be distribution for all cores presented in Hansson et al. (2014), and here, is therefore not a matter of smearing but a direct cause of downwash.

Our main objective here was to test whether downwashing, evidenced previously by ^{7}Be, affects the temporal accuracy of the CRS model in our cores, and thus, the quantitative record of trace metal accumulation rates — and more fundamentally also peat accumulation. Specifically, how might downwashing of the radioisotopes used for age–depth modeling influence our quantitative estimates of peat mass accumulation rates and also accumulation rates of metal pollutants? To test the quantitative accuracy of our accumulation records we compare our data to reported peat mass accumulation rates for similar sites and to deposition data available from monitoring programs in Sweden (forest moss biomonitoring and wet deposition; IVL.se). To do so we apply both a conventional CRS model and the newly proposed IP-CRS model (Initial Penetration — CRS; Olid et al., under revision) which considers vertical transport of ^{210}Pb in the uppermost peat layers.

2. Material & methods

The cores collected and analyzed for this study were assessed previously with regard to ^{7}Be (Hansson et al., 2014). Below is a brief description of the sites and sampling; more detailed information was published in Hansson et al. (2014) and in our previous studies (Bindler et al., 2004; Rydberg et al., 2010).

2.1. Site description and sampling

Store mosse is a large (~8000 ha) ombrotrophic bog in south-central Sweden (57° 15′ N, 13° 55′ E) and Rödmossamyran is a small (~7 ha) oligotrophic fen in northern Sweden (63° 47′ N, 20° 20′ E) close to the city of Umeå. From Store mosse we collected three surface cores (SM1–3) from hummocks in 2008 using a Wardenaar (1987) corer and from Rödmossamyran we collected two monoliths (S1c and F1c) in 2012 using a handsaw sharpened to a knife blade. The Rödmossamyran cores were retrieved from either side of the vegetation boundary separating an open *Sphagnum* lawn (S1c) from the main fen section (F1c). Cores were wrapped in plastic film and aluminum foil, taken back to the laboratory and stored either frozen at −18 °C (SM1–3) or at +4 °C (S1c and F1c) until processing. In a freezer room the outermost 1 cm of the Store mosse cores was removed, the surfaces hand-planed into even dimensions and the cores were cut into 2-cm-thick slices on a band saw with a stainless steel blade. The Rödmossamyran cores were cut into 2-cm slices using the sharpened handsaw the day after sampling. Samples were dried to constant weight at 30 °C (SM1–3 + S1c) or in a freeze drier (F1c), dry masses recorded and bulk density calculated, and finally ground in a ball mill before analysis.

2.2. Analytical methods

^{210}Pb, ^{137}Cs, ^{241}Am and ^{7}Be were measured using a Canberra Broad Energy 5030 (high-purity intrinsic Ge detectors with ultra-low background hardware and copper-lined 1000 kg lead shields). Detection limits were in the range of 2–3.5 Bq kg^{-1} for ^{210}Pb, 1–2 Bq kg^{-1} for ^{137}Cs, 0.25–1 Bq kg^{-1} for ^{241}Am and 2–4.5 Bq kg^{-1} for ^{7}Be based on mass and time counted. To keep counting errors below 8% (4% for ^{210}Pb and ^{137}Cs, and 8% for ^{241}Am and ^{7}Be), samples were counted for 48 h (for near-surface samples) to 100 h (for deeper samples) using 4.5–20 g. All ^{210}Pb measurements were corrected for self-attenuation using the point-source method (Cutshall et al., 1983). The ^{210}Pb activities in the deepest layers were considered as supported ^{210}Pb and subtracted from the total ^{210}Pb activity to determine the unsupported ^{210}Pb used for calculating peat ages. Further details can be found in Kaste et al. (2011).

Total Pb concentrations were measured using a Bruker S8 Tiger wavelength-dispersive X-ray fluorescence (WD-XRF) analyzer equipped with a Rh anticathode X-ray tube. A specific calibration was developed in order to optimize the WD-XRF for the matrix of the samples (see Supplementary information). The detection limit for Pb is 1 ppm, accuracy ±14% and reproducibility ±1.4 ppm.

Total Hg concentrations were analyzed with a thermal decomposition atomic absorption spectrometer (Perkin-Elmer SMS 100). Calibration of the mercury analyzer was done using different masses (10–200 mg) of NIST 1515, NCS ZC 73002 and NCS DC 73323. Analyses of two standard