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Perspectives on using peat records to reconstruct past atmospheric Hg levels

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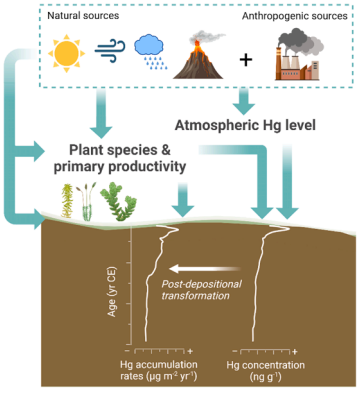
HIGHLIGHTS

- Peat cores can provide information on atmospheric Hg deposition temporally and spatially.
- The overlooked role of vegetational composition is identified in peat Hg accumulation.
- Recommendations are provided to improve the use of peat cores as atmospheric Hg archives.

GRAPHICAL ABSTRACT



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ABSTRACT

Anthropogenic mercury (Hg) emissions to the atmosphere have increased the concentration of this potent neurotoxin in terrestrial and aquatic ecosystems. The magnitude of regional variation in atmospheric Hg pollution levels raises questions about the interactions between natural processes and human activities at local and regional scales that are shaping global atmospheric Hg cycling. Peatlands are potentially valuable and widespread records of past atmospheric Hg levels that could help address these questions. This perspective aims to improve the utility of peatlands as authentic Hg archives by summarizing the processes that could affect Hg cycling in peatlands. We identify the overlooked role of peat vegetation species and their primary productivity in Hg sequestration under climatic and anthropogenic activities. We provide recommendations to improve the reliability of using peat cores to reconstruct the atmospheric Hg levels from past decades to millennia. Better information from peatland archives on regional variation in atmospheric Hg levels will be of value for testing

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hypotheses about the processes controlling global Hg cycling. This information can also contribute to evaluating how well international efforts under the UNEP Minamata Convention are succeeding in reducing atmospheric Hg levels and deposition in different regions.

1. Perspective

Mercury (Hg) is a global pollutant, with approximately 90 % of atmospheric Hg present in the form of gaseous elemental Hg (Hg^0) [1]. It has been recently found that there is a larger degree of regional variation (up to two-fold) in atmospheric Hg pollution levels than previously recognized [2,3]. This variation raises questions about how natural processes and human activities at local and regional scales interact to shape global atmospheric Hg cycling [4,5]. Regionally resolved, long-term Hg records would be of great value to fill in this gap and support the improvement of predictive modeling. Reconstruction of local and regional atmospheric Hg levels needs historical records that are widely distributed, easy to access and provide a direct response to atmospheric input, e.g., peatlands. Peatlands have been used as records of atmospheric Hg deposition to reconstruct background Hg levels and trace anthropogenic Hg pollution history [3,6]. Due to the global distribution of peatlands [7], these archives could be used more extensively to help evaluate the effectiveness of the *Minamata Convention on Mercury*, a global treaty ratified by over 140 countries since 2013, aiming to reduce Hg pollution to protect human health and the environment from potential harm caused by Hg.

The interpretation of peat-derived Hg records is, however, complicated by several processes internal to peatland ecosystems. First, there is the sequestration of Hg by different surface peat vegetation [8]. Second, there is the potential mobilization of Hg at the peat surface and during decomposition [9]. This includes both aerobic degradation of the peat above the groundwater table level which can remove 50 — 90 % of the peat biomass, and slower degradation below the water table. Understanding Hg deposition and post-depositional processes over the long term is a pre-requisite for reliable interpretation of peat Hg records [9]. The ability to understand key aspects of Hg inputs and outputs can be of further value for strategies to mitigate Hg export from peatlands to downstream environments.

The main pathways of atmospheric Hg deposition to peat bogs were clarified only recently with the help of the natural abundance of Hg stable isotopes. These isotopes provide a multi-dimensional fingerprint of Hg for tracing sources and transformation processes [8]. Based on the specific Hg isotope fractionation in the environment, these isotopic fingerprints revealed that 60 — 90 % of Hg is assimilated into the peat as gaseous Hg^0 [8–13]. This is taken up by the vegetation directly from the atmosphere with subsequent oxidation to Hg^{II} by enzymatic reactions or reactive oxygen species (dry deposition) [14]. The remainder is the deposition of oxidized Hg^{II} via precipitation and fog (wet deposition). Despite the important role of vegetation in Hg^0 sequestration, it has not yet been sufficiently recognized that Hg assimilation by vegetation greatly varies with plant species across seemingly uniform peatland landscapes (i.e. open peatlands without tree cover) [6].

Different plant species have different capacities to assimilate Hg^0, resulting in systematic differences in Hg concentrations of plant tissues between different species on the same peatland, even if the life forms appear similar (e.g. ground vegetation) [9,15]. For instance, Hg concentrations in sedge *Eriophorum vaginatum* are less than half of those in *Sphagnum* moss [9,15]. Furthermore, significantly different Hg concentrations are found among *Sphagnum* species (*Sphagnum subsecundum* vs *Sphagnum centrale* [15]; *Sphagnum fuscum* vs *Sphagnum balticum* [9]). Accordingly, if the dominant peatland plant community shifts from sedge to *Sphagnum* or from one *Sphagnum* species to another, a change in peat Hg concentration will likely be recorded due to the shift in the capacity of Hg^0 uptake by different peatland vegetation. This complicates the ability to relate Hg accumulated in peat to past atmospheric Hg concentrations at the time of peat formation.

Fortunately, the botanical evidence of surface plant species coupled with the downcore macrofossil assemblages enable reconstruction of the long-term vegetation composition. Macrofossil assemblages can be identified using a combination of low (magnification range ×10 – ×50) and high power microscopy (magnification range ×100 – ×600), based upon specimens of contemporary reference material [16–18]. When integrating the botanical evidence with Hg measurements in different surface plant species, one might be able to identify significant shifts in downcore peat Hg accumulation linked to vegetation changes. Changes in vegetation generally refer to the shift in average vegetational regime over the course of decades [16]. Such changes can be reflected in the potentially fluctuating peat Hg accumulation at least in the pre-industrial period with relatively stable atmospheric Hg levels. During industrial times with higher, but also more dynamic levels, the time-scale for vegetational changes impacting Hg accumulation could be shorter. Combined with a well-established peat chronology [9], it becomes possible to determine whether a shift in Hg accumulation rate is due to a change in atmospheric Hg level or a change in vegetation composition. This approach has the potential to give insight into Hg accumulation dynamics in peatlands under both natural conditions and anthropogenic pollution pressures. This information will improve the usefulness of peat as an archive for spatially and temporally resolved reconstructions of atmospheric Hg.

Climate variability, anthropogenic changes in the hydrological regimes of peatlands, or species succession through hummock and hollow phases at the peat surface can trigger a shift in the plant community [19], and hence the ability to sequester Hg^0 (Fig. 1). The plant Hg^0 sequestration capacity can be assessed by analyzing Hg concentrations and investigating biomass production in the surface vegetation species [9].

Changing climate and atmospheric circulation patterns can regulate the transport and deposition of aerosols [20], altering the input of both natural and anthropogenic nutrients to peatlands. Varying nutrient inputs affect primary productivity, which can also be altered by synoptic weather systems through their influence on temperature and humidity (precipitation: evaporation ratio). Changes in primary productivity also imply changes in total Hg accumulation rates, calculated by peat Hg concentration and accumulation rates.

The conversion of Hg concentrations into Hg accumulation rates solves the potential bias due to varying peat decomposition degrees (Fig. 1). Peat can lose up to 90 % of its initial organic matter as it decays [21], depending on redox (aerobic or anaerobic) conditions and residence time above the water table. This results in variations of Hg concentrations condensed in a given peat layer assuming no significant Hg mobility. Yet, efforts are needed to explore how the dominant plant species coupled with local environmental conditions influence Hg mobility over the long term, including decadal scale monitoring of the gaseous Hg^0 exchange flux between peat vegetation surfaces and the atmosphere. Hg mobility in peat [9,22–24] includes potential photochemical, biotic, and dark abiotic reactions (e.g., oxidized Hg^{II} being reduced to Hg^0 and/or converted into methyl-Hg), as well as adsorption-desorption under groundwater level fluctuations. The impact of vegetation composition on Hg^0 sequestration and the potential mobility of Hg during post-depositional processes also make peat a complex archive of atmospheric Hg deposition. However, plant macrofossils and Hg isotopic signatures [8,9] can possibly resolve peat Hg profiles into quantitative records of atmospheric Hg. Such information, combined with the widespread distribution of peatlands (Fig. 2), will be of great scientific value when investigating temporal and spatial Hg

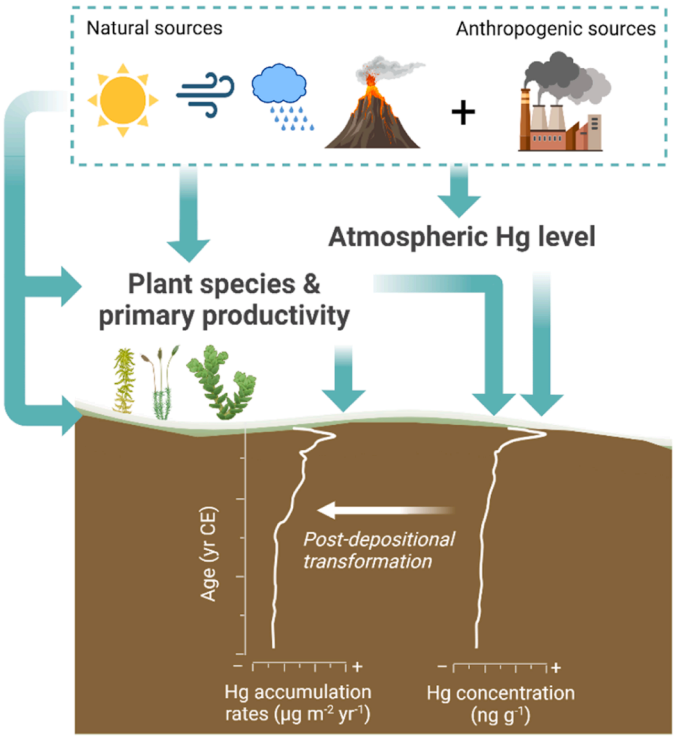

Fig. 1. The conceptual diagram shows the potential influence of natural and anthropogenic sources on plant species, primary productivity, and atmospheric Hg levels, hence Hg deposition to peatlands. Plant species, primary productivity, and atmospheric Hg level can affect initial Hg input (i.e. Hg concentration) [3,11]. During decomposition, Hg will be condensed into certain peat layers with vegetation composition involved (i.e. Hg accumulation rates) [3, 11]. Natural and anthropogenic drivers can also affect post-depositional Hg transformation processes, e.g., photochemical reaction, dark abiotic reaction, and peatland drainage. The peaks in both Hg accumulation rates and concentration profiles refer to net Hg deposition during the industrial period.

variability in the atmosphere. That is because regionally resolved Hg archives will help reveal relationships between global and local anthropogenic activities, as well as the effects of climate dynamics both directly (e.g. precipitation, temperature) or indirectly (e.g. plant species, primary productivity).

There has been marked progress in elucidating the dominant pathways of Hg deposition [8,9] and potential post-depositional mobility of Hg in peatlands [9,22]. Nevertheless, the critical roles of vegetation species and primary productivity under changing climate during the initial sequestration of atmospheric Hg need more recognition. This will enable an in-depth understanding of Hg inputs to peatlands and a better comparison of Hg accumulation within and between nearby peatlands. We suggest more investigation of surface vegetation composition and downcore plant macrofossils for better reconstruction of Hg deposition to peatlands as well as for understanding the fate of Hg in aquatic ecosystems where peatlands are present upstream. This is important for exploring both past Hg deposition and how regional differences in Hg deposition will develop in the coming decades if efforts to control emissions succeed in reducing atmospheric Hg levels.

CRediT authorship contribution statement

Chuxian Li: Writing – original draft & review & editing, Visualization, Investigation, Conceptualization, Funding acquisition. **Maxime Enrico:** Writing – review & editing, Visualization, Validation, Conceptualization. **Kevin Bishop:** Writing – review & editing, Validation, Conceptualization. **Stephen J. Roberts:** Writing – review & editing, Conceptualization. **Dominic A. Hodgson:** Writing – review & editing, Conceptualization. Mariusz Lamentowicz: Writing – review & editing, Conceptualization. **Dmitri Mauquoy:** Writing – review & editing, Conceptualization. **Adrien Mestrot:** Writing – review & editing, Conceptualization. **Martin Grosjean:** Writing – review & editing, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

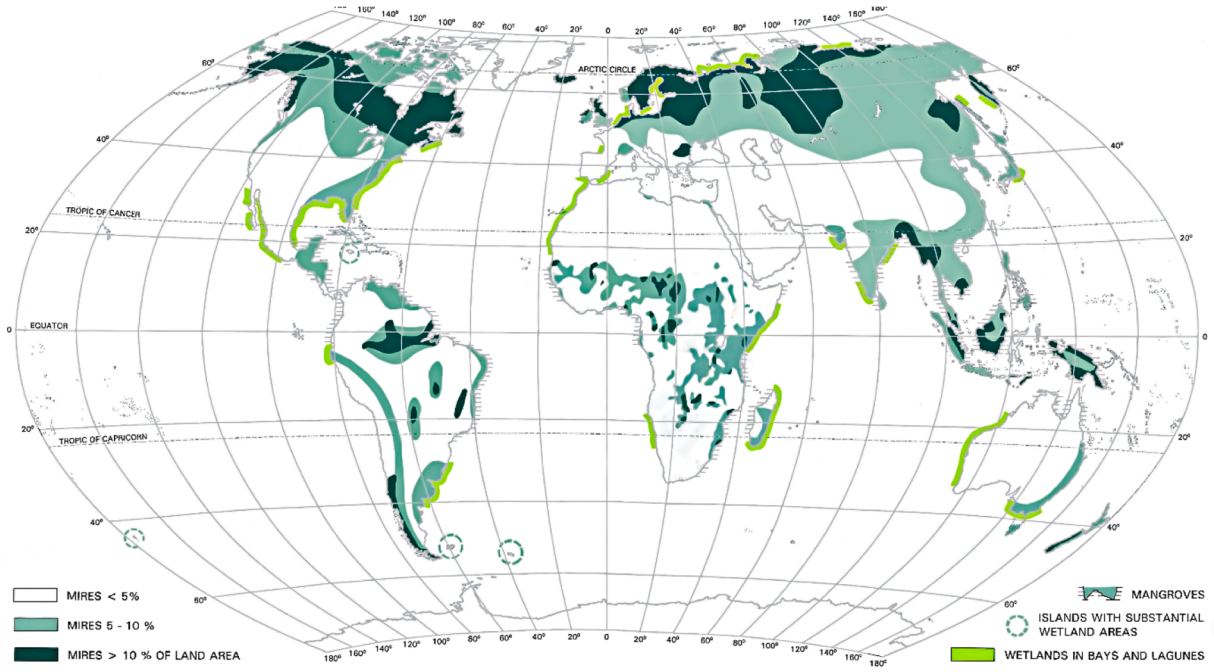

Fig. 2. Global distribution of peatlands (mires). Adapted from Lappalainen, (1996) [7], with permission to use from the *International Peat Society*.

the work reported in this paper.

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Data Availability

No data was used for the research described in the article.

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