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Modelling and mapping trace element accumulation in *Sphagnum* peatlands at the European scale using a geomatic model of pollutant emissions dispersion[☆]



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ABSTRACT

Trace elements (TEs) transported by atmospheric fluxes can negatively impact isolated ecosystems. Modelling based on moss-borne TE accumulation makes tracking TE deposition in remote areas without monitoring stations possible. Using a single moss species from ombrotrophic hummock peatlands reinforces estimate quality. This study used a validated geomatic model of particulate matter dispersion to identify the origin of Cd, Zn, Pb and Cu accumulated in *Sphagnum capillifolium* and the distance transported from their emission sources. The residential and industrial sectors of particulate matter emissions showed the highest correlations with the TEs accumulated in *S. capillifolium* (0.28(Zn)-0.56(Cu)) and (0.27(Zn)-0.47(Cu), respectively). Distances of dispersion varied depending on the sector of emissions and the considered TE. The greatest transportation distances for mean emissions values were found in the industrial (10.6 km when correlating with all TEs) and roads sectors (13 km when correlating with Pb). The residential sector showed the shortest distances (3.6 km when correlating with Cu, Cd, and Zn). The model presented here is a new tool for evaluating the efficacy of air pollution abatement policies in non-monitored areas and provides high-resolution (200 × 200 m) maps of TE accumulation that make it possible to survey the potential impacts of TEs on isolated ecosystems.

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1. Introduction

Trace elements (TEs) in the form of particulate matter (PM) can be transported long distances by atmospheric fluxes and deposited up to thousands of kilometres from the emission sources (Keating et al., 2007; NRC, 2009; Wagstrom and Pandis, 2011). Deposition of the TEs associated with PM on terrestrial and water surfaces can have negative impacts on ecosystem functionality due to TEs' toxicity to living organisms and their capacity to bioaccumulate throughout the food chain (Harmens et al., 2008). Although there is a particular interest in surveying TE dry and wet deposition in the

interest of protecting ecosystem functionality, monitoring TEs associated with PM in isolated areas and ecosystems remains difficult due to the lack of air quality monitoring stations, which are usually situated near populated zones because of the need to survey the impact of pollution on human health (Diaz-de-Quijano et al., 2014).

Both dry and wet deposition of TEs associated with PM in isolated areas can be effectively tracked by monitoring their accumulation on mosses (Agnan et al., 2013; Harmens et al., 2010). Ectohydric carpet-forming bryophytes have been widely used worldwide for the last 30 years to monitor the atmospheric deposition of TEs because they are distributed across a variety of ecosystems and obtain most of their nutrients directly from wet and dry deposition, with little uptake from soil (Holy et al., 2009; Onianwa, 2001). Moreover, bryophytes provide other advantages in TE deposition monitoring: they intercept pollutants that accumulate gradually over long periods of time, which is useful in the

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case of low depositions, and they permit higher sampling densities than conventional deposition measurements because of the low collection cost (Gusev et al., 2009).

TE analyses like the one presented here, conducted on a single moss species such as *Sphagnum capillifolium* from a relatively uniform ecosystem, should eliminate the ambiguities in TE concentrations that arise when different moss species are used indiscriminately. Indeed, significant differences have been found in the accumulation of several TEs when using different moss species (Fabure et al., 2010; Gombert et al., 2004). Ombrotrophic *Sphagnum* peatlands are characterized by a set of relatively similar conditions: waterlogging, anoxic and acidic water, low nutrient contents, and low temperatures (Rydin and Jeglum, 2006). In addition, the bumpy surface of hummock peatlands isolates *Sphagnum* from underground water and the nutrients dissolved in it, thus preventing the *Sphagnum* from absorbing from the substrate. For the quantification of total atmospheric deposition, *Sphagnum* may thus be an alternative to terrestrial bryophytes growing in a variety of ecosystems, which are problematic because they take up components from the substrate (Coskun et al., 2009).

Furthermore, the geographical distribution of *Sphagnum* peatlands all over Europe (Séneca and Söderström, 2009) makes this species of great utility to the study of atmospheric deposition at the European scale. Measurements derived directly from the accumulation of TEs on living mosses provide a complement to the conventional physicochemical techniques for acquiring depositional data (Aboal et al., 2010; Berg et al., 2003), facilitating future studies of the effects of TE deposition on ecosystem functionality. It is thus essential to obtain *Sphagnum* TE accumulation data as well as information on these TEs' emission sources and approximate distances traveled prior to deposition in order to improve the management and protection of isolated ecosystems far from the monitoring station network.

Consequently, the objective of this study is to identify the origin of TEs accumulated in *S. capillifolium* growing in peatlands in relation to the main emission sectors (industry, residential, roads, and agriculture) and the distance of transportation from emission sources. To achieve this, TE accumulation data from fifty-four isolated peatlands over an area of 160,000 km² in Central-Eastern France was applied to a previously validated geomatic model that describes the dispersion of TEs bound to PM10 (that is, PM with an aerodynamic diameter $\leq 10 \mu\text{m}$) according to the four main emissions sectors at the European scale (Diaz-de-Quijano et al., 2014). The application of this geomatic model produces new data in the form of TE accumulation maps, which could be used to evaluate policies for the reduction of pollutant emissions.

2. Materials and methods

2.1. Area of investigation and peatland sampling sites

The study area including the peatland zone and emission sources comprised a 653,600 km² square in the centre of Europe (Fig. 1, coordinates in RGF 1993 Lambert 93 are N: 7 069 780, W: 453 053, E: 1 213 053, S: 6 213 780). In the peatland zone (a square of 160,000 km² in the middle of the study area), 54 ombrotrophic peatlands were sampled across five mountain ranges of north-eastern France (Vosges, Jura, Morvan, Alps and Massif Central) at altitudes from 200 m to 2035 m (Fig. 1). The sampled peatlands were far from major pollution sources: on average 49 ± 31 km from any fixed pollution sources (industrial and urban) and 18 ± 10 km from any traffic sources (highway). Peatland selection is further described in Meyer et al. (2015).

2.2. Trace element concentrations in *Sphagnum capillifolium*

The study used concentrations of accumulated Cd, Zn, Pb and Cu in *S. capillifolium*, corresponding to a year's worth of total (dry and wet) atmospheric deposition accrued prior to September 2010. These four TEs were chosen because they represent atmospheric deposition from predominantly anthropogenic sources, as demonstrated by the elevated enrichment factors found by Meyer et al. (2015) in the same peatlands. Details of the sampling method and preparation for the analysis are thoroughly described in Meyer et al. (2015).

2.3. Emissions of trace elements associated to PM10

Maps of diffuse PM10-bound TE emissions for the year 2008 (latest year available) were obtained from the European Pollutant Release and Transfer Register (E-PRTR) ("Diffuse Air Releases under the E-PRTR," 2011). The maps depicted the total annual PM10 emissions for each main sector of activity (industrial, residential, road transport and agriculture) in tonnes per 5 by 5 km cells, which were resampled to express emissions in kg per 200 by 200 m cell, as in Diaz-de-Quijano et al. (2014); (a summary of emissions is in Table 1); the percentage of emitting pixels is 22, 97, 96 and 95% for the industrial, residential, roads, and agriculture sectors, respectively. For detailed information on the methodology used to estimate national PM10 emissions for each sector and the resulting spatial distribution, see the air pollutant emission inventory guidebook developed by the European Monitoring and Evaluation Programme (EMEP) (EEA, 2013) for the former, and the methodology developed for the spatial distribution of diffuse emissions in Europe (Theloke et al., 2011) for the latter. Our study treated each pixel in each of the E-PRTR's 4 PM10 emissions estimate maps as an emission source of the TEs associated with the PM10 emitted in the study area in 2008.

2.4. Dispersion of trace elements using a geomatic model

We used a previously published model of PM10 dispersion (Diaz-de-Quijano et al., 2014) to identify the main sectors emitting the TEs accumulated in *S. capillifolium* and the optimal distances of transportation from the emission sources to the peatlands. The model, which uses Euclidian distance and emissions weighted by a dilatation factor (F), represents the probability of the dissemination of PM10-borne TEs around the emission sources. Hence, the dispersion of PM10 and its associated TEs around a given pixel depends on the quantity of PM10 emitted by the pixel and on the applied F, which increases emissions levels, expressed in kg. This procedure expands the area of dispersion, the quantities dispersed declining linearly with distance (see Supplementary Information (SI) 1 for further explanation). Eight increasing dilatation factors (from 25 to 1500) were used to obtain different simulations of dispersion distances. The dilatation factors make it possible to represent the theoretical distances the PM10 and associated TEs may be transported around the emitting pixels. Table 1 presents PM10 dispersion distances based on the application of eight different Fs to several PM10 emission values in each sector. Further details on the geomatic model can be found in Diaz-de-Quijano et al. (2014).

2.5. Response of the accumulated trace elements according to emission sector after applying the dilatation factors, and prediction of trace element accumulation in *S. capillifolium*

In order to determine which emissions sectors and dilatation factors are most appropriate for explaining the accumulation of

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