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Climate-driven enrichment of pollutants in peatlands

A. Martínez Cortizas[1], H. Biester[2], T. Mighall[3], and R. Bindler[4]

[1]Dept. Edafología y Química Agrícola, Universidad de Santiago de Compostela, Campus Universitario s/n, 15782 Santiago de Compostela, Spain
[2]Institute of Environmental Geochemistry, University of Heidelberg, INF 236 69120 Heidelberg, Germany
[3]Department of Geography and Environment, School of Geosciences, University of Aberdeen, Elphinstone Road, Aberdeen, AB24 3UF, UK
[4]Dept. of Ecology and Environmental Science, Umeå University, 901 87 Umeå, Sweden

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Correspondence to: A. Martínez Cortizas (edantxon@usc-.es)

Abstract

Peatlands play an important role for global carbon dynamics, acting as a sink or source depending on climate. Such changes imply a series of additional effects because peatlands are also an important reservoir of atmospherically derived pollutants. Using a multiproxy approach (non-pollen-palynomorphs, δ^{15}N, C/N, Se, Br, I, Hg, Ti), we show a relationship between climate (wetter-drier) and peat decomposition, which affected element concentrations in a Spanish bog during the last 5500 years. Changes in superficial wetness played a critical role in the cycling of elements coupled to carbon dynamics. Dry phases caused increased peat mineralisation, resulting in a 2–3 times increase in concentrations of the analysed elements independent from atmospheric fluxes. Under the present trend of climate change large areas of northern peatlands are expected to be severely affected; in this context our findings indicate that the increase in carbon release, which leads to an enrichment of elements, may enhance the export of stored contaminants (Hg, organohalogens) to the aquatic systems or to the atmosphere.

1 Introduction

Peatlands cover about 5–8% of the earth's continental surface and contain one third of the total soil carbon stock (IPCC, 2000), playing an important role in the carbon cycle. Depending on climatic conditions they may act as sinks or sources of carbon and other elements. It has been estimated that at present the sink component is dominant, but recent modelling studies for boreal areas suggest that 59% of all peatlands area in Canada and a similar proportion in North Eurasia will be severely to extremely severely affected by climate change (Gorham, 1991; Tarnocai and Stolbovoy, 2006), leading to a higher release of carbon dioxide and methane to the atmosphere.

Although the predicted increase in temperature may enhance peat decomposition, the changes in hydrology coupled to changes in precipitation patterns and permafrost

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thawing are expected to have the highest impact (Tarnocai and Stolbovoy, 2006). In peatlands organic matter turnover is highest in the upper aerated layers, where more than 80% of the initial plant remains are mineralised and released as CO_2 or dissolved organic matter (DOM) (Worral et al., 2001; Reynolds and Fenner, 2001; Freeman et al., 2004). In the lower sections the decay rate decreases by two or three orders of magnitude due to the anoxic conditions and usually low pH, as well as by the inhibitory effect of the accumulation of some chemical substances like phenols (Kalviäinen and Karunen, 1984; Freeman et al., 2001). Although a slight release of carbon continues, the peat in the anoxic layers suffers only minor further degradation.

The cycle of biophyle elements (C, N, P and S) in peatlands has been the target of many studies (Ohlson, 1987; Ohlson and Okland, 2002), but not much is known about the cycle of other elements, particularly those of environmental concern such as toxic heavy metals like mercury. Elements are incorporated to peat by wet and dry atmospheric deposition (the dominant pathway in ombrotrophic mires, by definition), as well as by groundwater and surface runoff (in minerogenic mires). Once incorporated as solutes or particles, elements can be retained through plant uptake, physical entrainment, complexation by humified organic matter, or through enzymatic microbial mediated formation of organic species. Some elements can also be re-released to the atmosphere through evasion or to the hydrosphere in solution or coupled to DOM release. But the impact of present climate change on the accumulation/release of elements from peatlands to the atmosphere and aquatic ecosystems is still largely uncertain.

Heavy metals, particularly Pb and Hg, have been extensively investigated in peat cores to reconstruct long-term variations of their fluxes to the mires and thus to trace atmospheric pollution (Lee and Tallis, 1973; Shotyk, 1998; Martínez Cortizas et al., 2002). These investigations have rarely considered the role played by climate in the control of the deposition fluxes or the modification of element concentrations induced by changes in peat degradation.

In this study we compare climate variations since the middle Holocene (~5500 yrs) observed in a Spanish peatland with records of organically bound (e.g., Se, Br, I, Hg)

and lithogenic (Ti) elements. We aim to show that the geochemistry of peat is strongly related to climatically controlled carbon dynamics and that the geochemical record in peat is sensitive to Holocene climate changes. Our findings for past climatic changes, contextualized with present scenarios of climate change, may enable a better understanding of the role peatlands play in the global biogeochemical cycles of elements (nutrients and pollutants).

2 Material and methods

We investigated a peat core from Pena da Cadela (PDC), an oceanic ombrotrophic mire located on a mountain summit at 970 m of altitude in the Xistral Mountains (NW Spain), for historical records of trace elements (mercury, bromine, iodine, selenium, and titanium) and peat decomposition (C and N concentrations, C/N ratios and δ^{15}N). The core is 185 cm deep and has accumulated during the last 5500 cal. years BP. Sampling of the core, sample preparation, the ombrotrophic nature of this mire, as well as the records of some elements, such as Pb, are discussed elsewhere (Martínez Cortizas et al., 2002). Bromine, selenium and titanium, were measured using an energy dispersive miniprobe multielement XRF analyzer – its application to peat measurements is discussed in (Cheburkin and Shotyk, 1998). Mercury was determined in freeze dried samples by means of solid-phase thermal combustion atomic absorption spectrometry (LECO-Altec AMA-254); while iodine was determined following the procedure described by Biester et al. (2004). Carbon and nitrogen were analysed by means of a LECO-10000 CNH. δ^{15}N was obtained by mass spectrometry. Distribution of non-pollen palynomorphs (NPPs) and their relationship to climate in the same peat samples were discussed elsewhere (Mighall et al., 2006).

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3 Results

3.1 A link to climate change

Figure 1a shows the records of the non-pollen palynomorphs (NPPs) T18 and T306, and δ^{15}N and C/N ratios in the PDC core. The NPPs records show intense changes during the last 5,500 years, which indicate alternating wet (high T18 and low T306 abundances) and dry (low T18 and high T306 abundances) climate periods. There are five main wet phases in this peat record at 150–160 cm, 130–138 cm, 114–126 cm, 84–100 cm and 32–64 cm, which correspond to 3950–3590, 3090–2790, 2580–2260, 1760–1430 and 1030–530 yr cal BP respectively. The duration of each of these wet phases was about 300-360 years except for the most recent one that may have lasted 500 years.

In order to extract information on climatic effects on peat decomposition it is necessary to remove the secondary changes imposed by the gradual, long-term diagenetic effects that occur in the buried peat. For example, there is a general increase in the C/N ratio from around 20 in the surface peat to 37 at the base of the PDC core that is caused by the preferential mineralization of N relative to C during organic matter diagenesis, which has been documented in other studies for a number of sedimentary environments (Herczeg, 1988; Updegraff et al., 1995; Khury and De Vitt, 1996). In Fig. 1a we have detrended both the C/N ratio and δ^{15}N records and expressed them as standardized residuals ($\delta^{15}N_{detrend}$ and $C/N_{detrend}$, respectively). $\delta^{15}N_{detrend}$ and $C/N_{detrend}$ have a generally inverse behaviour with higher $\delta^{15}N_{detrend}$ values corresponding to relatively lower $C/N_{detrend}$. Low $C/N_{detrend}$ values are indicative of more decomposed and humified peat, while higher δ^{15}N values are indicative of higher N fractionation. The decomposition of the organic matter preferentially mineralizes ^{14}N, causing an enrichment in ^{15}N and thus in δ^{15}N (Létolle, 1980; Macko et al., 1993).

The variations in $\delta^{15}N_{detrend}$ and $C/N_{detrend}$ coincide with the changes in climate (wetter–drier) as indicated by the biological climate proxies (NPPs). Wetter phases are

characterized by positive $C/N_{detrend}$ values and low $\delta^{15}N_{detrend}$ values, whereas drier phases show the opposite – the decline in C/N ratios and increase in N fractionation are indicative of peat decomposition/mineralisation. In fact, the degree of humification of the peat was found to be a proxy for wet/dry cycles (Blackford and Chambers, 1993; Caseldine et al., 2000; Roos-Barraclough et al., 2004). Under saturated conditions oxygen availability in the peat is reduced, which inhibits the phenoloxidase enzyme activity controlling phenol accumulation (Freeman et al., 2001). Increased phenol accumulation under saturated conditions restricts the biological degradation of the organic matter; specifically, the elevated phenol concentrations restrain the activity of hydrolitic enzymes responsible for peat decomposition. In contrast, during dry periods when the water table is lower and formerly saturated peat levels are aerated, enzyme activity increases and promotes peat degradation. In ombrotrophic mires oxygen availability is connected to water table depth, which in turn depends on effective precipitation and thus on changes to wetter or cooler climates (Aaby, 1976; Barber, 1981; Chambers and Charman, 2004; Mauquoy et al., 2002).

3.2 Variations in elements concentrations

The concentration records of the organically bound (Br, I, Se and Hg) and lithogenic (Ti) elements are quite similar (Fig. 1b), with the only exception of Hg in the upper peat sections. The concentrations of bromine, selenium and titanium in the PDC core vary by 2.5–3 times (Br: 120–350 $\mu g\,g^{-1}$; Se: 0.9–2.2 $\mu g\,g^{-1}$; Ti: 100–250 $\mu g\,g^{-1}$), while mercury and iodine concentrations vary by 12–17 times (Hg: 19–230 $ng\,g^{-1}$; I: 3–50 $\mu g\,g^{-1}$). Below 36 cm the variation in mercury concentration (3.5-fold) is comparable to those of bromine and selenium, suggesting that mercury must be influenced by anthropogenic emissions in the upper section of the peat core as found in other peat records in the area (Martínez Cortizas et al., 1999). Notwithstanding this exception, the records of the analyzed elements show a remarkable similarity. The ocean is the main source of atmospheric selenium, bromine and iodine (Kabata-Pendias and Pendias,

1992; Görres and Frenzel, 1993), which are mostly incorporated into the bog through wet deposition. For example, concentrations of bromine in peat and pore waters have been used to trace differences in sea spray inputs (Shotyk, 1997), thus assuming a direct relationship with atmospheric fluxes – mainly wet deposition. Mercury deposition is also dominated by the wet component, and the oceans contribute to mercury emissions to the atmosphere, although there is an additional strong terrestrial source (weathering and degassing of the Earth's crust) (Schroeder and Munthe, 1998; Scholtz et al., 2003). Hg^0, the predominant form in the atmosphere, has a mean residence time of one year and it is subject to long-range transport (Mason et al., 1994; Nriagu y Pacyna, 1988; Schroeder and Munthe, 1998; Scholtz et al., 2003).

The similarity between the records of the organically-bound elements suggests a common source or deposition mechanism. However, titanium, a lithogenic element whose source is atmospheric dust deposition, also shows remarkably synchronous changes in concentration. Since the deposition of atmospheric aerosols increases in dry periods and decreases during wet periods (Mouline et al., 1997; Prospero and Lamb, 2003), the simultaneous increase in the concentration of organically bound and lithogenic elements is counterintuitive and has to be influenced by another factor than atmospheric deposition alone.

The comparison of the concentration records of selenium, bromine, iodine, mercury and titanium (Fig. 1b) with the climatic reconstruction reveals that, in the PDC core, elevated concentrations of these elements are found in sections corresponding to drier climate periods with greater humification (more positive $\delta^{15}N_{detrend}$ values and lower $C/N_{detrend}$ values), whereas lower concentrations correspond to wetter periods with less degraded peat (low $\delta^{15}N$ values and high C/N ratios). The elevated element concentrations are not the result of increased wet deposition, but instead indicate a relationship between the degree of peat transformation, i.e., decomposition, and the variations in the concentration of the elements. We do find a strong correlation between carbon accumulation and the accumulation of organically bound elements. The correlations between the net accumulation of carbon and selenium, bromine, iodine and mercury

(excluding the top 30 cm impacted by pollution) are highly significant (r^2=0.9; data not shown). For the organically bound elements this is a consistent result, because selenium, bromine, iodine and mercury all have a high affinity for binding to organic matter and are therefore usually enriched in organic-rich soils (Kabata-Pendias and Pendias, 1992; Görres et al., 1993; Biester et al., 2003). Selenium and mercury can form complexes with organic matter by abiotic mechanisms (Zhang and Moore, 1996; Xia et al., 1999), particularly when specific functional groups are availabe (e.g. S-groups for Hg). Bromine and iodine are retained in organic matter through formation of organohalogens by oxygen-dependent enzymatic processes involving haloperoxidases (Gribble, 2003; Keppler and Biester, 2003) and are enriched during the decomposition of the organic matter (Myneni, 2002; van Pée and Unversucht, 2003). Some authors have suggested that halogens in peat are fixed by organic matter, and recent studies have demonstrated that they exist mostly as halogenated organic compounds (Biester et al., 2004).

The coincidence of increased peat decomposition and higher titanium concentrations (titanium is a conservative lithogenic element that exists only in the deposited mineral dust) may also be indicative of wet/dry cycles, but for completely different reasons. Higher peat decomposition is typical of dry periods during which enhanced soil erosion and dust deposition on the bog surface are likely, which would result in higher titanium concentrations. However, a residual enrichment of titanium could also result from the degradation of the organic matter (i.e., a loss of substrate).

We suggest that changes in climate, here expressed as variations in wetness or dryness, exerted a fundamental control on the evolution of the decomposition of the peat at the surface of the bog and consequently on the concentrations of many elements. The lowering of the water table during dry periods increased the oxygenation of the superficial peat layer and triggered/enhanced microbial decomposition. This not only resulted in an increase in mineralisation but also in humification, enhancing the potential for metal and halogen retention (Meili, 1991; Mierle and Ingram, 1991; Yin et al., 1996). Thus, the increase in decomposition and humification and the enrichment of

metals and halogens are concurrent processes.

Contrary to what is commonly assumed, changes in concentrations, at least for organically bound elements, seem to depend to a larger extent on internal transformations of the peat matrix and related mass loss, and not on variations of atmospheric fluxes alone as some have suggested (Shotyk, 1997). This result is also consistent to that found at greater spatial scales for soil organic horizons and supports the hypothesis that the enrichment of organically bound elements is biologically mediated and climate dependent and would change with factors such as temperature and rainfall (Reimann and de Caritat, 2005).

4 Conclusions

Here, we have shown that there has been a clear relationship between climate (wetter-drier) and peat decomposition and its effect on element concentrations in a Spanish bog during the last 5500 years. Our findings have important implications for present day wetland environments. Projected changes in climate, particularly in areas of increased summer dryness, will increase rates of peat decomposition and thus loss of carbon as both CO_2 and DOC. During subsequent wetter periods dissolved organic matter, which is enriched in pollutants, is exported to surface waters. An increased export of dissolved organic matter has in fact already been observed in some peatlands. The surface layers of many bogs are artificially enriched in many metals, such as mercury, because of historical pollutant deposition and increased decomposition will magnify these already elevated concentrations. The changes we report here are from a mid-latitude bog, but the changes in carbon loss are expected to be much greater in the vast peatlands found at higher latitudes, particularly in permafrost areas that are or will be affected by severe thawing. Current climate research is focused largely on carbon dynamics, but these changes imply a series of additional effects. For example, a release of contaminants presently stored in high-latitude peatlands would exacerbate the current concern for high-latitude pollution.

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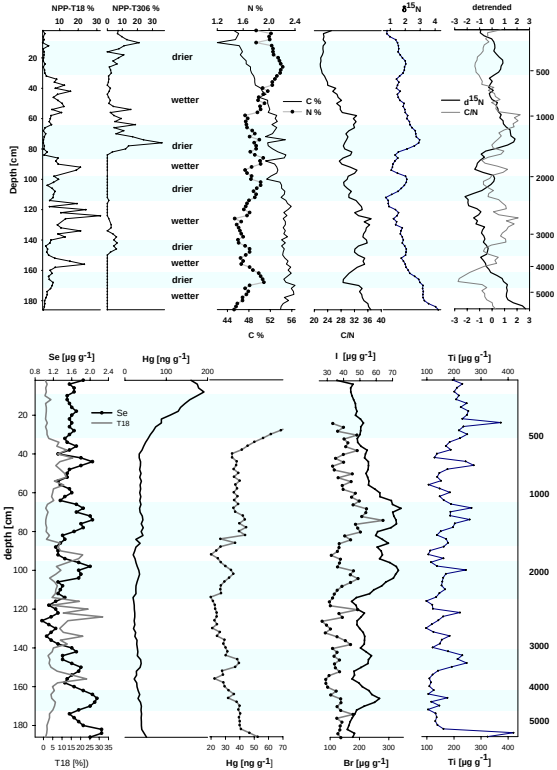

Fig. 1. (A) Records of NPPs T18 and T306, and δ^{15}N and C/N ratios in the PDC core. The detrended C/N ratios were calculated by substracting the C/N ratio of individual peat slices to the long-term trend modelled by a linear regression equation and are expressed as standardized residuals. **(B)** Concentrations records of organically-bound (Br, I, Se and Hg) and lithogenic (Ti) elements in the PDC core (all in $\mu g\,g^{-1}$ except for Hg in $ng\,g^{-1}$). The bottom axis of the mercury record was cut in $70\,ng\,g^{-1}$ so the variations in the deeper sections of the core are observable.

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