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Climate change enhances the mobilisation of naturally occurring metals in high altitude environments

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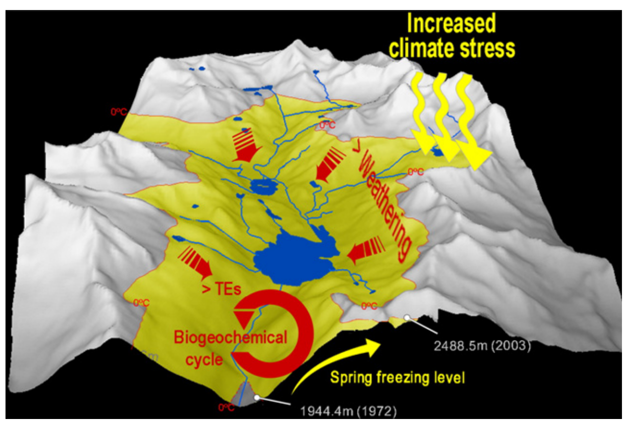
HIGHLIGHTS

- High geogenic metal levels were recorded in the sediments of a mountain catchment.
- The catchment experienced significant change in climate over the recent three decades.
- Metal increase matched changes in freezing line, precipitation and freezing events.
- This is important environmental risk, particularly if climate continues to change.
- Findings have wide implications for the natural and anthropic heavy-metal stores.

GRAPHICAL ABSTRACT



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ABSTRACT

Manmade climate change has expressed a plethora of complex effects on Earth's biogeochemical compartments. Climate change may also affect the mobilisation of natural metal sources, with potential ecological consequences beyond mountains' geographical limits; however, this question has remained largely unexplored. We investigated this by analysing a number of key climatic factors in relationship with trace metal accumulation in the sediment core of a Pyrenean lake. The sediment metal contents showed increasing accumulation trend over time, and their levels varied in step with recent climate change. The findings further revealed that a rise in the elevation of freezing level, a general increase in the frequency of drier periods, changes in the frequency of winter freezing days and a reducing snow cover since the early 1980s, together are responsible for the observed variability and augmented accumulation of trace metals. Our results provide clear evidence of increased mobilisation of natural metal sources - an overlooked effect of climate change on the environment. With further alterations in climate equilibrium predicted over the ensuing decades, it is likely that mountain catchments in metamorphic areas may become significant sources of trace metals, with potentially harmful consequences for the wider environment.

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1. Introduction

Crossing certain boundary of climate thresholds could have serious consequences for humanity, including major changes in the Earth's other systems, such as the water and geochemical cycles (Rockström et al., 2009). Similar to Polar Regions, mountain-top environments are generally more sensitive to climatic change and hence are likely to experience the consequential effects first. This sensitivity comes from stronger interactions between the major circulation patterns of the atmosphere and orography, resulting in greater changes in local environment, including cloud formation and precipitation, snow/ice level, surface moisture regime, soil erosion, heat transfer on the vertical and Foehn winds (Geiger, 1965; Beniston et al., 1997) – all these directly can influence the biogeochemical cycles (White and Blum, 1995; Camarero et al., 2004; Piao et al., 2010). This is supported by the rise in temperature observed in mountain regions in recent decades, which in some places is about five-fold greater than the global warming average (Beniston et al., 1997; Diaz and Bradley, 1997; Nogues-Bravo et al., 2007).

In high mountain catchments the biogeochemical cycling of trace elements (TEs) is generally governed by a weathering-limited regime (Stallard and Edmond, 1983). The geogenic inputs of TEs to mountain lakes may however be enhanced by climate change driven denudation of metal/metalloid-bearing minerals and their subsequent transport and burial in the sediments. Increased mobilisation of TEs can have adverse impacts on the wider environment, including water quality (Savage et al., 2000) and human health (Charlet and Polya, 2004). There is a considerable literature supporting increased accumulation of TEs and other contaminants in Polar and mountain regions, largely due to their long-range atmospheric transportation from anthropogenic activities (Macdonald et al., 2005; Outridge et al., 2005; Michelutti et al., 2009; Bing et al., 2016). Also, sediment core studies have suggested changes in land use and climate during the Holocene being responsible for erosion and geochemical changes seen in lake sediments (Augustsson et al., 2013). Despite various attempts to understand the influence of climate change on mountain geochemical processes (Psenner and Schmtdt, 1992), its potential impact on TEs release and accumulation in mountain catchments remains a major question, partly due to the complex nature of the processes involved and strong year-to-year variability. Secondly, any direct approach is not likely to provide reliable information on how weathering and transport of weathered material may have changed through time and its coupling with climate change.

Lake sediments are one of the endpoints for TEs emitted from both natural and anthropogenic sources; they can provide an accurate archive of changes in the surrounding landscapes (Yang et al., 2003), including geochemical cycling of TEs. Here we present data from a Pyrenean reservoir (Lake Bubal; Fig. 1), and examine the effects of changing climate patterns on TEs accumulation in a sediment profile covering >3 decades of historical record.

2. Methods

2.1. Hydrology and climate

Reservoir lakes are generally good sentinels of climate change. They can reflect changes in the catchment environment over known periods of time since their construction, usually by erecting dams (Williamson et al., 2009). Our study was conducted in Bubal lake catchment (headwaters of the River Gallego), a postglacial valley in the Central Pyrenees (42.68–42.85 N, 0.18–0.42 W, Spain; Fig. 1). This catchment, of about 305.5 km^2, follows a north-south orientation and is characterised by a series of interconnected lakes and typical altitude streams that drain water from the surrounding mountain slopes. Some of the lakes were transformed into reservoirs in the 1950s–60s and the main river in the catchment (Gellego) was dammed in 1971 to form Lake Bubal at 1085 m a.s.l., with its catchment peaks extending up to 3058 m a.s.l. The lake has a surface area of 234 ha, a used capacity of 64 hm^3 and annual water output of 382 hm^3 (Limnos, 1996). At about 6 water renewals each year the lake experiences a relatively short water residence time and frequent water level oscillations. Average depth of the reservoir is 27 m, which is also the depth at the sampling location. The thermocline depth varies between 5 and 12 m. All such lakes in the region have been used for both, hydroelectricity and supplying water for agriculture and potable purposes in the valley and the region further downstream.

The long-term mean annual precipitation for the catchment is 1077 mm (averaged over 1972–2005). The dominant air mass direction is from W-NW bringing precipitations mainly from the Atlantic (Dessens and Bucher, 1997). Hence, the long-distance aerial transport of contaminants must be limited in this region. This is also supported by low atmospheric deposition of metals found in the Central Pyrenees at the altitude of Bubal catchment, which is considered to be similar to European background levels (Bacardit-Penarroya, 2011). Moreover, the bulk precipitation in the Central Pyrenees is more neutral than in other Central European sites (e.g. Alps) mainly due to lower acidic pollutant levels (SO_x and NO_x) (Camarero and Catalan, 1993). There is also a reported decrease in acid depositions in the high Pyrenees in the last decades following the overall reductions in industrial emissions (Mosello et al., 2002). Therefore, the main potential source of TEs in core sediments is from the surrounding geology.

2.2. Geology

The geology of Bubal catchment is dominated by Devonian deposits (limestone, sandstone and lutite) shaped by Permian and Carboniferous materials at west and Cretaceous limestone at south (Fig. 1). This geology is marked by the extrusion of Cauterets-Panticosa granitic batholith at NE which generated a low-grade contact metamorphism aureole (Subias and Fernandez-Neto, 1995). This has significant presence of W-Au and F-Zn-Pb vein mineralisations formed in the Devonian limestone that contain fluorite (CaF_2), sphalerite, galena (PbS), pyrite (FeS_2), chalcopyrite ($CuFeS_2$), siderite ($FeCO_3$) and green and white fluorite (Subias, 1993). Pyrite in mineral deposits has As concentrations reaching 250 ± 40 mg kg^{-1}, while the hosting rock As content is relatively smaller and more consistent (87.5 ± 0.5 mg kg^{-1}) (Subias, 1993). This suggests that pyrite-containing materials (e.g. shale, schist) are potential geological source of As and other metals. Geogenic inputs of TEs have been reported in the waters and sediments of this zone, particularly in the east side (Garrido, 2002; Zaharescu et al., 2009b) (Fig. 1).

2.3. Samples collection and profile characteristics

The sampling strategy covered the sedimentary record from the major catchment depository: Lake Bubal, 1085 m a.s.l. (Fig. 1 and S1a). A 50 cm sediment core was extracted from the lake in bathymetric depression toward the edges of the former river (42°41′49.97″N, 0°18′ 51.65″W; see Interactive Map File), where the interferences from sediment slumping or turbidity were minimal. The sampling was conducted in August 2005 when the north half of the lake bed was exposed during an unusually extreme drought. A pit was dugout in an area free of potential disturbances to extract the sediment core (Fig. 1 and S1a). Therefore, no sediment compression/disturbance occurred during sampling. The cobles and forest soil at the bottom of the core clearly indicated the depth from where the new deposits were formed since 1971 when the dam was erected. The core was therefore expected to comprise a record from the year when this part of the river was transformed into reservoir.

The profile was brownish, indicative of oxic environment. Sediment organic content was low (3.39 ± 1.5%, as loss-on-ignition). The profile, visually comprised of fine alluvial sediments (silt–fine sand), showed continuous record of sedimentation and no important evidence of remixing or bioturbation during the pit excavation and sampling. It is