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Assessing the impact of long-term changes in climate and atmospheric deposition on a shallow alpine lake from southeast Tibet

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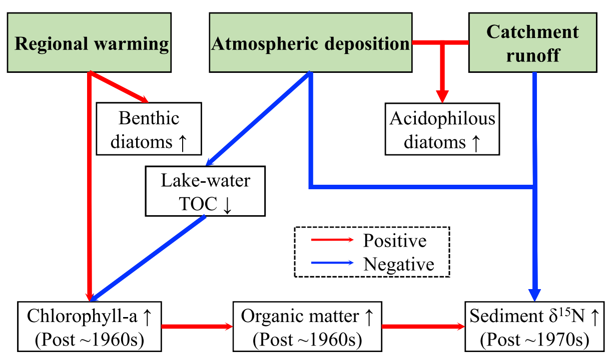
HIGHLIGHTS

- Anthropogenic Hg deposition increased from pre-industrial levels and accelerated from the 1960s
- Atmospheric acid deposition was linked to a consistent decrease in lake-water TOC from the 1960s.
- Increases in algal production and sediment OM were most closely linked to recent climate warming.
- Increased OM and reduced catchment erosion overrode acid deposition in enriching the $\delta^{15}N$ signal.
- Diatom community changes were linked to both acid deposition and warming climate.

GRAPHICAL ABSTRACT



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ABSTRACT

Regional warming and atmospheric nitrogen deposition have been widely recorded to impact remote catchments and alpine lakes; however, their independent roles and interactions have rarely been identified. Here, we combined down-core analyses of sedimentary mercury (Hg) and aluminum (Al) with multiple proxies (i.e. nitrogen stable isotope, chlorophyll *a* pigments, diatoms) for a radiometrically-dated sediment core of an alpine lake in southeast Tibet to track the atmospheric deposition of pollutants, and to examine possible effects of climate and catchment forcing over the past three centuries. The sediment data revealed that airborne deposition of Hg was recorded from the ~1860s, with an accelerating increase in anthropogenic Hg flux since the ~1960s. A synchronous decrease in reconstructed lake-water TOC indicated that acid deposition may have affected lake-water carbon concentrations and impaired catchment export of decomposed organic matter (OM). A moderate depletion of bulk sediment $\delta^{15}N$ started from the ~1820s, but was followed by an enriching trend after the ~1970s. This positive shift of $\delta^{15}N$ was associated with elevated sediment OM and decreased catchment runoff of clastic materials (as inferred by Al). Sediment OM content displayed an accelerating increase from the ~1960s, with an increased input of autochthonous sources (i.e. lower bulk sediment C:N ratios), such as algae (as inferred by sedimentary chlorophyll *a* pigments). Meanwhile, climate warming and decreased lake-water TOC enhanced the production of algae, which was characterized by a more enriched $\delta^{15}N$ signal than that of allochthonous OM. Furthermore, atmospheric acid deposition was significantly related to diatom assemblage changes, with an increase in acidophilous taxa. Our sediment evidence revealed the dominating impact of climate and catchment

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processes on lake-water chemistry and algal shifts in the context of atmospheric nitrogen deposition, and highlighted an increasing link of external forcing with in-lake processes in enriching sediment $\delta^{15}N$ signal over the last few decades.

1. Introduction

Climate warming and atmospheric nitrogen deposition have been recognized to cause considerable changes in limnological and catchment processes over regional to global scales (Catalan et al., 2013). There is growing evidence that the warming rate is often amplified at higher elevations and latitudes, with more pronounced warming and greater seasonal temperature anomalies (Pepin et al., 2015). In alpine and high-latitude lakes, warmer temperatures can result in shorter ice cover, prolonged growing season and stronger hydrological fluctuation (Smol and Douglas, 2007a, 2007b), thereby increasing primary productivity and favoring smaller diatoms (Catalan et al., 2013; Rühland et al., 2015). Meanwhile, human activities have indirectly altered catchment biogeochemical cycling in these remote regions via atmospheric emission and large-range deposition of pollutants such as reactive nitrogen (Nr; Galloway et al., 2008). The increased Nr deposition and influx can further alter lake-water nutrient availability and stoichiometry, which have been linked to changes in community structure and ecosystem productivity (Saros et al., 2005; Elser et al., 2009a). Furthermore, catchment processes (i.e. carbon sequestration) enhanced by warming can lead to the long-term change of lake biogeochemical cycling (McLauchlan et al., 2013). Therefore, there may exist a strong link of landscape processes with climate and atmospheric deposition that can be of significant consequences for alpine lakes.

The nitrogen stable isotope ratio ($\delta^{15}N$) has been widely applied to track anthropogenic nitrogen deposition and biogeochemical processes from natural archives, including ice cores (Hastings et al., 2009), corals (Ren et al., 2017), tree rings (Hietz et al., 2011) and lake sediments (Holtgrieve et al., 2011). The application of this proxy is grounded on the assumption that Nr derived from fossil fuel burning and fertilizer use is characterized by depleted $\delta^{15}N$ relative to the pre-industrial catchment N sources (Talbot, 2001). Nitrogen isotopic records derived from ice core nitrate-N in Greenland and lake sediments of North Hemisphere showed a consistent depletion in $\delta^{15}N$ over the past ~150 years, synchronous with the rise in atmospheric NO_3^- concentration (Hastings et al., 2009; Holtgrieve et al., 2011). However, it is now acknowledged that there exists spatial heterogeneity in the temporal trend of sedimentary $\delta^{15}N$ changes over the past few centuries at regional to global scales (McLauchlan et al., 2013; Hu et al., 2014). Firstly, the sediment $\delta^{15}N$ records often serve as a mixed signal for N cycling that can be modulated by transportation pathways and biogeochemical processes such as carbon burial (Gälman et al., 2009; Hu et al., 2017). Furthermore, the flux and temporal trajectory of atmospheric nitrogen deposition can vary depending on the spatial heterogeneity in regional Nr emission (Galloway and Cowling, 2002; Lü and Tian, 2007). Monitoring data demonstrated a reduction in Nr emission over the last few decades in many parts of North America and Europe, leading to a recovery of lake acidification with increased lake-water TOC (Meyer-Jacob et al., 2017). Simultaneously, a clear trend of increasing atmospheric Nr emission has persisted in east and south Asia (Geddes and Martin, 2017), regions that were predicted to receive increasing Nr flux in the coming decades (Bleeker et al., 2011).

Complementary proxies, such as atmosphere-borne trace metals, can potentially be used in paleolimnological studies to provide a fuller scope on the change of atmospheric deposition. Atmospheric emission can release not only Nr but also contaminants such as mercury (Hg; Zhang et al., 2016), which can be well preserved in natural archives (Hermanns and Biester, 2013; Eyrikh et al., 2017). The sediment records of Hg enrichment in alpine lakes were often found to be synchronized with an increase in the atmospheric Nr emission (Catalan et al., 2013).

The southeastern margin of the Tibetan Plateau lies between south and east Asia, the two global hotspots of atmospheric Nr deposition over the last few decades. The Hg accumulation rate in lake sediments from the Himalaya-Tibet region showed a rising trend at the onset of Industrial Revolution, followed by a striking increase after World War II, as recorded in ice cores (Kang et al., 2016), matching well with ice core NO_3^- change (Thompson et al., 2000). The monitoring data also indicated that there existed a clear trend of warming by >1.5 °C in this low-latitude and alpine region over the last five decades (Li, 2015), leading to obvious glacial retreats (Li et al., 2010), advancing tree line (Baker and Moseley, 2007) and enhanced algal production in alpine lakes (Lami et al., 2010). With continued warming and atmospheric deposition in SE Tibet, it is crucial to refine their independent and combined impacts on lake biogeochemical properties and their sediment signal that also integrate processes at the watershed scale.

To help tease out the relative roles of regional climate and atmospheric deposition in driving the sediment $\delta^{15}N$ signal and algal changes, we conducted multi-proxy analyses from a well-dated sediment core of an alpine headwater lake in SE Tibet, which included the proxies of nitrogen stable isotope, trace metals, algal pigments (i.e., inferred chlorophyll *a* and its main diagenetic products) and diatom assemblages. Firstly, we identify the signal of atmospheric deposition through combining the proxy analyses of sediment metal concentrations with lake-water TOC reconstruction over the last three centuries. Secondly, we tease out key environmental factors and quantify their strength in driving the temporal change in algal production and species shift. Finally, we aim to evaluate the mechanistic link among climate, acid deposition and catchment processes in regulating the temporal variations in sediment $\delta^{15}N$ signal.

2. Materials and methods

2.1. Study site

Taiji Lake (26.62877° N, 99.70935° E; 3978 m a.s.l.), formed by glacial erosion, is located on the southeastern margin of the Tibetan Plateau, southwest China (Fig. 1a). Taiji Lake, as a headwater lake (Fig. 1b), is located below the current tree line of ~4100 m a.s.l. (Xiao et al., 2014), with a small lake area (1.84 ha) and shallow water depth (max depth = 3.5 m). The catchment vegetation consists of conifer forests comprising mainly of *Tsuga* and *Abies*, as well as alpine shrub of *Rhododendron*, with alpine herb meadow and rock screes dominating above the tree line (Xiao et al., 2014). The geology of the region consists of weathering-resistant tertiary sandstone and conglomerate, as well as the underlying granite porphyry. Two ephemeral inlets and one outlet stream are located in the southwest, south and north part of the lake basin, respectively (Fig. 1c).

The climate in the study region is dominated by the Indian monsoon and East Asia monsoon in summer, as well as by the westerlies in winter (Li, 2015). Based on instrumental data (1950–2013) from the study region, the local annual mean temperature was estimated to be ~2.5 °C with an annual precipitation of ~910 mm (Xiao et al., 2014; http://data.cma.cn/). Around 80% of the annual mean rainfall occurs from May to October in the study region. The modern duration of ice cover based on the altitude and air temperature was inferred as around