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Limited acid deposition inferred from diatoms during the 20th century — A case study from lakes in the Tatra Mountains

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ABSTRACT

Mountain lakes are usually sensitive to the effects of global and regional environmental changes. Since the second half of the 20th century, surface-water acidification has become a significant ecological problem, and many lakes in Europe and North America have anthropogenically acidified. Additionally, following reduction in emissions of sulfur (S) and nitrogen (N) compounds, recovery from acidification has been observed in many lakes. In this study, we used changes in diatom communities to reconstruct the pH histories based on changes recorded in nine Tatra lakes (Western Carpathians, Poland) since approximately 1850 AD. Overall, results indicate that acidic precipitation had little influence on lake-water pH in the Tatra Mountain lakes. Changes in diatom-inferred pH (DI-pH) generally were small and showed little evidence of acidification during the time of the highest air pollution (since the 1960s), and have shown little change since the reduction of acidic deposition since the 1990s. Lakes that showed some evidence of acidification included dystrophic lakes with low acid neutralizing capacity. However, as illustrated by the PCA trajectories of the diatom assemblages, the majority of the lakes currently contain diatom assemblages that are unlike the diatom floras that existed *ca.* 1850.

Introduction

High mountain lake ecosystems are typically sensitive to environmental changes and are therefore of great interest for paleolimnological research. Many high-altitude lakes have been studied for the effects of acid rain and the recovery from human acidification (*e.g.*, Stoddard et al., 1999; Mosello et al., 2002; Kopáček et al., 2006a, 2006b; Sienkiewicz et al., 2006). Since the second half of the 20th century, many lakes in regions of Europe and North America receiving acidic deposition have become anthropogenically acidified (*e.g.*, Curtis et al., 2002; Battarbee et al., 2005). This acidification, which began particularly during the Industrial Revolution, intensified with time (Kopáček et al., 2001). According to historical data, in central-eastern Europe, the earliest industrial development occurred in Bohemia (after 1848) and in the Kingdom of Poland (*ca.* 1850–1890), which became the most economically developed areas of the Habsburg's Monarchy and the Russian Empire (Leslie et al., 1980). With increases in economic growth, environmental quality deteriorated as industry developed and emissions of pollution originating from the combustion of fossil fuels increased.

The most significant increase in air pollution occurred in the 20th century, particularly at the beginning of the 1960s, which was corroborated by geochemical and palaeobiological analyses of lake sediments and monitoring data (*e.g.*, Kopáček et al., 2001, 2003; Vrba et al., 2003). Emissions of gaseous forms of sulfur and nitrogen oxides are transformed into sulfuric and nitric acids, H_2SO_4 and HNO_3, respectively, when contacted by water. These acids are transported in the atmosphere and

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deposited as acid rain, often far from the primary source of pollution. Although most mountain lakes are remote ecosystems, they are also exposed to this type of pollution. The primary sources of increasing sulfur deposition in the High Tatra Mountains were heavy industrial production after World War II and the abrupt growth of energy consumption, originally based on lignite combustion in East Germany, the Czech Republic and Slovakia (Schöpp et al., 2003).

In Europe, in the early 1980s, due to political and economic transformations, the first steps were introduced to restrict sulfate emissions as the primary contributor to acidification. Following these restrictions, the European Commission then focused on limiting oxidized nitrogen compounds as a second factor in the acidification of terrestrial and aquatic ecosystems (Fowler et al., 2007). As a result of those regulations, recovery from acidification has been observed in recent decades (Vrba et al., 2003; Stuchlík et al., 2006). Based on the amount of sulfur and nitrogen bulk deposition, recovery in the Tatra Mountains began in the 1990s (Kopáček et al., 2001). Despite the rapid decrease in acidifying pollutants deposited in lake water, a biological response towards pre-acidification quality in European mountain lakes has not yet occurred in some cases, and recovery has not been observed in all places simultaneously, even in areas close to one another (Stoddard et al., 1999; Evans et al., 2001). The reversal of acidification in an individual lake depends on the catchment and lake characteristics, such as altitude, geology and morphology (Mosello et al., 2002). The current acid–base status of a lake, *i.e.*, the extent of acidification, determines the acid neutralizing capacity (ANC) (Hemond, 1990; Sullivan et al., 2007), and generally, chronic and/or episodic acidification was limited to lakes having an ANC ≤ 50 μeq/L (Sullivan et al., 1990).

Many lakes in the Tatra Mountains were not strongly acidified in comparison with some other mountain lakes in Europe (*e.g.*, Kopáček et al., 2003; Kawecka and Galas, 2003; Sienkiewicz et al., 2006), and in many cases, their recent ANCs are above 50 μeq/L. Generally, based on acid–base status, three primary groups of lakes are categorized in the Tatra Mountains: nonsensitive (NS) lakes with pH > 6 and ANC > 25 μeq/L; acid-sensitive (AS) lakes with 5 < pH < 6 and ANC 0–25 μeq/L; and extremely sensitive (ES) lakes with pH < 5 and ANC < 0 μeq/L (Kopáček et al., 2004).

However, in some of these lakes, acidic deposition lowered the diversity of zooplankton and caused the extinction of acid-sensitive taxa (Hořicka et al., 2006). In a few lakes, the process of eutrophication has also begun within recent decades (*e.g.*, Bombówna and Wojtan, 1996; Gliwicz, 1985; Kurzyca et al., 2009; Sienkiewicz and Gąsiorowski, 2014, 2016a, 2016b). Diverse analyses and observations are used to determine changes in lake ecosystems, including chemical and palaeobiological investigations and monitoring data. Although many scientists have studied the diatom flora of the Tatra Mountains (*e.g.*, Fott et al., 1999; Eloranta and Kwandrans, 2002; Kownacki et al., 2006; Kwandrans, 2007), investigations have primarily focused on epilithic diatoms, which are collected in different periods of vegetation. For this study, lake sediment cores were examined that were collected between April 2003 and March 2013. The diatom stratigraphy of these lakes, except for lake Czarny Staw Polski (CSP) (Appendix A Fig. S1), was presented in Gąsiorowski and Sienkiewicz (2010a, 2010b) and Sienkiewicz and Gąsiorowski (2014, 2016a, 2016b). Papers have focused on the trophic status of the lakes, the 20th century climate warming and changes of isotopic composition in lake sediments. Sedimentary diatom assemblages with taxa ordered by pH optima are shown in Appendix A Figs. S1–S4.

The aim of this paper was to examine the paleolimnological record to determine the intensity of acidity changes in lake ecosystems from the beginning of the Industrial Revolution (*ca.* 1880 AD) to *ca.* 2000 AD. This represents the synthesis of the acidification process recorded in most of the biggest lakes located in the Polish part of the Tatra Mountains. Diatom analysis is a primary tool among paleolimnological methods used to trace the pH and acidification history of lakes because diatoms are highly sensitive organisms that respond quickly to alterations in their environment.

The novelty of this study was the determination of changes in the distribution of algae grouped ecologically by pH preference to reconstruct diatom-inferred pH (DI-pH) from dated mountain lake sediments. Estimation of historical acidification can help determine catchment, surface water and diatom communities' response to acidic deposition. Additionally, these changes were correlated with the deposition of sulfur and nitrogen compounds. Changes in the diatom community and DI-pH every few years can be estimated because of the relatively low sedimentation rate, and for current lake management, this is helpful information because effective management requires long-term environmental data collected at the shortest possible time intervals (Smol, 1992). We selected nine lakes in the Polish section of the Tatra Mountains (central Europe) of which seven were oligotrophic and two dystrophic, with acidic to slightly alkaline waters. Most of the lakes are currently classified as nonsensitive water bodies (ANC > 25 μeq/L and pH > 6); however, the acid-neutralizing capacity and water pH can change, even within a relatively short time. From the beginning of the reduction in air pollution to approximately the present day, the values of both factors generally increase with time (Table 1). Although ANC data collected before the 1980s and pH measurements before the 1960s are available to some extent, the reliability is highly uncertain (Kopáček et al., 2004). Thus, with the exception of the last few decades, we do not have reliable information on changes in pH in recent lake history. Although diatom analysis does not provide an actual ANC value, the diatom-inferred pH can approximate estimated shifts in ANC and determine whether a specific lake was sensitive to acidification in the past.

1. Methods

1.1. Study sites

All studied lakes have a glacial origin and are in different valleys of the Tatra Mountain range (Poland, central Europe). The catchment bedrock consists primarily of crystalline rocks (granites) with quaternary glacial sediments and rock debris cones. We investigated the following lakes (Fig. 1): Zielony Staw Gąsienicowy (ZSG) and Czarny Staw Gąsienicowy (CSG) from the Gąsienicowa Valley; Morskie Oko (MOK) and Czarny

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