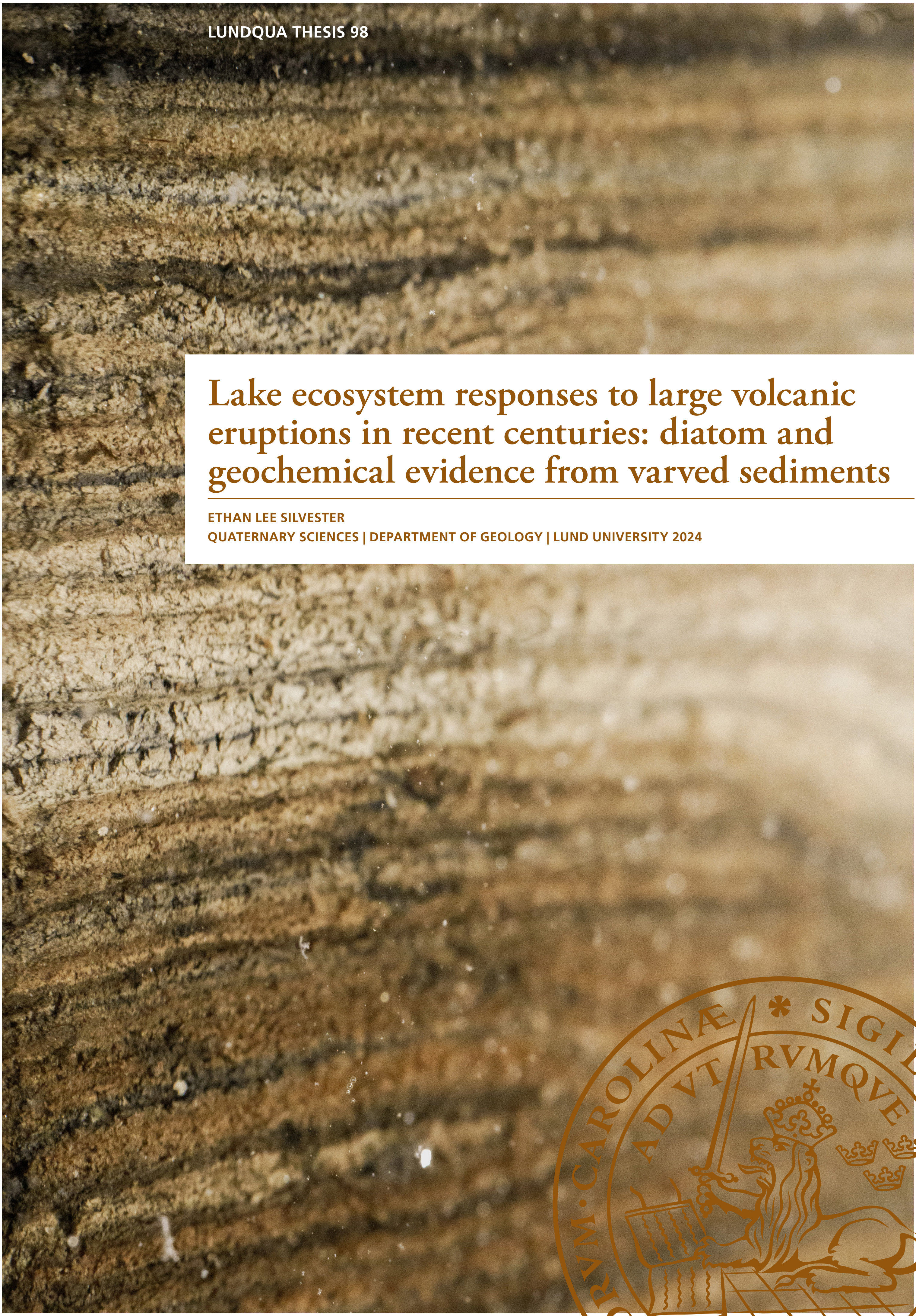
Lake ecosystem responses to large volcanic eruptions in recent centuries: diatom and geochemical evidence from varved sediments

Silvester, Ethan



LUNDQUA THESIS 98

Lake ecosystem responses to large volcanic eruptions in recent centuries: diatom and geochemical evidence from varved sediments

ETHAN LEE SILVESTER

QUATERNARY SCIENCES | DEPARTMENT OF GEOLOGY | LUND UNIVERSITY 2024

LUNDQUA THESIS 98

Lake ecosystem responses to large volcanic eruptions in recent centuries: diatom and geochemical evidence from varved sediments

Ethan Lee Silvester

Quaternary Sciences
Department of Geology

DOCTORAL DISSERTATION
by due permission of the Faculty of Science, Lund University, Sweden.
To be defended at Pangea, Geocentrum II, Sölvegatan 12. Date 18.12.2024 and time 13:15.

Faculty opponent
Prof. Helen Bennion
University College London, UK

Faculty of Science
Department of Geology

ISBN: Lundqua theses 978-91-87847-86-8 (print)

ISBN: Lundqua theses 978-91-87847-87-5 (e-)

ISSN: 0281-3033

Printed in Sweden by Media-Tryck, Lund University, Lund 2024

Printed matter 3041 0903

Media-Tryck is a Nordic Swan Ecolabel certified provider of printed material. Read more about our environmental work at www.mediatryck.lu.se

MADE IN SWEDEN

Organization LUND UNIVERSITY Department of Geology Sölvegatan 12 SE-22362, LUND Sweden Author: Ethan Lee Silvester	Document name DOCTORAL DISSERTATION Date of issue: December 18, 2024 Sponsoring organization
Title and subtitle: Lake ecosystem responses to large volcanic eruptions in recent centuries: diatom and geochemical evidence from varved sediments.	

Abstract

Volcanic eruptions are documented throughout geological history and are frequently observed and experienced in the modern world. Strong eruptions in recent decades have resulted in air and water pollution, acid snowfall, disruption in aerial transport and various illnesses in humans and livestock. Despite their consequences, these recent events are incomparable to eruptions of a much larger magnitude in recent centuries such as the Icelandic eruption of Laki (1783-84) and Tambora (1815) which are infamously known for massive losses in livestock and crop failure, resulting in famine and large losses of human life. While their impacts on society and agriculture are well documented and studied, little is known about how these major events impacted distant lake ecosystems.

This Ph.D. project focused on varved sediments from Swedish (Odensjön and Kassjön) and Finnish (Nautajärvi) lakes to investigate the effects of the eruptions of Laki and Tambora on distal lake ecosystems. Varved sediments are annually layered sediments, providing precise time control facilitating the investigation of changes in the past. Of particular interest to this project is the responses of diatoms – photosynthetic algae that often dominate lake biomass. Diatoms are great ecological indicators due to their high sensitivity to changes in the lake and the surrounding catchment. Small changes in environmental conditions can influence diatom productivity and community structure and they are widely documented to reflect changes in past environmental conditions. Past acid deposition and any chemical effects in lake waters can be represented by geochemical signals preserved in the sediments. Combined diatom and geochemical analysis of varved sediments should form a comprehensive view on whether and how distant volcanic eruptions affected lake ecosystems. This thesis has the following objectives: 1) assess the biogeochemical impacts of transient acidification and hydroclimatic perturbations related to these two eruptions, and 2) Investigate the type, duration and severity of impacts on the diatom community and terrestrial biota, and determine the degree of ecosystem resilience. These objectives were addressed in three subprojects.

Subproject 1 focused on Lake Odensjön in Southern Sweden. This study documented a new varved sediment record and detailed lake ecosystem dynamics during the last six centuries with the use of diatom and geochemical analysis. A minor response to Laki eruption may be reflected in the sediments, in the form of a minor decline in pH inferred by changes in the diatom assemblage. Odensjön represents the southernmost varved sediment record documented in Fennoscandia to date and although no strong signals of major volcanic eruptions were recorded, it offers a potential to study effects of environmental forcings on the lake ecosystem over several hundred years. Subproject 2 focused on Lake Kassjön in Northern Sweden. Although the main objective of this study was to identify eruptions of Laki and Tambora, we evaluated the potential confounding effects of land-use practices and human impacts in the catchment between 1644 and 1931. Substantial changes were recorded in the sediments in timing with the eruption of Laki, exhibited by: 1) a significant change in the diatom assemblage structure, 2) qualitative diatom evidence of complex changes in lake-water chemistry and nutrient and light availability and 3) enhanced chemical weathering indicated by deviations in the typical biogeochemical processes. No pronounced changes were observed during the eruption of Tambora. Subproject 3 focused on lake Nautajärvi in Southern Finland. Based on ultra-high resolution sampling of the varved sediments, we found evidence of ecological stress following the eruption of Laki, reflected by changes in the diatom assemblage and multiple other biotic indicators. Our records demonstrate how distal boreal ecosystems may undergo complex and yet poorly understood changes in soil chemistry in response to acid deposition.

This Ph.D. was an interdisciplinary approach combining geochemical proxies, diatom community dynamics and complex statistical methods to identify major volcanic eruptions during the past centuries. Annually layered sediments from Fennoscandian lakes recorded various changes in lake ecosystems in timing with the eruption of Laki, which were most pronounced in Kassjön and Nautajärvi, highlighting the potential of highly resolved studies of varved lake sediments.

Key words: diatoms, pH, volcanic eruption, lake acidification, lake catchment, land use		
Classification system and/or index terms (if any)		
Supplementary bibliographical information		Language: English
ISSN and key title: 0281-3033 LUNDQUA Thesis		ISBN: 978-91-87847-86-8 (print) 978-91-87847-87-5 (e-)
Recipient's notes	Number of pages: 141	Price
	Security classification	

Signature Date 2024-11-04

This is the real secret of life -- to be completely engaged with what you are doing in the here and now. And instead of calling it work, realize it is play.

Alan Watts

Contents

List of publications

This thesis is based on three publications listed below, which have been appended to this thesis.

Paper I

Silvester E.L., Ljung K., Bindler R., Hertzman H., Lodi G., Hammarlund D., 2024. **Diatom dynamics during the last six centuries in Lake Odensjön: a new varved sediment record from Southern Sweden.** *Journal of Paleolimnology,* 1–21.

Paper II

Silvester E.L., Bindler R., Bigler C., Björnerås C., Ljung K., Hammarlund D., 2024. **Diatom and biogeochemical changes during recent centuries in a small boreal lake: deciphering the influence of large volcanic eruptions.** Manuscript submitted to Quaternary.

Paper III

Silvester E.L., Ojala A.E.K., Kosonen E., Weckström J., Ljung K., Hammarlund D., 2024. **Diatom and biogeochemical evidence of the Laki 1783–84 eruption in highly-resolved varved sediments of a Finnish lake.** Manuscript.

Acknowledgements

First and foremost, I would like to thank my supervisors. I would like to thank **Dan**, for providing the incredible opportunity to study our natural environment, for your patience as a mentor, and for encouraging my development as a scientist. I would also like to thank **Kalle**, for helping me navigate the complexities of my research and for the fruitful scientific and philosophical discussions over the years.

I would like to thank all of my co-authors for their various contributions, guidance, positive and critical feedback and unwavering encouragement. I would like to thank **Caroline Björnerås**, whose advice on various and often disorienting aspects of geochemistry was essential. I would like to thank **Richard Bindler**, whose advice on data analysis and patience in receiving my many questions allowed me to grow as an analyst. I would like to thank **Christian Bigler** and **Jan Weckström** for encouraging my development as a diatomist and for their valuable advice on diatom-based reconstructions. I would like to thank **Antti Ojala** and **Emilia Kosonen** for their expert insight into the complexities of working with varved sediments and encouraging my development as a paleolimnologist.

I would like to thank **my colleagues and friends** from the department for creating an enthusiastic atmosphere and supporting my journey as a student. I am grateful to **all the PhD students** for our shared love of science, coffee, and cake. I am especially indebted to my office friends — **Bingjie**, **Ida**, and **Nikolas** — whose encouragement, shared passions, and support over these past years helped me endure the difficult periods and remember to celebrate and appreciate the moments of success.

I would like to thank **Petra Zahajská**, who helped me settle in Sweden, navigate and appreciate Swedish culture, and grow as a student. I also wish to thank **Gert** for his invaluable technical support, and the **Kansli** team for their patience and help with various administrative tasks over the years.

I would not be where I am today without my incredible fiancée, **Karolina**. Your support, encouragement, contagious enthusiasm, and shared love of diatoms had a great impact on my research and growth as a fellow diatomist. Your high standards, attention to detail and interest in so many different areas of research profoundly influenced my work. Thank you kochanie. I also wish to thank **Kajtek**, our loyal and loving dog, for making me take regular walks to clear my mind. Last but not least, I am deeply grateful to **my Family** for their constant support, encouragement, and interest in my work.

1 Introduction

Volcanic eruptions are documented throughout geological history and are frequently observed and experienced in the modern world. The environmental consequences of a given eruption depends on many factors, including the nature and scale of the event. In addition to the immediate and local impacts, the emissions released during an eruption can be transported in the atmosphere for thousands of kilometres and have various effects on the environment upon deposition. Eruptions in recent years provide some insight into the consequences for modern society, which include illness in humans and livestock, pollution of water, poor air quality and disruptions to aerial transport (Gudmundsson et al., 2012). However, the eruptions which have been experienced in recent decades are minor in comparison to those which have occurred in recent human history.

The major eruptions of Laki, Iceland in 1783–1784 and Tambora, Indonesia in 1815 are well documented in historical archives. Written accounts from the time describe the loss of human life in the range of thousands, major loss of livestock and famine resulting from the effects of Laki 1783–84 (Grattan et al., 2005) and the climatic impact of Tambora 1815 resulted in the following year being infamously known as “the year without a summer” (Oppenheimer, 2003). The consequences of both eruptions were felt throughout Europe. While historical accounts detail the effects of these two eruptions on agriculture and society, the wider ecological impacts of these events remain poorly understood.

As natural archives, lake sediments offer the opportunity to investigate the response of distal ecosystems to these events. Sediments that accumulate on the lakebed over time contain a record of changes within the lake system under past environmental conditions. Under certain conditions these sediments remain undisturbed and form a succession of annual layers called varves, facilitating the study of past environmental conditions with precise chronological control. As such, varved sediments are ideally suited to study the impacts of events of a known age, such as the eruptions of Laki 1783–84 and Tambora 1815. Changes in the geochemical properties of the sediments, in addition to the biological remains within, provide multiple insights into the dynamics of the lake ecosystem over time.

Diatoms are unicellular, photosynthetic eukaryotes and are present in the majority of water bodies on Earth. These microalgae constitute a significant proportion of the algal biomass in freshwater environments and are an important part of the lower food chain. Diatoms produce cell walls made of silicon dioxide, known as frustules, which are resistant to degradation and often remain well-preserved in lake sediments. As they rapidly respond to changes in their environment, such as the availability of light and nutrients, and water pH, diatoms are considered an indicator organism group and are commonly used in ecological research and water quality monitoring. As such, changes in the structure of the diatom assemblage preserved in lake sediments can provide valuable insights into past environmental conditions.

This Ph.D. project investigates the response of Fennoscandian lake ecosystems to the effects of the major volcanic eruptions of Laki 1783–84 and Tambora 1815, through multi-proxy studies of varved lake sediments incorporating both diatom and biogeochemical analyses.

2 Aims of the thesis

The aim of this thesis is to characterise lake ecosystem conditions during the period preceding, encompassing and following the eruptions of Laki 1783–84 and Tambora 1815 and assess the potential effects of the eruptions, with the following objectives:

1. Identify biogeochemical evidence of transient acidification related to the eruption of Laki 1783–84 and hydroclimatic perturbations related to the eruption of Tambora 1815.

2. Investigate the type of any disturbance to the lake ecosystem, with a focus on diatoms.

3. Assess the severity and persistence of any disturbance and the degree of ecosystem resilience.

To achieve these objectives, this project uses highly resolved, multi-proxy studies of varved lake sediments, which serve as precise natural archives of past environmental conditions.

3 Background

3.1 Impacts of recent volcanic eruptions

The observed consequences of recent volcanic events may provide us with some insight into the potential impacts of future eruptions many orders of magnitude larger. Recent eruptions have released between 100 and 1000 times less material and gas into the atmosphere than major eruptions, which are categorised as 6 or 7 on the Volcanic Explosivity Index (VEI) scale (Fig. 1) and have a recurrence frequency of 50–100 and 500–1000 years, respectively.

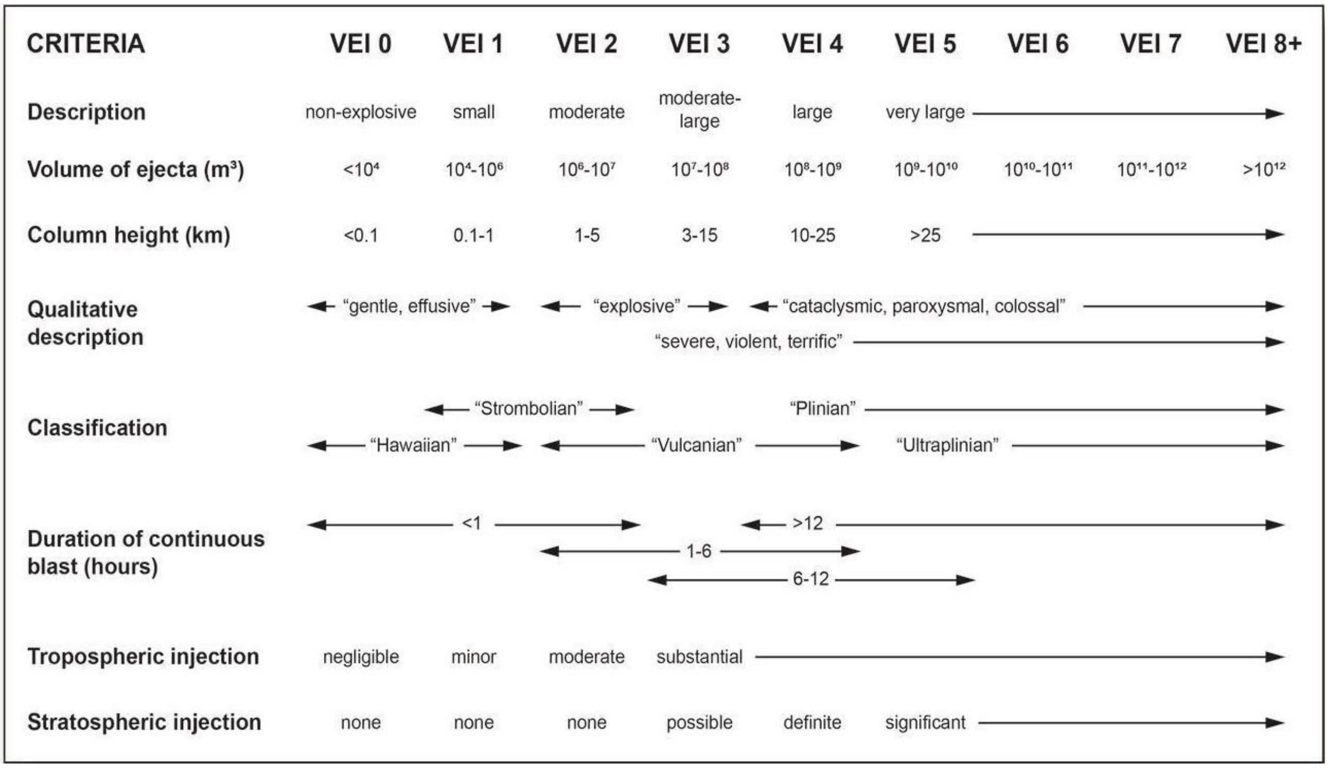


Figure 1. Criteria for estimation of the Volcanic Explosivity Index (VEI). Figure modified from Newhall and Self (1982) by Michael Poland (USGS).

The societal consequences of recent eruptions are well documented, from the disruption of air traffic and respiratory irritation resulting from the eruption of Eyjafjallajökull, Iceland, in 2010 (Petersen, 2010), to several hundred deaths associated with the eruption of Mount Merapi, Indonesia, in 2010, which is the largest example of human displacement caused by volcanic activity in recent times, having affected over 350,000 people (Mei et al., 2013). These eruptions released large volumes of ash and between 0.1 and 0.4 teragrams (Tg: 1 Tg = 1×10^{12} g) of sulphur dioxide (SO_2) gas to the atmosphere (Jousset et al., 2012; Boichu et al., 2013) and both were classified as VEI 4 eruptions. The 2014–2015 eruption of Bardarbunga (Iceland) is one of the more recent examples of volcanic SO_2 emissions affecting Europe. During this fissure eruption (an eruption occurring along a long crack in the Earth's surface), the plume distribution was tracked using modern satellites and air quality was recorded by surface monitoring stations. A model of SO_2 dispersal across Europe which incorporated these data indicated that the daily SO_2 output from Bardarbunga was at least 3 times greater than the combined daily average anthropogenic SO_2 emissions in Europe in 2010 (Schmidt et al., 2015). Fig. 2 shows surface SO_2 levels at different periods following the beginning of the eruption, with Iceland and areas of the British Isles exceeding the WHO threshold of air pollutant concentrations (WHO, 2006). Eruptions of this scale are relatively frequent events, and their consequences are minor in comparison with larger eruptions in recent centuries.

Historical written accounts provide some insight into the societal and climatic impacts of larger VEI 6–7 eruptions. A prime example of this is the VEI 6 fissure eruption of Laki (Iceland) (Fig. 3) in 1783–1784, which resulted in around 10,000 deaths in Iceland alone; around one fifth of the population at the time (Stone, 2004), in part due to the loss of livestock (~50%) and resulting famine. Estimates suggest there were over 25,000 deaths across NW Europe (>16,000 in France, >10,000 in Britain), resulting from respiratory illness associated with air pollution, followed by an anomalously cold winter (Grattan et al., 2005).

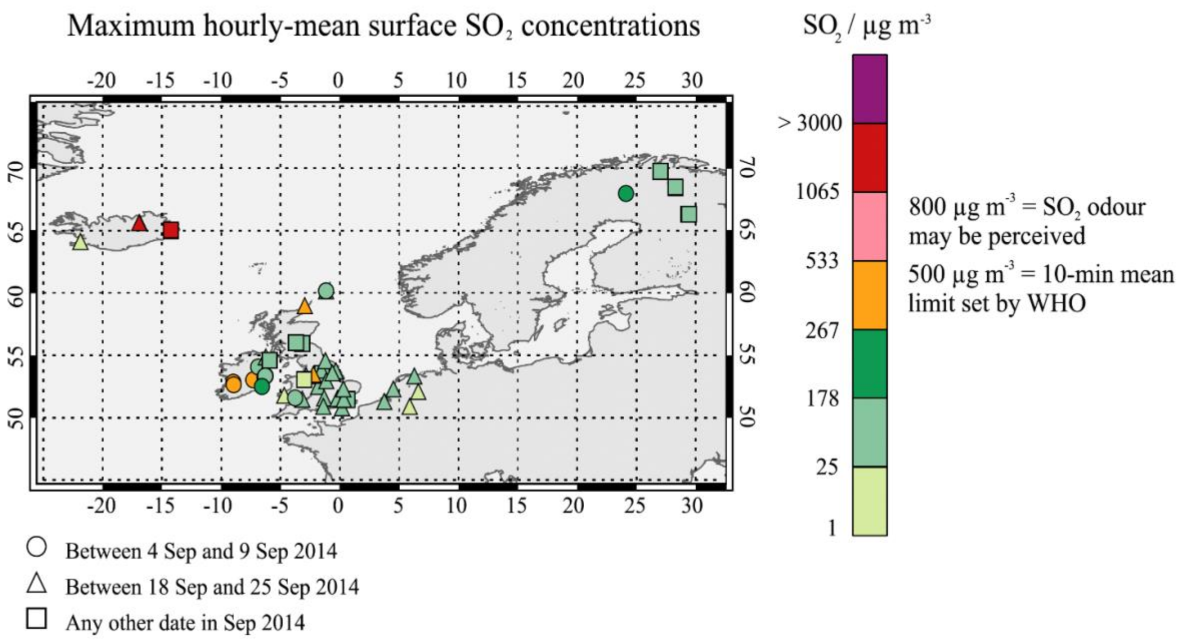


Figure 2. Maximum 1 hour mean concentrations of surface SO_2 recorded at monitoring stations across Europe during the 2014–2015 eruption of Bardarbunga (Schmidt et al. 2015). Data represented separately between 4th and 9th September 2014 (5–10 days after the eruption; indicated by circles), 18th and 23rd September 2014 (triangles) and all other days in September 2014 (squares). From Schmidt et al (2015).

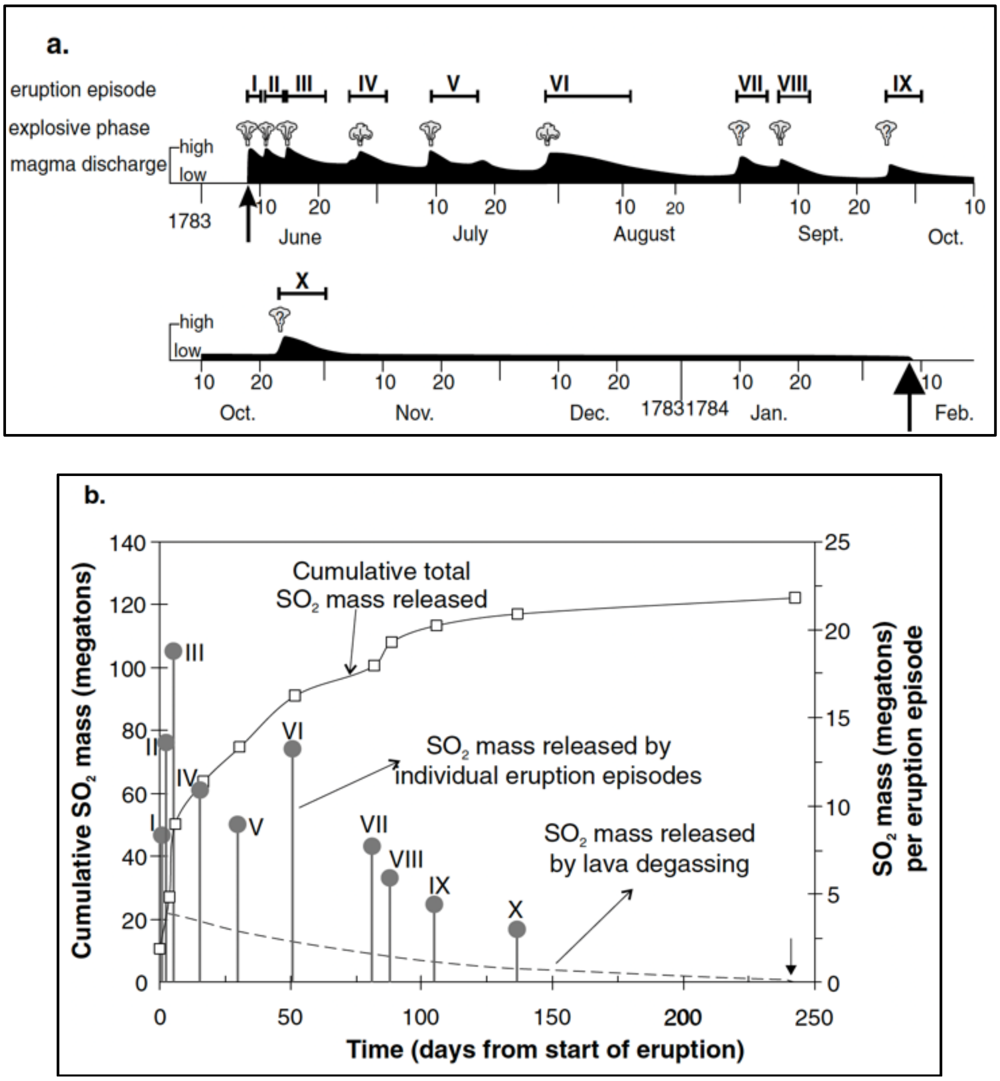


Figure 3. (a) Diagrammatic presentation of the sequence of events during the Laki 1783–1784 eruption showing the timing and duration of the 10 eruption episodes (labelled I–X) as well as the occurrence of explosive phases and fluctuations in the magma eruption rates. Arrows indicate onset and termination of the Laki eruption; eruption clouds denote an explosive phase at Laki fissures; the black curve shows qualitative changes in the magma discharge with time. Horizontal bars indicate the duration of each eruption episode. (b) Atmospheric SO_2 mass loading by the Laki eruption. Solid line and open squares show the cumulative SO_2 output (scale to the left). Vertical bars and filled circles show SO_2 mass released at the vents during each eruption episode and the broken line indicates sulphur mass released by the lava (scale to the right). The arrow to the lower right indicates the end of the Laki eruption. Figure and description from Thordarson and Self (2003).

In contrast to Laki 1783–84, the VEI 7 eruption of Tambora (Indonesia) in 1815 had far wider impacts on climate and severe societal implications. An estimated 100,000 people died due to the various impacts of this eruption, which included the loss of livestock, crop failure, famine and enhanced propagation of diseases including typhus and dysentery (Oppenheimer, 2003; Brönnimann and Krämer, 2016). The climatic forcing of this event caused a global cooling such that the following year is termed “the year without a summer” in many historical records (Harrington, 1992; Trigo et al., 2009; Klingaman and Klingaman, 2013).

3.2 Evidence of past volcanic eruptions

To date, multiple studies have presented quantitative documentation of sulphur peaks in dated ice cores corresponding with the timings of major volcanic eruptions (Toohey and Sigl, 2017; Sigl et al., 2015a; Kunasek et al., 2010). Figure 4 illustrates the concentrations of sulphur in polar ice cores, with peaks of S corresponding with multiple known eruptions, such as Samalas 1257 (both Greenland and Antarctica cores), Laki 1783 (Greenland core) and Tambora 1815 (both Greenland and Antarctica cores). These findings demonstrate how volcanic emissions are capable of reaching and influencing the environment at both regional and global scales.

Ice cores provide a valuable record of changes in atmospheric chemistry over time, from which peaks in sulphur concentrations can be associated with emissions from major volcanic events. Upon deposition to the terrestrial environment, sulphuric acid participates in complex geochemical processes and as a result, sulphur concentrations in lake sediments are not typically a reliable indicator of volcanic gas emissions (discussed further in section 3.2). However, particles of volcanic ash released during an eruption, known as tephra, can be widely dispersed and are found in lake sediments and other natural archives, such as peatlands. The geochemical composition of these particles can be used to determine the timing of the eruption and the volcano of origin by comparing with collected reference data from tephra layers found near the volcano, a method known as tephrochronology (Lowe, 2011). While a large proportion of the particulate material ejected during an eruption falls out of the atmosphere relatively quickly (depending on the size of the particles and the type and severity of the eruption), microscopic tephra known as cryptotephra remains in the atmosphere for longer and can be transported and dispersed over great distances. Icelandic cryptotephras have been found in North-west European and Scandinavian lakes and peatlands and shed light on the distribution of material from multiple VEI 4–6 events (Lawson et al., 2012).

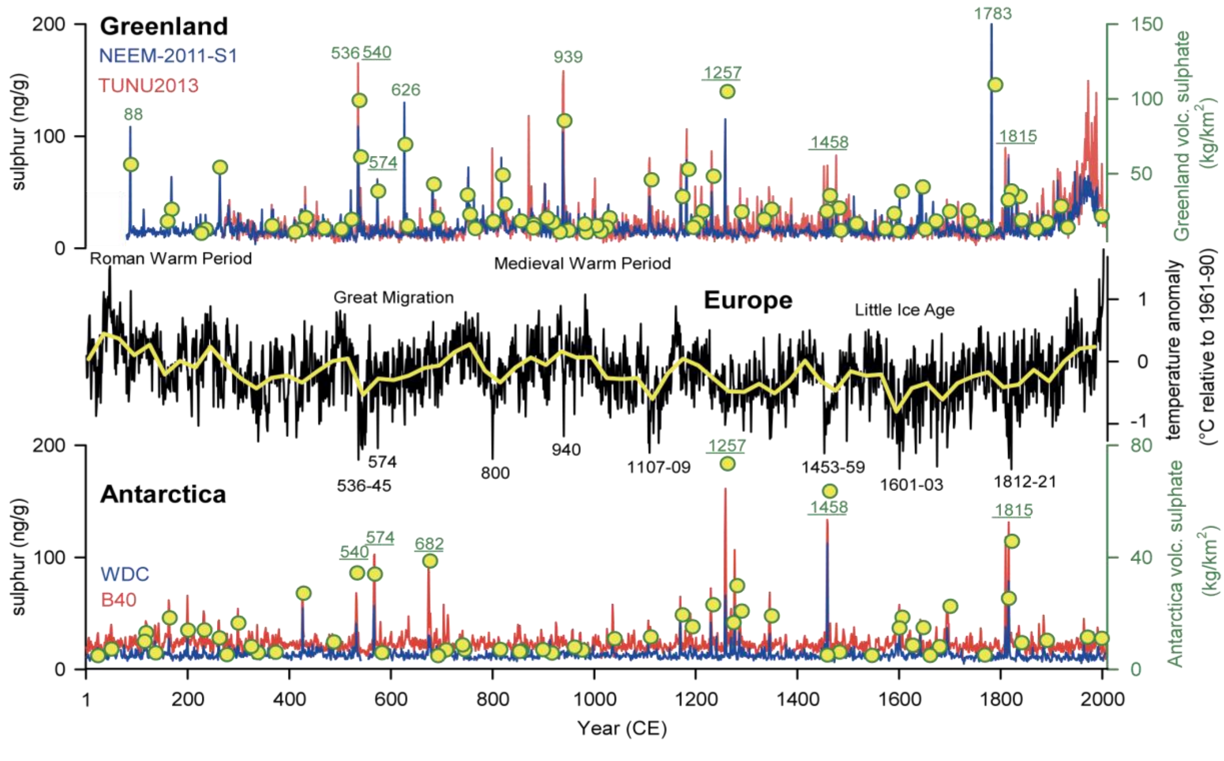


Figure 4. Sulphur concentration records from two ice cores in Greenland and two ice cores in Antarctica, shown together with a temperature reconstruction. From Sigl et al. (2015b).

The deposition of tephra has consequences for a lake and its catchment. Tephra deposition can result in the acidification of catchment soils and lake-waters depending on the buffering capacity of the catchment and the chemical composition of the tephra (Ayris and Delmelle, 2012). Likewise, tephra can also provide additional nutrients to the catchment, which may help to stimulate productivity during the recovery from acidification (Carrillo and Díaz-Villanueva, 2021). The physical effects of tephra deposition include an initial increase in lake-water turbidity (causing reduced light penetration) and in more extreme cases, the burial of vegetation in the catchment (Arnalds, 2013).

According to calculations by Thordarson and Self (1993), only 2.6% of the volume erupted by Laki in 1783–84 was in the form of tephra. Tephra dispersal from Laki was relatively localised around Iceland and evidence of cryptotephra from the eruption is limited, having only been found in ice cores in Greenland (Fiacco et al., 1994) and Svalbard (Kekonen et al., 2005). Likewise, tephra from the Indonesian eruption of Tambora 1815 has yet to be confirmed in Greenland ice-cores (Abbot and Davies, 2012), although ashfall was reported up to 1300 km away from the volcano (Stothers, 1984). While it is important to note that the deposition of large quantities of tephra can have serious ecological consequences, the impacts of Laki 1783–84 and Tambora 1815 in Europe were primarily related to acid deposition (resulting from gas emissions) and climatic impacts, respectively.

3.3 Volcanic emissions

Emissions from volcanic eruptions, as well as passive degassing during regular, more stable activity and magma flows, consist primarily of water, carbon dioxide and sulphur gases (mainly as sulphur dioxide, SO_2 and smaller amounts of hydrogen sulfide, H_2S). Smaller, but significant amounts of other gases are emitted, including chlorine gases (as hydrogen chloride, HCl, and less commonly, chlorine gas, Cl_2), hydrogen fluoride (HF) and minor amounts of nitrogen (N_2) and methane (CH_4).

The eruptions of Laki 1783–84 and Tambora 1815 released significant quantities of gas to the atmosphere. Unlike what may be envisioned as a typical eruption (i.e. originating from a localised cone or vent and lasting for only a brief period), the eruption of Laki 1783–84 occurred along a 27 km long fissure and lasted for 8 months (Fig. 3). As a result, Laki released massive quantities of gas, including an estimated 122 Tg of SO_2 (Thordarson and Self, 2003), in addition to around 15 Tg of HF and 7 Tg of HCl (Thordarson et al., 1996). The eruption of Tambora 1815 released up to 147 Tg of SO_2 and around 49 Tg of Cl and 20 Tg of F to the atmosphere (Pouget et al., 2023). Compared to Laki, the eruption of Tambora was particularly violent and resulted in significant stratospheric SO_2 loading, causing strong climatic impacts.

As the focus of this Ph.D. thesis is on the impacts of these two eruptions on distal ecosystems, it is important to understand how the gases released during an eruption behave in the environment. In the following sections, the environmental behaviour of acid-forming volcanic gases is first outlined, followed by a summary of their biological and ecological significance upon deposition to the terrestrial environment as acids. Acid deposition also has consequences for iron dynamics in the terrestrial environment, which is also discussed below.

3.3.1 Sulphur in the environment

The main sources of sulphur to the atmosphere are volcanic emissions, including those released during eruptive episodes and passive degassing, and fossil fuel emissions. Volcanic sulphur emissions mostly consist of SO_2, H_2S, and to a lesser extent, carbonyl sulphide [COS or OCS] and carbon disulphide [CS_2]. Other major sources of S are biogenic, where microbial decomposition releases H_2S and oceanic phytoplankton contribute large amounts of dimethyl sulphide [$(CH_3)_2S$] (DMS) and dimethyl disulphide [$(CH_3S)_2$] (DMDS) to the atmosphere – all eventually oxidise to H_2SO_4 (sulphuric acid) (Wallace and Hobbs, 2006). Unlike the relatively gradual changes in anthropogenic and biogenic S emissions over decadal time scales (Fig. 5), volcanic emissions of S are highly variable and are characteristically both chronic (passive degassing) and acute (eruptive expulsion) in nature.

Updated measurements of passive volcanic degassing using satellite instruments (as compared to the previous, intermittently and inconsistently obtained field measurements, e.g., Andres and Kasgnoc, 1998) reveal annual volcanic S emissions of approximately 23 ± 2 Tg over the period of 2005–2015 (Carn et al., 2017). This amounts to roughly 9% of all S emissions, along with biogenic and anthropogenic contributions, which account for 15% and 76%, respectively (Bleam, 2016). Figure 6 shows the contribution of both eruptive and passive volcanic SO_2 to the atmosphere between 2004 and 2016 (Carn et al. 2017). While passive SO_2 emissions are greater, eruptive contributions are highly variable. Should a major sulphur-rich eruption have occurred during this time, the eruptive SO_2 release would likely exceed passive emissions, with implications for the region(s) affected by its deposition. Key to considering these implications is an understanding of the mechanisms of transport of S and its behaviour in the environment upon deposition.

Wet deposition of S follows the dissolution of gaseous SO_2 into atmospheric water droplets (Fig. 6). The following equilibria then apply (Jones et al., 2015):

$$SO_2 \bullet H_2O \leftrightarrow HSO_3^- + H^+$$
$$HSO_3^- \leftrightarrow SO_3^{2-} + H^+$$

Following the formation of $SO_2 \bullet H_2O$, HSO_3^- and SO_3^{2-} in atmospheric water droplets, these compounds are oxidised to sulphate and eventually to sulphuric acid (H_2SO_4). The rates of these processes are dependent on the oxidant (common oxidisers include H_2O_2, O_3) and pH (Wallace and Hobbs, 2006). Once deposited, sulphuric acid dissociates into H^+ ions and SO_4^{2-}, leading to decreased pH in waters and soils. However, a decline in lake-water pH is not a given result of acid deposition. As shown by Siver et al., (2003), enhanced bacterial sulfate reduction in response to acid deposition may lead to increased lake-water alkalinity sufficient to negate some of the effects of acid deposition on algal communities.

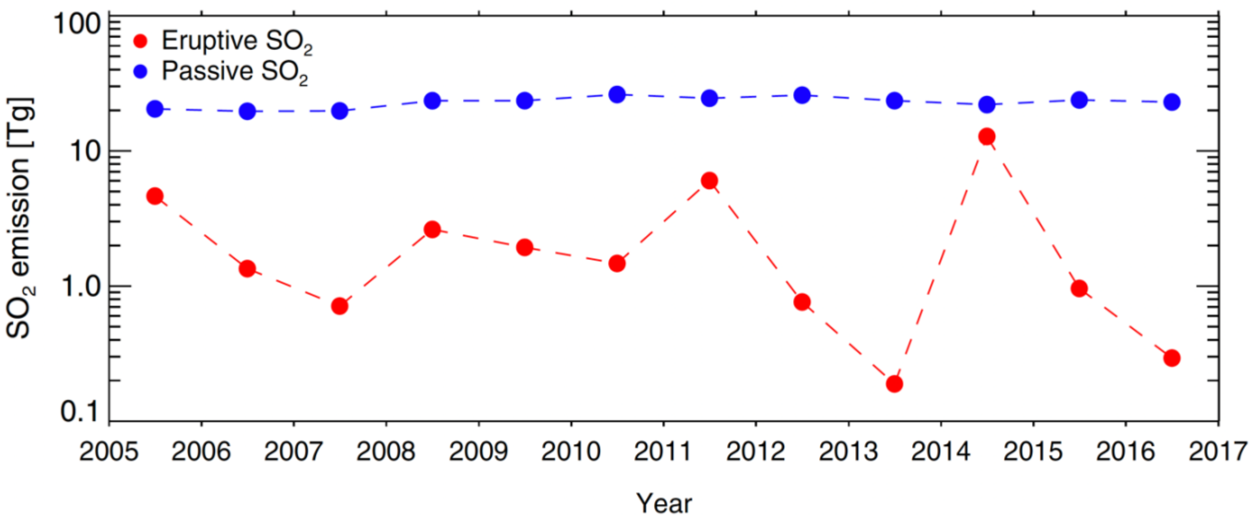


Figure 5. Total, global eruptive and passive volcanic SO_2 emissions in 2004–2016. From Carn et al. (2017).

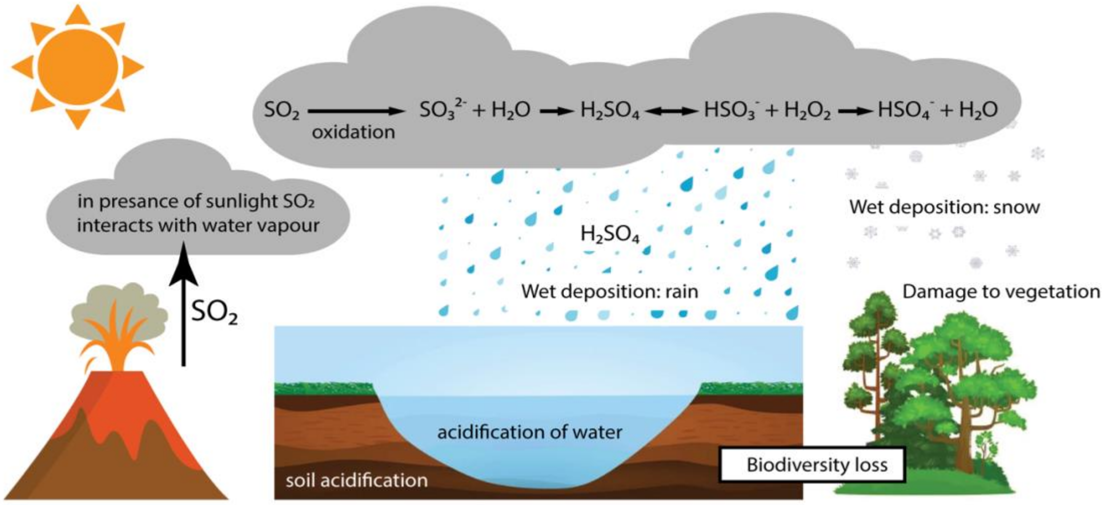


Figure 6. Schematic representation of the pathway of acid rain formation (wet sulphur deposition) and consequential effects. Redrawn from Prakash et al. (2023).

Volcanic injection of SO2 is the largest source of stratospheric sulphur and a significant contributor of volcanic climate forcing. A large explosive potential is required for plumes to penetrate the troposphere-stratosphere boundary (tropopause) and significant injections are generally restricted to ≥VEI 4 eruptions, as the plumes of less explosive eruptions are rarely capable of penetrating the tropopause. Major Volcanic Stratospheric Sulphur Injection (VSSI) accounts for a significant proportion of overall stratospheric loading; with a long-term (500 BCE–1900 CE) annual mean input of ca. 0.5 Tg, and background contributions amounting to ca. 0.014 Tg (Toohey and Sigl, 2017). While tropospheric ash/aerosol presence will also have an effect on albedo, S aerosols persist for much longer in the stratosphere and therefore possess a greater radiative forcing. As such, the climate forcing of major explosive eruptions (of VEI ≥4) is potent. Figure 7 shows the reconstructed temperature anomalies following the VEI 7 eruption of Tambora in 1815 compared to the years 1779–1808, a relatively stable period in terms of stratospheric sulphur loading.

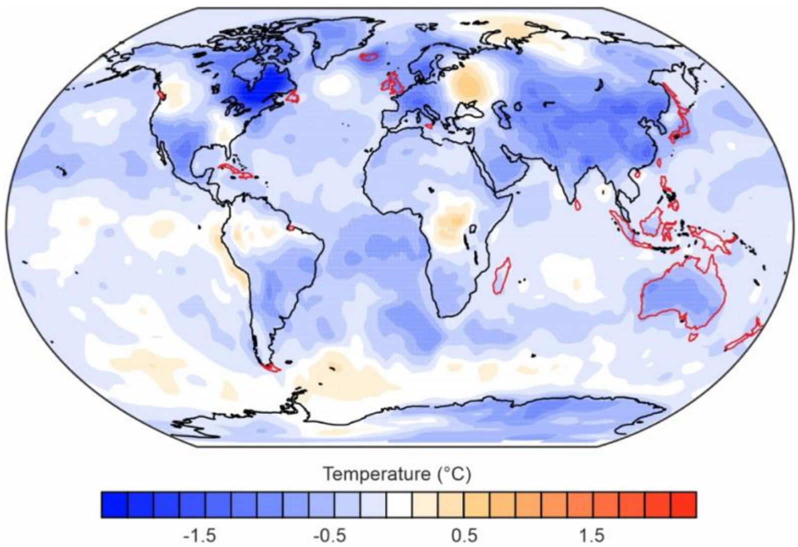


Figure 7. Reconstructed temperature anomalies in 1816 (“the year without a summer”) relative to the “non-volcanic” reference period (1779 to 1808). From Wilson et al. (2023).

Acidification of rainwater has abiotic consequences ranging from the weathering of surface elements (e.g., exposed rock faces) to significant geochemical changes in soils. Biotic consequences vary depending on the severity and duration of exposure. Acid scolding can cause the partial or complete loss of leaf tissue, resulting in impaired photosynthetic productivity and can ultimately lead to death of vegetation. Death can also occur following changes in the soil, with less tolerant vegetation responding poorly to minor changes in pH. Vegetation adapted to acidic soils may still succumb to changing nutrient dynamics and soil chemistry. A common reason for this is the leaching of aluminium from the soil following acid rain, which can be toxic towards plants and impair the growth of roots as well as the uptake of nutrients and water (Panda et al., 2009).

During more severe acid deposition such as may follow a large volcanic eruption, the death of vegetation can release additional nutrients to the soil in the short term. This can result in the input of organic carbon to lakes, transported mostly by water runoff, with input of both particulate organic matter (DOM) and dissolved organic carbon (DOC), as well as other bioavailable nutrients associated with detrital mineral matter supply to lakes. Acidification of soils is also known to enhance the mineralization of nitrogen and therefore increase its availability for vegetative uptake (Johnson et al., 1982). However, more extreme mortality (i.e., whole-organism death following the initial loss of leaves) will likely release an initial pulse of nutrients to the catchment followed by an extended period of disturbed nutrient cycling, in part due to reduced productivity until the vegetation recovers (Takahashi et al., 2007).

Acid rain is a significant contributor of freshwater acidification, with a range of consequences for aquatic biota (Schindler, 1988). Generally, ecosystem health and biodiversity decrease with increasing acidity in freshwater ecosystems. Freshwater fish in particular tend to have a narrow range of tolerance towards acidity (Baldigo and Lawrence, 2000; 2001; Tammi et al., 2003) and individual species may cease to inhabit freshwaters through death or migration following rapid changes in pH. Indirect effects on species of higher trophic levels can stem from changes in the biota of the lower trophic levels, with prey species including macroinvertebrates and planktonic biota sensitive to changes in pH.

3.3.2 Acid-forming halogens in the environment

Volcanic eruptions can release significant quantities of chlorine (Cl) and fluorine (F) to the atmosphere, both of which are halogens (salt-forming elements), in the forms of hydrogen chloride (HCl) and hydrogen fluoride (HF) gases, respectively. Both HCl and HF readily dissolve in water vapour, forming acidic aerosols:

Formation of hydrochloric acid:
$HCl + H_2O \leftrightarrow H^+ + Cl^-$

Formation of hydrofluoric acid:
$HF + H_2O \leftrightarrow H^+ + F^-$

The main source of chlorine in the atmosphere can be broadly categorized into natural and anthropogenic sources. By far the largest chlorine source is sea spray, primarily in the form of sodium chloride (NaCl) with an estimated emission of 5200–15000 Tg Cl Yr^{-1}, approximately 10% of which is deposited on land (Friend, 1990). Volcanic emissions of chlorine gases contribute ca. 0.4–11 Tg Cl Yr^{-1} (Friend, 1990). Anthropogenic sources such as coal combustion and industrial processes emit much smaller amounts of around 3 Tg Cl Yr^{-1} (Friend, 1990). Fluorine emissions are far smaller, with sea spray, soil dust and anthropogenic emissions accounting to less than 3 Tg Yr^{-1} combined (as both F and HF), although volcanic eruptions emit between 0.06 and 6 Tg HF Yr^{-1}.

The behaviour of sulphuric, hydrochloric and hydrofluoric acids upon deposition in the environment are related to their mobility - although all commonly cause acidification. Chloride ions are particularly mobile and are easily leached from soils, while sulphate ions tend to bind more readily to soil material, reducing their mobility. Fluoride ions rapidly bind to minerals in the soil and bioaccumulate in vegetation, causing lasting toxicity. Both Cl and F interact with nutrients and metals in soils, ultimately influencing biogeochemical processes, resulting in various ecological consequences (Friend, 1990). To date, no chlorine or fluorine signal in lake sediments has been directly attributed to a past volcanic eruption, nevertheless the potential impacts of these halogens in the terrestrial environment are significant.

3.3.3 Iron in the environment

The main sources of iron to the atmosphere are volcanic tephra, aeolian dust and particles produced by fossil fuel combustion. Iron in volcanic ash is primarily found in insoluble forms such as silicate glass, Fe-bearing silicates and iron-oxide minerals (mainly fayalite/forsterite ($(Fe,Mg)_2SiO_4$), magnetite (Fe_3O_4), ulvöspinel (Fe_2TiO_4) and ilmenite ($FeTiO_3$)) (Schmincke, 2004; Ayris and Delmelle, 2012) and comprises 2–8 wt% of ash, depending on the magma composition (Rogers and Hawkesworth, 2000). During an eruption, the mixing of magmatic gas, ash and ambient air, results (upon cooling) in the formation of a soluble layer containing ferric (Fe^{3+}) and ferrous (Fe^{2+}) iron on the surface of the otherwise insoluble Fe-bearing particles, which makes the iron more bioavailable (Hoshyaripour et al., 2014). This process is dependent on magma composition, temperature and chemical reactions in the ash cloud (Fig. 8) (Ayris and Delmelle, 2012). While ferric iron has low solubility and tends to precipitate as ferric oxyhydroxides (Liu and Millero, 2002), ferrous iron is more soluble and remains bioavailable for longer.

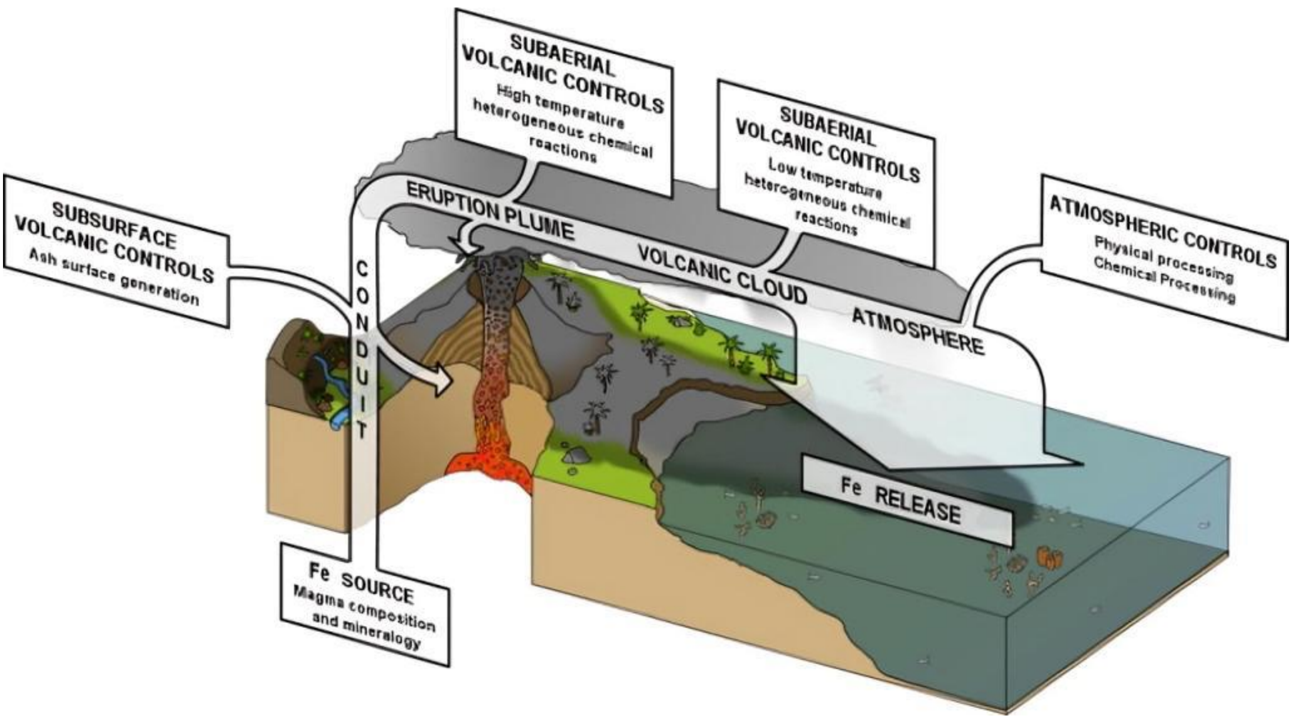


Figure 8. Volcanic and atmospheric control of iron solubility in volcanic ash. From Ayris and Delmelle (2012).

Large explosive eruptions have an important role in iron loading to the environment. The average volcanic ash emission is estimated to be ca. 176-256 Tg Yr $^{-1}$ (Durant et al., 2010). Approximately one third of the ocean is iron limited as well as some freshwater environments (Breitbarth et al., 2010; North et al. 2007, 2008). Several studies have highlighted the influence of iron input via the deposition of volcanic ash on the productivity of marine phytoplankton (Olgun et al., 2011; Duggen et al., 2010; Lindenthal et al., 2013). While neither Laki 1783–84 nor Tambora 1815 resulted in significant ash deposition outside of Iceland and Southeast Asia, respectively, the importance of volcanic ash deposition in relation to iron and nutrient availability highlights the broader environmental influence of volcanic ash on marine and iron-limited ecosystems.

Even in the absence of ash, acid deposition can have a significant influence on iron dynamics in the terrestrial environment. Acidic conditions increase iron solubility in soils, consequently increasing the amount of iron transported to freshwater ecosystems, including streams, rivers and lakes, where it is involved in various chemical and biological processes. While freshwater ecosystems tend not to be limited by iron, the release of iron to lakes can alter nutrient cycling, particularly in relation to phosphorus availability. Iron can bind with phosphorus (which is a key limiting nutrient), reducing its bioavailability, which can result in reduced primary productivity (Reynolds and Davies, 2001). However, in the scenario of volcanic acid deposition, increased iron mobility primarily results from sulphuric acid deposition, which brings greater levels of sulphur to the system. Under acidic conditions, shifts in this balance can increase the availability of P in the water column and ultimately stimulate primary productivity. The biogeochemical processes involving S, Fe and P are far more complex than described above, but these fundamental relationships highlight the sensitivity of freshwater ecosystems to acid deposition and the complexity of the processes affecting nutrient dynamics.

3.4 Diatoms as indicators of ecosystem functioning

3.4.1 Description of diatoms

Diatoms are single-cellular photosynthetic eukaryotes, present in nearly all water bodies on Earth (Armbrust, 2009). Diatoms form a significant proportion of the algal biomass in both marine and freshwater environments, contributing to the regulation of elemental cycles including those of silica, carbon, oxygen, nitrogen and phosphorus. Diatoms collectively account for an estimated 20% of global oxygen production (Chapman, 2013) and are significant producers of organic carbon, which makes them a vital food source for higher organisms (Armbrust, 2009). Belonging to the phylum Bacillariophyta, diatoms have silica-based (SiO_2) cell walls and are found in a variety of forms and colonial arrangements (Smetacek, 1985; Reynolds, 1984; Sournia, 1982). Diatoms consist of two valves, or halves, which join to form a frustule. The larger valve (epitheca) and smaller valve (hypotheca) are joined together by one or numerous hollow girdle bands, which line the connecting edge of each valve (Kröger, 2007). Diatoms can be benthic – located at the bottom of the water body, periphytic – attached to other flora or protrusions emanating from the bed, or planktonic – located within the water column (Fig. 9). Additionally, tychoplanktonic species are capable of growing either attached to a substrate or freely in the water column, but some species prefer one growth form; for instance, euplanktonic species are primarily adapted for growth in the plankton although they may occasionally be found attached to a substrate.

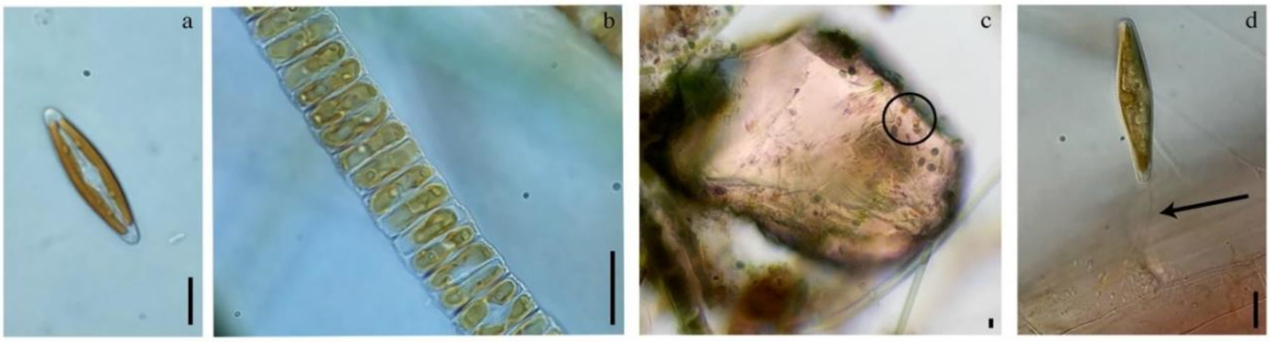


Figure 9. Different growth forms of freshwater diatoms: a) a single cell of *Navicula:* this diatom has a benthic life style and moves on a substrate through mucilage secretion from a raphe structure, b) a colonial chain of *Diadesmis*: one end of the colony is attached to the substrate while the other end floats freely in the water column, c) diatoms of the genus *Psammothidium* (black circle) attached to a grain of sand: its raphe structure allows it to move along the substrate surface, d) a diatom of the genus *Gomphonema* attached to brown algae via an extended stalk. Scale bar = 10 µm.

Diatoms are identified to species level by systematic observation of their morphological characteristics. However, species of the same genus can share similar characteristics with others, making conventional identification difficult. To simplify the taxonomic process, diatoms are split into two major morphological categories; centrics and pennates (Fig. 10). As the term suggests, centric valves are formed around a central point, whereas pennate valves exhibit a greater length in one plane than the other. Despite extensive research over the past century, very little was known about diatom evolutionary pathways until technologies in recent decades enabled more sophisticated analyses, including DNA sequencing, which has been used to investigate monophyletic relationships between divisions of Bacillariophyta and species exhibiting distinctive morphological characteristics (Medlin et al., 1993). Although DNA analyses have disproven the relationship between traditionally accepted clades, taxonomy still relies on visual identification using taxonomic keys (Medlin et al., 1996). Data-driven methods of automated diatom identification have long been desired (Du Buf, 2002; Du Buf et al. 1999) and methods of automatic identification using machine learning techniques are currently under development (Bueno at al., 2017; Pedraza et al., 2017).

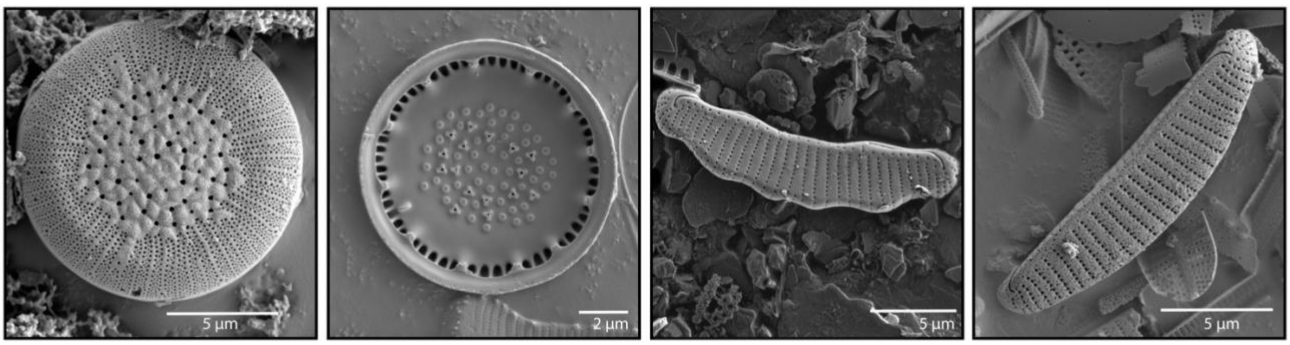


Figure 10. SEM pictures of (from left to right) *Lindavia* from Lake Odensjön, internal view of *Lindavia* from Lake Odensjön, *Eunotia* from Lake Nautajärvii, *Eunotia* from lake Kassjön. *Lindavia* species represent radially centric diatoms, whereas *Eunotia* represent raphid pennate diatoms.

Diatoms account for up to 80% of the species present in terrestrial waters (Sabater, 2010). As such, their prosperity is crucial for sustained oxygen and organic carbon production in a changing environment (Falkowski et al., 1998). Although environmental stress has been linked to the evolutionary development of phytoplankton (Falkowski et al., 2004), anthropogenic management of water resources presents unnatural levels of disturbance, which ecosystems must now endure (Hering et al., 2006), particularly in relation to pollutant input (Rimet and Bouchez, 2011) and increased sedimentation rates (Rowell, 1996).

3.4.2 Diatoms as indicator species

Diatoms have long been used as a tool for assessing the ecological impact of human activities, such as fertilizer and pesticide use in agriculture (Peterson et al., 1994), the monitoring of water supplies and ecological health in relation to oil-extraction practices (Polmear et al., 2015; Hsiao, 1978) and the monitoring of freshwater conditions near wastewater inputs that are both chronic (Moravcová et al., 2013) and acute (Mallin et al., 2007) in nature. They have shown the ability to rapidly respond to changes in the availability of carbon, nitrogen and phosphorus (Brzezinski, 1985). Other conditions diatoms are responsive to include pH, temperature, incident light and salinity. An important nutrient for diatoms, as the main component of their frustules, is silica (particularly in the ocean; Martin-Jézéquel et al., 2000), followed by P, N and C as for many primary producers (Armbrust, 2009).

Of critical importance for this project is the role of changing water acidity. Diatom communities are highly responsive to changes in pH (Fig. 11). Because of their widespread distribution and preference for a variety of conditions, the development and use of changes in diatom assemblages as proxies for lake acidity are well established (Battarbee, 2000; Battarbee et al., 2001). Strongly acidic conditions generally affect diatoms' ability to form frustules, since decreased water pH will decrease silica solubility and therefore its availability for diatoms for frustule formation. However, in freshwater environments, silica is not usually a limiting nutrient (Treguér and De la Rocha, 2013) so fluctuations in acidity may not directly impact diatom productivity in terms of silica, but as mentioned, can influence species composition (Fig. 11). Diatom records can also be used as proxies for both climatic conditions and anthropogenic disturbance (Fig. 12). The sensitivity of diatoms to changing pH may prove useful to detect acid deposition from historic volcanic eruptions. Kokfelt et al. (2016) analysed diatom community changes in a peat sequence in northern Sweden and found some evidence of blooming amongst opportunistic diatom species in response to two volcanic eruptions, Samalas (1257) and Laki (1783–84).

As previously mentioned, diatoms are a crucial source of energy for organisms in the aquatic ecosystem, providing vital fatty acids for smaller fauna (Sterner et al., 1993; Breteler et al., 2005). As one of the most prominent algal groups, changes in diatom communities, specifically their abundance and community composition, can impact energy transfer to higher trophic levels. Changes in diatom community characteristics such as those identified using common ecological metrics (e.g., biodiversity information) can be used as an indication of the health and functioning of the wider ecosystem. Specific proxy methods geared towards environmental variables such as acidity involve the use of transfer functions. These are weighted analyses based on contemporary studies investigating the sensitivities of individual species to changing environmental variables, incorporating data from multiple (often tens of) lake systems. Sweden in particular has been the focus of numerous diatom-incorporating, lake-based transfer functions (Bigler and Hall, 2002; Kahlert and Gottschalk, 2014; Roseacute et al., 2000; Bradshaw and Anderson, 2001; Kovács et al., 2006), which have provided valuable information about the preferences and tolerances of different species to various conditions. For more details on transfer functions, see section 4.8.3 below.

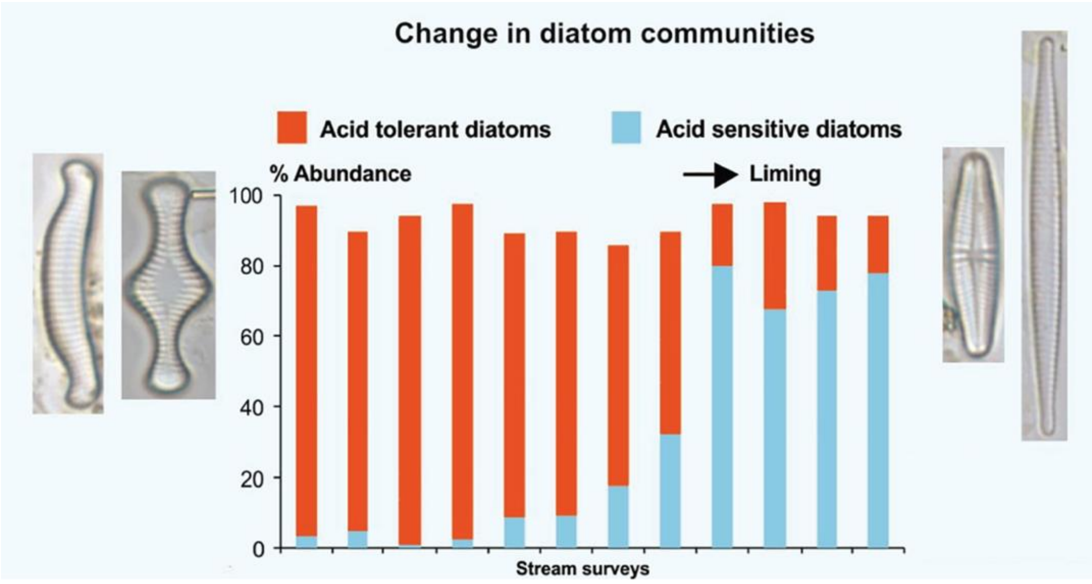


Figure 11. An example of pH effect on diatom community change. The graph represents diatom community change in response to liming (used to reduce stream acidity) in a stream in Wales, showing changes in the abundance of *Eunotia exigua*, an acid tolerant species to *Achnatidium minutissimum*, an acid sensitive species. Modified from Jüttner et al. (2021).

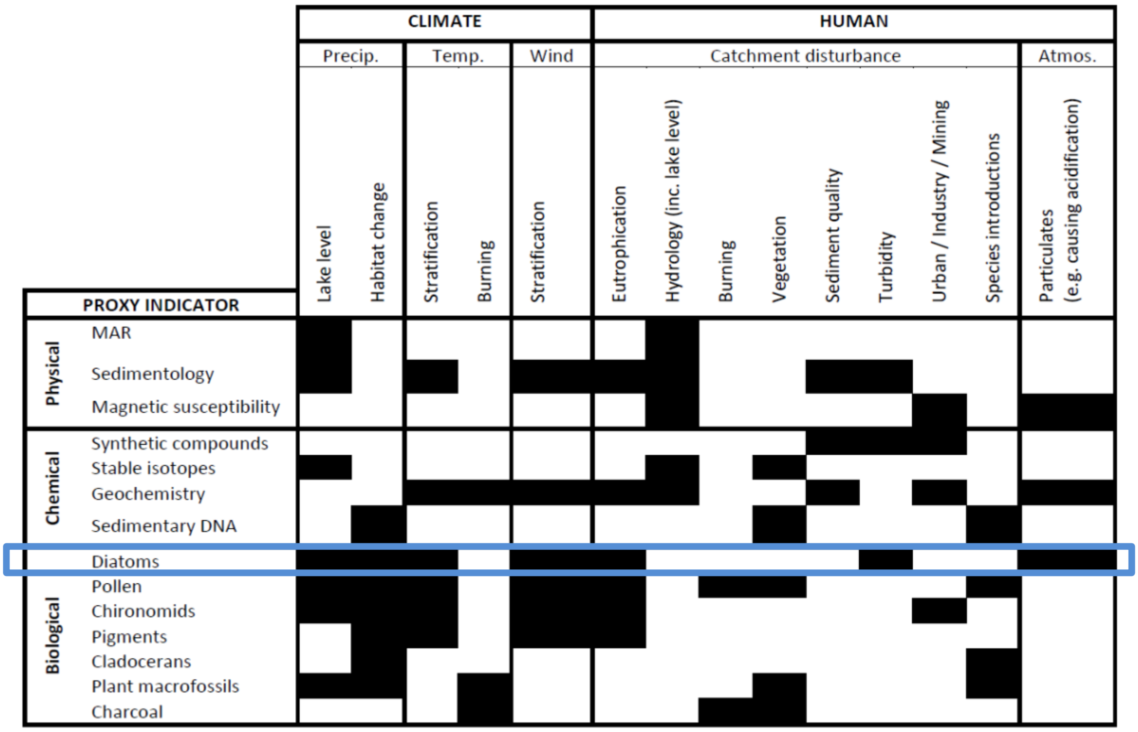


Figure 12. Proxy indicators commonly used in studies of lake sediments and the conditions they may be used to infer. Diatoms are indicated by the blue box. From Mills et al. (2017).

The response of diatom communities in lakes to the effects of volcanic eruptions has been studied previously, although many of these studies focus either on lakes close to the eruption centre or lakes which have been affected by tephra deposition. For example, the diatom population was virtually eliminated from Spirit Lake after the eruption of nearby Mount St. Helens in Washington, USA in 1980. Only a few opportunistic species survived in very low abundance and the community recovery to a state resembling those of similar nearby lakes took about six years (Larson et al., 2006). However, these consequences were connected to massive mudflow directly into the lake and not to fallouts from the traveling ash cloud. The consequences of non-tephratic volcanic acid deposition events on distal lake ecosystems are currently under-studied, which underscores the importance of the research presented in this thesis.

3.5 Varved sediment records

Sedimentary varves are annual layers of material deposited to a lakebed. The main requirement for continuous varve formation is an anoxic (depleted of oxygen / oxygen-free) bottom layer (hypolimnion) in the water column. As organisms requiring oxygen-rich waters are unable to survive under these conditions, this prevents bioturbation (biological disturbance) and allows the unmixed deposits to form sediments with little to no disturbance (Renberg, 1981). The formation and preservation of varved sediments depends on many factors, as illustrated in (Fig.13). Anoxia is not the only factor important in the formation of high-quality varves, for example, better preservation occurs where the system is thermally stratified and less gaseous disturbance occurs from methanogenesis in the sediment (Zolitschka et al., 2015).

Each varve consists of layers, called lamina, or laminae (plural), and commonly consists of two. Laminae contain the material deposited during the spring and summer, and the autumn and winter, respectively. Lake systems vary in complexity, and the varves in some lakes may contain more than two laminae, although the additional laminae tend to be reflected as part of a gradient or transition in the material deposited within one of the two main laminae. Varves may be either organic, clastic, or a mix of the two, depending on properties of the lake and its catchment (Fig. 14). Varved sediments offer a wealth of information about the annual changes in a lake system.

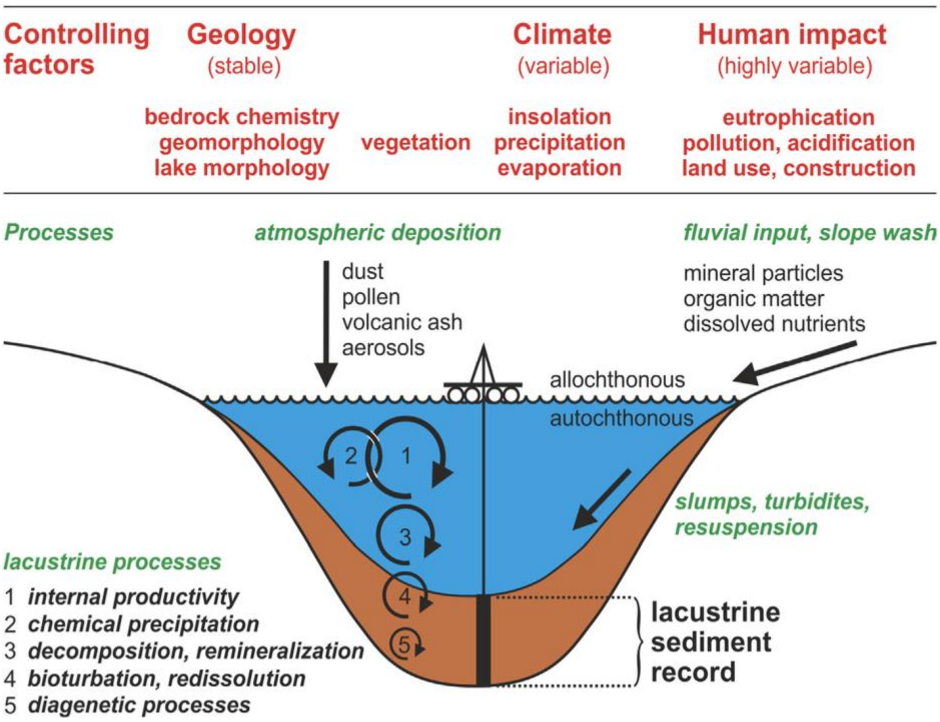


Figure 13. Factors and processes controlling formation of varved sediments. From Zolitschka et al. (2015).

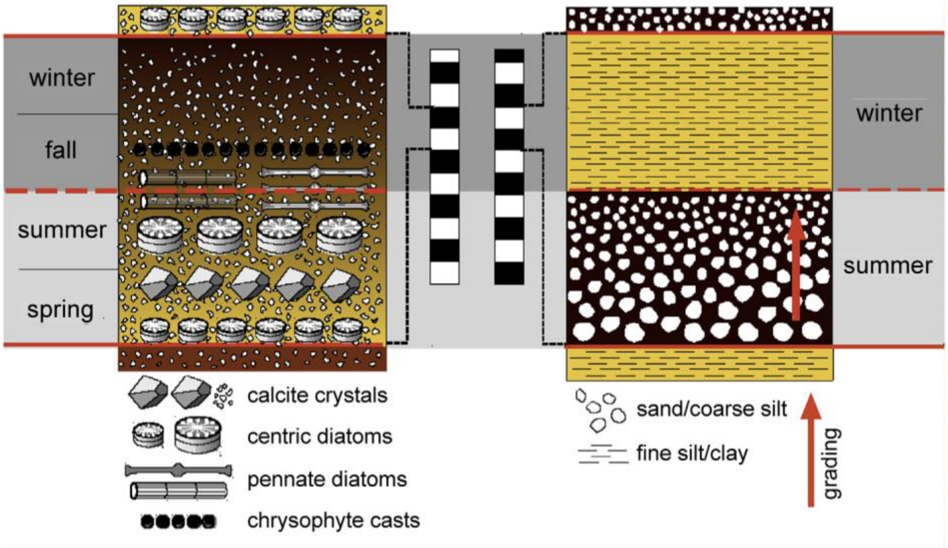


Figure 14. Composition of organic (left) and clastic (right) varved sediments with deposited components of their respective successions of laminae. From Zolitschka et al. (2015).

Varved sediments were described worldwide from 143 lakes and are predominantly localised at higher latitudes (Fig. 15) (Zolitschka et al. 2015). Varved sediments have frequently proven to be powerful archives for recording past volcanic eruptions. For example, tephra layers or cryptotephra horizons have been described from various records, which allowed both absolute age control and between-lake age correlation, contributing to what is now called tephrochronology (Zolitschka et al. 2015).

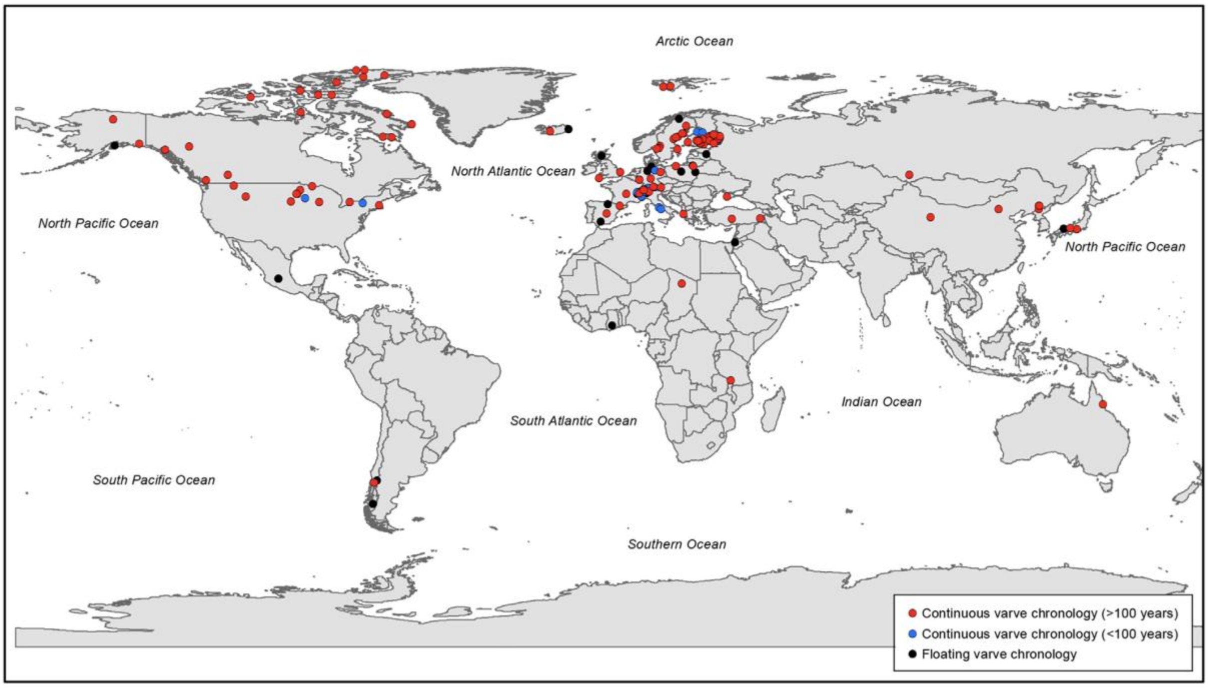


Figure 15. World map showing the distribution of 143 published records of Holocene and Pleistocene varved sediment records. From Zolitschka et al. (2015)

4 Materials and methods

This Ph.D. project focusses solely on the use of varved lake sediment records as these allow the detection of targeted eruptions (by varve counting) and quantification of related chemical and ecological responses at high temporal resolution. By combining the methods described below, the following eruptions were targeted in three lake sediment records:

● **1783-4 Laki, Iceland – VEI 6**. A fissure eruption lasting 8 months (June 1783 to February 1784), with extensive documentation of disruption to society and crop loss in Europe and Scandinavia (Grattan and Charman, 1994; Grattan and Brayshay, 1995). A total of 122 Tg of SO_2 (Thordarson and Self, 2003) and around 15 Tg of HF and 7 Tg of HCl was released to the atmosphere (Thordarson et al., 1996).

● **1815 Tambora, Indonesia – VEI 7**. A highly explosive tropic event which released injected around 60 Tg of SO_2 into the stratosphere (Oppenheimer, 2003; Clyne et al., 2020), resulting in global cooling in the following years. The estimated quantity of total emissions varies between studies. A recent reassessment suggests up to 147 Tg of SO_2 was released in addition to 49 Tg of Cl and 20 Tg of F to the atmosphere (Pouget et al., 2023), although this is amongst the higher end of the estimates.

We hypothesize that acid deposition following the eruption of Laki 1783–84:

1. damaged vegetation in the catchment leading to an increase in organic matter and nutrients supplied to the lake

2. increased the rate of mineral weathering and facilitated the leaching of mobile elements from catchment soils to the lake altered the physicochemical conditions of lake waters with immediate consequences for the biota of the lower-trophic levels, including diatoms

We hypothesise that cooling following the eruption of Tambora 1815 would reflect cooling in the Northern Hemisphere following the eruption and that its potential impacts may be dependent on the severity and persistence of any disturbance attributed to Laki and the subsequent state of recovery of the lake and its catchment.

4.1 Study sites

4.1.1 Odensjön

Odensjön is a small glacial cirque located 62 m above sea level (m a.s.l) in Söderåsen National Park, S. Sweden (Fig. 16). The lake is 140 m wide and is situated in a small catchment with some 200 m between the backwall of the basin, which tops at 95 m a.s.l, and its north-ward-flowing outlet. Odensjön is fed entirely by groundwater and has a maximum water depth of 21 m. The bedrock is composed exclusively of gneiss. The base of the backwall and the sides of the basin contain unsorted, angular scree of ≤0.7 m in diameter. The outlet flows initially through peatland and till. The lake is thermally stratified and contains around 5.5 m of Late Weichselian and Holocene sediments (Berglund and Rapp, 1988).

4.1.2 Kassjön

Kassjön is a small dimictic lake situated 84 m a.s.l in Umeå municipality, Västerbotten, Sweden (Fig. 16), with a maximum depth of 12.5 m. Kassjön has a small volume of 1.24 Mm^3 and covers a surface area of 0.22 km^2. It has four inlets and a well-defined outlet, and its catchment of 6.19 km^2 is dominated by clayey and silty deposits underlain by till and crystalline bedrock. Kassjön contains over 5 m of varved lake sediments at water depths greater than 10 m (Anderson, et al., 1994), dating to its origin around 6350 cal. BP (Petterson, 1999).

4.1.3 Nautajärvi

Nautajärvi is a small dimictic lake situated 103.7 m a.s.l. in Orivesi, southern Finland (Fig. 16). It has a volume of approximately 1.7 Mm^3, a maximum depth of 20 m and contains varved sediments at depths greater than 18.5 m (Ojala and Alenius, 2005). At maximum depth, the thickness of the sediment sequence is ca. 6.6 m and contains a continuous and uninterrupted varve record extending about 10,000 years (Ojala and Alenius, 2005). An outlet from the larger Lake Ristijärvi, which is located to the north-west, feeds one of three inlets from the north and there is one outlet which flows from the south. The catchment is dominated by till with glaciofluvial silt and clay near the lake margins, overlying coarse granitic bedrock.

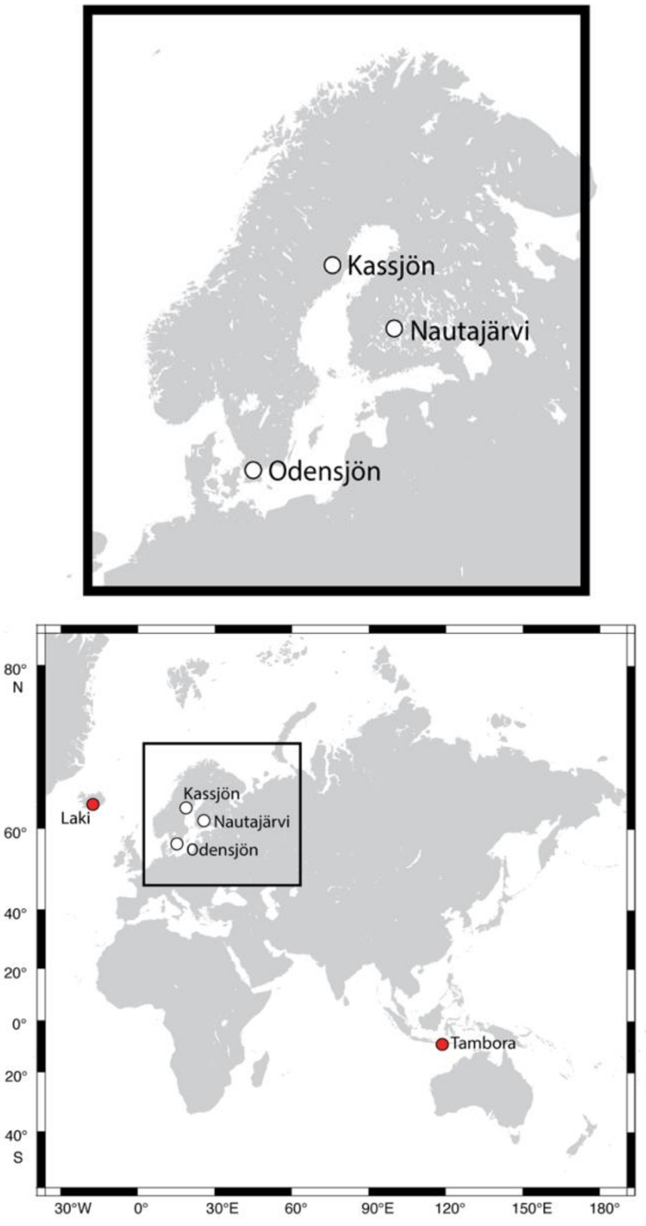


Figure 16. World map with marked study sites and investigated eruptions.

4.2 Core sampling procedure

Cores from all three study sites were obtained using the technique described below. Sediment cores were taken in February 2017 (Odensjön), March 2019 (Kassjön) and March 2021 (Nautajärvi). The sampling was made during winter, when the lakes were covered with a sufficiently thick layer of ice to allow a minimum of two people to safely conduct fieldwork. The areas of maximum depth were located using an echo sounder, assisted by available bathymetry data.

Sediment cores were obtained using a freeze coring device of the type described by Renberg and Hansson (2010) (Fig. 17). Ethanol was added to the hollow wedge of stainless steel situated in the lower section of the device (Fig. 18). Separated from the ethanol by a spring-release lever, the upper rounded section of the device was filled with dry ice. After identifying the absolute depth of the sediment surface using a weighted measuring tape, the device was carefully and slowly lowered such that all but the top 5–10 cm of the wedge sank into the sediments. The device was then fixed at this depth by a steel wire fitted to either a metal tripod or poles resting on the ice. A weight was then dropped, activating the spring lever, which released the dry ice into the ethanol, resulting in an endothermic reaction causing the wedge to rapidly cool to about −80°C and freeze the surrounding sediment sequence onto the outer surface of the wedge.

After a wait of 35–45 minutes the device with the frozen sediment record attached to it was retrieved. The wedge was then detached from the device and warm water poured on its inside surface to assist removal of the (effectively hollow) frozen sediment sequence attached to its outer surface. The outside surface of the frozen sequence was then wiped to remove superficial material collected during retrieval and a picture taken for later reference (Fig. 17). The sequence was then wrapped and stored in a vehicle-powered freezer box for transport to a freezer compartment in Lund.

The sequence was prepared for sub-sampling inside a workable freezer compartment. The sequence was first separated into two via its narrow edges using a bandsaw or crosscut saw. One side of the sequence was further worked on, and its 'twin' duplicate was moved to storage. Both inner (wedge-facing) and outer surfaces were 'cleaned' by removing the outer few millimetres of material using a suitable plane or thin blade; shavings always made in the direction of varves (laterally) for consistency and to reduce the risk of vertical contamination of material

Figure 17. The freeze corer (Renberg and Hansson, 2010) used for sampling (left) and the freshly retrieved sediment sequence from Kassjön in March 2019 (left).

4.3 Varve counting and sediment age determination

Prior to sub-sampling, the surfaces of the sediment sequences were cleaned and photographed. Varves were counted throughout the sequence and pictures of the sequence were taken (Fig. 18). The sediment record of Odensjön was documented in detail for the first time and radiometric dating was therefore also performed to compare with the varve chronology (for more details see Paper I). Both Kassjön and Nautajärvi have well-documented sediment records, which were used as a reference during varve counting. Specifically, Kassjön's sediments contain a distinctive clay layer resulting from a major ditching operation in the catchment in the years 1900–1902 (for more details see Paper II). Sediment sequences from Nautajärvi have been extensively studied (for more details see Paper III) and marker horizons of distinct varves defined in previous studies were marked on images of the core taken for the present study. The varve counting was performed by at least two observers and the associated uncertainties of the varve records were determined by comparison (further details are provided in each of the respective papers).

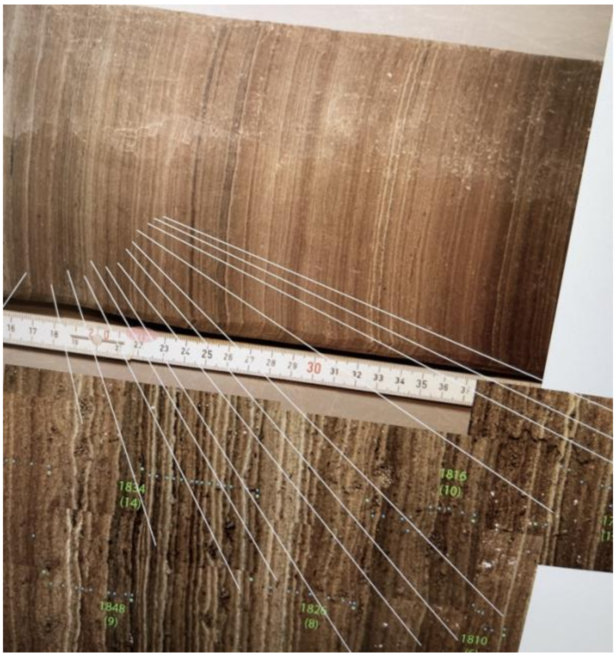


Figure 18. Example of a varve counting procedure.

4.4 Sub-sampling procedures

For the sediments of Odensjön, sampling was performed using a traditional approach. As detailed in Paper I, the core yielded two matching, adjacent slabs after cleaning and cutting, each containing approximately 92 cm of sediments. One of these slabs was used for imaging and varve counting and its sediments were used for radiometric dating. Using two thirds of the width of the second slab, a total of 46 sediment samples were obtained under frozen conditions at 2 cm contiguous intervals covering the full 92 cm. Using the remaining one third of the width of the second slab, an additional 24 samples were obtained at 1-cm intervals between the depths containing sediments encompassing the years of the eruptions of Laki 1783–84 and Tambora 1815 (32 to 56 cm), to assess diatom-assemblage dynamics at greater temporal resolution, of around 7 ± 2 years. To provide complementary information on the seasonal changes in the diatom community, material from seven individual laminae from the uppermost part of the sediment record was also obtained using a ceramic scalpel.

For the sediments of both Kassjön and Nautajärvi, all samples were obtained using a ceramic scalpel inside a freezer room (Fig. 19). Sampling was performed by gently scraping downwards one lamina/varve at a time. All samples were taken contiguously following the clearest visual varve boundary, which is at the boundary between the dark-coloured, predominantly organic material deposited during the winter and the light-coloured minerogenic material deposited during snow-melt when temperatures increase at the transition into spring. During sampling, images of the core were marked to record the exact sample boundaries for later reference. All samples were freeze-dried and homogenised before collection of various aliquots for further analyses.

Figure 19. Sub-sampling of the sediment record from Lake Kassjön.

4.5 Biogeochemical analyses

4.5.1 X-ray Fluorescence Spectroscopy

X-ray Fluorescence (XRF) Spectroscopy is a semi-quantitative technique used to measure the content of different elements in various types of material. For samples obtained from Odensjön and Kassjön, XRF measurements were performed to investigate the changes in the elemental composition of the sediments over time. A Bruker S1 Titan handheld XRF at the Department of Ecology and Environmental Science, Umeå University, was used to obtain measurements of ~200 mg freeze-dried material from each level.

4.5.2 Inductively coupled plasma optical emission spectroscopy and Ion chromatography

Due to the ultra-high resolution of the sub-sampling of the Nautajärvi sediment record, a small amount of material was obtained per sample, which warranted the use of geochemical analysis capable of measuring lower quantities of material. Although it is a destructive method, Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) analysis requires only around 60 mg of material for measurements and was therefore chosen. The measurements were performed at the Department of Biology at Lund University. Following sample digestion with 69% supra-pure HNO_3 in a MARS 6 Microwave Digestion System by CEM, samples were diluted with Milli-Q (ultra-pure deionized water) and analysed using an Optima 8300 ICP-OES system by Perkin Elmer to quantify the amounts of Aluminium [Al], Calcium [Ca], Iron [Fe], Potassium [K], Magnesium [Mg], Sodium [Na], Phosphorus [P], Sulphur [S], Titanium [Ti] and Zinc [Zn]. Additionally, following dilution using Milli-Q water, chloride content was determined using an 861 Advanced Compact Ion Chromatography (IC) system by Metrohm.

4.5.3 Elemental analysis

Total organic carbon (TOC), total inorganic carbon (TIC) and total nitrogen (TN) contents were determined using a Costech ECS4010 elemental analyser at the department of Geology, Lund University. For measurements of decalcified sediments, approximately 5 mg of freeze-dried material from each sample was loaded into Ag capsules and decalcification was achieved via iterative application of 50 µl of HCl in a fume hood until nil effect. Both unprocessed aliquots and decalcified samples (in Ag capsules) were wrapped in Sn capsules for analysis. Elemental TOC/TN ratios were converted to atomic ratios by multiplication with 1.167. This analysis provides information on the organic contributions to the lake sediments. Evidence of changes in the contribution of algal (lower TOC/TN ratio) and terrestrial (higher TOC/TN ratio) organic matter can be used to help identify changes in the functioning of the catchment and lake ecosystem (Meyers and Lallier-Verges, 1999).

4.5.4 Biogenic silica analysis

The measurement of biogenic silica (BSi) is used to determine the proportion of the sediments composed of remains of silicifying species such as diatoms. BSi content was measured following the procedures of DeMaster (1981) and Conley and Schelske (2001). First, a weak base of 1% Na_2CO_3 solution was used for digestion at 85 °C in a shaking water bath. Extractions were taken after 3 h, 4 h and 5 h, and neutralised with HCl before cooling, using frequent duplicates. To quantify the dissolved silicon (dSi) content, ammonium molybdate was added for spectrophotometric measurement of molybdate-blue using a Smartchem 200 (AMS System) wet chemical analyser. BSi concentrations were determined by the intercept of a least-squares regression of extracted dSi data.

4.6 Diatom sample treatment and slide preparation

4.6.1 Sediment digestion

Aliquots of around 15 mg of dry sediment were processed entirely in 50 ml centrifuge tubes in a water bath placed inside a fume hood. To remove organic material, 37% H_2O_2 was first added to the samples which were processed at 80°C for approximately 8 hours in a water bath, or until the organic material was visibly removed. The samples were then cleaned from H_2O_2 by repeated decanting of the supernatant and dilution with ultra-pure deionised Milli-Q water, allowing sediment to settle overnight between dilutions. To remove calcium carbonates, 10% HCl was then added to each sample and processed at 70°C for up to 8 hours in a water bath, followed by removal of HCl using repeated decanting and dilution as stated above.

4.6.2 Use of microspheres

To quantify the concentration of diatom frustules, a known concentration of divinylbenzene (DVB) microspheres was added to each sample, following sediment digestion and testing of pH to confirm neutrality. Diatom concentrations were determined for the studies of Kassjön and Nautajärvi, but not Odensjön, owing to the availability of DVB microspheres. Stock suspensions of a known concentration of DVB microspheres were produced by adding DVB microspheres (initially sourced in dry form) to deionised Milli-Q water, followed by several stages of magnetic stirring and sonication to ensure thorough mixing. The concentration of microspheres in the stock suspension was determined using a Beckman Coulter Z2 particle counter. At least five measurement runs were performed. For each run, 1000 μL (1 ml) of the stock suspension was added to 99 ml of ISOTON diluent (a buffered saline solution), and at least 10 counts (each of 1 ml) were made using the coulter counter. Prior to measurements the stock suspension was again subject to magnetic stirring and sonication, and so were the 100 ml solutions prior to each run to ensure thorough mixing and limit aggregation of the microspheres. Concentrations of the diluted solutions were then calculated and scaled accordingly to determine the concentration of microspheres per 1 ml of the stock suspension. Standard deviations were also calculated and confirmed to be low. Based on an initial inspection of the concentration of diatoms in the samples, a volume of (a known concentration of) the stock suspension was added to each sample. The number of microspheres observed, in addition to diatom valves, was recorded in each sample (detailed in section 4.7). Based on the weight of dry sediment processed (i.e., the weight of dry sediment added to each centrifuge tube), in addition to the number of diatom valves counted and microspheres observed, the concentration of diatom frustules per mg dry weight of sediment was calculated.

4.6.3 Preparation of microscope slides

The processed diatom samples were diluted with Milli-Q water until translucent. A bubble of the solution was placed on glass coverslips using a pipette and the coverslips were then covered to prevent dust contamination and allowed to dry overnight. The glass coverslips were mounted to the microscope slides using Naphrax, a mountant with a high refractive index (of 1.73), on a hotplate at temperatures of around 110–120°C to allow the solvent (toluene) in the Naphrax to disperse, setting the slide.

4.7 Diatom analysis

For Odensjön (Paper I), a minimum of 400 diatom valves were counted for the samples of the individual laminae and the 2-cm resolution samples, and a minimum of 350 valves were counted for the 1-cm resolution samples, achieving a diatom counting efficiency of over 92% (Pappas and Stoermer, 1996). For samples from Kassjön (Paper II), a minimum of 500 valves were counted and for samples from Nautajärvi (Paper III) a minimum of 400 valves were counted. For each slide, upon the encounter of a previously unobserved species within the slide, the number of diatom valves counted at that point was recorded. Species observation rates was plotted using these data and used as an aid to determine if any individual sample required additional counting to accurately reflect the diatom community. Siliceous chrysophycean stomatocysts (Fig. 20) were counted also to provide complementary information on ecosystem conditions (Smol, 1985), in addition to freshwater sponge spicules, synurophyte scales (Fig. 20) and diatom resting cells.

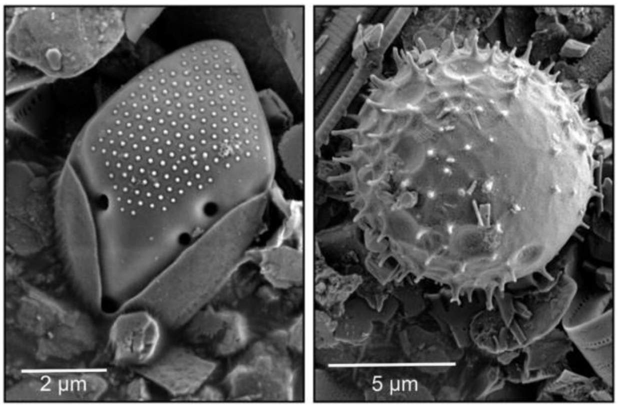


Figure 20. Left – synurophyte, right – chrysophycean stomatocyst, Lake Nautajärvi.

A combination of light microscopy (LM) and scanning electron microscopy (SEM) was used for an initial inspection and identification of the species present in the sediments of each lake. The use of SEM enabled the confident identification of small species which are difficult to identify using LM alone, including those of the order *Achnanthales* Silva. SEM was also used to confirm the identification of species within the genus *Aulacoseira* Thwaites. Multiple resources were consulted for species identification, including the modern guide "Freshwater Benthic Diatoms of Central Europe" by Lange-Bertalot et al. (2017), updated nomenclature including that of Nakov et al. (2015) and various other articles including that of Potapova et al. (2020), which described the illusive species *Discostella lacuskarluki* (Manguin ex Kociolek & Reviers) Potapova, Aycock & Bogan. Species of *Aulacoseira* are notorious for sharing similar morphological characteristics and have resulted in multiple publications and taxonomic workshops in attempts to seek consistency in identification

4.8 Data treatment and numerical analysis

4.8.1 Biogeochemistry

To simplify interpretation and identify patterns within the geochemical data, a principal component analysis (PCA) was performed in Paper I. PCA is a dimension reduction technique, used to capture the main directions of variation of complex data. In papers II and III, the analysis was refined by the use of factor analysis (FA). To assess the factors characterising the changes in sediment properties across samples, biogeochemical data were standardised and included in a principal component-based factor analysis using varimax rotation to maximise the variance of loadings within each factor. Variables were iteratively removed if the sum of squared factor loadings was less than 0.5. Factor analysis differs from PCA in that it focuses on the shared variance among variables, with the aim of revealing the underlying (latent) factors explaining the observed patterns within the data.

Titanium [Ti] is a stable component in clays and other silicate minerals, and is commonly considered to be representative of detrital input from the catchment. Titanium was therefore used as a constraining variable in elemental ratios to enable the comparison between the proportions of other elements, such as Aluminium [Al], Iron [Fe], Potassium [K], Sulphur [S], Silicon [Si] and Phosphorus [P]. Other elemental proxies were also used, for example, the K/Al ratio was assessed in Paper II as an indicator of erosion and chemical weathering, as K is more susceptible to chemical weathering than Al in clay minerals (Burnett et al., 2011).

4.8.2 Diatoms

In all studies, the relative proportion of diatom species in each sample was calculated. Microsphere-based diatom concentrations were calculated in the studies of Kassjön (Paper II) and Nautajärvi (Paper II). To assess changes in diatom community structure, the relative proportion of planktonic (inclusive of tychoplanktonic species) and benthic diatoms was calculated in addition to the relative proportion of centric and pennate species within the planktonic and tychoplanktonic fraction of the assemblage. For all three studies, the relative proportion of diatom frustules to chrysophycean stomatocysts was calculated following Smol (1985). The Shannon-Wiener Index (Shannon and Wiener, 1949) was used in the study of Odensjön (Paper I). For the studies of Kassjön and Nautajärvi, Hill's diversity (N2) of the diatom assemblage was estimated based on equal-coverage for robust comparison between samples (Roswell et al., 2021).

To characterise periods of distinct diatom community composition and define diatom zones, a stratigraphically constrained incremental sum of squares (CONISS) clustering of Hellinger-transformed diatom data was performed, based on the euclidean distances between samples.

4.8.3 Diatom-inferred pH reconstruction

The previous acidity of a lake cannot be directly measured from the sediment or by any other means but can be indirectly inferred from sediment-based proxies, including the composition of the diatom assemblage, which can be used to produce diatom-inferred pH (DI-pH) reconstructions. To enable this, transfer functions are developed by contemporary sampling of surface sediments for diatom observations along with environmental variable measurements at multiple (generally over 20) lakes in a specified geographic region. Based on these data, the statistically significant tolerance and optima for pH and other conditions (including temperature, water physiochemistry and the availability of nutrients such as phosphorus and nitrogen) are determined for each species. This information is then used to reconstruct past lake conditions based on changes in the composition of the diatom assemblage over time.

Diatom-inferred pH reconstructions are typically performed using transfer functions developed near the study site, as this helps to account for the regional differences in species optima. Two pH transfer functions were identified as suitable for this project, a 167-lake training dataset from the Surface Water Acidity Program (SWAP; Battarbee and Renberg, 1990), encompassing England, Scotland, Wales, Sweden and Norway, and a 100-lake training dataset from northern Sweden (Bigler and Hall, 2002). Both reconstructions use species codes according to the European Diatom Database Initiative (EDDI) (Battarbee et al. 2001). The contemporary nomenclature of each of the species observed during this project was compared with the nomenclature used by the EDDI database for harmonisation between the data collected here and that of the transfer functions. Further details are provided for each specific reconstruction in papers I−III.

5 Summary of papers

5.1 Paper I

Silvester E.L., Ljung K., Bindler R., Hertzman H., Lodi G., Hammarlund D., 2024. ***Diatom dynamics during the last six centuries in Lake Odensjön: a new varved sediment record from southern Sweden.*** *Journal of Paleolimnology*, 1–21.

In this article we present a new varved sediment record from Lake Odensjön, southern Sweden, which is the southernmost varved sediment record in Fennoscandia documented to date. As we documented the record for the first time, we produced an age model based on radiometric dating techniques in addition to counting the varves in the record. We identified discrepancies between the two chronologies and ultimately showed sediment ages based on the radiometric age model, in part because the varves were poorly visible in some sections of the record. The sediments were sampled at a sub-decadal resolution, with greater temporal resolution at sediment depths encompassing the eruptions of Laki 1783–84 and Tambora 1815.

The uppermost sediments present varves appearing as couplets of light and dark laminae composed of greater amounts of biogenic silica and organic matter, respectively. The analysis of intact, freeze-dried varves using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) revealed that the varves are predominantly organic in composition. The light laminae represent diatom-rich material deposited in spring through autumn and the dark laminae contain a greater proportion of organic material deposited in late autumn through spring. Diatom analysis of a succession of laminae in the top of the core revealed that *Asterionella formosa* dominates the diatom community in spring and summer and *Lindavia comensis* and *Lindavia radiosa* thrive in late summer and autumn, *Fragilaria saxoplanktonica* is present year-round. No seasonal pattern was found in the proportion of benthic relative to pennate diatom species or the proportion of chrysophyte cysts relative to diatom frustules. Our results show a seasonal pattern in the proportion of centric relative to pennate species in the planktonic fraction of the diatom assemblage. The seasonal diatom trends are primarily influenced by temperature and light availability.

Throughout the record, the diatom assemblage was heavily dominated by planktonic species. Between the late 1400s and mid-1500s, the dominant species in the assemblage shifted from pennates (e.g., *A. formosa*, *F. saxoplanktonica*) to centrics (e.g., *L. comensis*), likely due to improved light availability and stratification associated with a reduction in forest cover, as indicated by pollen data from the same sediment record. Forest reestablishment in the late 1800s reversed this trend, favouring pennate species. Since the 1900s, the diatom community has resembled that of the 1400s, with *A. formosa* and *F. saxoplanktonica* dominating the assemblage, likely due to nutrient enrichment associated with forest regrowth. Both sediment geochemistry and diatom community composition reflect changes in forest cover, nutrient input, and oxygenation, which also impacted varve preservation and dating accuracy.

Finally, we aimed at identifying signals of Laki and Tambora in the sediments. The diatom inferred pH (DI-pH) reconstruction revealed stable, mildly alkaline conditions since the early 1400s in the range of 7.1–8.0 pH units, indicating high buffering capacity in the catchment. We concluded that Odensjön's small catchment and well-buffered waters make it relatively resilient to acid deposition, including the assumed episode following the eruption of Laki in 1783–84. We also concluded that changes in the diatom community around the timing of Laki and Tambora were largely related to anthropogenic changes in land use around the catchment, although a minor response to the Laki eruption may be recorded in the diatom stratigraphy. While no strong signals of major volcanic eruptions were recorded, Odensjön's varved sediments offer the potential to investigate past environmental changes with high temporal precision in future studies.

5.2 Paper II

Silvester E.L., Bindler R., Bigler C., Björnerås C., Ljung K., Hammarlund D., 2024. ***Diatom and biogeochemical changes during recent centuries in a small boreal lake: deciphering the influence of large volcanic eruptions****. Manuscript submitted to Quaternary.*

In this study, we present a multi-proxy analysis of the varved sediments of Lake Kassjön, northern Sweden, with the aim of assessing the impacts of the eruptions of Laki 1783–84 and Tambora 1815. The sediments contain a distinct clay layer, which is associated with a major ditching operation conducted in the catchment in 1900–02. Using this as a marker horizon, we performed a varve count extending to the mid-1600s, with minor uncertainties of ±1 year for varves dated to the period of 1712–1931 and ±3 years for older sediments. Precise sub-sampling of the varves was performed, yielding an average sample resolution of 6 years and we analysed the sediments spanning the years 1641 to 1932. In addition to our main aims, we also assessed the impacts of the major ditching operation in 1900–02 to provide site-specific process understanding.

In the decades prior to the eruption of Laki, the structure of the diatom assemblage and the geochemical properties of the sediments were generally stable. Our findings indicate clear and immediate changes in the diatom community in timing with the eruption of Laki, with a decline in the concentrations of *Tabellaria flocculosa* and an increase in *Aulacoseira nivaloides*. In the decades following Laki, numerous changes were indicated by the biogeochemical records, including enhanced mineral weathering, reduced detrital input and greater iron, sulphur and phosphorus contents of the sediments, reflecting elevated chemical weathering of catchment soils, altered lake-water chemistry and increased nutrient availability. These changes were accompanied by increases in diatom concentration and BSi content, indicative of greater diatom productivity. We produced two diatom-inferred pH (DI-pH) reconstructions and found a general agreement between the two throughout the record. A minor decline in DI-pH was recorded in timing with Laki, although we assessed that the decline was not sufficient to indicate significant acidification of lake waters according to the modern threshold of a decline of 0.4 pH units.

Paper II highlights the value offered by varved sediments for highly resolved studies of targeted time periods with precise chronological control. Our findings underscore the importance of changing land-use in recent centuries for lake ecosystem functioning and highlight the need to consider potentially confounding factors when assessing the hypothesised impacts of natural events.

5.3 Paper III

Silvester E.L., Ojala A.E.K., Kosonen E., Weckström J., Ljung K., Hammarlund D., 2024. ***Diatom and biogeochemical evidence of the Laki 1783–84 eruption in highly-resolved varved sediments of a Finnish lake.*** *Manuscript.*

In this study, we investigated the varved sediments of Lake Nautajärvi, located in southern Finland, with the aims of 1) assessing the impacts of the eruptions of Laki 1783–84 and Tambora 1815, and 2) validating the findings we observed in Kassjön at greater distances from Laki. Facilitated by existing high-quality documentation of the varve chronology, the sediments were sampled at ultra-high resolution, of 1- to 2-years per sample, covering the period 1766–1844.

Through a combination of diatom and geochemical analyses we documented multiple changes in the lake ecosystem occurring in timing with the eruption of Laki 1783–84. Specifically, we observed an increase in the concentrations of diatom resting cells and sponge spicules, predominantly gemmules, suggesting unfavourable conditions in the lake. No indications of enhanced chemical weathering in the catchment were inferred by geochemical proxies. Changes in nutrient dynamics following the eruption of Laki were subtle and did not fully explain changes in the diatom assemblage or concentrations in the sediments.

A transient pulse of chlorine occurred in timing with the eruption of Laki and persisted for the duration of changes in diatom community and other biotic indicators of stress. We infer that this pulse of chlorine is likely a result of enhanced bacterial chlorination in catchment soils triggered by acid deposition, resulting in the preservation of organic chlorine compounds in the lake sediments. Further investigation is needed to characterise the forms of chlorine and understand the processes influencing chlorine mobility in the catchment. Nevertheless, our findings show potentially significant evidence of a biological signal of acid deposition which is at present poorly understood.

Paper III highlights the potential of highly resolved sampling of varved sediments to reveal fine-scale temporal changes in lake ecosystems. Our findings underscore the importance of using a variety of geochemical analyses when investigating or attempting to identify poorly documented past environmental changes which may be reflected by subtle signals.

Table 2. Author’s contribution to the papers

	Paper I	Paper II	Paper III
Planning and study design	D. Hammarlund K. Ljung E. L. Silvester	E. L. Silvester D. Hammarlund	E. L. Silvester D. Hammarlund
Literature review	E. L. Silvester	E. L. Silvester	E. L. Silvester
Fieldwork and sediment core collection	D. Hammarlund K. Ljung Y. Rohozin* E. L. Silvester	E. L. Silvester D. Hammarlund R. Bindler	A.E.K. Ojala J. Weckström
Core imaging and sub-sampling	E. L. Silvester	E. L. Silvester	E. L. Silvester
Age model and varve counting	E. L. Silvester D. Hammarlund K. Ljung	E. L. Silvester C. Björnerås	A.E.K. Ojala E. Kosonen E. L. Silvester
Geochemical measurements	E. L. Silvester R. Bindler G. Lodi K. Ljung H. Hertzman	R. Bindler E. L. Silvester K. Ljung G. Lodi*	E. L. Silvester E. Kosonen K. Ljung G. Lodi* S. M. Wisén*
Lab work and diatom sample preparation	E. L. Silvester	E. L. Silvester	E. L. Silvester
Data analysis	E. L. Silvester	E. L. Silvester	E. L. Silvester
Figures	E. L. Silvester Rohozin Y.*	E. L. Silvester	E. L. Silvester E. Kosonen A. E. K. Ojala
Data interpretation and discussion	All authors	All authors	All authors
Lead author	E. L. Silvester	E. L. Silvester	E. L. Silvester
Comments and editing of manuscript	All authors	All authors	All authors

**Contributions from those who are not co-authors*

6 Discussion

The overall motivation of this Ph.D. was to identify and assess the potential impacts of the eruptions of Laki 1783–84 and Tambora 1815 on distant lake ecosystems. With this objective we studied the varved sediments of three Fennoscandian lakes using a multi-proxy study design incorporating both diatom and biogeochemical analyses. Our aims were addressed through the use of statistical methods and paleoecological analyses to uncover the underlying relationships in the data and assess whether the observed changes resulted from volcanic forcings. Through the collection of three papers, we investigated three lake settings within a variable distance from the epicentre of Laki

In Paper I, we documented the upper varved sediments of lake Odensjön in southern Sweden for the first time, covering sediments spanning the past six centuries. Our findings revealed long-term changes in diatom assemblage structure in response to forest cover in the catchment. This study revealed limited evidence of changes in diatom community or sediment properties attributable to Laki, and no evidence attributable to Tambora. Nevertheless, this study details relatively stable conditions in the lake ecosystem in recent centuries. With its small catchment providing a stable setting and its limited exposure to the types of anthropogenic disturbance typically facing lake systems in the modern era (e.g., enhanced soil erosion, chemical pollution or agricultural eutrophication), Odensjön's predominantly organic varved sediments are of high value for future paleoecological studies.

In Paper II, we investigated the varved sediments of lake Kassjön in northern Sweden. In mind of our objectives and in comparison to Paper I, we improved upon our experimental design by 1) sampling the sediments at higher temporal resolution, facilitated by Kassjön's well-presented, unbroken varve sequence, 2) achieving greater chronological control by the use of varve counts and comparison of our data with the well-established varve chronology, 3) calculating the concentrations of diatom frustules in the sediments, facilitated by the use of divinylbenzene microspheres, and 4) producing two diatom-based pH reconstructions for a comparison and assessment of any discrepancies to evaluate their potential to reflect acute and potentially short-lasting acidification. This study revealed significant changes in the structure of the diatom assemblage in timing with the eruption of Laki, coinciding with biogeochemical changes in the sediments, indicative of increased chemical weathering in the catchment and enhanced nutrient availability in the lake. We found little evidence of changes potentially attributable to Tambora, nevertheless, the longevity of changes following Laki may have obscured its signal in our data, if indeed it was present.

In Paper III, we investigated the varved sediments of lake Nautajärvi in southern Finland. In this study, we intended to build upon our findings from Paper II and assess whether they could be validated elsewhere and, in this case, at a further distance from Laki. Building upon the methodology applied in Paper II, we improved our experimental design by 1) sampling the sediments at even higher temporal resolution, facilitated by the unbroken varve sequence in addition to existing high quality documentation of the varve chronology, including high-resolution imagery, x-ray images of epoxy-embedded sediments, numerous marker horizons and a comparison with existing varve counts, 2) counting of additional siliceous sub-fossils, including synurophyte scales, freshwater sponge spicules and diatom resting cells, in addition to diatoms and chrysophycean stomatocysts, and 3) given the low sediment yield at 1 to 2 year resolution per sample, we used ICP-OES and IC analyses to ensure that high-quality geochemical measurements could be obtained despite limited available material, in addition to the other analyses used in Papers I and II. As with Paper II, this study revealed changes in the structure of the diatom assemblage in timing with the eruption of Laki (within acceptable uncertainties of the varve ages). While our findings did not reveal evidence of enhanced chemical weathering in the catchment (as found in Paper II), instead we identified an increase in chlorine concentrations in timing with Laki, coinciding with the changes in the diatom assemblage and multiple other indications of stress in the lake ecosystem.

Several aspects discussed in the papers and further technical aspects are considered in the discussion below.

6.1 Identification of volcanic eruptions in varved lake sediments

The targeted study of sediments pertaining to a specific interval of time in the past requires accurate chronological control. Varved sediments can provide this control, but even an uncertainty of a few years can mean the difference between a confident interpretation of cause-and-effect relationships—such as linking an eruption with observed biogeochemical changes—and the uncertain interpretation of potentially unrelated changes. With the high temporal resolution of sampling achieved in the studies of Papers II and III, we were able to document past environmental changes with great precision. Because no tephra was expected to have been deposited following either of the eruptions, chronological accuracy was required for confidently assessing the substantial ecological and biogeochemical changes we observed in the studies of Papers II and III as potential responses to the eruptions. It was therefore essential, that any potentially significant biogeochemical changes we observed, occurred within an acceptable margin of error (according to the varve chronologies), which was the case for the developments we associated with the eruption of Laki in Papers II and III. Our evaluation was further complicated by the challenge of confidently ruling out other factors, such as human activities in the catchment, which are a major driver of changes in lake ecosystems (Anderson, 2014). Indeed, in the case of Odensjön (Paper I), we found that the primary driver of changes in the lake ecosystem was anthropogenic modulation of forest cover in the catchment.

In the case of Kassjön (Paper II), significant human activity occurred in the region during the period studied (1641–1931) and the surrounding terrain was continuously modified for agriculture. The area of the catchment under cultivation increased significantly from the mid-late 1600's, during a major agricultural expansion in northern Sweden (Segerström, 1990). In previous studies of Kassjön's sediments, this was corroborated by a significant increase in cereal and grass pollen from the 1700's (Segerström et al., 1984; Anderson et al., 1995; Anderson et al., 1996). However, because the previous studies were conducted at coarser resolution than was required for our study, they provide only a general context for the changes in vegetation and land-use inferred from (their) pollen data during the period we studied. The few historical maps available detail an increase in the area of the catchment under cultivation, almost doubling between 1786 and 1852 (Lantmäteriet, 2024). The findings of Paper II show numerous changes attributable to acid deposition following the eruption of Laki (discussed further in sections 6.2 and 6.3), however, given the various indications of human developments in the region from the 1700's, we conclude that the confounding effects of local-land use changes may have simultaneously influenced the lake ecosystem and therefore cannot be fully excluded. In the case of Nautajärvi (Paper III), historical maps detail limited developments in land use during the period we studied, adding confidence to our interpretation of the changes observed in timing with the eruption of Laki.

6.2 Biogeochemical indicators of volcanic disturbance

Changes in the biogeochemical properties of lake sediments reflect the types of materials deposited to the land bed, indicating shifts in catchment inputs and biological and physicochemical processes within the lake. Paper I revealed substantial geochemical changes associated with deforestation in the early 1400s and the return of forest cover from the early 1800s. Specifically, a decline in forest cover in the 1400s (as evidenced by pollen and macrofossil data and corroborated by substantial changes in the structure of the diatom assemblage) was reflected in the sediments by a decline in total organic carbon, representing a reduction in the delivery of organic material from the catchment. Likewise, the return of forest cover in the catchment in the 1800s was reflected by an increase in organic carbon. Because of the relatively well-buffered waters and the stable environment provided by the small catchment, we found limited evidence of the impacts of the eruptions in our study of Odensjön.

In Paper II we found multiple indications of enhanced chemical weathering following the eruption of Laki. We assessed the K/Al ratio as an indicator of erosion and chemical weathering, because K is more susceptible to chemical weathering than Al in clay minerals (Burnett et al., 2011) and lower values of the ratio suggest enhanced chemical weathering. To support our interpretations, we also inspected the K/Ti and Al/Ti ratios to assess how the proportions of K and Al relative to the proportion of detrital minerals in the sediment, for which we considered Ti as a stable component of minerals (resistant to chemical weathering) and used it as a constraining variable. Using these ratios, we showed how changes in the K/Ti and Al/Ti ratios were largely aligned during the period before Laki, suggesting the dominance of physical (erosion) versus chemical weathering, supported by relatively stable, high values of the K/Al ratio. This allowed us to demonstrate that variability and even minor declines in the K/Al were still largely indicative of stable weathering conditions (i.e., no significant change in the rate of chemical versus physical weathering) when the K/Ti and Al/Ti ratios are aligned. In timing with Laki, the K/Al ratio declined to its lowest values, suggesting the dominance of chemical versus physical weathering. At the same time, we observed a divergence of alignment between the K/Ti and Al/Ti ratios, where the K/Ti ratio declined. Based on the above reasoning we were able to argue that together these results reflect a significant increase in the delivery of weathered minerals (depleted of K) from the catchment over the following decades. We associated a rapid change in the Zr/Rb ratio with changes in the source of minerals delivered to the lake, resulting from ditching efforts in the catchment. Accordingly, the stability of the Zr/Rb ratio following Laki provided some confidence that there were no significant human modifications in the catchment which may have influenced the changes we observed. Our findings demonstrate the importance of using multiple proxies to provide complementary support when interpreting geochemical data.

In contrast to Paper II, in Paper III we did not find evidence of chemical weathering using the K/Al, K/Ti and Al/Ti ratios. As mentioned, land-use in Nautajärvi's catchment during the period studied was rather limited and our biogeochemical data indicate stable, gradual changes. The main finding of Paper III was an increase in Cl in timing with the eruption of Laki, concurrent with changes in the structure of the diatom assemblage, which also recovered in timing with the decline in Cl around two decades later. This finding is significant, as it shows two well-aligned, independent types of data indicating changes in timing with the eruption of Laki. To our knowledge, this is the first report of an increase in Cl in lake sediments, unrelated to tephra, associated with a volcanic eruption. Chlorine is not established as a marker for past volcanic acid deposition because it is highly soluble and tends to be poorly incorporated in a stable form in sediments. Further research is needed to identify the specific forms of chlorine present in the sediments and to unravel the processes influencing its mobility in the catchment, incorporation to the lake sediments and post-depositional stability.

6.3 Diatoms as indicators

Diatom-inferred pH (DI-pH) reconstructions typically rely on calibration data obtained in a region near the study site, because this helps to account for the differences in species optima at spatial scales. In all three studies, we produced DI-pH reconstructions based on the transfer function developed as part of the Surface Water Acidity Program (SWAP; Battarbee and Renberg, 1990). Because our objective differs somewhat from the typical use of pH reconstructions, in which trends over longer time periods are investigated, in Paper II we also produced an additional DI-pH reconstruction based on a transfer function developed in northern Sweden (Bigler and Hall, 2002), for an assessment any potential short-term discrepancies. The direction and magnitude of the shifts were more critical to our investigation than an agreement in the values of reconstructed pH, acknowledging that no two pH transfer functions will yield the same species optima. Our results showed that both reconstructions were robust, with low error margins and similar proportions of the diatom populations covered within samples. Despite large changes in diatom assemblage structure in timing with Laki, neither pH reconstruction indicated significant acidification of the lake waters, based on the criterion of a change of 0.4 units following the threshold described by Fölster et al (2007). Overall, DI-pH did not indicate significant acidification in any of the three lakes in timing with the eruptions, despite showing minor declines in timing with Laki.

In Paper II we found a decline of *Tabellaria flocculosa* and an increase in *Aulacoseira nivaloides* in timing with Laki. The decline of *T. flocculosa*, which is generally well adapted to mildly acidic conditions, was an interesting find considering the geochemical indications of enhanced chemical weathering in timing with Laki. However, as shown by Siver et al. (2003), enhanced bacterial sulfate reduction in response to acid deposition may lead to increased lake-water alkalinity sufficient to negate some of the effects of acid deposition on algal communities. Supporting the findings of Caraco et al. (1989), Siver et al. (2003) also suggest that enhanced sequestration of Fe resulting from sulfate reduction can lead to increased availability of phosphorus. The initial increase in *A. nivaloides* in timing with an increase in P/Ti indicated a response to increased nutrient availability in the lake triggered by acid deposition and increased delivery of organic matter from the catchment. Accordingly, *A. nivaloides* responded more positively to increased nutrient availability compared to *T. flocculosa*. In the years following, the increase of *A. pusilla* was suggestive of some recovery in the lake waters and decreased delivery of terrestrial organic matter, indicated by a slight decrease in TOC/TN ratio.

Compared to our findings in Paper II, in Paper III we instead observed an increase in *T. flocculosa* in timing with Laki and a decline in *A. pusilla*, somewhat contrary to our findings in Paper II. While Kassjön and Nautajärvi share similar dominant species, there are significant differences in the proportion and composition of the benthic species in their assemblages, partly related to lake depth. Nevertheless, both findings demonstrate significant changes in their diatom assemblages in timing with the eruption of Laki. In Nautajärvi, we did not observe any indication of chemical weathering in the catchment or a significant increase in nutrient availability. Together considered, our results reflect site-specific responses to the eruptions, likely influenced by catchment settings, soil properties and buffering capacity. An important metric in all three studies was the relative proportion of centric to pennate diatoms within the planktonic (and tychoplanktonic) fraction of the diatom assemblage. Despite different species being dominant in the assemblages of the lakes, this metric captured changes in the assemblage associated with key events including deforestation (Paper I), and the eruption of Laki (Papers II and III).

6.4 Challenges of working with frozen varved lake sediments

Varved sediments facilitate highly resolved studies of past environmental changes with precise chronological control. Multiple obstacles are faced when working with these sediments, from imaging the sediments to counting the varves and ultimately, sampling. After retrieval of the frozen core, it is customary to take images which are then used as a reference when sampling the sediments and for varve counting. Imaging of sediments with thick varves can be achieved easily at room temperatures (for a short period) with normal equipment, as was the case in Paper I. We attempted a varve count of Odensjön's sediments, however in some areas the varves were poorly presented which prevented counting in some sections of the core, and we therefore adjusted for this (Fig. 21). We then compared our adjusted varve chronology to our radiometrically based age-model, finding some discrepancies.

Our findings from Paper I suggest that predominantly organic varved sediments like those of Odensjön may not present clearly distinguishable varves, despite containing well-preserved sediments, partly because of the small mineral component. The use of surface-scanning techniques would be particularly beneficial when establishing the chronologies of heavily organic varved sediments. Techniques compatible with methodologies which require sediments to remain frozen for later sub-sampling, including CT scanning and magnetic susceptibility, would allow for high-resolution, non-destructive analysis capable of complementing traditional varve counting approaches in future research.

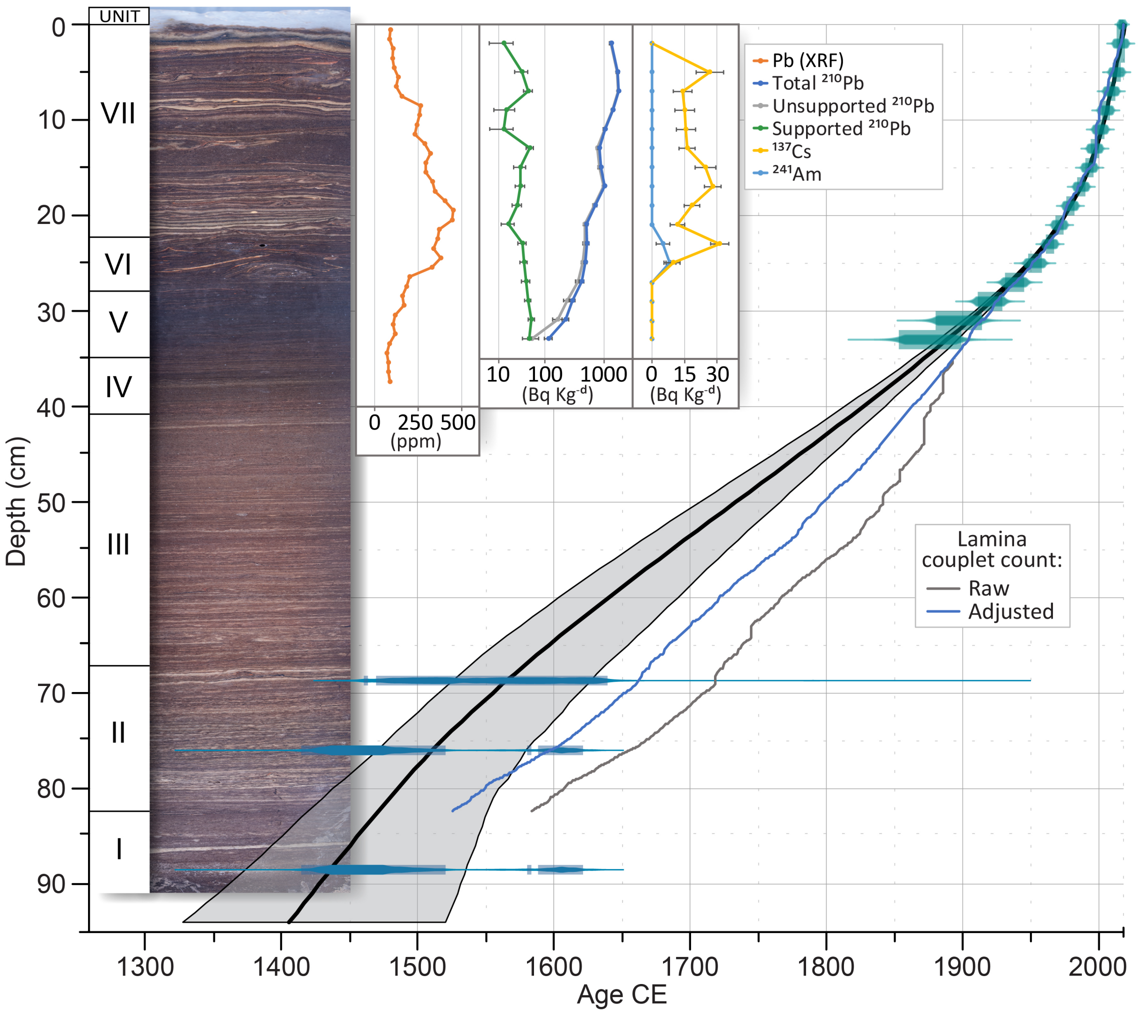


Figure. 21. Age-depth model (black line with uncertainty envelope at double standard deviation level in grey) for the studied sediment sequence from Odensjön based on 210Pb (green bars with uncertainty envelopes) and 14C dates (blue bars and lines representing 68% and 95% probability envelopes, respectively). Lithostratigraphic units I–VII are shown next to a photograph of the sediments. Inset graphs show 210Pb, 137Cs and 241Am data used for constraining the 210Pb CRS model in addition to XRF-based Pb data for the uppermost 37 cm, yielding independent support to the radiometrically based age model. Grey and blue lines represent ages based on cumulative counts of lamina couplets, where the grey line is the raw count and the blue line represents the adjusted lamina record.

7 Conclusions

1. The newly documented varved sediments of Lake Odensjön offer large potential for future studies of past environmental changes. The record reveals gradual, yet substantial changes in the diatom assemblage during the past six centuries reflecting changes in forest cover.

2. Odensjön's small catchment provides relatively stable and contained conditions, largely unaffected by external influences. Our findings suggest that the conditions for varve formation were consistent throughout the record, although, the predominantly organic varves are not visually distinguishable in some areas. Future research should make use of scanning techniques capable of detecting changes in sediment composition to assist varve counting.

3. The proportion of centric to pennate species within the planktonic fraction of the diatom assemblage is a powerful metric capable of reflecting changes in the assemblage associated with disturbance in the catchment and physicochemical changes in the lake waters. Nevertheless, interpretation requires careful consideration of the species present and their preferences.

4. Substantial changes in the diatom assemblage occurred in lakes Kassjön and Nautajärvi in timing with the eruption of Laki, with elevated productivity among dominant species persisting for multiple decades. Nevertheless, the ecological effects were different, with *Tabellaria flocculosa* responding negatively in Kassjön, but positively in Nautajärvi. Likewise, species of *Aulacoseira* responded positively in Kassjön, but negatively in Nautajärvi.

5. Our findings suggest that similar diatom assemblages may have different responses to the effects of acid deposition depending on the catchment settings. The extent to which acid deposition affects nutrient dynamics will influence the type and persistence of its impacts on the aquatic ecosystem.

6. In Lake Kassjön, changes in nutrient dynamics consistent with acid deposition resulted in multiple decades of succession in the diatom assemblage following the eruption of Laki, and geochemical proxies indicated enhanced weathering of minerals in the catchment.

7. In Lake Nautajärvi, changes in the diatom assemblage were observed in timing with the eruption of Laki, coinciding with other biotic indicators of stress. A pulse of Cl in the sediments suggests complex changes in the catchment warranting further research.

8. This research provides valuable insights into how acid deposition resulting from major volcanic eruptions can affect lake ecosystems, demonstrating that responses vary across different environmental settings, depending on various catchment characteristics. The findings demonstrate the importance of using a multi-proxy approach to decipher the influence of natural and human disturbances. Our findings demonstrate the potential of high-resolution sampling of varved lake sediments to reconstruct past environmental changes with precise temporal resolution.

8.1 Popular summary

Volcanic eruptions are powerful natural events which have been documented throughout geological history. Recent eruptions have shown how volcanic activity can impact our lives and can have significant consequences for the natural environment. However, while we might consider these recent eruptions to be disastrous, they are small in comparison to the eruptions which affected societies only a few hundred years ago and may occur again. These massive events had far more severe consequences and resulted in widespread crop failures and the loss of thousands of human lives. While written accounts of these historical eruptions tell us about impacts on past societies, the wider environmental impacts are still poorly understood.

This Ph.D. project aimed to improve our understanding of the environmental consequences of these large historical eruptions. The study focused on two eruptions which are known to have impacted past societies. The eruption of Laki, in Iceland, began in the year 1783 and lasted for 8 months, releasing large amounts of acid to the atmosphere which was transported for long distances and eventually deposited over Europe and Scandinavia. The eruption of Tambora, in Indonesia, occurred in the year 1815 and released large amounts of sulphur gases to the upper atmosphere, causing a volcanic winter. In historical records, the year 1816 is called "the year without a summer" because of the lasting cooling effect on the climate caused by Tambora.

To investigate how acid deposition following the eruption of Laki and cold temperatures following the eruption of Tambora impacted the environment in the past, we studied lake sediments, which contain a record of environmental changes in the past. Changes which occurred within the lake ecosystem are represented by the remains of algae called diatoms which are found in the lake. Diatoms serve as ecological indicators, meaning their presence and abundance can reveal how healthy the ecosystem is. Changes in the chemistry of the lake and its catchment are also reflected by chemical markers in the sediments. By examining both diatom changes and chemical markers in the sediments, we aimed to understand if and how the volcanic eruptions of Laki and Tambora affected lakes in Sweden (Odensjön and Kassjön) and Finland (Nautajärvi).

Each lake provided unique insights. In Lake Odensjön, changes in the sediment suggested a potential response to the Laki eruption, however because the lake is located in a stable setting, the changes were minor and difficult to associate with the eruption. In Lake Kassjön, the changes were significant. Chemical markers in Kassjön's sediments indicated increased chemical weathering caused by the deposition of acid following the eruption of Laki. At the same time, diatom changes indicated changes in nutrient availability triggered by acid deposition. Diatom changes in Lake Nautajärvi were less pronounced. However, we found interesting chemical markers in Nautajärvi's sediments which may represent a new and poorly understood effect of volcanic acid deposition. We did not identify any significant ecological consequences of the cooling triggered by the eruption of Tambora.

Overall, this research combinds methods from geology, chemistry and ecology to improve our understanding of how large volcanic eruptions can impact lake ecosystems over long distances. By studying these sediments, we gain a better understanding of how future eruptions might impact aquatic ecosystems and their surroundings.

8.2 Populärvetenskaplig sammanfattning

Vulkanutbrott är våldsamma naturkatastrofer som har förekommit under jordens hela historia. Isländska utbrott under de senaste decennierna har visat vilka konsekvenser de kan få för våra liv och hur de kan påverka miljön. Men även om dessa händelser kunde te sig katastrofala var de små i jämförelse med mycket större vulkanutbrott som inträffade under 17- och 1800-talen och som resulterade i omfattande missväxt och tusentals dödsoffer. Medan skriftliga redogörelser vittnar om effekterna på tidigare samhällen är de vidare miljökonsekvenserna fortfarande dåligt kända trots att liknande utbrott kan inträffa när som helst i framtiden.

Detta doktorandprojekt syftar till att förbättra vår förståelse av miljökonsekvenserna av stora vulkanutbrott under de senaste seklerna. Studien fokuserar på två utbrott som man vet har haft omfattande påverkan på tidigare samhällen. Lakis utbrott på Island 1783−84 ledde till mycket stora försurande utsläpp som transporterades långa sträckor och nådde Europas och Skandinaviens landområden. Tamboras utbrott i Indonesien 1815 frigjorde stora mängder svavelgaser till den övre atmosfären, vilket påverkade klimatet över hela jorden till den grad att året efter kom att kallas ”året utan sommar”.

För att undersöka försurningseffektera efter Laki och klimateffekterna efter Tambora studerades sediment från botten av små sjöar som fungerar som en typ av naturliga arkiv över miljöförändringar i det förflutna. Utvecklingen av sjöarnas ekosystem analyserades med hjälp av kiselalger som bevaras i sedimenten och som reagerar känsligt på förändringar i vattenkemin. På motsvarande sätt användes geokemiska analyser av sedimentlagerföljderna för att klarlägga miljöförändringar i sjöarna och deras avrinningsområden.

Sjöar med förekomst av varviga (årslaminerade) sediment valdes ut för att möjliggöra högupplösande tidsserier med exakt åldersprecision, och varje enskild studie gav unika insikter. I sediment från Odensjön i Skåne finns en antydan till påverkan till följd av Lakis utbrott men eftersom sjöns ekosystem är relativt stabilt var förändringarna små och svåra att koppla till utbrottet. I Kassjön utanför Umeå i norra Sverige var förändringarna mer betydande. Geokemiska förändringar efter Laki-utbrottets syranedfall tyder på ökad kemisk vittring av kringliggande jordmåner som fortgick under en period av drygt 50 år. Samtidigt indikerade ändringar av kiselalgernas sammansättning förändringar i näringstillgången till följd av det sura nedfallet. Förändringarna av kiselalgsfloran i Nautajärvi i södra Finland var mindre uttalade men en samtidig ökning av mängden klor i sedimenten som sammanföll med Lakis utbrott tyder på mikrobiell bildning av organiska klorföreningar i kringliggande jordmåner under en tjugoårsperiod efter det isländska utbrottet. Detta är en möjlig miljöeffekt av stora vulkanutbrott som inte har dokumenterats tidigare. Inga betydande ekologiska konsekvenser av den nedkylning som utlöstes av Tamboras utbrott kunde identifieras i de tre sedimentstudierna.

Sammanfattningsvis kan man konstatera att en kombination av biologiska och geokemiska analyser av varviga sjösediment kan ge oss ökad förståelse av hur stora vulkanutbrott kan påverka miljön i och kring sjöar på stora avstånd från vulkansystemen. Denna typ av forskning kan ge oss fördjupad kunskap om hur framtida utbrott kan påverka miljön och människors hälsa.

8 References

Abbott, P.M. and Davies, S.M., 2012. Volcanism and the Greenland ice-cores: the tephra record. Earth-Science Reviews, 115(3), 173-191.

Anderson, N.J., 2014. Landscape disturbance and lake response: temporal and spatial perspectives. Freshwater Reviews, 7(2), 77-120.

Anderson, N. J., Korsman, T., and Renberg, I., 1994. Spatial Heterogeneity of Diatom Stratigraphy in Varved and Non-Varved Sediments of a Small, Boreal-Forest Lake. Aquatic Sciences, 56(1), 40-58.

Anderson, N.J., Odgaard, B.V., Segerström, U. and Renberg, I., 1996. Climate-lake interactions recorded in varved sediments from a Swedish boreal forest lake. Global Change Biology, 2(4), 399-403.

Anderson, N.J., Renberg, I. and Segerstrom, U., 1995. Diatom production responses to the development of early agriculture in a boreal forest lake-catchment (Kassjon, Northern Sweden). Journal of Ecology, 809-822.

Andres, R.J. and Kasgnoc, A.D., 1998. A time-averaged inventory of subaerial volcanic sulfur emissions. Journal of Geophysical Research: Atmospheres, 103(D19), 25251-25261.

Armbrust, E.V., 2009. The life of diatoms in the world's oceans. Nature, 459(7244), 185-192.

Arnalds, O., 2013. The influence of volcanic tephra (ash) on ecosystems. Advances in agronomy, 121, 331-380.

Ayris, P. and Delmelle, P., 2012. Volcanic and atmospheric controls on ash iron solubility: A review. Physics and Chemistry of the Earth, Parts A/B/C, 45, 103-112.

Baldigo, B.P. and Lawrence, G.B., 2000. Composition of fish communities in relation to stream acidification and habitat in the Neversink River, New York. Transactions of the American Fisheries Society, 129(1), 60-76.

Baldigo, B.P. and Lawrence, G.B., 2001. Effects of stream acidification and habitat on fish populations of a North American river. Aquatic Sciences, 63(2), 196-222.

Battarbee, R.W., 2000. Palaeolimnological approaches to climate change, with special regard to the biological record. Quaternary Science Reviews, 19(1-5), 107-124.

Battarbee, R.W., Juggins, S., Gasse, F., Anderson, N.J., Bennion, H., Cameron, N.G., Ryves, D.B., Pailles, C., Chalie, F. and Telford, R., 2001. European Diatom Database (EDDI): an information system for palaeoenvironmental reconstruction (p. 94). Environmental Change Research Centre.

Battarbee, R.W. and Renberg, I., 1990. The surface water acidification project (SWAP) palaeolimnology programme. Philosophical Transactions of the Royal Society of London. B, Biological Sciences, 327(1240), 227-232.

Berglund, B.E. and Rapp, A., 1988. Geomorphology, climate and vegetation in north-west Scania, Sweden, during the Late Weichselian. Geographia Polonica, 55, 13-35.

Bigler, C. and Hall, R.I., 2002. Diatoms as indicators of climatic and limnological change in Swedish Lapland: a 100-lake calibration set and its validation for paleoecological reconstructions. Journal of Paleolimnology, 27(1), 97-115.

Bleam, W.F., 2016. Soil and environmental chemistry. Academic Press, Cambridge.

Boichu, M., Menut, L., Khvorostyanov, D., Clarisse, L., Clerbaux, C., Turquety, S. and Coheur, P.F., 2013. Inverting for volcanic SO 2 flux at high temporal resolution using spaceborne plume imagery and chemistry-transport modelling: the 2010 Eyjafjallajökull eruption case study. Atmospheric Chemistry and Physics, 13(17), 8569-8584.

Bradshaw, E.G. and Anderson, N.J., 2001. Validation of a diatom–phosphorus calibration set for Sweden. Freshwater Biology, 46(8), 1035-1048.

Breitbarth, E., Achterberg, E.P., Ardelan, M.V., Baker, A.R., Bucciarelli, E., Chever, F., Croot, P.L., Duggen, S., Gledhill, M., Hassellöv, M. and Hassler, C., 2010. Iron biogeochemistry across marine systems–progress from the past decade. Biogeosciences, 7(3), 1075-1097.

Breteler, W.K., Schogt, N. and Rampen, S., 2005. Effect of diatom nutrient limitation on copepod development: role of essential lipids. Marine Ecology Progress Series, 291, 125-133.

Brönnimann, S. and Krämer, D., 2016. Tambora and the" Year Without a Summer" of 1816. A Perspective on earth and human systems science. Geographica Bernensia, Bern.

Brzezinski, M.A., 1985. The Si: C: N ratio of marine diatoms: interspecific variability and the effect of some environmental variables 1. Journal of Phycology, 21(3), 347-357.

Bueno, G., Deniz, O., Pedraza, A., Ruiz-Santaquiteria, J., Salido, J., Cristóbal, G., Borrego-Ramos, M. and Blanco, S., 2017. Automated diatom classification (Part A): handcrafted feature approaches. Applied Sciences, 7(8), 753.

Burnett, A.P., Soreghan, M.J., Scholz, C.A. and Brown, E.T., 2011. Tropical East African climate change and its relation to global climate: a record from Lake Tanganyika, Tropical East Africa, over the past 90+ kyr. Palaeogeography, Palaeoclimatology, Palaeoecology, 303(1-4), 155-167.

Carn, S.A., Fioletov, V.E., McLinden, C.A., Li, C. and Krotkov, N.A., 2017. A decade of global volcanic SO 2 emissions measured from space. Scientific reports, 7, 44095.

Chapman, R.L., 2013. Algae: the world's most important "plants"–an introduction. Mitigation and adaptation strategies for global change, 18(1), 5-12.

Clyne, M., Lamarque, J.F., Mills, M.J., Khodri, M., Ball, W., Bekki, S., Dhomse, S.S., Lebas, N., Mann, G., Marshall, L. and Niemeier, U., 2020. Model physics and chemistry causing intermodel disagreement within the VolMIP-Tambora Interactive Stratospheric Aerosol ensemble. Atmospheric Chemistry and Physics Discussions, 2020, 1-43.

Conley, D.J. and Schelske, C.L., 2001. Biogenic silica. Tracking environmental change using lake sediments: terrestrial, algal, and siliceous indicators, 281-293.

DeMaster, D.J., 1981. The supply and accumulation of silica in the marine environment. Geochimica et Cosmochimica acta, 45(10), 1715-1732.

Du Buf, H., 2002. Automatic diatom identification (Vol. 51). World Scientific, London.

Du Buf, H., Bayer, M., Droop, S., Head, R., Juggins, S., Fischer, S., Bunke, H., Wilkinson, M., Roerdink, J., Pech-Pacheco, J. and Cristóbal, G., 1999. Diatom identification: a double challenge called ADIAC, Proceedings 10th International Conference on Image Analysis and Processing, Venice, Italy, 734-739

Duggen, S., Olgun, N., Croot, P., Hoffmann, L., Dietze, H., Delmelle, P. and Teschner, C., 2010. The role of airborne volcanic ash for the surface ocean biogeochemical iron-cycle: a review. Biogeosciences, 7(3), 827-844.

Durant, A.J., Bonadonna, C. and Horwell, C.J., 2010. Atmospheric and environmental impacts of volcanic particulates. Elements, 6(4), 235-240.

Falkowski, P.G., Barber, R.T. and Smetacek, V., 1998. Biogeochemical controls and feedbacks on ocean primary production. Science, 281(5374), 200-206.

Falkowski, P.G., Katz, M.E., Knoll, A.H., Quigg, A., Raven, J.A., Schofield, O. and Taylor, F.J.R., 2004. The evolution of modern eukaryotic phytoplankton. science, 305(5682), 354-360.

Fölster, J., Andrén, C., Bishop, K., Buffam, I., Cory, N., Goedkoop, W., Holmgren, K., Johnson, R., Laudon, H. and Wilander, A. (2007) 'A novel environmental quality criterion for acidification in Swedish lakes—an application of studies on the relationship between Biota and water chemistry', in Acid Rain - Deposition to Recovery. Dordrecht: Springer Netherlands, 331–338.

Fiacco Jr, R.J., Thordarson, T., Germani, M.S., Self, S., Palais, J.M., Whitlow, S. and Grootes, P.M., 1994. Atmospheric aerosol loading and transport due to the 1783-84 Laki eruption in Iceland, interpreted from ash particles and acidity in the GISP2 ice core. Quaternary Research, 42(3), 231-240.

Friend, J.P., 1990. Natural chlorine and fluorine in the atmosphere, water and precipitation. NASA, Washington, Scientific Assessment of Stratospheric Ozone: 1989, Volume 2. Appendix: AFEAS Report.

Grattan, J. and Brayshay, M., 1995. An amazing and portentous summer: environmental and social responses in Britain to the 1783 eruption of an Iceland volcano. Geographical Journal, 125-134.

Grattan, J. and Charman, D.J., 1994. Non-climatic factors and the environmental impact of volcanic volatiles: implications of the Laki fissure eruption of AD 1783. The Holocene, 4(1), 101-106.

Grattan, J., Rabartin, R., Self, S. and Thordarson, T., 2005. Volcanic air pollution and mortality in France 1783–1784. Comptes Rendus Geoscience, 337(7), 641-651.

Gudmundsson, M.T., Thordarson, T., Höskuldsson, Á., Larsen, G., Björnsson, H., Prata, F.J., Oddsson, B., Magnússon, E., Högnadóttir, T., Petersen, G.N. and Hayward, C.L., 2012. Ash generation and distribution from the April-May 2010 eruption of Eyjafjallajökull, Iceland. Scientific reports, 2, 572.

Harrington, C.R., 1992. The Year Without a Summer? World Climate in 1816. Ottawa: Canadian Museum of Nature, 37.

Hering, D., Johnson, R.K., Kramm, S., Schmutz, S., Szoszkiewicz, K. and Verdonschot, P.F., 2006. Assessment of European streams with diatoms, macrophytes, macroinvertebrates and fish: a comparative metric-based analysis of organism response to stress. Freshwater Biology, 51(9), 1757-1785.

Hoshyaripour, G., Hort, M., Langmann, B. and Delmelle, P., 2014. Volcanic controls on ash iron solubility: New insights from high-temperature gas–ash interaction modeling. Journal of volcanology and geothermal research, 286, 67-77.

Hsiao, S.I., 1978. Effects of crude oils on the growth of arctic marine phytoplankton. Environmental Pollution (1970), 17(2), 93-107.

Johnson, D.W., Turner, J. and Kelly, J.M., 1982. The effects of acid rain on forest nutrient status. Water Resources Research, 18(3), 449-461.

Jones, O., Preston, M.R., Fawell, J., Mayes, W., Cartmell, E., Pollard, S., Harrison, R.M., Mackenzie, A.R., Williams, M., Maynard, R. and Ashmore, M., 2015. Pollution: causes, effects and control. Royal Society of Chemistry, Cambridge.

Jousset, P., Pallister, J., Boichu, M., Buongiorno, M.F., Budisantoso, A., Costa, F., Andreastuti, S., Prata, F., Schneider, D., Clarisse, L. and Humaida, H., 2012. The 2010 explosive eruption of Java's Merapi volcano—a '100-year'event. Journal of volcanology and geothermal research, 241, 121-135.

Jüttner, I., Kelly, M.G., Evans, S., Probert, H., Orange, A., Ector, L. and Marsh-Smith, S., 2021. Assessing the impact of land use and liming on stream quality, diatom assemblages and juvenile salmon in Wales, United Kingdom. Ecological Indicators, 121, 107057.

Kahlert, M. and Gottschalk, S., 2014. Differences in benthic diatom assemblages between streams and lakes in Sweden and implications for ecological assessment. Freshwater science, 33(2), 655-669.

Kekonen, T., Moore, J., Perämäki, P. and Martma, T., 2005. The Icelandic Laki volcanic tephra layer in the Lomonosovfonna ice core, Svalbard. Polar Research, 24(1-2), 33-40.

Klingaman, W.K. and Klingaman, N.P., 2013. The year without summer: 1816 and the Volcano that Darkened the World and Changed History. St. Martin's Press, New York.

Kokfelt, U., Muscheler, R., Mellström, A., Struyf, E., Rundgren, M., Wastegård, S. and Hammarlund, D., 2016. Diatom blooms and associated vegetation shifts in a subarctic peatland: responses to distant volcanic eruptions? Journal of Quaternary Science, 31(7), 723-730.

Kovács, C., Kahlert, M. and Padisák, J., 2006. Benthic diatom communities along pH and TP gradients in Hungarian and Swedish streams. Journal of Applied Phycology, 18(2), 105-117.

Kröger, N., 2007. Prescribing diatom morphology: toward genetic engineering of biological nanomaterials. Current opinion in chemical biology, 11(6), 662-669.

Kunasek, S.A., Alexander, B., Steig, E.J., Sofen, E.D., Jackson, T.L., Thiemens, M.H., McConnell, J.R., Gleason, D.J. and Amos, H.M., 2010. Sulfate sources and oxidation chemistry over the past 230 years from sulfur and oxygen isotopes of sulfate in a West Antarctic ice core. Journal of Geophysical Research: Atmospheres, 115(D18).

Lange-Bertalot, H., Hofmann, G., Werum, M., Kelly, M. and Cantonati, M., 2017. Freshwater benthic diatoms of Central Europe: over 800 common species used in ecological assessment (Vol. 942, 1-908). Schmitten-Oberreifenberg: Koeltz Botanical Books.

Lantmäteriet (2024) Lantmäteriet: Mapping, property information and land survey. Available at: https://www.lantmateriet.se (Accessed: 11 May 2024).

Larson, D.W., Sweet, J., Petersen, R.R. and Crisafulli, C.M., 2006. Posteruption response of phytoplankton and zooplankton communities in Spirit Lake, Mount St. Helens, Washington. Lake and Reservoir Management, 22(4), 273-292.

Lawson, I.T., Swindles, G.T., Plunkett, G. and Greenberg, D., 2012. The spatial distribution of Holocene cryptotephras in north-west Europe since 7 ka: implications for understanding ash fall events from Icelandic eruptions. Quaternary Science Reviews, 41, 57-66.

Lindenthal, A., Langmann, B., Pätsch, J., Lorkowski, I. and Hort, M., 2013. The ocean response to volcanic iron fertilisation after the eruption of Kasatochi volcano: a regional-scale biogeochemical ocean model study. Biogeosciences, 10(6), 3715-3729.

Liu, X. and Millero, F.J., 2002. The solubility of iron in seawater. Marine Chemistry, 77(1), 43-54.

Lowe, D.J., 2011. Tephrochronology and its application: a review. Quaternary Geochronology, 6(2), 107-153.

Mallin, M.A., Cahoon, L.B., Toothman, B.R., Parsons, D.C., McIver, M.R., Ortwine, M.L. and Harrington, R.N., 2007. Impacts of a raw sewage spill on water and sediment quality in an urbanized estuary. Marine Pollution Bulletin, 54(1), 81-88.

Martin-Jézéquel, V., Hildebrand, M. and Brzezinski, M.A., 2000. Silicon metabolism in diatoms: implications for growth. Journal of Phycology, 36(5), 821-840.

Medlin, L.K., Kooistra, W.H., Gersonde, R. and Wellbrock, U., 1996. Evolution of the diatoms (Bacillariophyta). II. Nuclear-encoded smallsubunit rRNA sequence comparisons confirm a paraphyletic origin for the centric diatoms. Molecular Biology and Evolution, 13(1), 67-75.

Medlin, L.K., Williams, D.M. and Sims, P.A., 1993. The evolution of the diatoms (Bacillariophyta). I. Origin of the group and assessment of the monophyly of its major divisions. European Journal of Phycology, 28(4), 261-275.

Mei, E.T.W., Lavigne, F., Picquout, A., de Bélizal, E., Brunstein, D., Grancher, D., Sartohadi, J., Cholik, N. and Vidal, C., 2013. Lessons learned from the 2010 evacuations at Merapi volcano. Journal of Volcanology and Geothermal Research, 261, 348-365.

Meyers, P.A. and Lallier-Vergès, E., 1999. Lacustrine sedimentary organic matter records of Late Quaternary paleoclimates. Journal of Paleolimnology, 21(3), 345-372.

Mills, K., Schillereff, D., Saulnier-Talbot, É., Gell, P., Anderson, N.J., Arnaud, F., Dong, X., Jones, M., McGowan, S., Massaferro, J. and Moorhouse, H., 2017. Deciphering long-term records of natural variability and human impact as recorded in lake sediments: a palaeolimnological puzzle. Wiley Interdisciplinary Reviews: Water, 4(2), e1195.

Moravcová, A., Rauch, O., Lukavský, J. and Nedbalová, L., 2013. The response of epilithic diatom assemblages to sewage pollution in mountain streams of the Czech Republic. Plant Ecology and Evolution, 146(2), 153-166.

Nakov, T., Guillory, W., Julius, M., Theriot, E. and Alverson, A., 2015. Towards a phylogenetic classification of species belonging to the diatom genus Cyclotella (Bacillariophyceae): Transfer of species formerly placed in Puncticulata, Handmannia, Pliocaenicus and Cyclotella to the genus Lindavia. Phytotaxa, 217(3), 249-264.

Newhall, C.G., and Self, S., 1982, The volcanic explosivity index (VEI): An estimate of explosive magnitude for historical volcanism. Journal of Geophysical Research, v. 87, no. C2, 1231-1238.

North, R.L., Guildford, S.J., Smith, R.E.H., Havens, S.M. and Twiss, M.R., 2007. Evidence for phosphorus, nitrogen, and iron colimitation of phytoplankton communities in Lake Erie. Limnology and Oceanography, 52(1), 315-328.

North, R.L., Guildford, S.J., Smith, R.E.H., Twiss, M.R. and Kling, H.J., 2008. Nitrogen, phosphorus, and iron colimitation of phytoplankton communities in the nearshore and offshore regions of the African Great Lakes. Internationale Vereinigung für theoretische und angewandte Limnologie: Verhandlungen, 30(2), 259-264.

Ojala, A.E. and Alenius, T., 2005. 10 000 years of interannual sedimentation recorded in the Lake Nautajärvi (Finland) clastic–organic varves. Palaeogeography, Palaeoclimatology, Palaeoecology, 219(3-4), 285-302.

Olgun, N., Duggen, S., Croot, P.L., Delmelle, P., Dietze, H., Schacht, U., Óskarsson, N., Siebe, C., Auer, A. and Garbe-Schönberg, D., 2011. Surface ocean iron fertilization: The role of airborne volcanic ash from subduction zone and hot spot volcanoes and related iron fluxes into the Pacific Ocean. Global Biogeochemical Cycles, 25(4), GB4001.

Oppenheimer, C., 2003. Climatic, environmental and human consequences of the largest known historic eruption: Tambora volcano (Indonesia) 1815. Progress in Physical Geography, 27(2), 230-259.

Panda, S.K., Baluška, F. and Matsumoto, H., 2009. Aluminum stress signaling in plants. Plant Signaling and Behavior, 4(7), 592-597.

Pappas, J.L. and Stoermer, E.F., 1996. Quantitative method for determining a representative algal sample count 1. Journal of phycology, 32(4), 693-696.

Pedraza, A., Bueno, G., Deniz, O., Cristóbal, G., Blanco, S. and Borrego-Ramos, M., 2017. Automated diatom classification (Part B): a deep learning approach. Applied Sciences, 7(5), 460.

Peterson, H.G., Boutin, C., Martin, P.A., Freemark, K.E., Ruecker, N.J. and Moody, M.J., 1994. Aquatic phyto-toxicity of 23 pesticides applied at expected environmental concentrations. Aquatic Toxicology, 28(3–4), 275-292.

Petterson, G. 1999: Image analysis, varved lake sedi- ments and climate reconstruction. PhD Thesis. Umeå University. Sweden.

Petersen, G.N., 2010. A short meteorological overview of the Eyjafjallajökull eruption 14 April–23 May 2010. Weather, 65(8), 203-207.

Polmear, R., Stark, J.S., Roberts, D. and McMinn, A., 2015. The effects of oil pollution on Antarctic benthic diatom communities over 5 years. Marine Pollution Bulletin, 90(1-2), 33-40.

Pouget, M., Moussallam, Y., Rose-Koga, E.F. and Sigurdsson, H., 2023. A reassessment of the sulfur, chlorine and fluorine atmospheric loading during the 1815 Tambora eruption. Bulletin of Volcanology, 85(11), 66.

Potapova, M.G., Aycock, L. and Bogan, D., 2020. Discostella lacuskarluki (Manguin ex Kociolek & Reviers) comb. nov.: a common nanoplanktonic diatom of Arctic and boreal lakes. Diatom research, 35(1), 55-62.

Prakash, J., Agrawal, S.B. and Agrawal, M., 2023. Global trends of acidity in rainfall and its impact on plants and soil. Journal of Soil Science and Plant Nutrition, 23(1), 398-419.

Renberg, I. and Hansson, H., 2010. Freeze corer No. 3 for lake sediments. Journal of Paleolimnology, 44(2), 731-736.

Renberg, I., 1981. Formation, structure and visual appearance of iron-rich, varved lake sediments: With 3 figures in the text. Internationale Vereinigung für theoretische und angewandte Limnologie: Verhandlungen, 21(1), 94-101.

Reynolds, C.S., 1984. Phytoplankton periodicity: the interactions of form, function and environmental variability. Freshwater Biology, 14(2), 111-142.

Reynolds, C.S. and Davies, P.S., 2001. Sources and bioavailability of phosphorus fractions in freshwaters: a British perspective. Biological reviews, 76(1), 27-64.

Rimet, F. and Bouchez, A., 2011. Use of diatom life-forms and ecological guilds to assess pesticide contamination in rivers: lotic mesocosm approaches. Ecological Indicators, 11(2), 489-499.

Rogers, N. and Hawkesworth, C., 2000. Composition of magmas. Encyclopedia of volcanoes, 115-131.

Roseacute, P., Hall, R., Korsman, T. and Renberg, I., 2000. Diatom transfer-functions for quantifying past air temperature, pH and total organic carbon concentration from lakes in northern Sweden. Journal of Paleolimnology, 24(2), 109-123.

Roswell, M., Dushoff, J. and Winfree, R., 2021. A conceptual guide to measuring species diversity. Oikos, 130(3), 321-338.

Rowell, H.C., 1996. Paleolimnology of Onondaga Lake: the history of anthropogenic impacts on water quality. Lake and Reservoir Management, 12(1), 35-45.

Sabater, S., 2010. The diatom cell and its taxonomical entity. Plankton of Inland Waters, 149.

Segerström, U., 1990. The natural Holocene vegetation development and the introduction of agriculture in northern Norrland, Sweden: studies of soil, peat and especially varved lake sediments (Doctoral dissertation, Umeå Universitet).

Segerström, U., Renberg, I. and Wallin, J.E., 1984. Annual sediment accumulation and land use history; investigations of varved lake sediments: With 7 figures in the text. Internationale Vereinigung für theoretische und angewandte Limnologie: Verhandlungen, 22(3), 1396-1403.

Schindler, D.W., 1988. Effects of acid rain on freshwater ecosystems. Science, 239(4836), 149–157.

Schmidt, A., Carslaw, K.S., Mann, G.W., Wilson, M., Breider, T.J., Pickering, S.J. and Thordarson, T., 2010. The impact of the 1783–1784 AD Laki eruption on global aerosol formation processes and cloud condensation nuclei. Atmospheric Chemistry and Physics, 10(13), 6025-6041.

Schmidt, A., Leadbetter, S., Theys, N., Carboni, E., Witham, C.S., Stevenson, J.A., Birch, C.E., Thordarson, T., Turnock, S., Barsotti, S. and Delaney, L., 2015. Satellite detection, long-range transport, and air quality impacts of volcanic sulfur dioxide from the 2014–2015 flood lava eruption at Bárðarbunga (Iceland). Journal of Geophysical Research: Atmospheres, 120(18), 9739-9757.

Schmincke, H.U., 2004. Volcanism (Vol. 28). Springer Science & Business Media, Berlin.

Shannon, C.E., Wiener, W., 1949. The Mathematical Theory of Communication. University of Illinois Press, Urbana.

Sigl, M., Mcconnell, J., Toohey, M., Plunkett, G., Ludlow, F., Winstrup, M., Kipfstuhl, S. and Motizuki, Y., 2015b. The history of volcanic eruptions since Roman times. PAGES News, 23, 48-49.

Sigl, M., Winstrup, M., McConnell, J.R., Welten, K.C., Plunkett, G., Ludlow, F., Büntgen, U., Caffee, M., Chellman, N., Dahl-Jensen, D. and Fischer, H., 2015a. Timing and climate forcing of volcanic eruptions for the past 2,500 years. Nature, 523(7562), 543-549.

Siver, P.A., Ricard, R., Goodwin, R. and Giblin, A.E., 2003. Estimating historical in-lake alkalinity generation from sulfate reduction and its relationship to lake chemistry as inferred from algal microfossils. Journal of Paleolimnology, 29, 179-197.

Smetacek, V.S., 1985. Role of sinking in diatom life-history cycles: ecological, evolutionary and geological significance. Marine biology, 84(3), 239-251.

Smol, J.P., 1985. The ratio of diatom frustules to chrysophycean statospores: a useful paleolimnological index. Hydrobiologia, 123, 199-208.

Sournia, A., 1982. Form and function in marine phytoplankton. Biological Reviews, 57(3), 347-394.

Sterner, R.W., Hagemeier, D.D., Smith, W.L. and Smith, R.F., 1993. Phytoplankton nutrient limitation and food quality for Daphnia. Limnology and Oceanography, 38(4), 857-871.

Stone, R., 2004. Iceland's doomsday scenario? Science, 306(570), 1278–1281.

Stothers, R.B., 1984. The great Tambora eruption in 1815 and its aftermath. Science, 224(4654), 1191-1198.

Takahashi, M., Furusawa, H., Limtong, P., Sunanthapongsuk, V., Marod, D. and Panuthai, S., 2007. Soil nutrient status after bamboo flowering and death in a seasonal tropical forest in western Thailand. Ecological Research, 22(1), 160-164.

Tammi, J., Appelberg, M., Beier, U., Hesthagen, T., Lappalainen, A. and Rask, M., 2003. Fish status survey of Nordic lakes: effects of acidification, eutrophication and stocking activity on present fish species composition. AMBIO: A Journal of the Human Environment, 32(2), 98-105.

Thordarson, T. and Self, S., 2003. Atmospheric and environmental effects of the 1783-1784 Laki eruption: A review and reassessment. Journal of Geophysical Research: Atmospheres, 108(D1), AAC-7.

Thordarson, T., Self, S., Oskarsson, N. and Hulsebosch, T., 1996. Sulfur, chlorine, and fluorine degassing and atmospheric loading by the 1783–1784 AD Laki (Skaftár Fires) eruption in Iceland. Bulletin of Volcanology, 58, 205-225.

Toohey, M. and Sigl, M., 2017. Volcanic stratospheric sulphur injections and aerosol optical depth from 500 BCE to 1900 CE. Earth System Science Data, 9, 809-831.

Tréguer, P.J. and De La Rocha, C.L., 2013. The world ocean silica cycle. Annual review of marine science, 5(1), 477-501.

Trigo, R.M., Vaquero, J.M., Alcoforado, M.J., Barriendos, M., Taborda, J., García-Herrera, R. and Luterbacher, J., 2009. Iberia in 1816, the year without a summer. International Journal of Climatology: A Journal of the Royal Meteorological Society, 29(1), 99-115.

USGS, 2022, Criteria for estimation of the Volcanic Explosivity Index (VEI) at https://www.usgs.gov/observatories/yvo/news/volcanic-explosivity-index-a-tool-comparing-sizes-explosive-volcanic (accessed July 15, 2024)

Wallace, J. M. and P. V. Hobbs, 2006: Atmospheric Science: An Introductory Survey. Elsevier, Amsterdam.

Wilson, N., Valler, V., Cassidy, M., Boyd, M., Mani, L. and Brönnimann, S., 2023. Impact of the Tambora volcanic eruption of 1815 on islands and relevance to future sunlight-blocking catastrophes. Scientific Reports, 13(1), 3649.

World Health Organization, 2006. Air quality guidelines: global update 2005: particulate matter, ozone, nitrogen dioxide, and sulfur dioxide. World Health Organization.

Zolitschka, B., Francus, P., Ojala, A.E. and Schimmelmann, A., 2015. Varves in lake sediments–a review. Quaternary Science Reviews, 117, 1-41

Quaternary Sciences
Department of Geology
Lund University
Sölvegatan 12
SE-223 62 Lund, Sweden
Telephone +46 46 222 78 80

ISSN 0281-3033
ISBN 978-91-87847-86-8

LUND UNIVERSITY