ACID CONTENT OF SNOW FROM A MID-TROPOSPHERE SAMPLING SITE ON MOUNT LOGAN, YUKON TERRITORY, CANADA

by

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ABSTRACT

An ice core 103 m long was extracted in 1980 from an altitude of 5340 m on the icefield plateau of Mount Logan, Yukon Territory (lat 60°35’N, long 140°30’W). The firn-ice transition occurs at a depth of 65 m, corresponding to about the year 1880.

The chemistry of this upper 65 m is apparently dominated by acid-ion species, the peaks in which are provisionally identified with several documented volcanic events. Although the analyses cover only selected discontinuous intervals, it appears that there is no significant long-term trend in the background acidity level of the precipitation at this location over the past century, in contrast to the results from the North American Arctic and Greenland.

Nitrate ion concentration shows pseudo-seasonal variations, which may be associated with stratospheric-tropospheric interactions, although other seasonally linked mechanisms are possible. This result has also been reported for ice-core sequences from Greenland.

Other nitrate pulses are tentatively associated with local volcanic events and a possible meteorite event (the entry of Tunguska in 1908). One of the largest short-term sources of sulfate ions is probably from volcanic activity on the north Pacific rim. Background volcanically-quiet nitrate and sulfate ion concentrations are compared with similar Greenland data in an attempt to throw further light on the origin of the acids.

Since the moisture for this precipitation originates primarily in the Gulf of Alaska, the data has particular relevance to that region. Short-term climatic changes, as reflected by the oxygen isotope (δ18O) record, show some response to the major volcanic-acid events. The influences affecting the δ18O record are listed but not discussed.

INTRODUCTION

The monitoring of acidity of precipitation in remote areas of the world is now being undertaken, because the results are expected to be of great significance to the issue of the long-range transport of atmospheric pollutants. Delmas and Aristarain (1978) presented results for snow from the summit of Mount Blanc which showed an increase of 20 μeq l⁻¹ in H⁺ concentration of strong acids from 1940 to 1975 (pH<5.0). These measurements excluded the effects of CO₂. Dyurgerov and others (1980) measured the pH of snow from 5 to 6 km altitude in the Pamirs and obtained values of from 6.3 to 6.9 for the years from 1975 to 1977. The method of measurement was not stated.

Barrie and others (1981) showed that at stations in the Canadian Arctic north of about 70°N the acidity of winter aerosols is such that the pH values of snow are in the range from 5.0 to 5.2 in the absence of neutralization by local dust particles. Such values are reported from the snowpack at Barrow, Alaska.

Galloway and others (1982), working within the Global Precipitation Chemistry Project, reported a mean pH value of 5.0 for precipitation at Poker Flats, Alaska during 1979-81, although values varied from about 4.1 to 5.6 over about one year. They estimated that the mean natural pH for the region was probably >5.

Koerner and Fisher (1982) reported pH values of recent snow from 2000 m altitude on the Agassiz Ice Cap, Ellesmere Island in the range from 4.8 to >6.0. When seasonal variations are averaged out, a mean value of 5.23 is obtained (1954-80). These pH values were obtained using a standard instrument without elimination of CO₂. The authors deduced from the data that there had been a decrease in pH at that site since the early 1930s when the general Holocene mean value of 5.48 was assumed to apply.

An ice core 103 m long was obtained from the icefield plateau of Mount Logan, Yukon Territory. It was originally intended for studies of climatic change but has been used in studies of trace chemistry and the implications for atmospheric processes (Delmas and others unpublished). That study showed, for the period from 1953 to 1966, that the trace chemistry of the snow was dominated by the strong mineral acids H₂SO₄, HNO₃ and HCl. The above study has subsequently been expanded in an attempt to cover as much of the last 100 years as possible. Even though the resulting time series contain large gaps, some useful information may still be deduced from them.

Some additional data are presented from a shallow firn core taken from a site near Mount Logan but further inland, where the net accumulation rate is about four times as high. Some comparisons could then be made with, and some extensions made to, comparable data from the Greenland ice sheet presented by Herron (1982[b]).

GEOGRAPHICAL DESCRIPTION OF THE SITES

Mount Logan (lat 60°35’N, long 140°30’W, altitude 5951 m) is situated in the St Elias Mountains, south-west Yukon Territory, within 100 km of the Pacific Ocean. The upper plateau, from which the cores were obtained in 1980, lies at an average altitude of 5300 m, covering an area of about 20 km². This plateau is just below the average altitude of the 500 mbar pressure level (Fig.1). Outcrops of granodiorite occur in the immediate vicinity.

The mean annual air temperature at the core site (5340 m) is expected to be close to ~29°C, since the mean annual air temperature at 10 m is ~28.5 and ~28.9°C, respectively, in two bore holes on the north-west col for the period from 1976 to 1980. The net accumulation rate has been determined using pole measurements, gross β-activity, tritium and oxygen isotope profiles (Holdsworth and others 1984) and is close to 380 kg m⁻² a⁻¹ on average.

A second sampling site (Eclipse) at lat 60°50’N, long 139°50’W, altitude 3017 m, is also shown in Figure 1. This site is close to outcrops of sedimentary rocks...
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Fig. 1. Vertical cross-section through the Gulf of Alaska and the St Elias Mountains showing the sampling sites at Mount Logan (ML) and Eclipse (E). The principal domains of marine aerosols (MA) and continental aerosols (CA) are also shown. The position of the tropopause is an average under stable conditions. (Modified after Junge (1977.))

which include limestone. Cores were obtained from here in 1982 and 1983. The mean annual firn temperature at 10 m depth is close to -6°C, although the mean annual air temperature is expected to be about -15°C. The net mean annual accumulation rate determined from oxygen-18 is quite marked, near-surface variations reaching 18/0. (b) Relatively confident dating of the upper 26 m of the C-core was carried out using gross beta-activity levels, tritium concentration and 18/0 oscillations together (Holdsworth and others 1984). This procedure established a mean net accumulation rate, from which, knowing the density profile and the total ice depth, an approximate theoretical time scale may be established for the deeper ice. (c) Volcanic events of sufficient size cause perturbations in electrolytic conductivity and pH (Hammer and others 1980) and the presence of detection mineral acids. Using the theoretical time scale as a guide these events can be used to date absolutely a particular section of ice. In this way, the 1912 Novarupta (Mount Katmai) event and other less certain events were identified with an overall accuracy of ±1 a. Mainly volcanic events from the north Pacific rim seem to be recorded.

ANALYSES

Electrolytic conductivity and pH of meltwater

Samples ranging from 5 to 10 cm long were cut from the core in a cold room at -15°C in which an air filter unit was operating. Samples were transferred to a clean air enclosure at about 25°C. In a laminar flow cabinet, the electrolytic conductivity of the sample was measured with a Radiometer CDM 3 conductivity meter, using a CDC 314 conductivity cell, and the pH of the sample measured with an Orion 399A meter, using initially a 91-03 combination electrode and later a 91-62 electrode designed for use with low-conductivity water. An equilibrium reading was achieved without final stirring of the ion chromatography. It is possible that pH values obtained using different electrodes could differ by as much as ±0.05 units. The pH of distilled, deionized water was routinely checked after each calibration and values of from 5.55 to 5.7 were usually found for water with a specific electrolytic conductivity of <1 mS m⁻¹. This agrees approximately with the expected value for pure water at low altitudes, in equilibrium with atmospheric CO₂ (Charlson and Rodhe 1982).

Earlier pH measurements made in Whitehorse with a portable field pH meter seem to be unreliable and are not given here. This is true for water samples in equilibrium with atmospheric CO₂ in the high mountains, which is the case for nearly all meltwater samples. The pH measurements are mainly used to identify the larger acid events so that the expensive and time-consuming nitrate, sulfate and chloride ion determinations could be carried out at strategic points. Koerner and Fisher (1982) obtained some significant results for the eastern Arctic using only pH and conductivity data, and it is therefore convenient to compare the present partial data with their results. The estimated error in the pH values given is thought to be about ±0.05 units. Since a complete ion balance has not been carried out we have not attempted to compare the measured pH values with the concentrations of anions that were measured.

Acid anion measurements

Nitrate

Nitrate concentrations in the meltwater were determined by two instrumental methods: (a) a colorimetric method using a Technicon Auto-Analyzer (TAA) and (b) ion chromatography (IC). For the first method, about 3 ml of sample were used in order to obtain duplicate values. In all cases
Fig. 2. Plot of (a) oxygen isotope data 

$$\delta^{18}O = \left[\frac{\left([^{18}O/^{16}O\right)_{\text{sample}}}{\left([^{18}O/^{16}O\right)_{\text{SMOW}}} - 1\right]$$

where SMOW = Standard Mean Ocean Water, (b) specific electrolytic conductivity (μS cm⁻¹) and (c) pH, for the upper 37 m of the C-core. The $\delta^{18}O$ is used to identify annual layers. The $\delta^{18}O$ values were provided by the Stable Isotope Laboratory, University of Calgary. Acid spikes are marked where they are considered to correspond to volcanic events (AUG = St Augustine, Alaska, KLI = Kliuchevskoi, Kamchatka, RAI = Raikoke, Kuril Islands).

\[\text{[NO}_3^+]_N \leq 2 \text{ ng g}^{-1}.\] The detection limit for \([\text{NO}_3^-]_N\) is about 3 ng g⁻¹, or 13 ng g⁻¹ [NO₂⁻]. In all cases, control blanks showed no values above detection limit.

The IC method was used when a Dionex 2020A ion chromatograph with a concentrator system became operational at the University of Calgary. With this system \([\text{NO}_3^-]_N\), \([\text{SO}_4^{2-}]_N\), and the concentrations of other anionic species could be determined on one sample with a detection limit of about 1 ng g⁻¹. The concentrator system was found to be necessary for analyzing samples of concentration <100 ng g⁻¹.

Nitrate concentration values for some split samples have been obtained from three independent laboratories. The two TAA determinations showed nearly random scatter in values which were almost always less than the IC values. It appears that the average background \([\text{NO}_3^-]_N\) in the core samples is near the detection limit of the TAA method.

**Sulfate and chloride**

Sulfate and chloride concentrations in the meltwater were determined by the IC method. Contamination appears to be more of a problem with these ions than with \([\text{NO}_3^-]_N\).

Of all the anions measured so far, sulfate variation shows the largest span, from a background of about 20 ng g⁻¹ to almost 600 ng g⁻¹ in 1912, corresponding to the Novarupta event. Concentrations of the other halides are not reported. Fluoride is not retained by the concentrator system and most bromide values are near or below detection.

**Particulate analyses**

Selected samples were taken at certain event horizons corresponding to certain acid events (e.g. the 1912 Novarupta eruption). Samples were analyzed on a Coulter counter at the Ice Physics Laboratory in Ottawa (Koerner 1977).

**RESULTS**

(1). Electrolytic conductivity and pH data

Figures 2 and 3 show oxygen isotope, conductivity and some pH data. The conductivity profile is assumed to reflect approximately the pH profile (Koeraer and Fisher 1982) in the absence of significant neutralization (Delmas and others unpublished), but there seem to be intervals where this assumption may not be accurate.

Figure 4 shows some recent electrolytic conductivity and pH data for the Eclipse site (Fig. 1). The $\delta^{18}O$ data has been used to identify the annual layers from seasonal oscillations which occur with lower amplitude here than on Mount Logan. The slightly higher mean pH of the Eclipse core (5.45) compared with the Mount Logan core (5.3) may be due to acid neutralization at the former site which lies within a region of sedimentary rocks containing limestones.

![Fig. 4](image-url)

**Fig. 4.** Plot of (a) $\delta^{18}O$, (b) specific electrolytic conductivity, and (c) pH for an 11 m core at the Eclipse site. The top of the core is at the 1982 summer.
(2). Nitrate

Figures 5 and 6 show the variation of nitrate and other anions in an upper and a lower section of the Mount Logan core. A limited depth profile covering about two years at the Eclipse site is shown in Figure 7. It is apparent that the background levels of \([\text{NO}_3^-]\) at the two sites are comparable and that, for the Mount Logan site, the background levels have not risen systematically over the last century.

From this data it is possible to recognize that \([\text{NO}_3^-]\) is strongly modulated by several processes. (a) A seasonal process operates at both sites. The 1981-83 Eclipse cycles are very clear and point to spring or summer peaks, whereas the cycles on Mount Logan are frequently obscured or modified by other processes. The sections where the seasonal oscillations are best developed are between 10 and 21 m (Delmas and others unpublished), 47.5 to 54 m and 61 to 66 m, although they would be unreliable for annual layer differentiation if used independently.

(b) According to Foley and Ruderman (1973), atmospheric nuclear weapons tests (between 1951 and 1980) generate and introduce significant quantities of NO into the stratosphere. This is a precursor gas for HNO₃ which is contained in the precipitation. For the interval from 10 to 21 m (representing 1953 to 1966) (Delmas and others unpublished), \([\text{NO}_3^-]\) concentrations show an elevated background which seems to correspond to the smoothed background variations in gross β-activity. These have been correlated with contemporary tests of nuclear weapons (Holdsworth and others 1984). Furthermore, several of the peaks in nitrate coincide in time with the peaks in gross β-activity, and, significantly, the maximum \([\text{NO}_3^-]\) peak for this era occurs in 1963. The average \([\text{NO}_3^-]\) concentration in this period is about 12% higher than for a similar length of time in a volcanically quiet period (Fig. 6).

(c) Volcanic eruptions along the north Pacific rim are thought to be responsible for some of the nitrate spikes in the time series. This is certainly true for the 1912 Novarupta (Mount Katmai) eruption which has proved an unequivocal chemical signature in the ice core (Figs. 5 and 6). The \([\text{NO}_3^-]\) levels reached four times the background values for a significant fraction of the 1912 interval. Moreover, the shape of the perturbation is compatible with a sudden injection of \([\text{NO}_3^-]\) and a slow decay, which suggests fallout of stratospheric aerosols.

Other nitrate pulses in the series are provisionally identified with a specific volcanic eruption (Figs. 5 and 6), using Simkin and others (1981) as a reference. Use was also made of pH, conductivity, other anion and particulate data.

(3). Sulfate and chloride

The \([\text{SO}_4^{2-}]\) and \([\text{Cl}^-]\) data (Figs. 5 and 6) have been determined mainly across known or suspected volcanic intervals and the series are extremely gapped. Nevertheless, the data have been useful in lending support to the preliminary identification of volcanic events. Herron (1982[b]) has shown the often characteristic signatures of a number of volcanic events in terms of the pulses of \([\text{SO}_4^{2-}], [\text{Cl}^-]\) and \([F^-]\) but, with the exception of one case, not \([\text{NO}_3^-]\). This may be a significant observation in terms of the differential transport of volcanic gases and derived acids. Usually, but not invariably, volcanically contaminated snow contains \([\text{SO}_4^{2-}]\) concentrations in the highest proportion, followed by \([\text{Cl}^-]\) or \([\text{NO}_3^-]\) in similar amounts, and then \([F^-]\) which is lowest.

Background levels of \([\text{SO}_4^{2-}]\) appear not to have changed substantially over the past century.

Limited \([\text{SO}_4^{2-}]\) analyses for the Eclipse site were carried out. From these data an average value was obtained for the purpose of comparing ionic concentrations with altitude and precipitation rate at different sites.
Particulates

Particulate data were obtained for two intervals: between 24 and 37 m in the C-core and between 43 and 60 m in the D-core. The raw data are not presented here. Instead, prominent peaks found in the particulate data are marked on Figures 5 and 6 as arrows, which are appropriately identified in the figure captions. The particle counts reveal an unusual range of concentrations ranging from $10^4$ to over $10^6$ particles $>1 \mu$m $^{-1}$. There appears to be pseudo-annual spikes in the data.

Two non-volcanic intervals of 27 to 28 m and 44.8 to 45.2 m were sampled and found to contain the lowest particulate counts.

DISCUSSION OF RESULTS

From the limited data on pH and anions, and the more continuous data on electrolytic conductivity, we conclude that recent snow in the St Elias Mountains is not, on average, any more acidic than snow deposited over the last century. The absence of a long-term trend in the data over this period implies that there is no detectable anthropogenic input of major acid ions (sulfate and nitrate).

The overall data suggest that any significant influence of volcanically derived acids on the chemistry of snow is very short-term. It is likely that only the north Pacific rim volcanoes cause most of these acid signatures, the largest and most definite of which was the 1912 eruption of Novarupta (Mount Katmai). Further processing of the core will reveal whether several earlier volcanic events of global significance are recorded in the core.

Relatively rapid recovery from these polluting events indicates the efficiency of the local north Pacific troposphere in terms of cleansing processes. Thus the apparent absence of evidence for an anthropogenic acid trend originating from the Asiatic industrial complex is hardly unexpected (Rahn 1981). This result contrasts with the snow and ice chemistry data for the much lower sites along the edge of the Arctic Ocean, where anthropogenic components in precipitation have been identified (Barrie and others 1981, Rahn 1981). The climatic conditions in that region, however, are entirely different from the present one.

It should be stressed that this study is incomplete, but it is not expected that further data will alter materially the main conclusions deduced from the results so far. In addition, more detailed sample analyses, resulting in an ion balance, may be desirable (Legrand and Delmas 1984).

Other important but more speculative implications of the data are now discussed.

(1). Atmospheric-precipitation processes

Seasonal variations in chemical species present an interesting but evidently very complex problem. Herron (1982[b]) discussed the occurrence of seasonal peaks in chemical time series and concluded that for different chemical species and locations, pulses in the concentration of a particular species may occur at different times of the year.

Annual variations in $[NO_3]$ were recognized in ice-core sequences first from Greenland (Hammer and others 1980, Risbo and others 1981, Herron 1982[b]) and now from the Antarctic (Legrand and Delmas 1984). Nitrate maxima generally occur about mid-year, although this is not invariably, possibly due to the interaction of several processes. Data from Greenland showing long regular sequences in parallel with $\delta^{18}O$ data indicate that the $[NO_3]$ peak may tend to occur there in the spring showing essentially the same timing as peak concentrations in other chemical species including particulates (Koerner and Fisher 1982).

The $[NO_3]$ data for Mount Logan seems to contain significant noise, some of it volcanic, frequently making identification of the exact time of annual peaks uncertain. Data from the Eclipse core shows convincing annual maxima and minima.

There are at least three possible mechanisms which might account for the observed variations in $[NO_3]$.

(a). A mechanism of biogenic origin (Junge 1963) with maximum activity in the spring or summer. The sites of biological activity would be coastal-marine.

(b). A mechanism involving a photochemical process, possibly within the troposphere (Legrand and Delmas 1984) but with an unknown primary source for the gaseous precursors.

(c). A mechanism controlled by the seasonal variations in the position and speed of rotation of the atmospheric Ferrel cell (Newell and others 1969). In this case the primary source of $NO_x$ is still uncertain, but Huebert and Lazarus (1980) have identified a natural gradient-free layer of $NO_x$ at 5 to 6 km altitude between 70°N and 55°S over western North America and the Pacific Ocean. This layer might have a stratospheric origin. Tropopause folding (Shapiro and others 1984) is one way in which stratospheric air may be injected into the troposphere and vice versa (Shapiro 1980). Risbo and others (1981) suggested seasonal variations in the stratospheric-tropospheric exchange to account for the seasonal oscillations in their $[NO_3]$ data from Greenland, which tends to show spring peaks. Shapiro (1980) and Shapiro and others (1984) have described instances of strong tropopause folding in the spring.

(2). Depositional processes near the snow surface

The data on species concentration discussed above have not been corrected for seasonal variations in precipitation rate because, generally, this is not known. In the case of Mount Logan, meteorological data from stations on either side of the St Elias Mountains show slightly greater precipitation in winter than in summer for Yakutat, Alaska and the reverse for Whitehorse. Therefore it is unlikely that the apparent seasonal variations in the concentration of $NO_3$ can be explained in terms of the precipitation regime. This leads us to a consideration of the effects at different sites of the altitude and the precipitation rates on the concentration of chemical species in the snow. The precipitation rates are derived from the net annual snow accumulation and its density.

Herron (1982[b]) has presented data from before AD 1900 relating concentrations of chloride, nitrate and sulfate to altitude and precipitation rate at several sampling sites in Greenland. Data on chloride concentration are presented in Figure 8, which shows that for volcanically quiet periods the chloride value lies essentially on the altitude depletion curve described by the equation:

$$C = C_0 \exp(-z/H)$$

where $C$ is the concentration of the ion at height $z$ above
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Fig.8. Plot of mean chloride concentration against altitude of snow-sampling site. Greenland data (open squares) are from Herron (1982[b]). Values for the Mount Logan (ML) and Eclipse (E) sites are for the background average in a short volcanically quiet period (filled triangles) and for a longer period (open triangle). The curve is the theoretical altitude depletion for chloride (see text).

Fig.9(a). Plot of mean long-term concentration versus altitude. Greenland data (open squares) as in Figure 8. Values for the Mount Logan (ML) site are for the long term value in a volcanically quiet period (filled triangle) and for the mean during 1953-66 (CNRS data) (open triangle).

Fig.9(b). Plot of $\left[NO_3^\cdot\right]$ data against accumulation rate. Empirical dilution curve shown is from Herron (1982[b]).

Fig.10(a). Plot of mean non-marine sulfate concentration against altitude. Greenland data (open squares) as in Figures 8 and 9. Values for the Mount Logan (ML) site are the mean for a pre-1900 interval of 10 a (filled triangle) and for 1953-66 (CNRS data) (open triangle). The Eclipse (E) value is the mean for the period summer 1981 - summer 1982 and may be higher than a volcanically quiet background mean. Non-marine sulfate $= [SO_4^{2\cdot}] - 0.14 \left[Cl^\cdot\right]$ (Herron 1982[b]).

Fig.10(b). The same sulfate data as in Figure 10(a) plotted against accumulation rate. Note apparent inverse relationship in data up to 500 kg m$^{-2}$ y$^{-1}$.

the sea level. $C_0 = 135$ ng.g$^{-1}$ and $H = 1.52$ km (Herron 1982[b]). Higher values of $[Cl^\cdot]$ are attributed in several cases to volcanic sources (Figs.5 and 6). The Eclipse value is the mean of 15 values obtained from samples covering summer 1981 to summer 1982. This is an insufficient period of time for obtaining a reliable volcanically quiet background value. We have eliminated the three highest values which are spikes to obtain a mean of 19.6 ng.g$^{-1}$, which plots virtually on the theoretical curve.

Nitrate concentrations in a volcanically quiet period for Mount Logan and for the period from 1982 to 1983 for Eclipse are plotted versus altitude in Figure 9(a). The points for the Greenland sites are the same as in Figure 8. The Mount Logan point (ML) is comparable to the Dye 3 (D3), Greenland value. Both sites have comparable accumulation rates, which suggests therefore that $[NO_3^\cdot]$ is independent of altitude. All points, except Eclipse, lie on an empirical dilution curve described by

$$C = C_0 \cdot A^{-n}$$

where $C$ is the concentration of nitrate, $C_0$ is a constant, $A$ is the accumulation rate and $n$ is an exponent. Herron
particulate fallout indicates that the immediate tropospheric particle fallout was very local and the 1913 pulse must then be from stratospheric fallout. (5) Climatic implications

It is relevant to point out a climatic connection to the chemical data presented here. The climatic information assumed to be contained in the $\delta^{18}O$ time series (Fig.2 and 3) is interpreted in the following way. Whereas seasonal variations in $\delta^{18}O$ are fairly reliable, long-term averages do not cross-correlate well with station temperature data. Some of the complicating factors to be considered are: (a) variations in the relative proportions of seasonal precipitation, (b) variations in relative removal of snow in different seasons, (c) changes in storm path lengths, (d) changes in the speed of progression of storms, (e) shifts in the maritime versus continental climatic regimes, and (f) changes in the mode and intensity of the jet stream, which may be the fundamental cause of any of the above factors. These and possibly other factors must also influence the content of trace chemicals in the snow.

An interesting observation is the amplitude modulation of $\delta^{18}O$ after the occurrence of the 1912 Novarupta eruption (Fig.3). The time series also shows a shift towards more negative $\delta^{18}O$ values, suggesting hemispheric cooling. It is thus of interest to see if a similar phenomenon occurs in the $\delta^{18}O$ series after the Krakatau eruption of 1883. In fact there is an isotopic cooling after this year and some reduction in amplitude of the $\delta^{18}O$ oscillation. The chemical evidence identifies the existence of the Krakatau eruption is unclear, and further analyses need to be done.

Finally, based on a knowledge of the long-term accumulation rate, core density, vertical strain-rate and ice thickness, a time span of 300 a is possible for the 103 m core. This provides the opportunity to identify the chemical and isotope signatures of the 1815 Tambora volcanic event and the Lakagigar (Laki) volcanic event of 1783-84 (Hammer and others 1980, Simkin and others 1981).

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