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(Bolivia)**

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Volcanic eruptions recorded in the Illimani ice core (Bolivia): 1918–1998 and Tambora periods

M. De Angelis¹, J. Simões^{1,2}, H. Bonnaveira¹, J.-D. Taupin³, and R. J. Delmas¹

¹LGGE, CNRS and Université Joseph Fourier, Grenoble-Saint Martin d'Hères, France

²Institute of Geosciences, Federal University of Rio Grande do Sul, Porto Alegre, Brazil

³LGGE, IRD, Grenoble-Saint Martin d'Hères, France

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Correspondence to: M. De Angelis (ange@lgge.obs.ujf-grenoble.fr)

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Abstract

Acid layers of volcanic origin detected in polar snow and ice layers are commonly used to document past volcanic activity on a global scale or, conversely, to date polar ice cores. Although most cataclysmic eruptions of the last two centuries (Pinatubo, El Chichon, Agung, Krakatoa, Cosiguina, Tambora, etc.) occurred in the tropics, cold tropical glaciers have not been used for the reconstruction of past volcanism. The glaciochemical study of a 137 m ice core drilled in 1999 close to the summit of Nevado Illimani (Eastern Bolivian Andes, 16°37' S, 67°46' W, 6350 m a.s.l.) demonstrates, for the first time, that such eruptions are recorded by both their tropospheric and stratospheric deposits. An 80-year ice sequence (1918-1998) and the Tambora years have been analyzed in detail. In several cases, ash, chloride and fluoride were also detected. The ice records of the Pinatubo (1991), Agung (1963) and Tambora (1815) eruptions are discussed in detail. Less important eruptions located in the Andes are also recorded and may also disturb background aerosol composition on a regional scale.

1. Introduction

Volcanic eruptions emit large amounts of water vapor, ash particles and acid species (in particular sulfur and halogen gases), which change the chemical composition of the atmosphere. It has been observed that the global-average surface temperature may decline by 0.2–0.3°C for one to three years following a major eruption (Lamb, 1970; Briffa et al., 1998; Robock, 2000). This cooling effect may differ in the upper and lower troposphere. Due to the high elevation of volcanic emissions, the impurities are generally introduced into the free troposphere but the main volcanic cloud is efficiently removed by settling and rain- or washout processes before being transported far from the emitting volcano. However, some of the emitted particles and gases remain in the troposphere where they can cause additional regional effects for weeks or months following the eruption. It is estimated that volcanoes contribute about 36% to the global

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tropospheric sulfur burden (Graf et al., 1997).

On the other hand, cataclysmic events inject most of their products directly into the stratosphere. A sulfuric acid veil is then formed which may persist for 1 to 3 years, affecting the global climate. In this case, chemical compounds may be spread on a global scale. Note that it has been demonstrated that volcanic gases are a significant constituent of the Earth's stratosphere. Most recent eruptions have been extensively studied by satellite observation. The 1991 eruption of Pinatubo (Philippines) is particularly well documented [Soden et al., 2002]. Moreover, the recent volcanic eruptions of El Chichon (1982) and Pinatubo (1991) were accompanied by El Niño-type climatic perturbations which prolonged the environmental consequences of the disturbances (Self et al., 1997).

Ice-core studies have demonstrated that major tropical eruptions of the past are clearly recorded in polar snow and ice, generally by their large H₂SO₄ deposits (Hammer, 1977, 1980; Clausen et al., 1997; Delmas et al., 1992; Palmer et al., 2001). Conversely, the acid layers of volcanic origin (H₂SO₄ spikes) serve as reference horizons for dating polar ice cores (Schwander et al., 2001). Several continuous long-term records of past volcanic activity have been deduced from acidity measurements along deep ice cores recently recovered in both polar regions (Zielinski et al., 1996; Clausen et al., 1997). Large volcanic eruptions of global significance may sometimes also be detected in polar ice cores by their glass or silicate grain deposits (de Angelis et al. 1985; Palais et al., 1990; Silva and Zielinski, 1998; Zielinski et al., 1997). However, even in the case of very large halogen-rich volcanic eruptions, significant amounts of halogens have been detected in polar ice layers only when the eruption occurred at a high latitude in the same hemisphere, allowing rapid tropospheric transport (De Angelis and Legrand, 1995).

Cataclysmic events, which are particularly frequent at tropical latitudes, are recorded through their acidic deposits both in Greenland and Antarctica (Langway et al., 1988). On the other hand, little is known about volcanic records from tropical glaciers. The only record of a large volcanic eruption in a tropical ice core has been reported by

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Thompson et al. (1986) for the case of the 1600 AD eruption of Huaynaputina (Peru).

In 1993, in his book “Volcanoes, A Planetary Perspective”, Peter Francis wrote (p. 373): “There are a few ice-caps on high mountains in tropical regions which also contain potentially excellent records of equatorial eruptions.... Ice cores, therefore, have the potential to provide long-term records of world-wide eruptions. But how should we read these curious records, inscribed in acid on ice?”.

This study is an answer to this question. We have investigated for the first time how large eruptions in the tropics are recorded in glacier ice collected at very high sites located in the same latitude band as the eruptive center. Records are complex but rich in information about the environmental impact of major tropical eruptions.

The study site is Nevado Illimani (6350 m a.s.l.). It is located in the Eastern Bolivian Andes (16°37' S, 67°46' W). Its summit is covered by a permanent ice cap directly impacted by moist air masses coming from the Amazon Basin (Vuille, 1999; Vuille et al., 2000). More than 80% of annual precipitation occurs during the austral summer (DJF), whereas during the winter, the site is mostly influenced by air masses circulating in the free troposphere. In addition to seasonal variations due to the alternating wet season (WS) and dry season (DS), along with seasonal changes in atmospheric circulation patterns, chemical species deposited in the snow are expected to be influenced by atmospheric phenomena of global significance, such as major volcanic eruptions.

2. Experimental

In June 1999, two parallel ice cores (136.7 and 138.7 m) were drilled down to bedrock very close to the summit by a joint expedition involving scientists from the French Institut de Recherche pour le Developpement (IRD) and the Swiss Paul Scherrer Institute (PSI). One of these cores was transported frozen to France and sub-sampled at the Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), Grenoble-Saint Martin d'Hères. We present here the volcanic signal inferred from a first set of proxies including major ionic species.

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Before any sub-sampling, the core was studied by ECM (Electrical Conductivity Measurement) in the cold-room facilities of the LGGE (-15°C). This technique is non-destructive and was expected to provide information on volcanic acid spikes (see Sect. 3-1). Next, continuous sub-sampling of the upper 50 m of the core was carried out. At greater depths, samples were taken only along a few specific sections of the core corresponding to very high or to typical background ECM values.

The outer part of the core, contaminated in the field by drilling and handling, was removed using a band saw. The remaining part was either: i) mechanically decontaminated using first an electrical plane and then a scalpel, and cut in 10 cm long sub-sections (this procedure concerned firn samples above 30 m depth) or, ii) below close-off, lower than 30 m depth, cut in successive samples which were rinsed in ultra pure water. In all, 744 samples were prepared, among them 654 for the upper 50 m. A few samples are missing from 12.35 to 12.78 m (brittle core). Samples were allowed to melt in polyethylene vials before being divided in several aliquots respectively devoted to insoluble particle counting, Ionic Chromatography (IC) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The aliquots were stored in frozen state in tightly closed polycarbonate containers and melted just before analysis. Ice decontamination and sample processing took place under clean room conditions. The reliability of this cleaning procedure has already been proven for polar ice cores (Legrand et al., 1993) where concentrations are much lower. This paper mainly deals with IC data measured for 13 inorganic ions (F^{-} , $\text{CH}_3\text{SO}_3^{-}$, Cl^{-} , NO_3^{-} , SO_4^{2-} , Na^{+} , NH_4^{+} , K^{+} , Mg^{2+} , Ca^{2+}) and 3 carboxylic ions ($\text{CH}_3\text{COO}^{-}$, HCOO^{-} , $\text{C}_2\text{O}_4^{2-}$). Due to IC problems, SO_4^{2-} and $\text{C}_2\text{O}_4^{2-}$ data are missing from 14.37 m to 14.70 m. Chemical data are more thoroughly discussed in De Angelis et al. (in preparation).

3. Results and discussions

3.1. Identifying the volcanic contribution

The method for characterizing volcanic deposits by their acidic signature (mainly H_2SO_4 but also halogenated gases such as HF or HCl) or ash input is not a trivial issue. In particular, such deposits may be partly or totally overlapped by continental dust and anthropogenic inputs of local, regional or semi-global origin, as observed in recent Greenland snow by de Angelis and Legrand, 1995. Up to now, Electrical Conductivity Measurements (ECM) have been widely used on polar ice cores as a non-destructive tool for the detection of acid layers presumably of volcanic origin. Indeed, although the identity of the associated anion (Cl^- , NO_3^- or SO_4^-) influences the strength of ECM response to proton concentration (Legrand et al., 1987, Wolff et al., 1995, 1997), most ECM spikes detected in pre-anthropogenic polar ice correspond to large H_2SO_4 excesses related to major volcanic eruptions. This has been demonstrated for Greenland ice cores (Hammer, 1980, Clausen et al., 1997) as well as for Antarctic ice cores (Legrand et al., 1987, Wolff et al., 1999). However, while significant correlation between ECM and acidity has been established for acidic snow containing small amounts of continental dust, the ECM signal drops nearly to zero in snow heavily charged with alkaline dust (Ram and Koenig, 1997). This neutralizing effect made debatable the use of ECM as an unambiguous detection method of volcanic events in Andean ice cores, where the combination of dry deposition and sublimation was expected to lead to high dust loads at the snow surface during the dry season, and required careful comparison of ECM with major ionic species. Thus, ECM data were considered as preliminary indications of acidity along the Illimani ice core, to be compared with ionic profiles. In this study, the proton concentration, denoted H^+ and expressed in $\mu\text{Eq l}^{-1}$, was not measured but calculated from the ionic balance [$\sum \text{anions} (\mu\text{Eq l}^{-1}) - \sum \text{cations} (\mu\text{Eq l}^{-1})$]. It has been demonstrated that the ionic balance is positive in acidic or slightly acidic snow and ice from various areas, where it is very close to the proton concentra-

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tion (Legrand, 1987, Maupetit and Delmas, 1994), while negative ionic-balance values correspond to alkaline snow.

3.2. ECM-Chemistry relationship for the Illimani ice core

Calcium and sulfate profiles of the 50 upper meters of the core (continuous sampling) are reported in Figs. 1a and 1b, respectively, and compared with the ECM record (Fig. 1c). Data obtained from 72.32 up to 71.01 m depth and corresponding to the highest ECM values of the profile are also presented and will be discussed later. Sulfate and calcium exhibit regular variations which can be related to seasonal patterns. At the Illimani site, preliminary studies of snow accumulation, of freshly fallen snow and of the evolution in the composition of surface snow during the dry season have been carried out in 2001 and 2002. The results, summarized in Table 1, show that the sublimation rate measured for a few sunny days during the dry season in 2001 and 2002 varies from 0.8 to 1.2 mm w.e./day, whereas significant snow accumulation (21 g.cm^{-2}) occurred (stake measurements) from May to October 2001, which corresponds to the whole dry season. In relation to wet-season compositions, the concentrations of most species were higher than expected in dry-season snow layers, base on potential sublimation rates and snow accumulation heights. These results strongly suggest that sublimation remains of minor importance at this site and that large concentration peaks occur during the dry season (austral winter, April to November) for most of studied species, in relation to dry deposition processes and precipitation of heavily loaded snow at this time. Roughly 70% of annual precipitation occurs during the wet season (austral summer) and correspond to less concentrated snow layers. Thus, the wide peaks observed along the core have been considered dry-season deposits alternating with less concentrated layers deposited during the wet season. Such a seasonal pattern is consistent with satellite observations showing maximum regional dust emissions from the Bolivian Altiplano occurring from September to November (Prospero et al., 2002). Detailed chemical stratigraphy provides an independent dating of the core (reported every 10 years in Fig. 1, de Angelis et al., in preparation). According to this

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dating, the upper 50 m of the core cover the 1919–1998 time period. As explained in Part 2, a few data are missing (all ions at the beginning of 1990, sulfate and oxalate data in mid 1988).

No direct link between ECM and sulfate can be inferred from Fig. 1, whereas well marked calcium peaks are associated with most of the sulfate peaks. The two highest calcium peaks of the profile are particularly interesting and are detailed in Fig. 2, where total SO_4^{2-} concentrations are reported (thinner line) along with $[\text{SO}_4^{2-}] - [\text{Ca}^{2+}]$ (thicker line). The deeper peak ($\text{Ca}^{++} = 636 \text{ ng g}^{-1}$) denoted B and centered around 43.56 m corresponds to a negative ionic balance ($-3.8 \mu\text{Eq l}^{-1}$). In this first layer, it appears from stoichiometric considerations (De Angelis et al., in preparation) that sulfate (1340 ng g^{-1}) could be entirely associated with 90% of total calcium as CaSO_4 , the remaining 10% of calcium being present most likely as CaCO_3 . This peak was attributed to a very large dust event of regional origin containing large amounts of gypsum and halite. In the second major calcium peak ($\text{Ca}^{++} = 726 \text{ ng g}^{-1}$) denoted A and centered around 13.68 m, only 70% of total sulfate ($\sim 1700 \text{ ng g}^{-1}$) could be explained by “gypsum-like” material, while the remaining 30% ($\sim 620 \text{ ng g}^{-1}$) could be partly related to a moderate peak of the ion balance ($2.9 \mu\text{Eq l}^{-1}$) and partly present as other salts, such as NH_4SO_4 and Na_2SO_4 . Such a pattern strongly suggests that atmospheric H_2SO_4 reacted with various aerosols of mineral and biogenic continental origin (process discussed in detail in De Angelis et al., in preparation). It may be assumed that calcium containing aerosols is a mixture of pure gypsum and carbonate-rich dust having previously reacted with atmospheric H_2SO_4 . Considering that Ca^{2+} is entirely deposited as CaSO_4 , it is then possible to calculate excess sulfate ($\text{SO}_4^{2-}_{\text{exc}}$) with respect to the gypsum-like fraction. Interestingly, our calculation leads to the satisfactory correlation ($r = 0.8$, $n = 654$) between calculated H^+ and $\text{SO}_4^{2-}_{\text{exc}}$, both expressed in $\mu\text{Eq l}^{-1}$ (Fig. 3). The slope of the regression line (1.1) strongly suggests that most $\text{SO}_4^{2-}_{\text{exc}}$ is acidic.

Once these calculations have been made, the correspondence between H^+ , $\text{SO}_4^{2-}_{\text{exc}}$

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and ECM becomes more obvious, as displayed in the upper part of Fig. 1. The ion balance is positive for most samples. It varies between background values in the 1–5 $\mu\text{Eq l}^{-1}$ range and peak values higher than 10 $\mu\text{Eq l}^{-1}$. A detailed examination of these profiles shows that most ECM peaks are associated with H^+ and SO_4^{2-} peaks, but without any obvious quantitative relationship between ECM peak heights and the two other acidity indicators (H^+ and SO_4^{2-}).

Nevertheless, the three highest ECM spikes of the whole record, observed at 71.46 m (1), 33.43 m (2), and 9.12 m (3), correspond to high positive ion-balance values (11.4, 10.04, and 24.8 $\mu\text{Eq l}^{-1}$, respectively), and to narrow and pure SO_4^{2-} peaks (482, 458, and 1234 ng g^{-1} , respectively). These peaks are found for samples where all other species are very low, i.e. most likely for wet-season deposited snow. This supports the idea that, even for positive ion balances, the ECM signal is significantly buffered by high-concentration dust events of various origins (de Angelis et al., in preparation), mostly observed in dry-season precipitation. Based on glaciological considerations, Knüsel et al. (2003) propose that the ECM peak at 71.46 m is linked to acidic fallout from the cataclysmic Tambora eruption (1815). According to our dating, the two other ECM maxima correspond to snow layers deposited in early 1964 and early 1993, respectively, i.e. very close to, but later than, the time of the Agung (March 1963) and Pinatubo (June 1991) eruptions. This suggests that the ECM spikes are not linked to the arrival of tropospheric volcanic clouds, assumed to occur a few weeks after the eruption. What does the Illimani record tell us about the contributions of these three major eruptions to the tropical free troposphere? Several indicators are considered, namely SO_4^{2-} and H^+ as strong acidity markers, along with HF and HCl, the two main halogenated species encountered in volcanic eruptions (Sumonds et al., 1988) respectively denoted by F^- and volcanic chloride (Cl_{exc}^-). Volcanic chloride is estimated by subtracting the soil fraction and the possible (probably very low, de Angelis et al., in preparation) sea-salt derived fraction of Cl^- :

$$\text{Cl}_{\text{exc}}^- = (\text{Cl}_{\text{total}}^- - \text{Cl}_{\text{soil}}^-) - 1.7 * (\text{Na}_{\text{total}}^+ - \text{Na}_{\text{soil}}^+).$$

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We assume that the soluble part of continental input at the Illimani site is mainly of regional origin and is mainly composed of gypsum and halite, as observed for the huge dust peak at 43.56 m. $\text{Cl}^-_{\text{soil}}$ and $\text{Na}^+_{\text{soil}}$ are calculated from Ca^{2+} concentrations, using the $\text{Cl}^-/\text{Ca}^{2+}$ and $\text{Na}^+/\text{Ca}^{2+}$ mass ratio measured at 43.56 m. However, volcanic debris are likely different in composition compared to regional soils in which most of the Na^+ comes from halite. They should be composed of partly solubilized tephra and glass closer to mean soil or mean crustal compositions, and thus probably chloride depleted. For this reason, in ice samples suspected to contain very large amounts of volcanic dust (positive ion balance associated with a Ca^{2+} concentration greater than $(\text{Ca}_{\text{max}})_{\text{mean}} + \sigma$, $(\text{Ca}_{\text{max}})_{\text{mean}}$ being the mean value of peak maximums calculated for the past 20 years and σ the corresponding standard deviation), soil and marine contributions may be calculated using Ca^{2+} and Na^+ concentrations of adjacent samples. Main results concerning the relative influence of these three eruptions on sulfate, halogens and dust budgets at the Illimani site are summarized in Table 2, where they are compared to typical background mean values calculated for annual snow layers deposited the year before or a few years after the eruption (bgd_{mean}). Before any discussion, it must be kept in mind that annual layer thicknesses at the Tambora level (about 70 m depth) are close to 40 cm of snow, whereas they are 2 or 3 times higher at the Pinatubo level (about 10 m). The ice core was sub-sampled every 10 cm, so that sampling resolution decreases with increasing depth, which leads to a progressive smoothing of chemical records. This effect has to be taken into account when comparing the three data sets.

3.3. Major volcanic eruptions

3.3.1. Tambora (8°25 S, 118° E, April 1815)

Tambora (1815), known as the most powerful eruption of global significance since 1259 AD, was responsible for the “year without a summer” (Stommel and Stommel, 1979). The amount of H_2SO_4 produced by this eruption is estimated to be in the 52-175 Tg

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range (Devine et al., 1984, Sigurdsson and Carey, 1992). Moreover, it is known that TAMBORA emitted very large amounts of halogenated species (100–200 Tg and 70–125 Tg of HCl and HF, respectively). Contrary to sulfate, these compounds are not significantly transported to high latitudes and Antarctic snow and ice glaciochemical studies show that their concentrations in the snow did not increase during the TAMBORA years, whereas for the last millennium, the sulfate perturbation is second only to that of the 1259 eruption (Delmas et al., 1992).

For that reason and according to firn densification studies for the Illimani site, the strong ECM spike between 70.8 and 71.5 m, with an absolute maximum at 71.46 m is expected to be linked to TAMBORA acidic fallout. As shown in Fig. 4., very high peaks of SO_4^{2-} (3080 ng g⁻¹), $\text{SO}_4^{2-}_{\text{exc}}$ (900 ng g⁻¹), Ca^{2+} (910 ng g⁻¹), Cl^-_{exc} (140 ng g⁻¹) and F^- (10 ng g⁻¹) are observed at 71.75 m. They are associated with high H^+ value (10.8 μ Eq l⁻¹), while the ECM signal drops to zero. The highest concentration of each peak, denoted x_{max} , is reported in Table 2. Despite the poor sampling resolution at this depth, calcium and fluoride spikes are respectively more than one hundred times and twenty times higher than corresponding background values and represent the highest concentrations encountered in this study. This is also true for total sulfate, partly (70%) neutralized by carbonate. These peaks are as high or even higher than those observed in layers assumed to contain Pinatubo tropospheric debris (see next section) and are very likely to correspond to the arrival of aerosols and gases emitted in the troposphere by the TAMBORA eruption. The huge mass of volcanic glass and tephra injected in the atmosphere certainly contributed to fixing halogen species, preventing them from being removed by large quantities of water present in the plume or excluded from strongly acidic snow layers. Two other series of peaks are observed at 71.46 and 71.07 m. Even if quite low in absolute terms, calcium values (28 and 30 ng g⁻¹, respectively) significantly higher than background are observed, $\text{SO}_4^{2-}_{\text{exc}}$ concentrations (410 to 370 ng g⁻¹, respectively) remain high although decreasing, and halogenated species are more than 5 (Cl^-_{exc}) or 10 (F^-) times higher than the background at this depth. H^+ is slightly higher (11.4 μ Eq l⁻¹) in the peak at 71.46 m where the ECM maximum was

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observed than before or later ($9.3 \mu \text{Eq l}^{-1}$), which confirms the bias induced in ECM measurements by large amounts of dust, leading to misdetection of tropospheric fallout of major eruptive events. The time interval between the three sets of spikes is close to annual layer thicknesses at this depth, which probably means that the arrival of

5 Tambora tropospheric debris at 71.75 m was followed by two successive stratospheric inputs. Arrival of tropospheric and stratospheric debris are denoted “T” and “S” in Fig. 4, respectively.

In both polar regions, the ECM ice records show that Tambora was preceded by another large (but weaker than Tambora) eruption of unknown location, which probably

10 occurred in 1809–1810 (Delmas et al., 1992; Palmer et al., 2001; Mayewski et al., 1996; Clausen et al., 1988). The Illimani ECM record does not show any marked signal 6 years before Tambora, i.e. around 73 m depth.

The subsidence of halogenated compounds from the stratosphere has not been observed in polar ice cores. In the case of Tambora, the top of the plume reached 43 km

15 elevation, while the main column reached 30 km (Sigurdsson and Carey, 1989), so that large quantities of HCl and HF should have been transported through the tropopause, and later uptaken by water vapor and volcanic glass or silicate grains traveling through the lower stratosphere. However, because larger particles would tend to fall out faster than the smaller ones, H_2SO_4 aerosols are expected to reach more southern latitudes

20 than most of the ash particles. The finest grains may reach polar sites as demonstrated by the observation of glass shards emitted by major volcanic eruptions in ice layers corresponding to stratospheric sulfate arrival (de Angelis et al., 1985; Cole-Dai et al., 1997; Zielinsky et al., 1997), but they are too small in quantity to have carried significant amounts of halogens. This could explain large differences in composition

25 between stratospheric fallout over tropical areas and over polar regions. Comparison between background mean values (bgd_{mean}) and mean values calculated for the year of the eruption and for years after ($\text{year}_{\text{mean}}$) clearly shows that halogen and SO_4^{2-} budgets were significantly enhanced until at least the end of 1817 over southern tropical areas. Further analysis is required to determine exactly when the perturbation

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ended.

3.3.2. Pinatubo (15° N, 120° E, June 1991)

The Pinatubo eruption in June 1991 injected 12 to 20 Tg of SO₂ into the middle stratosphere (Mc Peters, 1993). This estimate is 2 or 3 times lower than the figures published for the 1815 Tambora eruption. Due to wind shear, aerosols stratified very quickly in several layers ranging from 17 to 26 km in altitude with different physical or chemical signatures (Winker and Osborn, 1992). Several successive events are detected in the Illimani core during and after this period. As shown in Fig. 5., a first SO₄²⁻_{exc} peak is observed at 10.75–10.94 m. The first part of this peak (~1000 ng g⁻¹) corresponds to pure H₂SO₄, while the second part (~1400 ng g⁻¹) represents only 50% of the total sulfate, because of very high calcium concentrations (~600 ng g⁻¹). Because they correspond to rather different chemical fingerprints, both parts are presented in Table 2. The SO₄²⁻_{exc} peak is associated with well marked Cl⁻_{exc} and F⁻ peaks, although Cl⁻_{exc} and F⁻ are very low in the sample containing pure H₂SO₄. This could be due to HF and HCl exclusion by strong acidity at grain boundaries, as previously observed on Greenland ice cores by de Angelis and Legrand, 1995. Halogenated species have been partly fixed by dust in the calcium-rich layer corresponding to the second part of the SO₄²⁻_{exc} peak, either during transport or later, after snow deposition. This post-depositional effect was taken into account, when reporting fluoride maxima in Table 2. In tropical ice cores, the seasonality of the isotope-atmospheric temperature relationship is opposite to what was observed in polar ice cores, with lowest isotope content occurring during the austral summer (Thompson et al., 1986, 2001). Considering the relatively high δD value at this time (Simões et al., in preparation), it can be concluded that this deposition occurred during the dry season in 1991, i.e. from May to October. This is consistent with the month of the eruption (June) and suggests that concentration peaks of all considered species were caused by the upper tropospheric transport of the dust and gaseous products emitted by the initial Pinatubo eruptive phase. Thereafter,

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at around 10m depth, a second SO_4^{2-} peak (450–700 ng g⁻¹) is found, accompanied by large amount of fluoride and Cl_{exc}^- , apparently fixed on adjacent Ca^{2+} and NH_4^+ (not reported here) layers. These peaks could correspond to the first subsidence of stratospheric clouds by the beginning of the year following the eruption (1992). Our data concerning the Tambora period support the idea that such halogen-rich deposits from the lower stratosphere may occur at tropical latitudes after cataclysmic halogenated eruptions. Two narrow peaks of almost pure H_2SO_4 are observed during the tropical summer 1992-1993. In the first one at 9.5 m, SO_4^{2-} represents ca 85% of total sulfate, while in the second one at 9.12 m, it represents more than 97% of total sulfate, leading to a H^+ value close to $25 \mu \text{Eq l}^{-1}$ which is the highest value in our data. Taking into account dating uncertainty, month by month dating is somewhat arbitrary, but these two peaks clearly occurred by the end of the tropical winter, suggesting that the deposition of the highest stratospheric cloud occurred near the end of 1992. It is known that stratospheric/tropospheric interchanges occur through tropopause-foldings at mid-latitudes and in the spring (Holton et al., 1995), i.e. in September for the southern hemisphere, which satisfactorily aligns with our observations. At various Antarctic sites, the sulfate perturbation linked to the Pinatubo (and Cerro Hudson) eruption was also observed in snow layers deposited between 1991 and 1993 (Dibb and Whitlow, 1996; Legrand and Wagenbach, 1999; Cole-Dai and Mosley-Thompson, 1997). It appears from Fig. 5 and from yearly mean concentrations presented in Table 2. that sulfate input at the Illimani site was perturbed for two years, until mid 1993, while noticeable effects lasted only one year for halogens. The duration of sulfate perturbation is consistent with stratospheric aerosol measurements in the tropics (20° S to 30° N, Grant et al., 1996). A particularly negative δD level is found at 9.2 m depth. This value (−181.6‰) is even the lowest one of the whole record. Contrary to seasonal patterns, on the decadal to millennium scale, similar relationships are observed in polar and tropical ice cores, suggesting that the absolute value of δD is partly linked to air temperature (Thompson et al., 2000). If true, lower δD values in early 1993 would indicate a temperature lowering at this time. This observation suggests a significant cooling

of the tropical troposphere as previously observed at mid latitudes some 18 months after the eruption (Robock, 2000, Robock, 2002, Soden et al., 2002). No climatic or chemical perturbation can be detected later in our profile after this date, suggesting that the major environmental impact of the Pinatubo eruption was located in the years 1991 and 1992.

3.3.3. Agung (8°34 S, 115°51 E, March 1963)

According to Devine et al. (1984) and Sigurdsson et al. (1985), the total mass erupted during the 1963 Mount Agung eruption (2.4×10^{12} kg) was 2 orders of magnitude lower than that during the Tambora eruption, similar to the estimated masses of HCl (1.5×10^6 kg) and HF (8×10^5 kg). Estimated H_2SO_4 emissions were 2 to 4 times greater than HCl and HF emissions, respectively. Consequently, we expect very low impact, if any, of the Agung eruption on the Illimani ice-core profiles of these two elements. Data confirm this prediction (see Fig. 6 and Table 2). No definite Ca^{2+} , F^- or Cl_{exc}^- trends are observed in the 1963–1964 ice layers, when compared with previous or following years in the 1960s. On the other hand, SO_4^{2-} and H^+ concentrations are higher by a factor of 2 to 3 than background values from mid-1963 to late 1964. Four relative maxima of both species are observed. Although moderate when compared to the second peak at 33.43 m, the first peak (33.82 m) at the beginning of the dry season in 1963 corresponds to strong acidity markedly higher than background mean values.

Although tropospheric aerosol life is expected to be rather short, in the range to 1 to a few weeks (Robock, 2002), the first two peaks could correspond to two successive precipitation events rather close in time. Both of them could tentatively be attributed to the arrival of the tropospheric cloud in mid-1963. The third peak, in late-1963 or early 1964, corresponds to a very high ECM value and could be related to initial stratospheric fallout. The fourth peak, dated at the end of the 1964 dry season, could correspond to a second-year stratospheric input. Contrary to the halogen budget which did not significantly vary, both the sulfate and acidity budgets remained perturbed from mid 1963 until the end of 1964.

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3.3.4. Fingerprint comparison

A very rough comparison of the relative impact of these three major eruptions on the tropical atmosphere can be inferred from the yearly mean concentrations presented (see Table 2). For each studied species, the ratio $R = \text{year}_{\text{mean}} / \text{bgd}_{\text{mean}}$ of the annual and background means provides a first estimate of an eruption on the deposition flux. Volcanic Explosivity Index (VEI) is commonly used as a means to classify eruptions according to parameters such as plume injection height and the total mass of material injected into the atmosphere (Simkin and Siebert, 1994). The greatest impact, by far, on the Illimani ice core chemistry is observed in 1815, the year of the Tambora eruption (VEI 7). The value of R is 31, 5, and 9 for calcium, sulfate and fluoride concentrations, respectively. For Pinatubo (VEI 6), these figures are 2, 3.5 and 1.4 and for Agung (VEI 4), only ~ 1 , 1.8 and 1.6. Significant amounts of halogen compounds probably entrapped on the finest ash grains seem to have remained in the tropical stratosphere for more than two years after the Tambora eruption, as shown by F^- and Cl_{exc}^- mean concentrations still markedly higher in 1817 than during the reference time period. A significant increase in strong acidity indicators (H^+ and $SO_4^{2-}_{\text{exc}}$) are observed for at least two years after the 3 eruptions, which suggests annual subsidence of stratospheric sulfate linked to seasonal folding and/or gravitational settling. Further studies of ice layers above 71 m are required to determine exactly the end of the Tambora perturbation. Concerning Pinatubo, it is obvious from Figs. 5 and 6 that background values were reached in mid-1993 after the June 1991 eruption. For Agung (March 1963), this occurred by the end of 1964. This is consistent with a stratospheric residence time ($1/e$) of approximately 1 year for sulfate aerosols at these latitudes (Grant et al., 1996).

3.4. Other volcanic eruptions

It has been demonstrated that tropical explosive eruptions with VEI greater than 5 could be detected in the Illimani ice core through both their tropospheric (very high gypsum-like concentration associated with strong remaining acidity) and stratospheric

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(strong, pure acidity spikes) fallout. High concentrations of F^- and Cl_{exc}^- were also measured in ice layers containing volcanic debris likely to have scavenged gaseous HCl and HF. However, a quantitative relationship between the estimated strength of a given eruption (VEI) and its impact on the chemistry of high-elevation Andean ice is not expected since other parameters such as the location of the eruption, the season it occurred, or atmospheric circulation patterns strongly influence cloud diffusion and removal. Sulfate (SO_4^{2-} and $SO_4^{2-}_{exc}$) background values as well as sulfate peak heights depict large variability in the Illimani ice over the past 80 years (Fig. 1) with a noticeable increase since the mid 1950s, but this variability may not simply be related to other well known eruptions of global importance and requires more detailed study into potential sulfate sources. Several hypotheses may be put forward. Quiescent degassing and moderate eruptions have a long-term effect on the atmospheric composition and several high-altitude volcanoes located in the southern tropical zone (Peru, Ecuador) or even farther south (northern Chile, northern Argentina) can provide large amounts of tephra and acid gases on a regional scale even without being cataclysmic. Regional anthropogenic activities, biomass burning (H_2SO_4 , F^-) and DMS oxidation (H_2SO_4) also influence ice chemistry. Figure 7 clearly shows that the annual sulfate deposition rate increased starting in the mid 1950s while methanesulfonate (MSA) to total or excess-sulfate weight ratios were very low ($<1\%$ and $\sim 2\%$, respectively) compared to values measured over tropical oceans (4–10%, Saltzman et al., 1986), and have slightly decreased since the 1970s. This suggests that DMS does not significantly contribute to the sulfur budget at the Illimani site (De Angelis et al., in preparation). As shown in Fig. 1, a significant part of the sulfate trend is related to long term fluctuations of the background contribution (in particular in wet season deposition) and could be linked to anthropogenic emissions from regional north-eastern countries, while most of the spikes correspond to sporadic events occurring during the dry season. At that time of the year, volcanic emissions from Andean volcanoes or dust plumes from the arid Altiplano may contribute significantly to the chemical content of the regional free troposphere. Graf et al., (1997) have estimated the relative contribution of volcanic SO_2

emissions to the atmospheric sulfur budget, demonstrating that volcanic emissions represent more than 50% of sulfur deposition in the tropics, west of South America. We agree with this figure.

Detailed temporal profiles of F^- , SO_4^{2-} _{exc} and H^+ are reported in Fig. 8. In this figure, an attempt to remove the trend linked to changing background contributions for wet season ice was made with the aid of a spline function (smoothed values were subtracted from individual sulfate concentrations, as explained in more detail elsewhere, de Angelis et al., in preparation). Two main time periods corresponding to apparently increased volcanic activity may be observed. During the first period, from 1919 until late the late 1960s, SO_4^{2-} _{exc} remained rather low and more or less constant. Moderate peaks, roughly 2 times higher than background values and generally associated with F^- and Cl^- _{exc} peaks (not reported here), occurred rather regularly during dry seasons, denoted by high Ca^{2+} concentrations. A few periods of slightly higher activity seem to have occurred around 1919–1920, 1933–1936, 1941–1943, 1951, 1959, and 1963–1964. Five peaks, denoted “1” to “6”, correspond to very low Ca^{2+} and F^- concentrations. According to our criteria, they may tentatively be attributed to stratospheric fallout during the wet seasons 1919–1920, 1933–1934, 1936–1937, 1942–1943, 1951–1952 and 1963–1964 respectively, bearing in mind dating uncertainties at this depth (± 1 year). An increase in peak heights is observed since the mid 1970s, with 10 peaks related to possible stratospheric sulfate subsidence events, i.e. pure H_2SO_4 peaks occurring during wet seasons. Nine of them, dated in 1975–1976, 1976–1977, 1979–1980, 1982–1983, 1985–1986, 1989–1990, and 1992–1993 respectively, were preceded by dust arrival. Although located in dry season ice layers, the first SO_4^{2-} _{exc} peak of the second period in mid 1971, corresponds to very low Ca^{2+} concentrations with no halogens and could tentatively be attributed to stratospheric deposits. If that is true, the absence of significant tropospheric contributions the year before suggests that the eruption took place in the northern hemisphere. From the comprehensive study of relevant observational data – especially pyrheliometry – from both hemispheres, it has been stated that, “during the long period 1881–1992, about 80% of all stratospheric aerosols generated

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by the largest sulfur-producing eruptions were emplaced during the two short time intervals 1883–1902 and 1982–1991” (Stothers, 1996). Our results showing a significant increase in the number of stratospheric inputs at Illimani from the mid 1970s to the mid 1990s are reasonably consistent with this statement.

5 Taking into account VEI from Simkin and Siebert, 1994, and volcano locations, volcanic eruptions with possible impact on snow chemistry at the Illimani site were selected and are reported in Table 3. Comparison between periods of enhanced volcanic activity as recorded in Illimani ice (Fig. 8) and eruptions referenced in Table 3 suggests that, despite rather modest VEI (<4), South American volcanoes and in particular those
10 located at low latitudes may significantly contribute to volcanic input over the Illimani site, examples being Tungurahua (1918), Kelut (1919), Sangay (1934), Reventador (1936, 1973, 1976), Quizapu (1932), Nevado de Ruiz (1985), or Sabancaya (activity starting in 1986 with a major halogenated eruption in 1990), and may be responsible of most of the events observed in the ice core, while the 1982 El Chichon eruption had moderate impact probably because it occurred at a rather high northern latitude.
15

4. Conclusions

An exhaustive glaciochemical study carried out on a high-elevation tropical ice core from Nevado Illimani (Eastern Bolivian Andes) shows for the first time that large volcanic events in tropical latitudes are recorded in the snow and ice layers. The study
20 covers the time period 1918 to 1998. The Tambora period was also investigated. It is possible to interpret the chemical data in terms of tropospheric and stratospheric deposits of volcanic origin. Volcanic products are particularly well identified for Tambora, the strongest investigated event. Shortly after the eruption time (1815), calcium-sulfate, as well as halogenated-species deposits are observed. During the following two years (1816–1817), sulfur and halogen acids of stratospheric origin are present in the ice.
25 For the eruptions of Pinatubo (1991) and Agung (1963), less pronounced spikes are observed than for Tambora, but the sequence of deposited species in Illimani snow

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seems to be similar. In particular, the recent Pinatubo eruption provided an excellent opportunity to calibrate the timing. Our data shows that cataclysmic eruptions in tropical latitudes have a strong impact on the aerosol content of the upper tropical troposphere, not only for sulfuric but also for F and Cl acids and crustal dust.

5 The volcanic acid sulfur species transported in the stratosphere during the few months following the eruption most probably react with particulate volcanic material of crustal origin. Finally, our study points out that moderate eruptions, which are relatively frequent in the Andean region, have a strong impact on the regional free tropospheric budget of sulfur, confirming other estimates on the impact of Andean volcanism on this
10 cycle.

Further studies will include the comparison of the obtained volcanic record with the climatic parameters recorded in this ice core.

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15 Développement, France) and Swiss PSI (Paul Scherrer Institute, Switzerland). The help of the Peruvian and Bolivian porters was highly appreciated for the transfer of the ice under cold conditions. The authors wish to thank L. Liu for his helpful contribution to IC analysis.

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Table 1. Snow accumulation rate, snow sublimation rate and chemical composition of recent snow deposited during wet and dry season at the Illimani site. Except precipitation event, all samples were taken along the wall of a 2 m deep pit dug in February 2002 and covering the end of Wet Season (WS) 2000–2001, the complete Dry Season (DS) 2001, and the beginning of WS 2001–2002

Measured snow accumulation (w.e.)		Dry season 2001 (May–October)		21 cm		
		May 2001–May 2002		80 cm		
Sublimation rate (w.e./d)		Dry season		.8 - 1.2 mm		
		Mean concentrations (ng g ⁻¹)				
		F ⁻	Cl ⁻	SO ₄ ²⁻	Na ⁺	Ca ²⁺
Wet Season	February 2002 (Precipitation event)	.98	7.0	67	2.0	19
	End of WS 2000- 2001(Pit 156-200cm)	.08	7.6	29	3.8	2.8
Dry season	Beginning of DS 2001 (Pit 130-154cm)	2.6	68	671	37	166
	Mean value DS 2001 (Pit 102-154cm)	2.0	74	534	40	116

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Table 2. Evolution of ionic balance H^+ , fluoride, excess chloride, sulfate, excess sulfate and calcium concentrations over three time periods respectively centered around the Tambora (1815), Agung (1963), and Pinatubo (1991) volcanic eruptions. Excess sulfate, denoted $SO_4^{2-}_{exc}$, is the “gypsum-free” component of SO_4^{2-} (see text), and excess chloride, denoted Cl^-_{exc} , was calculated removing the possible halite contribution from Cl^- concentration (see text). Background reference concentrations (mean values denoted bgd_{mean} , and corresponding standard deviations, σ) were calculated over 1- year time-periods apparently free of volcanic influence and chosen as close as possible to eruption dates. They must be compared to annual mean values ($year_{mean}$) and corresponding standard deviations (σ) calculated along ice sections containing volcanic fallout. x_{max} are the highest concentrations measured in every peak attributed either to tropospheric or to stratospheric volcanic fallout (see text and Figs. 4, 5, and 6)

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Table 2.

Tentative dating		H ⁺ μEq.l ⁻¹	F ⁻ ng.g ⁻¹	Cl ⁻ _{exc} ng.g ⁻¹	SO ₄ ²⁻ ng.g ⁻¹	SO ₄ ²⁻ _{exc} ng.g ⁻¹	Ca ²⁺ ng.g ⁻¹
TAMBORA							
Background (below 72.3m)	bgd _{mean} (σ)	3.8 (.9)	.45 (.14)	4.3 (2.5)	184 (48)	166 (43)	7.8 (6.8)
1815	year _{mean} (σ)	5.6 (3.7)	4.0 (3.5)	64 (42)	909 (1250)	325 (330)	245 (386)
	X _{max}	10.8	10.0	136	3075	898	914
1816	year _{mean} (σ)	6.1 (3.0)	3.0 (1.9)	47 (62)	282 (123)	237 (110)	19 (6.0)
	X _{max}	11.4	6.2	152	481	415	28
1817	year _{mean} (σ)	5.7 (2.3)	2.9 (51.7)	59 (31)	263 (110)	232 (85)	13 (12)
	X _{max}	9.3	5.6	114	452	373	33
AGUNG							
Background (1962)	bgd _{mean} (σ)	2.9 (.8)	.22 (.39)	6.7 (9.9)	166 (105)	102 (42)	26 (36)
1963	year _{mean} (σ)	4.7 (2.0)	.35 (.29)	9.8 (7.2)	304 (158)	204 (145)	23 (11)
DS 1963	X _{max} (1)	6.0	.91	16.6	428	334	40
DS 1963	X _{max} (2)	8.5	.36	22.2	494	440	23
1964	year _{mean} (σ)	5.2 (2.2)	.24 (.45)	6.3 (6.6)	265 (172)	213 (136)	22 (31)
WS 63-64	X _{max} (1)	10	-	16.3	458	435	9.6
DS 1964	X _{max} (2)	7.2	-	13.4	463	430	14
PINATUBO							
Background (1995)	bgd _{mean} (σ)	3.8 (1.4)	.75 (.67)	22 (11)	282 (109)	164 (82)	50 (24)
1991	year _{mean} (σ)	7.9 (3.9)	1.02 (1.11)	30 (53)	650 (750)	434 (368)	90 (177)
DS 1991	X _{max}	14.7	2.89	37	995	905	38
		15.0	2.49	188	2781	1327	610
1992	year _{mean} (σ)	3.7 (3.2)	1.8 (2.2)	57 (60)	589 (382)	342 (191)	100 (83)
Early 1992	X _{max}	5.6	6.8	145	1360	737	261
1993	year _{mean} (σ)	7.7 (5.5)	.56 (.89)	6.9 (8.5)	449 (316)	372 (290)	32 (38)
WS 92-93	X _{max} (1)	12.2	.61	11.3	906	759	62
	X _{max} (2)	24.8	.18	15.5	1233	1203	13

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Table 3. Volcanic eruptions with possible impact on snow chemistry at Illimani, as compiled by Simkin and Siebert (1994). Major events studied in detail are indicated in red. Fourteen volcanic events (with stratospheric impacts dated in 1919–20, 1933–34, 1936–37, 1942–43, 1951–52, 1963–64, mid-1971, 1975–76, 1976–77, 1979–80, 1982–83, 1985–86, 1989–90, 1992–93) were recorded at Illimani. They may have occurred in the following locations: tropical latitude belt. Eruption products (gases and fine ash) injected in the free tropical troposphere may encircle the earth in a few weeks, as shown by satellite observations. If the injection is significant ($VEI \geq 4$), the signature of the eruption will be recorded in the Illimani ice core. Typical case: Agung.

regional. For eruptions of moderate strength ($VEI \geq 3$), the ash and gas plume injected at high altitudes may reach the Illimani region and contaminate the snow. Typical cases: Reventador in Ecuador or Sabancaya in Chile. In the table, we indicate only the major eruptive phase of the eruptions (for instance, the volcanic activity of Sabancaya started in 1986, with a major halogenated eruption occurring in 1990). Even minor events ($VEI \leq 3$) of the most nearby volcanoes (Cotopaxi, Reventador, Sabancaya, Sangay, Tungurahua) may also influence the composition of the regional atmosphere.

remote. Andean eruptions from mid-latitudes (Chile, Mexico, Columbia) to nearby tropical regions (Peru, Ecuador) can produce huge amounts of sulfur gases that can potentially cause acid spikes in Illimani snow

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Table 3.

Eruption	Date (mo/yr)	Latitude(°)	Longitude (°)	Elevation (m)	VEI
Lascar	01/1993	23.4 S	67.7 W	5590	4
Cerro Hudson	08/1991	45.9 S	73 W	1900	5
Pinatubo	06/1991	15.1 N	120 E	1600	6
Sabancaya	12/86-06/90	15.8 S	71.8 W	5967	3
Nevado del Ruiz	09/1985	4.9 N	75.3 W	5320	3
El Chichon	03/1982	17.4 N	93 W	1060	5
Reventador	11/1973	0 S	77.7 W	3560	3
Fernandina	06/1968	0.4 S	91.5 W	1495	4
Awu	08/1966	3.7 N	125 W	1320	4
Kelut	04/1966	7.9 S	112 W	1730	4
Taal	09/1965	14 N	121 W	400	4
Fuego	09/1963	14.5 N	91 W	3760	3
Agung	03/1963	8.5 N	115.5 E	3140	4
Reventador	06/1960	0 S	77.7 W	3560	3
Cordon Caulle	05/1960	40.5 S	72.2 W	1800	3
Reventador	11/1958	0 S	77.7 W	3560	3
Carran los Venados	07/1955	40.3 S	72.1 W	1114	4
Ambrym	1951	16.2 S	168 E	1334	4
Reventador	02/1944	0 S	77.7 W	3560	3
Sangay	1937	2.0 S	78.3	5320	3
Reventador	08/1936	0 S	77.7 W	3560	3
Sangay	08/1934	2.0 S	78.3 W	5320	3
Quizapu	04/1932	35.7 S	70.8 W	3790	5
Reventador	1929	0 S	77.7 W	3560	3
Reventador	01/1926	0.1 S	77.7 W	3560	3
Galeras	10/1924	1.2 N	77.4 W	4280	3
Cordon Caulle	12/1921	40.5 S	72.2 W	1800	3
Manam	08/1919	4.1 S	145 E	1725	4
Kelut	05/1919	7.9 S	112 W	1730	4
Tungurahua	04/1918	1.5 S	78.4 W	5020	4
Tambora	04/1815	8.2 S	118 E	2850	7

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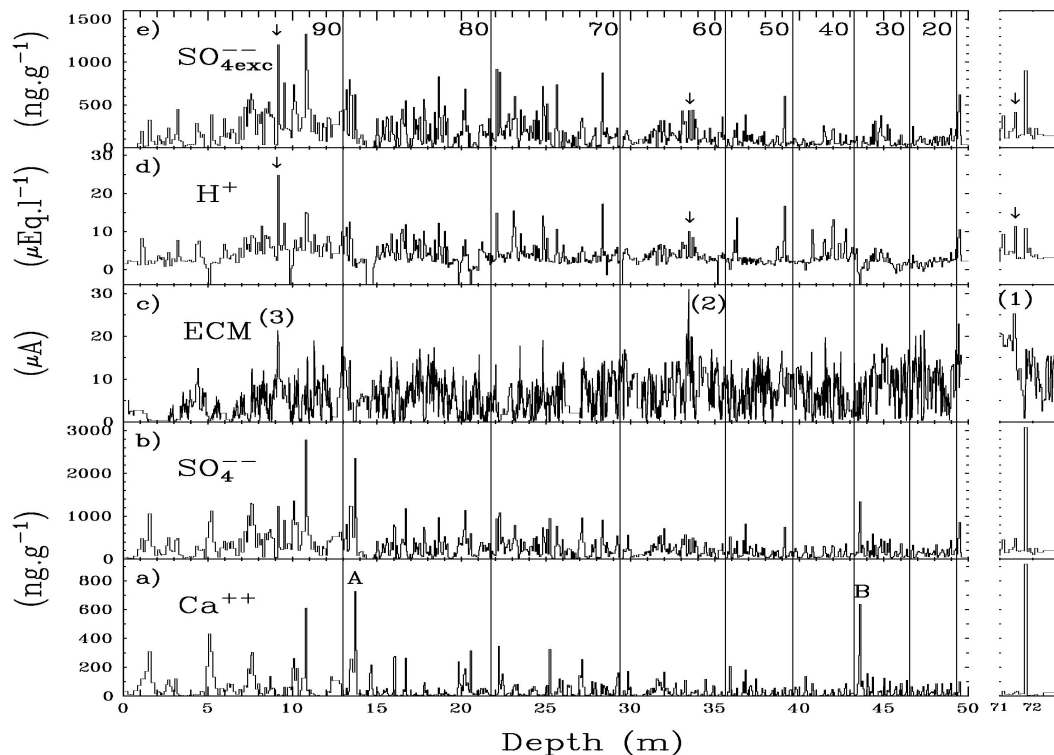


Fig. 1. Detailed depth profiles of selected chemical species for the upper 50 m of the Illimani ice core and from 71 to 72.3 m depth (“Tambora level”). Calcium and sulfate concentrations are reported in **(a)** and **(b)**, respectively. Original ECM (Electrical Conductivity Measurements) data (Simoes et al., in preparation) have been averaged every 10 values **(c)**. In **(d)**, H^+ is the calculated ionic balance (see text). Excess-sulfate ($SO_4^{2-}_{exc}$) is the “gypsum-free” fraction of SO_4^{2-} (see text). Vertical arrows indicate the acidity peaks related to the three highest ECM spikes of the profile ((1), (2), and (3) in (c)). Thin vertical lines correspond to the beginning of each decade from 1920 to 1990.

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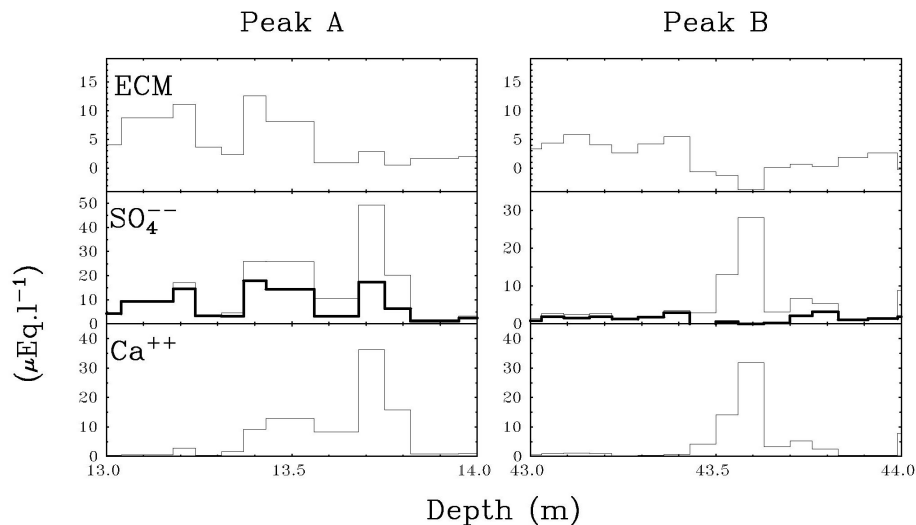


Fig. 2. Sulfate (SO_4^{2+} , thinner lines), its gypsum-free component ($\text{SO}_4^{2-}_{\text{exc}}$, thicker lines), and ECM profiles of ice layers corresponding to the two highest calcium peaks of the first 50 m of the core, denoted A and B in Fig. 1a.

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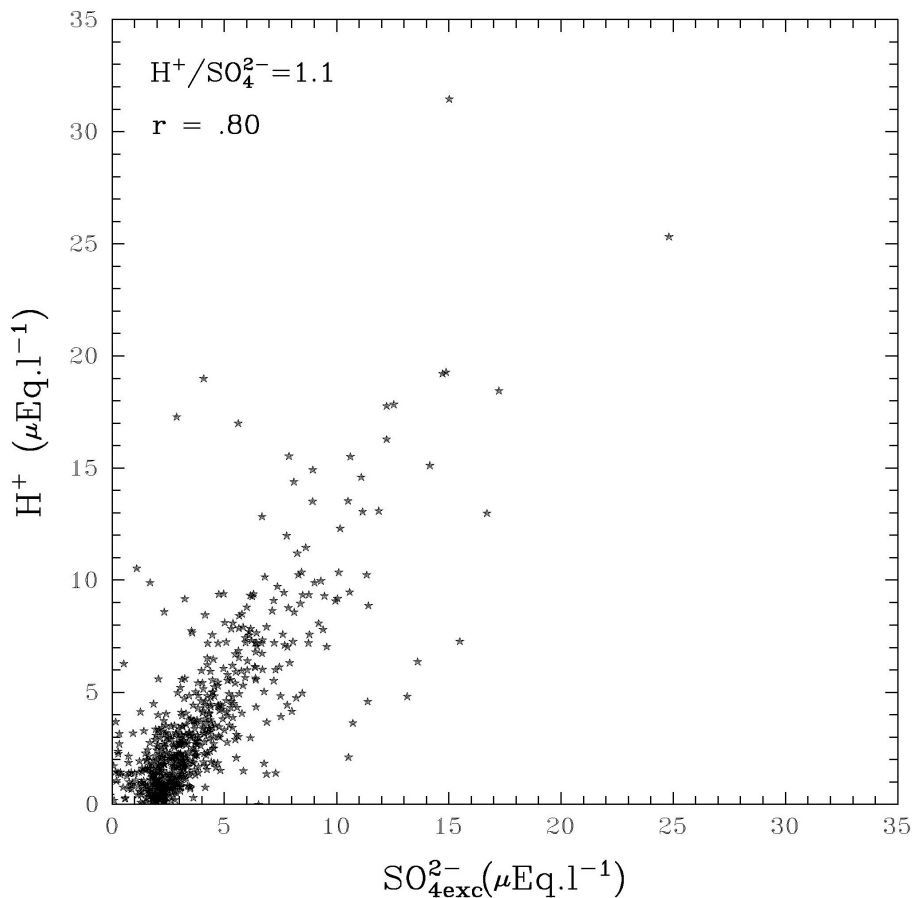


Fig. 3. Ion balance H^+ as a function of the gypsum-free fraction of sulfate ($\text{SO}_4^{2-}_{\text{exc}}$).

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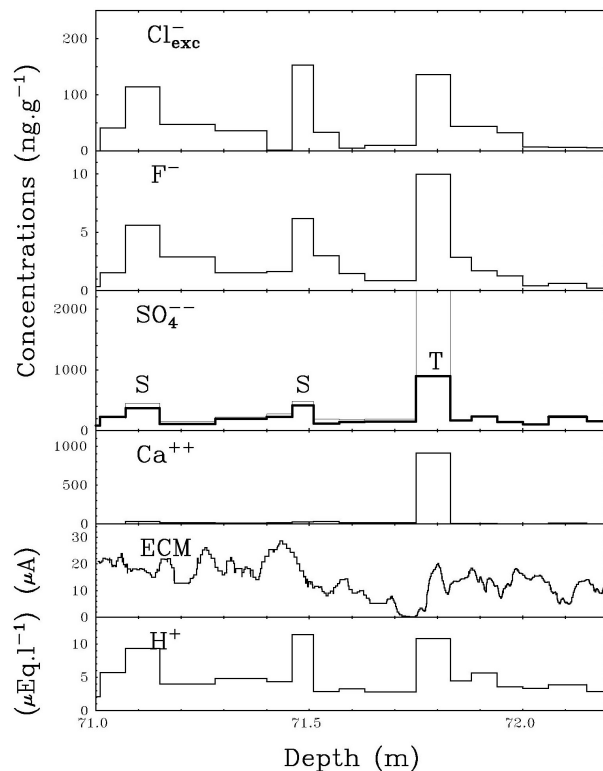


Fig. 4. Detailed depth profiles of H^+ , ECM, Ca^{2+} , SO_4^{2-} (thinner line), $\text{SO}_4^{2-}_{\text{exc}}$ (thicker line), F^- and Cl^-_{exc} in the ice layers including ECM peak (1) in Fig. 1, and likely to correspond to the “Tambora” period. $\text{SO}_4^{2-}_{\text{exc}}$ is the gypsum-free fraction of sulfate, and Cl^-_{exc} was calculated removing the possible halite contribution to Cl^- (see text). Letters T and S denote tropospheric or stratospheric potential input, respectively.

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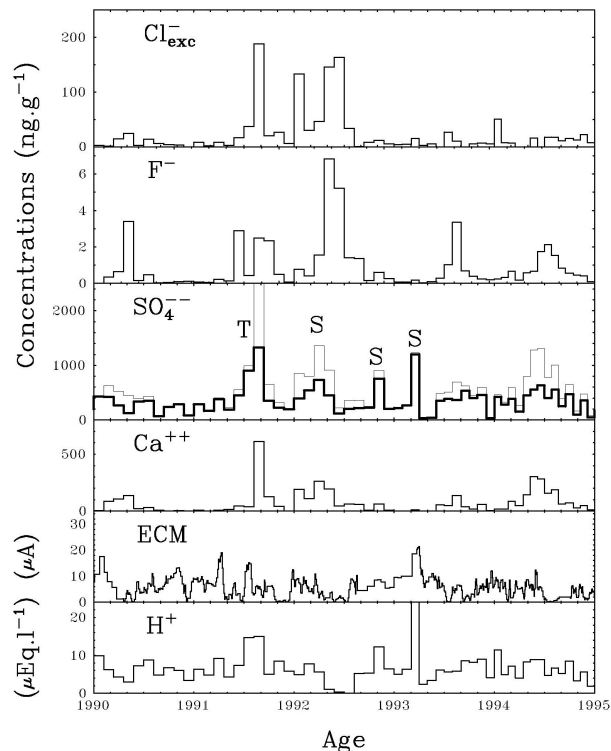


Fig. 5. Detailed time profiles of H^+ , ECM, Ca^{2+} , SO_4^{2-} (thinner line), $\text{SO}_4^{2-}_{\text{exc}}$ (thicker line), F^- and Cl^-_{exc} during the time period corresponding to ECM peak (3) in Fig. 1, and likely to correspond to the Pinatubo eruption. $\text{SO}_4^{2-}_{\text{exc}}$ is the gypsum-free fraction of sulfate, and Cl^-_{exc} was calculated removing the possible halite contribution to Cl^- concentration (see text). Letters T and S denote tropospheric or stratospheric potential input, respectively.

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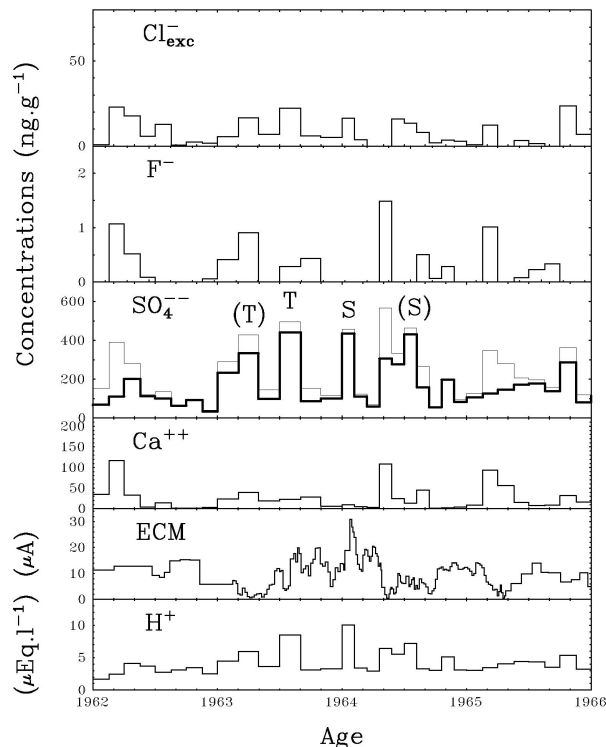


Fig. 6. Detailed time profiles of H^+ , ECM, Ca^{2+} , SO_4^{2-} (thinner line), $\text{SO}_4^{2-}_{\text{exc}}$ (thicker line), F^- and Cl^-_{exc} during the time period corresponding to ECM peak (2) in Fig. 1, and likely to correspond to the Agung eruption. $\text{SO}_4^{2-}_{\text{exc}}$ is the gypsum-free fraction of sulfate, and Cl^-_{exc} was calculated removing the possible halite contribution to Cl^- concentration (see text). Letters T and S denote tropospheric or stratospheric potential input, respectively.

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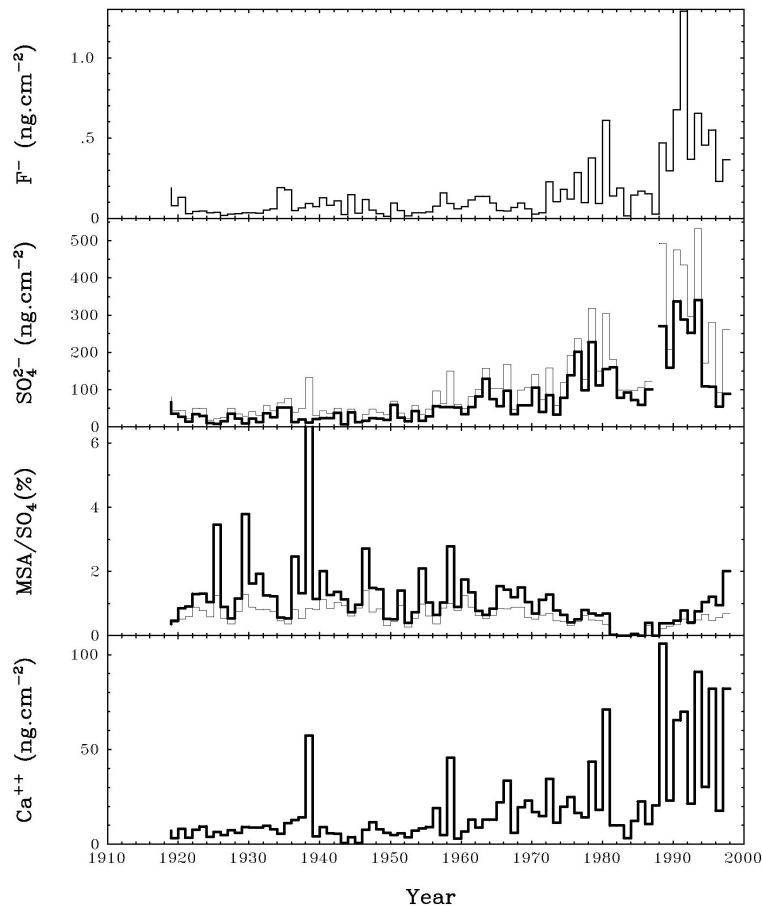


Fig. 7. Annual deposition fluxes of SO_4^{2-} (thinner line), $\text{SO}_4^{2-}_{\text{exc}}$ (thicker line) and F^- calculated from 1918 to 1988. Yearly averaged methane sulfonate (MSA) to SO_4^{2-} (thinner line) and to $\text{SO}_4^{2-}_{\text{exc}}$ (thicker line) are also reported.

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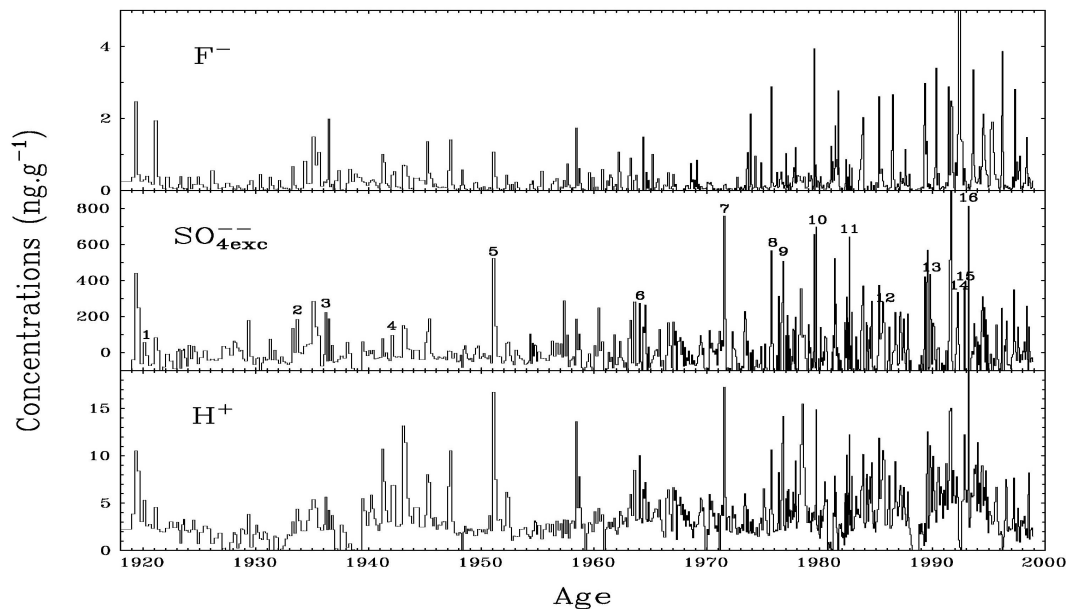


Fig. 8. Detailed time profiles of H^+ (expressed in $\mu\text{Eq.l}^{-1}$), $\text{SO}_4^{2-}_{\text{exc}}$ and F^- , considered as possible volcanism proxies, over the 1918–1998 time period. Numbers 1 to 16 denote peaks likely to be produced by the subsidence of stratospheric H_2SO_4 . All concentrations are in ng.g^{-1} , except H^+ which is in $\mu\text{Eq.l}^{-1}$.

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