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Ammonium record over the last 96 years from the Muztagata glacier in Central Asia

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As a major alkaline gas in the atmosphere, ammonia (NH_3) plays an important role in atmospheric chemistry. However, there is little knowledge about NH_3 variations in the Central Asia. Here we analyzed the ammonium (NH_4^+) history recorded in an ice core from the East Pamir in Central Asia, which was drilled on the Mt. Muztagata at the elevation of 7010 m a.s.l. in 2003. The core was carefully dated and NH_4^+ concentration history during 1907—2002 was reconstructed. The result shows that NH_4^+ concentration remained approximately constant until the 1930s after a sudden decrease at the very beginning of the 20th century, followed by a minimum in 1940 before increasing steadily to the peak at the end of 1990s. It is found that the annual mean NH_4^+ concentration was strongly associated with the Northern Hemisphere temperature, suggesting the impact of temperature on NH_3 emissions in the Central Asia. Moreover, an increase of NH_4^+ concentration after 1940 also reflects the enhancement of NH_3 emissions from anthropogenic sources such as fertilizer applications and livestock wastes in the 20th century.

Muztagata, ice core, ammonium concentration, ammonia emission, deposition, source

As the primary gaseous base in the atmosphere[1], NH_3 influences the oxidation rate of SO_2 in cloud[2,3], and neutralizes the acids generated in the atmosphere by the oxidation of SO_2 and NO_x ($NO + NO_2$). The resulting NH_4^+ is a major component of atmospheric aerosols and precipitation[4−8]. Atmospheric NH_3 is derived from a variety of sources, including livestock wastes, fertilizer applications, biomass burning, and so on[1,9]. Globally, domestic animals are the largest source of atmospheric NH_3, comprising ~40% of natural and anthropogenic emissions combined, while synthetic fertilizers and agricultural crops together account for ~23% of total emissions[5,8,10,11]. NH_3 is one of the most important N-containing compounds in the global nitrogen cycle. Excessive nitrogen deposition may lead to the eutrophication of terrestrial and aquatic ecosystems, altering both their ecological functioning and the diversity of plant species[4,5,12,13], and to some extent, affecting global carbon cycle[14−16]. In addition, atmospheric NH_3 is considered to be acidifying after deposition because of the nitrification processes by Nitrosomas and Nitrobacteria bacteria in the soil where H^+ is produced[2,17−19]. The control measures for NH_3 emissions are much later implemented than that for SO_2 and NO_x, which has caused the constantly high level of NH_3 emission rate during the recent decades. The total NH_3 emissions in Asia, for example, showed a dramatic increase from 24.6 Tg in 1989/1991 to 27.5 Tg in 2000[20,21].

However, due to a lack of continuous long-term

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measurements of atmospheric NH_3 or NH_4^+ in precipitation, it was difficult to further understand the impacts of NH_3 deposition on human health and biological environment. To find an alternative approach is therefore impending for investigating the atmospheric NH_3 content of the past. The high-resoluted ice core record thus outstands as one of the ideal proxies for paleoenvironmental and paleoclimatic reconstruction[22 − 24]. The variations of atmospheric NH_3 content can be retrieved from the NH_4^+ concentration in ice cores, thereby reflecting the changes of NH_3 emission strength from regional sources. The global estimates of NH_3 emissions by some researchers differed from each other due to large uncertainties in emission factors for source categories[1,2,8]. Furthermore, NH_3 emission shows a high spatial and temporal variability, which makes it very difficult to estimate global distribution of atmospheric NH_3 content from measurement alone. It is therefore necessary to reveal the regional atmospheric NH_3 content. Bearing this in mind, in this paper we will discuss the changes of atmospheric NH_3 content in the Central Asia over the last 96 years by analyzing the variations of NH_4^+ concentration in a Muztagata ice core.

1 Methodology

1.1 Ice core drilling and analytical procedure

Mt. Muztagata (75°06′E, 38°17′N, 7546 m a.s.l.) is located on the East Pamir Plateau, near the border of Tajikistan, Kyrgyzstan and Kazakhstan in Central Asia. In the summer of 2003, five ice cores (diameter 9.4 cm) were drilled at the elevation of 7010 m a.s.l. on the Muztagata glacier. The borehole temperature is –23.09℃ at 10 m depth and –25.73℃ near the bottom. The mean annual accumulation rate is ~56 cm (w.e.a^{-1}), and the firn/ice transition is observed at a depth of ~33 m[25]. The high accumulation rate and low englacial temperature ensure the preservation of a climatic and environmental record. The ice cores were transported frozen to the Cold and Arid Regions Environmental and Engineering Research Institute (CAREERI), Chinese Academy of Sciences (CAS) for analysis.

This study focuses on the 54.6 m long Core3 nearly down to the bedrock. The ice core was sectioned continuously into 1223 samples, with a resolution of 3—5 cm per length. For each of the sample, an outer part of 1—1.5 cm thick was removed with pre-cleaned stainless bistouries for stable oxygen isotope analysis, leaving the inner portion in pre-cleaned polyethylene sample container for further chemical analysis. All sampling procedures were performed in a cold (–5℃) clean room and were strictly quality-controlled to avoid any possible contamination. The samples were afterwards stored frozen in a cold room (–20℃) till analysis.

Analyses of stable oxygen isotope ratios ($\delta^{18}O$) and soluble anion species (Cl^-, NO_3^-, SO_4^{2-}) were performed in the Laboratory of Environment and Process of the Institute of Tibetan Plateau Research, CAS, using a MAT-253 mass spectrometer and a Dionex ICS-2500 ion chromatography, respectively. $\delta^{18}O$ values are expressed as the relative deviation of heavy isotope content of standard mean ocean water (SMOW), with the precision of the $\delta^{18}O$ measurement better than ±0.2‰. The soluble cation species (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) were analyzed by a Dionex DX-600 ion chromatography in the State Key Laboratory of Cryospheric Science, CAREERI, CAS. Anions were analyzed with an AS11 analytical column, AG11 guard column, ASRS-ULTRA Ⅱ4-mm suppressor, 0.5 mL sampling loop and 25 mmol/L KOH eluent. Cations were analyzed with a CS12A analytical column, CG12A guard column, CSRS-ULTRA 4-mm suppressor, 25 μL sampling loop and 25 mmol/L MSA eluent. Samples were melted immediately prior to ion chromatographic analysis for inorganic ion compositions. The analytical uncertainty for NH_4^+ concentration is <3%, with others <1%. The detection limits for all ions are less than $1 ng \cdot g^{-1}$. Detailed ion chromatography techniques were described by Buck and others[26]. Baseline values for NH_4^+ were subtracted from the measurement data.

1.2 Ice core dating

This ice core was dated by counting the seasonal variations of the stable oxygen isotopic profile, with verification in 1954 and 1963 by double β activity peaks as the results of the atmospheric thermonuclear tests in the early 1950s and the early 1960s. The upper 51.6 m of this core, including 1122 samples, was dated to span the period of 1907—2002 (Figure 1). The lowest 3 m section was not dated due to layer thinning near the bottom. Moreover, the profile of $\delta^{18}O$ in this core is strongly consistent during the overlapping period with the one in another 41.6 m ice core[27] (figure omitted) drilled at the same site, thus verifying our dating result.

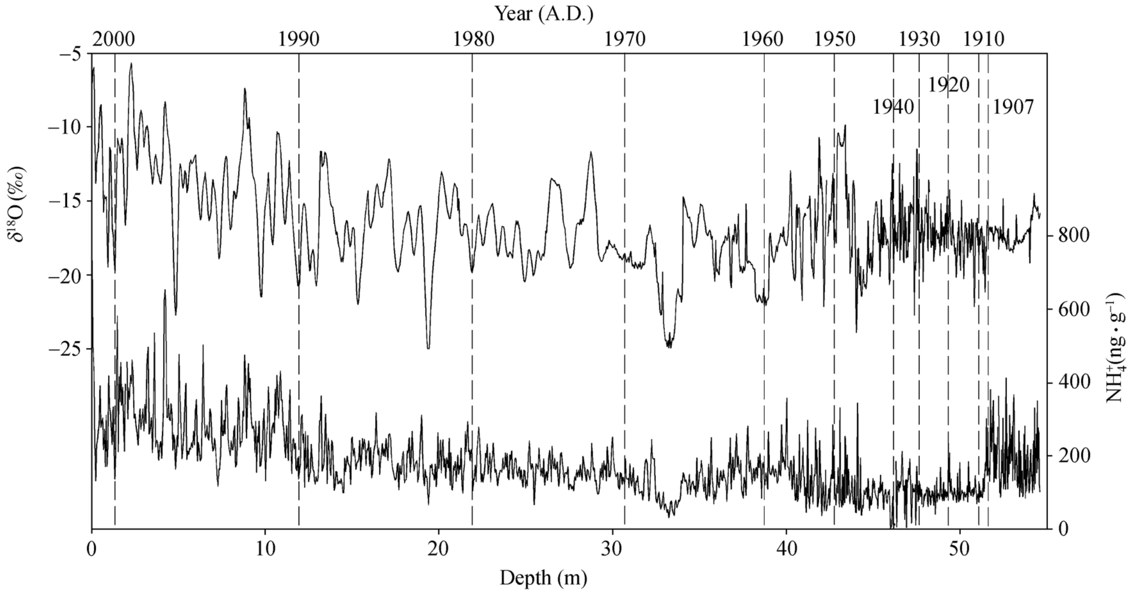

Figure 1 Variations of $\delta^{18}O$ and NH_4^+ concentration (original sampling) in Muztagata ice core and dating result.

2 Results and discussions

2.1 Record of NH_4^+ concentration in Muztagata ice core

The original NH_4^+ record is shown in Figure 1. Large short-term concentration fluctuations are observed, and attributed to seasonal effects. The concentration of the aerosol species in snow reflects the pronounced seasonality of the atmospheric aerosol concentration. In the case of NH_4^+, the seasonality is influenced by the seasonal variations of NH_3 emissions from animal excreta, soil and natural plant which are remarkably dependent on the ambient temperature[4,5,8]. The agricultural activities, which are maximal in summer, also augment the seasonal variations[28]. Figure 2 shows the variations of annual mean NH_4^+ concentration in the period of 1907—2002, from which we can see that NH_4^+ concentration remained approximately constant until in the 1930s after a sudden decrease at the very beginning of 20th century, followed by a minimum in 1940 before increasing steadily to the peak at the end of 1990s. Though a weak drop can be observed around 1967/1968, the decadal average NH_4^+ concentration in the 1990s generally increased by a factor of three compared to the 1940s.

The profiles of NH_4^+ concentration from Muztagata ice core and East Rongbuk ice core[9,29,30] in the Himalayas both displayed a decline trend at the beginning of 20th century and a rising trend after 1940 (Figure 2). However, the two series peaked at different periods of time, with Muztagata ice core series peaking in the late 1990s, while the East Rongbuk series in the 1980s. East Rongbuk NH_4^+ variations reflect the changes of agriculture activities in the East and Southeast Asia and its relationship with atmospheric circulation[9,29,30]. These four time series of NH_4^+ concentration in Figure 2 all showed significant increase trends in the 20th century. It is also noteworthy, however, that NH_4^+ concentration in Colle Gnifetti ice core[6] had substantially increased since as early as the end of 19th century, earlier by about 50 years than that in Muztagata ice core. NH_4^+ in Belukha ice core from the Siberian Altai[31] displayed a decline trend since the 1880s, then experienced a strong

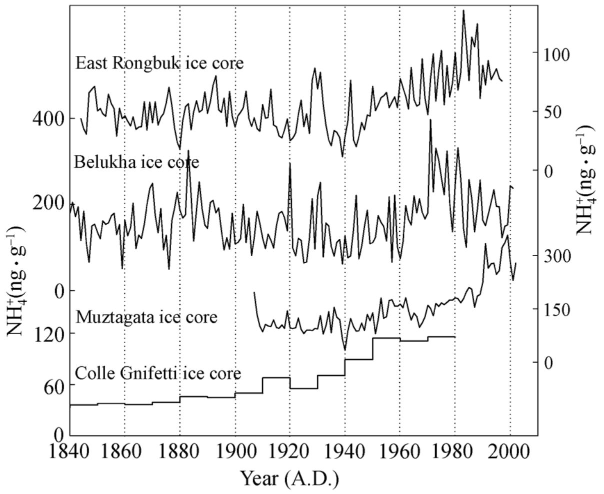

Figure 2 Variations in annual mean NH_4^+ concentration in ice cores from East Rongbuk[9,29,30], Belukha[31], Muztagata and Colle Gnifetti[6].

increase in the 1940s until peaked in the 1970s, though a slight decrease was apparent during the 1980s—1990s. The different NH_4^+ variations in ice cores may be caused by the regional differences in emission source strength and the atmospheric transport and deposition for NH_x ($NH_3 + NH_4^+$).

2.2 Relationship between NH_4^+ concentration and temperature

As NH_3 emission is mainly influenced by temperature, NH_3 content in the atmosphere therefore exhibits pronounced seasonal and diurnal cycles[32]. Figure 3 shows the profiles of annual mean NH_4^+ concentration and Northern Hemisphere temperature anomaly[33]. For a comparison, annual mean air temperature record (data available at http://222.82.227.218:81/scbweb/index.htm) is also given from Taxkorgen Meteorological Station (75°14′E, 37°47′N, 3100 m a.s.l.), the nearest meteorological station to the ice core drilling site. Similar trends exist between NH_4^+ concentration and air temperature during the overlapping period. The series of NH_4^+ concentration is significantly correlated with that of the Northern Hemisphere temperature anomaly, especially after 1940 with the correlation coefficient of 0.70 (significant at 0.01 level, $P < 0.0001$). Global temperature fluctuations can alter the plant community structure in many ecosystems, thereby affecting the emission strength of atmospheric NH_3 from natural sources such as plant, soil and ocean. The high temperature may speed up NH_3 emissions from breakdown of animal excreta and dead plant in soil by ammoniafication[30] (Defined as the breaking down of organic nitrogen compounds with releasing NH_3 or NH_4^+). The different responses of NH_4^+ concentration to temperature before and after 1940 may be associated to some extent with nitrogen fertilizer applications. Synthetic fertilizer were not widely used until the middle of the 20th century[30,34], causing more NH_3 volatilization from fertilizer applications accompanied by rising temperature[24,35]. Besides, as a long-term nitrogen reservoir, soil has released more and more nitrogen compounds under a rapid warming over the past decades, thus contributing significantly to the total amounts of atmospheric NH_3.

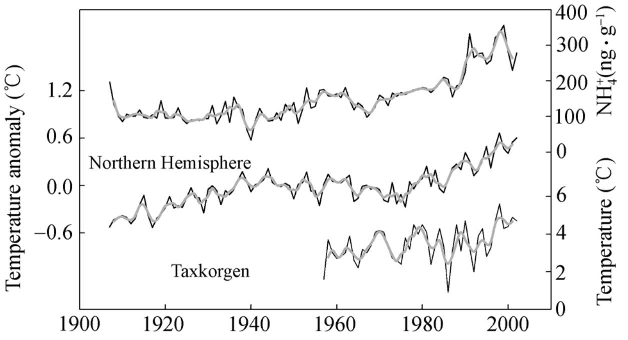

Figure 3 Comparison of annual mean NH_4^+ concentration with air temperature from Taxkorgen Meteorological Station and Northern Hemisphere temperature anomaly. The thick grey line represents the 3-year smoothing average.

2.3 Relationship between NH_4^+ and acid species

A substantial part of the acid in the atmosphere is neutralized by NH_3[4−8]. The resulting $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3 are important constituents of airborne fine particulate matter[36]. In order to explore the possible effect of acid species on NH_4^+ concentration in the Muztagata ice core, annual mean concentrations are compared between NH_4^+ and acidic ions ($SO_4^{2-} + NO_3^-$) (Figure 4). The correlation coefficients between the two time series are 0.61 during 1907—2002 and 0.76 during 1940—2002 (All significant at 0.01 level, $P < 0.0001$), respectively. This suggests that the three species may share the same sources or transportation and deposition processes[37,38]. It is notable that annual mean NH_4^+ concentration correlates more strongly with NO_3^- than with SO_4^{2-}, with the correlation coefficients of 0.69 and 0.50 (All significant at 0.01 level, $P < 0.0001$), respectively. This is also the case with the East Rongbuk ice core[29] and Belukha ice core[38] (figures omitted), given the fact that atmospheric NH_3 and NO_x have lots of common sources such as fertilizer applications, natural soil, and biomass burning.

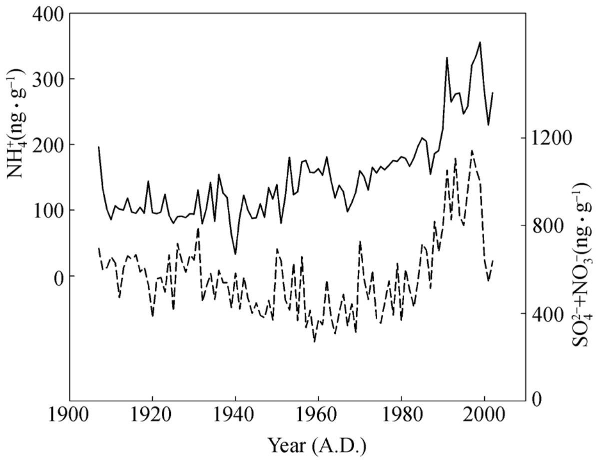

Figure 4 Comparison of annual mean concentrations between NH_4^+ and acidic ions ($SO_4^{2-} + NO_3^-$) in Muztagata ice core.

Virtually all NH_x emissions occur in the form of NH_3, with NH_4^+ in the atmosphere originated from chemical reactions of NH_3. Some of the emitted NH_3 return to the surface by dry deposition, while the others diffuse upwards over the source areas and then react with acid species. The reaction rate of NH_3 and acid species depends on the acid concentration, atmospheric humidity and air temperature[4]. Also, the reaction rate has a large influence on the deposition and long-distance transport of NH_x because NH_3 is usually dry deposited at a high rate, whereas NH_4^+ is deposited much more slowly. Thus the amount and the temporal and spatial distribution of atmospheric NH_x depend not only on atmospheric turbulence but also on the degree to which the air mass is polluted and acidified[30]. Based on these analyses, it is reasonable to conclude that NH_4^+ concentration increase in the Muztagata ice core can be partly attributed to enhanced atmospheric acidification.

2.4 Possible sources of NH_4^+ in ice core

In the global emission inventories for NH_3, about 60%—70% of the global NH_3 emission is estimated to come from the anthropogenic sources[4,5]. The regions with the highest emission rates are found in Europe, the Indian Subcontinent, eastern China[4,5], and parts of U.S[39]. The rising NH_4^+ concentration in Muztagata after 1940, partly due to the enhanced emission from natural sources caused by global warming, probably originated mainly from the increasing anthropogenic emissions. A growing population has boosted the agricultural activities, fertilizer applications and energy consumption, all of which would contribute to an increasing amount of NH_3 emission year by year[30,31]. The similar trends of annual mean concentrations between NH_4^+ and acid species suggest not only that NH_4^+ concentration was affected by the acid content in the atmosphere, but also that the precursor gases (NH_3, SO_2 and NO_x) were all associated with anthropogenic pollution.

Arguably, dry deposition of NH_3 near the sources is most important for its short atmospheric lifetime. On the other hand, NH_3 can be converted to NH_4^+ in cloud droplets or on aerosol particles, resulting in a longer atmospheric lifetime than gaseous NH_3. NH_4^+ can therefore be transported over long distances away from the sources and be mainly deposited by wet deposition[4,9]. The climate of Mt. Muztagata region is affected by the prevailing westerlies. Accordingly, precipitation in this region is largely contributed by water vapor transport from the westerlies and local moisture circulation[25,40]. Countries in Europe and Central Asia are therefore attributed as the main source regions for NH_4^+ in Muztagata ice core, given their locations in the upwind direction of Mt. Muztagata. The rising trend of NH_4^+ concentration in Colle Gnifetti ice core showed that NH_3 emissions in Europe had substantially increased in the 20th century[6]. Yet NH_3 emission rates in European countries either showed a somewhat decline trend or remained stable during the 1980s—1990s[19,34]. It seems that the variations of NH_4^+ concentration in Muztagata ice core were slightly, if not at all, affected by NH_3 emissions in Europe based on the different trends between them.

The variations of NH_4^+ concentration in Muztagata ice core differ from those in Belukha ice core, although they are all subjected to the control of westerlies. This indicates that local emission sources around the regions had more contributions to the two ice cores. NH_4^+ concentration in Belukha ice core was attributable to biogenic emissions, biomass burning and agricultural activities in southern Siberia and its surrounding areas[31]. Mt. Muztagata is adjacent to Central Asia regions, where agriculture and animal husbandry are well developed and play an important role in their economy[41,42]. Emissions from agricultural activities, both crop and animal, are exactly the major sources for atmospheric NH_3[1,9,10]. Otherwise, the total NH_3 amount emitted from Commonwealth of Independent States presented a persistent increase during the recent decades[34]. Based on the above discussions, we suggest that NH_4^+ in Muztagata ice core is mainly originated from NH_3 emissions in Central Asia and its surrounding regions. The contribution from Europe was trivial, though it does exist. As a confirmation of our result, Kreutz et al.[43] analyzed the soluble ionic composition of a shallow firn core, Inilchek glacier, central Tianshan Mts. and also suggested that an increase of NH_4^+ concentration was related to regional agricultural activities.

3 Conclusions

Variations of NH_4^+ concentration have been recovered over the last 96 years using an ice core record from Mt. Muztagata. An increase of NH_4^+ concentration after 1940 reflects the enhancement of NH_3 emissions from anthropogenic sources (e.g. fertilizer applications, livestock wastes) and natural sources (e.g. soil, natural plant) in the 20th century. Ice core NH_4^+ record is correlated significantly with Northern Hemisphere temperature anomaly, reflecting a large scale temperature effects on NH_3 emissions and NH_x deposition under global warming. Due to the discrepancy in spatial distribution of atmospheric NH_3, the post-depositional effects for NH_4^+ in ice core, and the shortage of detailed emission inventory in the upwind and surrounding regions, more available data are needed for elucidating the detailed characteristics of NH_4^+ variations in Muztagata ice core and recovering the NH_3 emission history.

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