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Nitrate concentrations in snow from remote areas: implication for the global NO_x flux

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Abstract. Increased emissions of nitrogen compounds to the atmosphere by human activities have been well documented. However, in order to better quantify these anthropogenic emissions, better knowledge of natural emissions rates must be known. In addition, variation in natural emissions through time should be documented. In this note we present data collected and/or analyzed by us for NO_3^- in recent snow from remote regions of the world. We also summarize existing data sets from other remote regions. This is done to establish a better understanding of NO_3^- deposition rates in these regions as well as to add more information to our global understanding of NO_3^- deposition.

Nitrogen compounds are emitted to the atmosphere by both naturally occurring biogenic and geologic sources as well as by human activity. Due to the potential effect of N_2O in Greenhouse warming and the deterioration of aquatic and terrestrial ecosystems by the introduction of HNO_3 via acid precipitation, an understanding of natural fluxes of N compounds is of utmost importance. Nitrogen gas emissions from biogenic sources undoubtedly vary with such regional and global ecological changes as variations in terrestrial biomass and photosynthetic rates (Bowden & Bormann 1986) and soil acidification (Firestone et al. 1980; Martikainen 1985). Recent calculations of natural emission rates of N into the troposphere suggest that the 'best' previous estimates could be off by greater than a factor of two (Galbally 1985; Mooney et al. 1987). The importance of gaseous nitrogen emissions from various ecosystems is poorly understood (Bowden 1986). However, budgetary information indicate that present day anthropogenic activities such as fossil fuel combustion and biomass burning exceed natural sources of nitrogen to the troposphere (Logan 1983). In addition, recent ice core work from Southern Greenland verifies this increase in fossil fuel combustion source in the past 30–40 years in the Northern Hemisphere (Mayewski et al. 1986a). Indeed, the anthropogenic flux of NO_x into the atmosphere is proceeding at a more rapid rate of increase than that of SO_2 input (Galloway & Likens 1981; Mayewski et al. 1986a). If assessments of the anthropogenically produced nitrogen species into the troposphere are to be made, measurements of nitrate in precipitation from remote areas far from anthropogenically affected areas are needed. Information on depositional fluxes from remote locations can improve our estimates in determining the various natural source strengths for nitrogen compounds.

In this note we present recent measurements of NO_3^- concentrations for combined wet and dry deposition from surface snows collected from remote locations. This is done to add additional information regarding natural N depositional fluxes. These samples include those collected and analyzed by us as well as samples analyzed in our laboratory but collected by others. Samples include previously unpublished results from Hofsjokull, Iceland (collected by us), and Dominion Range and Southern Victoria Land in Antarctica (collected by us) as well as samples from Mt. Kilimanjaro in Central Africa (collected by E. Friedman). From these data and published data anlayzed in our laboratory from other locations in Antarctica, Greenland, Peru, the Canadian Rockies, the Indian Himalayas, the Nepalese Himalayas, the Pakistan Karakoram, Heard Island in the Southern Indian Ocean and the Tibetan Himalayas (Butler et al. 1980; Lyons & Mayewski 1983; Allen et al. 1985; Lyons et al. 1985; Spencer et al. 1985; Mayewski et al. 1987; Wake 1987; Jenkins et al. 1987), depositional fluxes of nitrate have been calculated and compared.

Surface samples were collected by pushing precleaned polyethylene or polycarbonate containers along the snow surface. Deeper samples were collected from hand dug snowpit faces. The sampled snowpit face was thoroughly cleaned with precleaned plexiglass scrapers prior to sampling (Twickler et al. 1986; Mayewski et al. 1987), and in all cases, the sampling was carefully undertaken by individuals wearing polyethylene gloves and particle masks and in most cases non-particulating clothing. Sample containers were precleaned according to Lyons & Mayewski (1983) and Mayewski et al. (1986a). In all cases blanks containing distilled-deionized water in sample collection containers were analyzed as samples. Blanks were always extremely low compared to the sample values and in most cases were at or below our analytical detection limits. During the nine-year period of sample collection and analysis, two different analytical methods were utilized: a colorimetric technique using an auto-analyzer (AA) system (Lyons et al. 1985) and ion chromatography (IC) (Mayewski et al. 1986a). To ensure that these two techniques yielded comparable results, a series of samples were analyzed by both methods. In general, the colorimetric method yielded values 3% higher than the IC method (Fig. 1). However, it

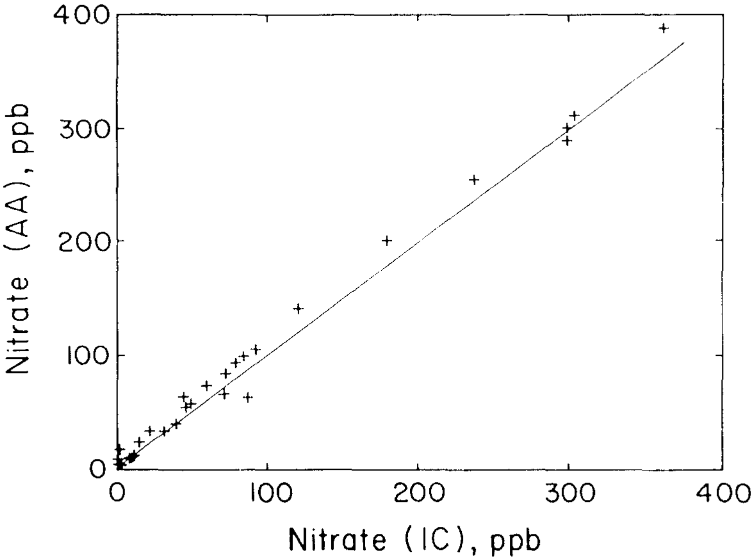

Fig. 1. **Comparison of nitrate concentrations of samples measured by colorimetric (AA) and ion chromotographic (IC) techniques.**

can be seen that there is very good agreement between the two methods and therefore the data from the two different techniques can be directly compared. This is similar to what has been observed by other workers analyzing remote precipitation samples (Johnson & Parnell 1986).

For the past ten years our group has been analyzing snow and ice samples from high elevation and/or high latitude glaciated areas throughout the world for acidic anions as well as other chemical species. These samples include those collected by our group as well as those collected by others using our protocol, sampling equipment and techniques. In all cases the analyses were done at UNH. Our NO_3^- data from surface (top 1 m) and other NO_3^- data from recent snow from glaciated areas are presented in Table 1. These samples represent arithmetic means of the analysis of the upper meters of snow from freshly dug snowpits or surface snow (top few cms). These data along with the published data from other remote areas provide an important set of 'background' NO_3^- values that can be compared to data from areas in the world that have been more acutely affected by anthropogenically introduced NO_3^-. In addition, these remote area data provide an indirect means of better quantifying the tropospheric N budget. The studies of Neftel et al. (1985), Mayewski et al. (1986) and Finkel et al. (1986) have shown unequivocally that the chemistry of NO_3^- in Greenland ice cores mimics that of Northern Hemispheric anthropogenic emissions of NO_x over the past century. Thus, glaciochemical records reflect the relative

Table 1. Nitrate concentrations in remote area snow.

Location	Mean nitrate, μM	Reference
Northern Greenland		
Camp Century	1.9	[13]
Southern Greenland		
pre 1940	0.8	[1]
present	1.6	
Iceland	0.7	This work
Tibet	6.8	[2]
Nepal	~8.0	[3]
Indian Himalayas	1.4	[4]
Pakistan Karakoram	2.9	[5]
Canadian Rockies, Alberta	1.1	[6]
Cascade–Sierra Nevadas	1.8	[7]
Quelccaya, Peru	1.1	[8]
Mt. Kilimanjaro	27.6	This work
Heard Island	~0.3	[9]
Spitzbergen	< 1.5	[10]
Mt. Logan, Yukon Territory	0.8	[15]
Colle Gnifetti, Switzerland	4.5	[19]
Antarctica		
James Ross Island	0.4	[11]
N. Victoria Land	0.7	[12]
S. Victoria Land	1.1	This work
Dominion Range	1.6	This work
Ross Ice Shelf	0.7	[13]
Byrd Station	0.6	[13]
Vostok	0.3	[13]
Dome C	0.2	[14]
Gomez Nunatak	0.4	[16]
South Pole	1.9	[14, 17]
Law Dome	0.5	[18]

References
1. Mayewski et al. (1986a, 1987)
2. Mayewski et al. (1986b)
3. Jenkins et al. (1987)
4. Lyons and Mayewski (1983)
5. Wake (1987)
6. Butler et al. (1980)
7. Laird et al. (1980)
8. Lyons et al. (1985)
9. Spencer et al. (1985)
10. Semb et al. (1984)
11. Aristarain et al. (1982)
12. Allen et al. (1985)
13. Herron (1982)
14. Delmas et al. (1982)
15. Holdsworth and Peake (1985)
16. Mulvaney and Peel (1988)
17. Legrand (1987)
18. Ivey et al. (1986)
19. Wagenbach et al. (1988)

changes in atmospheric chemistry through time at any one location. The comparison of similar time horizons on glaciers in different locations should yield a spatial distribution of nitrate fluxes if these glaciers are not subject to 'melt out' of the glaciochemical record and the snow accumulation rate is known or can be determined. This spatial distribution would then allow a temporal depositional history to be estimated at various locations not easily sampled for continuous precipitation chemistry.

We have divided the data into four general groupings: polar, subpolar, temperate and tropical (Table 1). The highest values observed are from the Himalayan–Karakoram regions in Asia, Mt. Kilimanjaro and Colle Gnifetti glacier in Switzerland. The Antarctic data are generally lower than 0.7 μM with the maximum values at or close to the South Pole (i.e. South Pole, Dominion Range). The Greenland values are approximately two times higher than the Antarctic concentrations.

Mayewski et al. (1983) have shown an increase in NO_3^- concentration with elevation in freshly fallen snow in the Himalayas. The higher values of the NO_3^- in the samples from Tibet, Nepal and Pakistan collected at 6300 m, 5500–5700 m and 5450 m, respectively, may thus represent the input of NO_3^- from an upper tropospheric or even a stratospheric source. Wake (1987), however, observed no trend in NO_3^- concentration with elevation (3900–5660 m) in the Karakoram. The samples with the highest nitrate values from Nepal also have high calcium concentrations (up to 3.8 μM). Wake (1987) observed that the highest concentrations of nitrate in a snowpit at 5450 m are associated with above background concentrations of microparticles (0.5–12.7 μm). The NO_3^- concentrations in the Peru snowpit also show a positive relationship with such crustally derived species as Si (Lyons et al. 1985). There is evidence of high aerosol nitrate concentrations in long traveled desert dust particles over both the western south Atlantic (Talbot et al. 1986) and the northern Indian Ocean (Savoie et al. 1987). The higher nitrate data observed in the Asian samples may thus represent the transport of soil from the arid Tibetan Plateau. The higher African concentrations may also represent input of nitrogen-rich soil particles from the surrounding semi-arid countryside. Wolff (1984) has demonstrated that coarse continental aerosols have strong correlations with NO_3^- concentrations. He believes that the source of NO_3^- is the reaction of HNO_3 with the airborne crustal constituents. The Columbia Icefield (Alberta) samples are associated with high concentrations of crustal components (Si and Fe) and probably also reflect a high concentration of continental aerosol. Although detailed data are lacking, Bowden (1986) reports that compared to other ecosystems, the natural fluxes of fixed nitrogen gases from undisturbed arid and semi-arid regions are high. This too may be a factor in the higher concentrations of NO_3^- observed in aerosols derived from these regions.

The Colle Gnifetti, Switzerland data represent snow that has been seriously influenced by anthropogenic nitrogen emissions from Western Europe (Wagenbach et al. 1988). These smaples should not therefore be termed ‘remote’.

The regions primarily influenced by oceanic air rather than continental air such as Mt. Logan, Heard Island, James Ross Island and Southern Greenland, pre 1940 all have relatively low NO_3^- concentrations especially compared to the snows dominated by continental aerosols. These results are supported by recent aerosol measurements made over the Indian Ocean indicating that the 'pure' oceanic background aerosol concentration of NO_3^- is at least one factor or lower than continental aerosol values and may be as much as a factor of 5 times lower than aerosols dominated by desert dust (Savoie et al. 1987). Even the aerosol sampled in Barbados is dominated by NO_3^- associated with Saharan dust (Savoie 1989). Therefore, areas that are 'continentally' influenced should have higher depositional fluxes of NO_3^- than areas that are more influenced by oceanic air or by stratospheric input.

The Antarctic data suggest that the highest concentrations of NO_3^- on the continent are closest to the South Pole. At or near the Pole stratospherically injected material is transfered onto the Earth's surface either through a stratospheric-tropospheric exchange or directly through stratospheric cloud precipitation (Jouzel et al. 1979). Recent measurements of ^{7}Be fluxes in snow collected at South Pole and in Southern Victoria Land support the notion that there is an increased input of stratospherically derived aerosol closer to the Pole (J. Dibb, pers. comm.). Other studies have come to the same conclusion (Meanhaut et al. 1979; Ganapathy 1983; Pourchet et al. 1983). If this is indeed true, the major source of the elevated concentrations of NO_3^- closer to the geographic pole is the stratosphere and therefore represents a long traveled, high altitude aerosol. The samples from Southern Victoria Land come from a valley glacier in Wright Valley. The higher NO_3^- concentrations are associated with elevated Si and Fe values suggesting a significant crustal component is present (Mayewski & Lyons 1982).

We have calculated the depositional fluxes from our data by multiplying the mean snow concentrations by the water equivalent accumulation rates. The accumulation rates have been determined by various means and are reported in the publications cited. The accumulation rates for the Southern Victoria Land, Dominion Range and Iceland data were determined by chemical seasonality techniques as described in Allen et al. (1985) and Lyons et al. (1985). We are unable to determine the Nepal or African fluxes due to lack of accumulation rate information. We understand that due to the small data base any global extrapolation is extremely tentative at best. Yet there are very few regions of the world where remote NO_3^- in precipitation samples have been obtained. The glacier medium provides a small, but extremely important, source of NO_3^- flux information. Therefore we have chosen to present the flux calculations with the caveat that they represent a first approximation that can be added to as more remote sites are visited and data obtained.

Table 2. Calculated nitrogen precipitation fluxes as nitrate–nitrogen.

Nitrogen fluxes kg ha^{-1} yr^{-1}

Location	Lat.		Flux
Polar			
Antarctica			
Northern Victoria Land	~71°S		~0.007
	~71°S		~0.033
Southern Victoria Land	~78°S		~0.006
Dominion Range	~85°S		~0.017
Greenland			
Dye 3	~65°N		~0.05
		Mean	~0.024
Subpolar			
Heard Is	~53°S		~0.045
Iceland	~64°N		~0.1
		Mean	~0.07
Temperate			
Alberta	~52°N		~0.3
India	~35°N		~0.06
Tibet	~29°N		~1.7–3.4
Karakoram	~36°N		~2.9
		Mean	~1.2
Tropical			
Peru	~14°S		~0.17

It is quite evident that the polar regions have the lowest fluxes (Table 2). This is due in part to lack of substantial continental dust input, the fact that rain is a more efficient scavenger of NO_x than snow (Barrie 1985) and that polar regions are more distant from the sources of NO_3^-. Although the source of NO_3^- to polar snow is a topic of much debate (Herron 1982; Parker et al. 1982; Mayewski et al. 1986), the primary source is probably stratospheric. If this is the case, one would suspect very low depositional fluxes due to the lack of continentally derived aerosol enriched in dust and/or biogenically produced NO_3^- as well as anthropogenically introduced NO_3^-. The temperate region mean is higher than the other geographic areas. This is due to the fact that the mean flux value is dominated by the very high fluxes in Tibet and Pakistan associated with continental debris. These results are supported by recent aerosol measurements made over the Indian Ocean indicating that the 'pure' oceanic background aerosol concentration of NO_3^- is at least a factor or 2 lower than continental aerosol values and may be as much as a factor of 5 times lower than aerosols dominated by desert dust

Table 3. Nitrogen fluxes in remote areas in $Kg\,ha^{-1}\,yr^{-1}$.

Location	Lat.	Flux
Poker Flat [1]	67°N	0.076
Amsterdam Is [1]	32°S	0.3
Hawaii [2]	19°N	0.15
Samoa [3]	14°S	0.19
Australia [1]	14°S	0.55
Venezuela [1,4]	1°N	1.42
	10°N	1.3
Costa Rica [5]	10°N	0.5

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1. Galloway et al. (1982)
2. Harding & Miller (1982)
3. Pszenny & Duce (1982, 1986)
4. Lewis (1981)
5. Hendry et al. (1984)

(Savoie et al. 1987). It is not surprising that areas that are 'continentally' influenced should have higher depositional fluxes of NO_3^- than areas that are more influenced by oceanic air or by stratospheric input. Certainly our concentration data from Central Africa (Table 1) indicates that continental dust influence may also be very important there.

Using our NO_x deposition rates as well as published fluxes in other remote areas dominated by rain rather than snow (Table 3) we calculated a non-anthropogenic depositional flux of $19 \times 10^{12}\,g\,N\,yr^{-1}$ on a global basis. This calculation was accomplished by multiplying the mean oceanic, continental and polar fluxes by their respective geographic areas. This value corresponds quite well with the NO_x flux calculations of Logan (1983) even though many fewer data points exist for glacier snow, suggesting that the earlier N fluxes are, indeed, quite reasonable. The fossil fuel emission rate is estimated to be $21 \times 10^{12}\,g\,N\,yr^{-1}$ (Logan 1983). Therefore the non-anthropogenic fluxes are quite similar to the fossil fuel emissions. Because most of the fossil fuel emissions occurs in mid-latitudes in the Northern Hemisphere these are the regions that are most affected. This can be seen by comparing depositional fluxes in truly remote areas to fluxes in non-urban locations downwind from anthropogenic sources such as Bermuda and the Colorado Rockies as well as southern Greenland. The depositional fluxes of NO_3^- are 1.2 and $1.8\,kg\,ha^{-1}\,yr^{-1}$ in Bermuda and the Colorado Rockies, respectively (Church et al. 1982; Kauer 1986). The Bermuda value is an order of magnitude higher than our calculated remote oceanic value, while the Colorado value, is a factor of two higher than our mean remote continental flux.

In this discussion we have only dealt with NO_3^- deposition. Ammonium is also a component of glacier ice and snow. What little data that are available suggest concentrations in polar snow ranging from 0.1 to $1.0\,\mu M$

(Delmas et al. 1982; Legrand & Delmas 1984; Palais & Legrand 1985). Less information is available from temperate and tropical glaciers but what little data there are indicate higher concentrations (Mayewski et al. 1984; Laird et al. 1986). In order to better quantify the global atmospheric nitrogen cycle more NH_4^+ measurements must be made on remote precipitation. This undertaking provides a difficult challenge due to the ease with which ice and snow can be contaminated by NH_3 (Legrand et al. 1984).

The need for an assessment of both natural and anthropogenic fluxes of nitrogen into the environment is of extreme importance for the reasons stated in the beginning of this paper. As our data base from remote areas becomes larger, our ability to better quantify natural fluxes of nitrogen will improve. Although precipitation chemical data from geographic regions unaffected by anthropogenic activities is sparse, this information coupled with historic records preserved in ice cores (Neftel et al. 1985; Mayewski et al. 1986; Finkel et al. 1986) can indicate the relative importance of anthropogenic emissions of NO_x into the atmosphere on a regional, hemispheric and global scale.

The natural flux of N to the atmosphere is $\sim 20 \times 10^{12}\,N\,yr^{-1}$ (Logan 1983). Data from snow samples from remote areas around the world confirm this previous estimate. Our data indicate that the polar N flux is probably dominated by a stratospheric source, while crustal input may be of great importance to the N fluxes in arid and semi-arid regions. Many so-called remote areas are in fact affected by anthropogenic emissions of N. These areas are downwind from large region sources such as North America and Western Europe. Ice cores and snowpit sampling provide a relatively untapped source of detailed historic information regarding both N and S emissions. More effort should be given to obtaining data from high elevation ice masses in Asia and South America in order to better understand the regional and global fluxes of these elements.

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