

Atmospheric nitrous oxide observations above the oceanic surface during the first Chinese Arctic Research Expedition

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Abstract 339 gas samples above oceanic surface were collected on the cruise of “Xuelong” expeditionary ship and nitrous oxide concentrations were analyzed in the laboratory. Results showed that Atmospheric average N_2O concentration was $309 \pm 3.8 \text{ nL/L}$ above the surface of northern Pacific and Arctic ocean. N_2O concentrations were significantly different on the northbound and southbound track in the range of the same latitude, $308.0 \pm 3.5 \text{ nL/L}$ from Shanghai harbor to the Arctic and $311.9 \pm 2.5 \text{ nL/L}$ from the Arctic to Shanghai harbor. N_2O concentration had a greater changing magnitude on the mid- and high-latitude oceanic surface of northern Pacific Ocean than in the other latitudinal ranges. The correlation between the concentrations of the compositions in the aerosol samples and atmospheric N_2O showed that continental sources had a great contribution on atmospheric N_2O concentration above the oceanic surface. Atmospheric N_2O concentration significantly increased when the expeditionary ship approached Shanghai harbor. The average N_2O concentrations were $315.1 \pm 2.5 \text{ nL/L}$, $307.2 \pm 1.4 \text{ nL/L}$ and $306.2 \pm 0.7 \text{ nL/L}$, respectively, at Shanghai harbor, at ice stations and at floating ices. The distribution of N_2O concentrations was related with air pressure and temperature above the mid- and high-latitude Pacific Ocean.

Key words Arctic Ocean, Northern Pacific Ocean, Nitrous oxide, Atmosphere.

1 Introduction

Nitrous oxide (N_2O) is one of the most important gases for heat budget of the terrestrial atmosphere, and its radiative effect on climate change is thought to be about 200 times as strong as CO_2 on a per molecule basis (IPCC 1990). In addition, N_2O plays an important role in the depletion of stratospheric ozone (Ramanathan 1988). From the results of polar ice core analyses and recent direct atmospheric measurements, it is well known that the tropospheric N_2O concentration has been increasing due to human activities, and its increase rates for the last few decades are observed to be $0.5\text{--}2 \text{ nL L}^{-1} \text{ yr}^{-1}$ (IPCC 1990). Such a concentration increase is generally ascribed to direct emissions of N_2O into the atmosphere through fertilizer production, crop production, fossil fuel combustion, biomass

burning, nitric acid production and sewage, as well as to enhancement of biogenic N_2O emissions by additional fixation of nitrogen by cultivation of leguminous plants and deposition of anthropogenic nitrogen compounds in the open atmosphere (Xing 1998; Xing and Zhu 1997; Mosier *et al.* 1997; IPCC 1990). Of the sources of atmospheric N_2O , the oceans are considered significant, though not dominant. McElroy and Wofsy (1986) calculated the oceanic contribution to be about 13% of all sources, with an estimated error of about 50%. Cohen and Gordon (1979) estimated oceanic N_2O concentration arrived at a mean supersaturation of 9%. At the other extreme, studies by Elkins (1978) and Singh *et al.* (1979) reported mean saturation anomalies of 25% and 34%. Butler *et al.* (1989) reported N_2O in the surface water and marine troposphere showed a latitudinally weighted, mean inter-hemispheric difference of 0.97 nL/L, which suggested that 2/3 of the global flux of N_2O into the atmosphere derived from sources in the northern hemisphere. Zhu *et al.* (2003) estimated that 2/5 of the global flux of N_2O is derived from the sources in the southern hemisphere. Stratospheric N_2O has been observed above the ocean in the past few years (Strahan *et al.* 1999; Kondo *et al.* 1999).

Climatic and environmental characteristics, physical and chemical processes in Arctic areas have good connections with global climatic and environmental changes. This area corresponds and feedbacks to global climatic and environmental changes very sensitively, being one important background area for global atmospheric environmental monitoring, and therefore it is considered important for the studies about human activities and global climatic and environmental changes. At present, the data for N_2O concentrations from mid- and high-latitude oceanic surface of northern Pacific Ocean to Arctic Ocean are very sparse. During the first Chinese Arctic Research Expedition (FCARE), we collected gas samples above oceanic surface from Shanghai harbor to Arctic Ocean and analyzed atmospheric N_2O concentrations in the lab. The purpose of this study is to research how oceanic atmospheric N_2O concentrations change with the latitudes and the affecting factors.

2 Gas sampling and N_2O determination

“Xuelong” expeditionary ship set out from Shanghai harbor in China on July 1, 1999. Its cruise was Shanghai harbor—Japanese Sea—Bering Sea—Chukchi Sea—Areas approaching Arctic ices—Chukchi Sea—Bering Sea—Japanese Sea—Shanghai harbor. Latitudinal and longitudinal ranges included 31°N – 7°N , 123°E – 33°W (i. e. Northern Pacific Ocean and the Arctic Ocean). Bering Sea, Chukchi Sea, Canadian Oceanic Basin and areas approaching Arctic ices were the main expedition and research areas (Fig. 1).

We used vacuum vials made in the institute of Japanese agricultural environment, which had been vacuumized to close to zero Pa in advance to collect atmospheric samples above oceanic surface on the cruise. The sampling height was about 2 m above the ship deck. To avoid the effects of anthropogenic factors and expeditionary ship, we collected gas samples upwind on the fore. 3 duplicate gas samples were collected every time to ensure the accuracy of determination and sampling time was 8:00 AM and 20:00 PM every day. The air temperature and pressure on the oceanic surface were simultaneously recorded to investigate the effects of these climatic factors on N_2O concentrations. In addition, more gas sa –

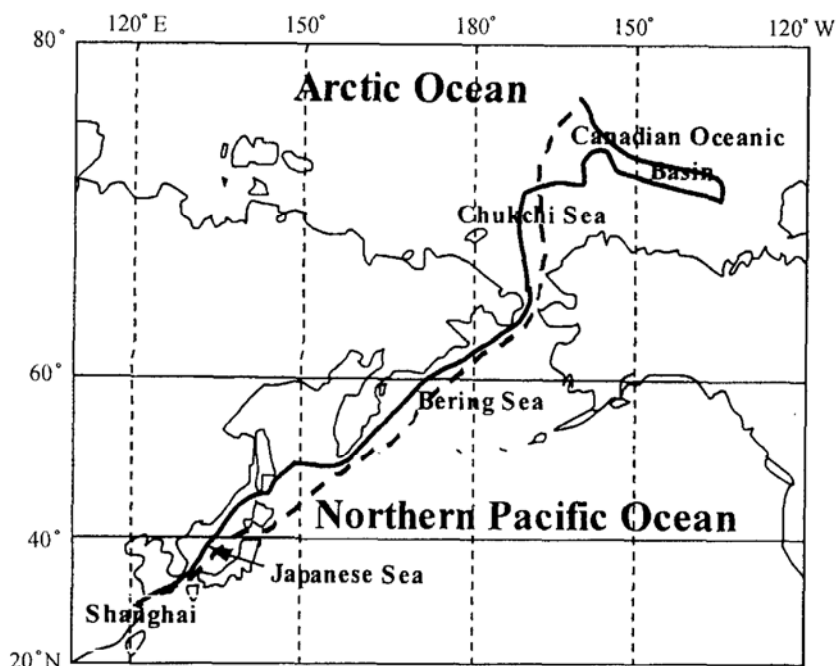


Fig. 1 The cruise track of "Xuelong" expeditionary ship during the first Chinese Arctic Research Expedition. The bold line indicates the northbound track and the ordinary line indicates southbound track.

mples concentrated in the areas approaching Arctic ice, ice stations and floating ices. 339 gas samples on the oceanic surface were collected on the cruise (including northbound and southbound tracks) from July 2 to September 9, 1999. These vials were sealed with a butyl rubber septum and then covered by a plastic cap. High vacuum inside the vial can maintain at least for a year (Xing 1998; Xing and Zhu 1997). In addition, standard gas samples stored in the vials showed no significant change in concentration during 3 months of storage in the laboratory or during transport from the field site to the laboratory, suggesting that the quality of sample air in the vials did not change during the sampling and transport period as well (Sun *et al.* 2000).

The collected gas samples were brought back to China and analyzed in the laboratory of Material Cycling in Pedosphere, Institute of Soil Science, Academia Sinica of China, for the N_2O concentrations. N_2O concentrations were determined by HP5890 II GC using a ^{63}Ni electron capture detector (ECD) (Xing and Zhu 1997; Sun *et al.* 2000). GC-ECD was equipped with back flush system with 10-port valves. A pre-column (2m) and a main-column (Porapak Q, 100 mesh) were used with an argon-methane (95%: 5%) mixture as the carrier gas at a flow rate of 30mL/min. The detector and column temperatures were 330°C and 85°C, respectively. The back flush time was 2.8 min. The injecting gas volume was set up as 3.0ml by using an adjustable pressure syringe injector. Compressed air was used as standard gas with the value of 303 nL/L demarcated by the National Institute of Japanese Agricultural Environment. The variance coefficient for standard samples was within 0.1%–0.4% in ten hours.

To identify the sources for atmospheric N_2O , aerosol sampling was conducted using a

high-volume bulk aerosol sampler (Tianhong Instruments, Wuhan, China) on the cruise of the expeditionary ship. Bulk aerosol samples were collected on $20 \times 25 \text{ cm}^2$ Whatman 41 filters at a flow rate of $1.05 \text{ m}^3 \text{ min}^{-1}$. All filter samples were retrieved from the sampler with strict contamination control procedures. They were sealed into clean plastic bags and kept at 4°C until lab analysis. Aerosol sample preparation and chemical analysis (including MSA , Cl^- , Na^+ , SO_4^{2-} etc) were conducted at the Byrd Polar Research Center of the Ohio State University (Columbus, OH, USA) using DX500 (Dionex, Sunnyville, CA) ion chromatography (IC). The experimental procedures were described in details by Xie *et al.* (2002).

3 Results and discussion

3.1 The latitudinal distribution of atmospheric N_2O above oceanic surface

Fig. 2 is a scatter plot of atmospheric N_2O for the cruise, which had measured mixing ratios between 301 and 318 nL/L for the entire cruise and a mean N_2O mixing ratio of $309.0 \pm 3.8 \text{ nL/L}$. Atmospheric N_2O mixing ratios above oceanic surface may be divided into three areas according to the latitudes on the northbound track of expeditionary ship: The first area was in the range from $30\text{--}35^\circ\text{N}$, where N_2O mixing ratios were above 309.0 nL/L and the average was 312.0 nL/L ($n=9$); This latitudinal range contained noticeably higher mean N_2O mixing ratios. The second area was in the range of $35\text{--}65^\circ\text{N}$, where N_2O mixing ratios were generally lower than 307.0 nL/L and the average fell down to about 304.0 nL/L ($n=60$); This range was the area in which the inflow and outflow regions of Typhoon Mireille were sampled (Newell *et al.* 1996; Collins *et al.* 1996). Therefore the lower N_2O levels may be attributed to encountering some air masses brought from the mid-Pacific near the equator within the typhoon. In addition, Collins *et al.* (1996) proposed that this latitudinal range appeared to be preferentially influenced by the stratosphere/troposphere exchange, which would reduce the N_2O mixing ratio at higher latitudes. The third area was from 65°N to the Arctic, where N_2O concentration significantly increased and the average was about 309.3 nL/L ($n=123$). Atmospheric N_2O concentrations generally appeared a linearly rising trend on the southbound track from the Arctic back to Shanghai harbor and most of the values were above 309 nL/L with the exception of N_2O concentrations in the range of $40\text{--}45^\circ\text{N}$ (Fig. 2). The distribution of atmospheric N_2O concentrations was also different in the same latitudinal range on the cruise. Atmospheric N_2O concentrations on the northbound and southbound tracks had no apparent distinction in the ranges of $31\text{--}35^\circ\text{N}$ and $70\text{--}77^\circ\text{N}$ (2.6 nL/L and 0.5 nL/L, respectively), whereas the greater differences appeared in the other latitudinal ranges, especially in the range of $35\text{--}45^\circ\text{N}$ (10 nL/L), which may be relate with the effects of continental sources because of the different sampling time. The wind direction was from southeast and the continental sources had little effects on atmospheric N_2O concentrations above the oceanic surface on the northbound track, whereas the wind direction switched, and was from the Eur-Asia continent (northwest) and the concentrations were more greatly affected by continental sources on the southbound track.

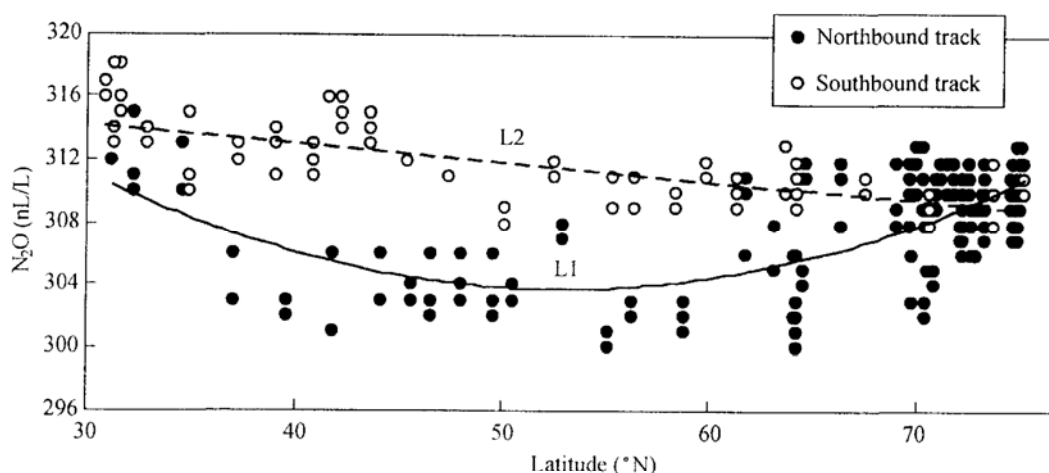


Fig. 2 The latitudinal distribution of N_2O above the oceanic surface along the cruise of "Xuelong" expeditionary ship ($L_1: y_1 = 0.0142x_1^2 - 1.5064x_1 + 343.64$, $r_1 = 0.67$ $n = 198$; $L_2: y_2 = -0.1217x_2 + 318.15$, $r_2 = 0.68$ $n = 81$).

The atmospheric average mixing ratio of nitrous oxide were listed in table 1 above the seas in continental margin, Arctic Ocean and on Antarctic Fildes Peninsula. The atmosphere above the East China Sea (31-35°N) contained noticeably higher N_2O mixing ratios than those above the rest oceanic regions. During September-October 1991 as part of the NASA Pacific Exploratory Mission (PEM)-West A, airborne N_2O observations over the western Pacific Ocean were conducted between the equator and the 40°N latitude. This latitude survey indicated that the latitude ranges between 20°N and 35°N contained significantly higher mean N_2O mixing ratios due to the continental sources (Collins *et al.* 1996). Our observation was consistent with the results reported by Collins *et al.* (1996). Mean N_2O mixing ratios above Japanese Sea were above 2 nL/L higher than those above Bering Sea. Japanese Sea is located in the mid-latitudes and closer to the Asian continent. Since the sampled air mass were significantly influenced by Asian continental sources (biogenic and urban-anthropogenic), contained larger mean N_2O and other pollutant mixing ratios. Bering sea is located out of more remote locations, and the lower mean N_2O mixing ratio is indicative of (1) being farther away from the dominant land sources and (2) stronger center Pacific influences. High atmospheric N_2O concentrations were also observed above Chukchi Sea and Canadian Oceanic Basin due to the effects of continental sources from Siberian and northern American. In addition, the mean N_2O concentration on Antarctic Fildes Peninsula was slightly higher than that above high-latitude oceanic surface of northwestern Pacific Ocean, and almost equal to that above Arctic Ocean. The atmospheric N_2O concentration on Antarctic Nelson Icecap was 0.8 nL/L higher than those for Arctic floating ices and ice stations, indicating that atmospheric N_2O concentrations in two pole regions had no significant distinctions.

3.2 Comparison of N_2O concentrations above ocean surface at 8:00 AM and 20:00 PM

The changing trend of atmospheric N_2O concentrations above oceanic surface is con-

sistent at 8:00 AM and 20:00 PM (Fig. 3). The average concentrations were 307.8 ± 3.3 nL/L ($n=96$) and 307.9 ± 1.5 nL/L ($n=96$), respectively, for the observations at 8:00 AM and 20:00 PM on the northbound track, and the difference of N_2O concentrations was insignificant according to t test ($t=0.21 < t_{0.1, 62} = 1.64$); The average concentrations were 312.3 ± 2.2 nL/L ($n=42$) and 311.9 ± 2.7 nL/L ($n=42$), respectively, on the southbound track. The difference of N_2O concentrations was also insignificant by t test ($t=0.24 < t_{0.1, 26} = 1.71$), indicating that the daily variation of atmospheric N_2O concentrations may be very small above northern Pacific and Arctic Ocean surface. However, N_2O concentrations on the southbound track were significantly higher than those on the northbound track at 8:00 A.M or 20:00 P.M, suggesting that the seasonal variations may be large in the study areas.

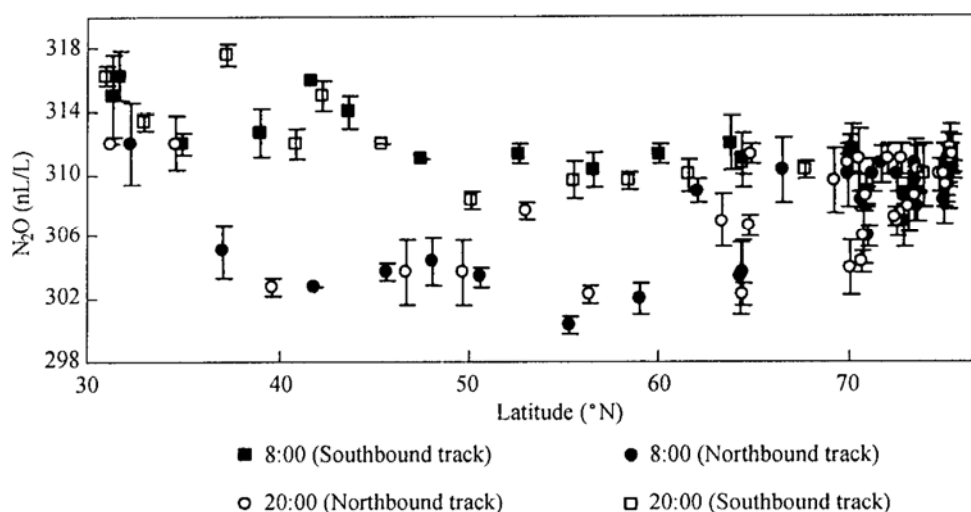


Fig. 3 The comparisons of average N_2O concentrations above oceanic surface at 8:00 AM and 20:00 PM.

3.3 Influential factors of N_2O concentrations above oceanic surface

3.3.1 Source analyses and assessment for atmospheric N_2O

The concentrations for $nss-SO_4^{2-}$, SO_4^{2-} , MSA, Cl^- , Na^+ in atmospheric aerosol were also observed. The correlations were analyzed between average N_2O concentrations and the content of these aerosol compositions in the ranges of corresponding latitudes. The weak correlations were observed between N_2O concentrations and the content of Na^+ , Cl^- (Fig. 4a and Fig. 4b), indicating that atmospheric N_2O had different sources. However, the negative correlation was significant between N_2O and MSA (Fig. 4c). Study showed that biogenic sulfur in atmospheric aerosol above oceanic surface was mainly from $nss-SO_4^{2-}$ and MSA, which were produced by the processes of biological oxidation of marine Dimethyl sulfide (DMS). DMS production in oceans has been seen to covary with biological activity. N_2O production in oceans is also related to biological activity, particularly microbial metabolism (Elkins and Wofsy 1978; Collins *et al.* 1996). However, N_2O can accumulate in the marine boundary layer for days, while DMS has a lifetime of less than a day

in the marine boundary layer, which means that DMS is not a perfect tracer for determining the N_2O source from the oceans. MSA production by DMS is related with the productivity for marine organisms and their activities and MSA is more stable in the marine boundary layer (Gao *et al.* 1996). Therefore MSA is a more rational tracer for determining the N_2O source for the oceans. The negative correlation indicates that atmospheric MSA and N_2O have the different sources and N_2O is an artifact of the continental sources. The positive correlation between N_2O and $nss-SO_4^{2-}/MSA$, SO_4^{2-} , $nss-SO_4^{2-}$ are also obtained, indicating that the emissions from the continental sources have a great contribution to atmospheric N_2O above oceanic surface. Anthropogenic sources from Eur-Asian continent may have an important effect on N_2O concentration above oceanic surface. The concentrations significantly increased while the expeditionary ship approached Shanghai harbor on the cruise. N_2O concentrations in the range of 30-35°N were higher than those in the other areas above oceanic surface (Fig. 2), 3-5 nL/L higher than the areas approaching Arctic ices (Table 1). In addition, the concentrations in Shanghai harbor were also significantly higher than those at Arctic ice stations and floating ices (Table 2). The average N_2O concentration for three different sites in Shanghai harbor was 315.1 ± 2.5 nL/L. The averages for ice stations and floating ice were 307.2 ± 1.4 nL/L and 306.2 ± 0.7 nL/L, respectively, 7.9 nL/L and 8.9 nL/L lower than that in Shanghai harbor, further suggesting that continental sources may have a significant contribution to the global atmospheric N_2O .

Table 1. The comparisons of atmospheric mean N_2O concentrations in the ranges of different latitudes for the cruise and Antarctic Fildes Peninsula.

Latitudinal range	Sampling areas	Mean N_2O concentration for Northbound track (nL/L)	Mean N_2O concentration for southbound track (nL/L)	The cruise (nL/L)
31 ~ 35°N	East China Sea	312.0 ± 1.9	314.6 ± 2.4	314.3 ± 2.6
35 ~ 45°N	Japanese Ssea	303.6 ± 1.9	313.3 ± 1.8	309.2 ± 5.2
45 ~ 55°N		304.4 ± 1.9	310.5 ± 1.5	306.5 ± 3.6
55 ~ 65°N	Bering Sea	304.1 ± 3.3	310.6 ± 1.3	307.2 ± 4.0
65 ~ 70°N	Chukchi Sea	307.9 ± 1.5	309.7 ± 1.2	308.1 ± 2.9
	Canadian oceanic basin			309.9 ± 1.6
70 ~ 77°N	Areas approaching Arctic ices	309.3 ± 2.3	309.8 ± 1.3	307.2 ± 1.4
	Ice stations and floating ices			307.2 ± 1.7
31 ~ 77°N	The cruise	308.0 ± 3.5	311.9 ± 2.5	309.0 ± 3.9
62°12'S	Antarctic Fildes Peninsula		310.7 ± 4.4	
62°12'S	Antarctic Nelson Icecap		308.0 ± 0	

Note: The data for atmospheric N_2O concentrations in Antarctica are from Sun *et al.* (2001).

Table 2. Comparisons of atmospheric N_2O concentrations at Shanghai harbor, ice stations and floating ices.

Sampling area	Latitude	Longitude	Average N_2O concentration (nL/L)
Shanghai harbor 1	31°14.4'N	123°14.48'E	312.0 ± 0
Shanghai harbor 2	30°59.45'N	122°26.38'E	316.0 ± 0.6
Shanghai harbor 3	31°23.38'N	121°36.1'E	317.3 ± 0.6
Ice station 1	73°6.05'N	165°22.48'W	306.3 ± 0.6
Ice station 2	73°24.93'N	164°54.05'W	308.7 ± 1.2
Ice station 3	73°27'N	165°1.8'W	306.7 ± 1.2
Floating ice 1	73°25'N	158°57'W	306.3 ± 1.2
Floating ice 2	73°28.13'N	158°56.97'W	305.7 ± 0.6
Floating ice 3	72°28'N	155°30.03'W	306.7 ± 0.6
Floating ice 4	72°26.20'N	153°29.42'W	306.0 ± 0.6

Note: Sampling numble n = 3

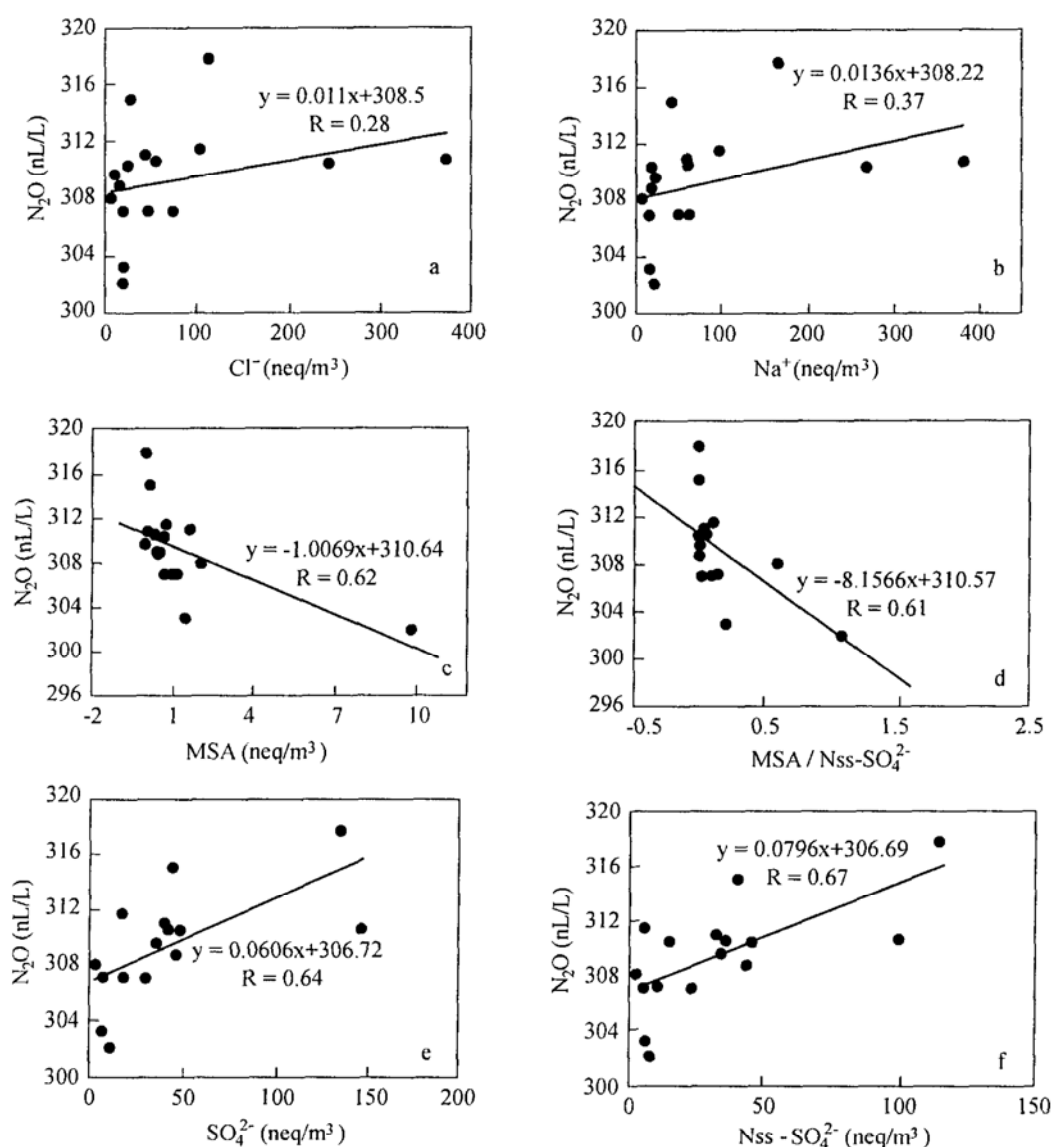


Fig. 4. The identification of atmospheric N_2O sources above the oceanic surface. Note: The data for N_2O concentrations are the average values corresponding to the same latitudinal range of the aerosol compositions.

3.3.2 The relationship between the distribution of N_2O concentrations and air pressure, air temperature above oceanic surface

The distribution of atmospheric N_2O concentration may be related with air pressure above the oceanic surface. Air pressure was lower in the range from 30-35°N, but the N_2O concentrations were the highest, which was mainly related with anthropogenic emissions because the sampling sites approached the continent. However, N_2O concentrations and air pressure had a good corresponding relationship in the range from 35-70°N where their changing trends were generally consistent, only a few values existing in advancing or lagging response while their changes were relatively smaller in the range from 70-77°N (Fig. 5a and

Fig. 5b). Therefore air pressure may be one of the main factors affecting atmospheric N_2O concentrations above mid-and high-latitude oceanic surface. The movement of atmospheric circumfluence is controlled by polar high pressure and subpolar low pressure in the Arctic areas approaching the sea ice. The areas from $65^\circ N$ to $77^\circ N$ were located in the confluent zone of prevailing western wind and polar eastern wind and N_2O strong exchange in marine boundary layer may occur, which was responsible for the increased N_2O concentrations.

The solubility for N_2O increases in the seawater under the conditions of low air temperature, which prevents N_2O in deeper water from diffusing and transferring to the surface water, therefore its exchanging amount decreases at the interface between the ocean and atmosphere (Butler *et al.* 1989). Therefore atmospheric N_2O concentration is indirectly affected by air temperature. Higher N_2O concentration may be related with the emissions from oceanic sources as a result of high air temperature in the range of $30-35^\circ N$ besides the effects of continental sources. The variations of N_2O concentration and air temperature above the oceanic surface were generally simultaneous in the range of $40-60^\circ N$, especially for the southbound track (Fig. 5c and Fig. 5d). The changes of air temperatures above the oceanic surface were consistent on the northbound and southbound tracks, suggesting the effects of ocean on the air temperatures. However N_2O concentrations were significantly different, indicating that they were influenced by other environmental factors.

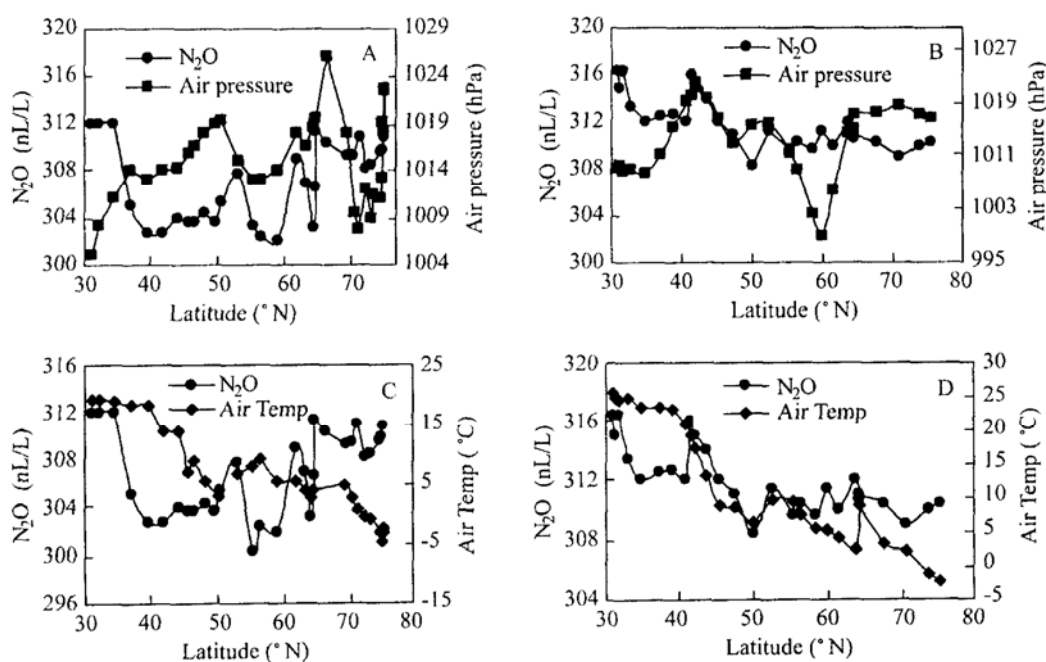


Fig. 5 The changing relationships between atmospheric average N_2O concentration, air pressure and temperature above the oceanic surface with the latitude. Fig A and C indicate the northbound track and Fig B and D southbound track.

4 Conclusions

On the cruise of "Xuelong" expeditionary ship, atmospheric average N_2O concentra-

tion was 309.0 ± 3.9 nL/L above the surface from mid- and high-latitude Northern Pacific Ocean to Arctic ocean. Atmospheric average N_2O concentrations in Arctic ocean, at ice stations and floating ices were 309.3 ± 2.3 nL/L, 307.2 ± 1.4 nL/L and 306.2 ± 0.7 nL/L, respectively. N_2O concentrations were significantly different in the same latitudinal area of Northern Pacific Ocean on the northbound and southbound. In addition, the distribution of atmospheric N_2O concentration was related with the effects of continental sources, air temperature and pressure.

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