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Monitoring atmospheric nitrous oxide background concentrations at Zhongshan Station, east Antarctica

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ARTICLE INFO

Article history:

Received 2 September 2015

Revised 19 December 2015

Accepted 21 December 2015

Available online 16 April 2016

Keywords:

Antarctica

Zhongshan station

N₂O background concentration

N₂O characteristics

Greenhouse Gases

ABSTRACT

At present, continuous observation data for atmospheric nitrous oxide (N₂O) concentrations are still lacking, especially in east Antarctica. In this paper, nitrous oxide background concentrations were measured at Zhongshan Station (69°22'25"S, 76°22'14"E), east Antarctica during the period of 2008–2012, and their interannual and seasonal characteristics were analyzed and discussed. The mean N₂O concentration was 321.9 nL/L with the range of 320.5–324.8 nL/L during the five years, and it has been increasing at a rate of 0.29% year⁻¹. Atmospheric N₂O concentrations showed a strong seasonal fluctuation during these five years. The concentrations appeared to follow a downtrend from spring to autumn, and then increased in winter. Generally the highest concentrations occurred in spring. This trend was very similar to that observed at other global observation sites. The overall N₂O concentration at the selected global sites showed an increasing annual trend, and the mean N₂O concentration in the Northern Hemisphere was slightly higher than that in the Southern Hemisphere. Our result could be representative of atmospheric N₂O background levels at the global scale. This study provided valuable data for atmospheric N₂O concentrations in east Antarctica, which is important to study on the relationships between N₂O emissions and climate change.

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Introduction

Nitrous oxide (N₂O) is one of the important greenhouse gases, with a long atmospheric lifetime of about 120 years, and its greenhouse effect is estimated to be 298 times greater than that of carbon dioxide (CO₂) on a molecular basis over a 100-year-time horizon (Artaxo et al., 2007). Although N₂O is one of the trace gases in the atmosphere, it plays an important role in global climate warming, and also takes part in the destruction of stratospheric ozone (Mosier, 1998). At present, the mean N₂O concentration in the global atmosphere is about 324 nL/L, 20% higher than that during the pre-industrial era (IPCC, 2013). The increase in N₂O concentrations and its

contributions to global warming have been becoming a serious concern. Anthropogenic sources, including agricultural soils with nitrogen fertilizer addition, combustion of fossil fuels and biomass, human waste etc., are regarded as the main sources for atmospheric N₂O (Xing and Zhu, 1997; Huttunen et al., 2003). The emissions from these sources have led to a continuous increase in atmospheric N₂O concentrations (Skiba et al., 1994; Regina et al., 2004; Takakai et al., 2006; Jungkunst and Fiedler, 2007; Thompson et al., 2014). Therefore, in order to evaluate its sources and sinks, it is very important to conduct long-term measurements of atmospheric N₂O background concentrations.

In the past decades, observation stations for measuring the concentrations of atmospheric trace gases including N₂O have

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been established at many sites on the globe. Some observation stations have also been set up in the Arctic and Antarctic regions. The long-term observation of atmospheric N_2O concentrations at Barrow (71.3°N , 156.6°W) in the Arctic showed that the mean annual N_2O growth rate was 0.22% during the period 1998 to 2014. The overall atmospheric mean N_2O concentration in the Northern Hemisphere was 0.7 ± 0.04 nL/L higher than that in the Southern Hemisphere, and the difference in N_2O concentrations between the Arctic and Antarctic regions was about 1.2 ± 0.1 nL/L (Khalil et al., 2002). Antarctica, which is remote from anthropogenic disturbance, is one of the key regions for the study of global climate change, for example in several international projects such as WCRP (World Climate Research Program) and IGBP (International Geosphere Biosphere Program). In addition, Antarctica has a harsh climate and a fragile ecological environment, and it is very sensitive to global climate change. The concentrations of atmospheric greenhouse gases in Antarctica might reflect their background levels at global scale (Priscu, 1997; Sun et al., 2000, 2001; Zhu et al., 2003a, 2003b, 2005, 2008a; Zhan and Chen, 2007). However, continuous observation data are still lacking for atmospheric N_2O background concentrations, especially in east Antarctica.

During February 2008 to January 2013, atmospheric N_2O background concentrations were observed at Chinese Zhongshan Station, east Antarctica. In this paper, the interannual and seasonal variations of atmospheric N_2O concentrations at Zhongshan Station were analyzed, and their influencing factors were discussed. We also compared the data for N_2O concentrations at Zhongshan Station with those at observation stations in other global areas.

1. Materials and methods

1.1. Study area

The observation site for atmospheric N_2O concentrations was set up at Zhongshan Station ($69^\circ22'25''\text{S}$, $76^\circ22'14''\text{E}$), which is located on the Millor Peninsula, east Antarctica (Fig. 1). Millor Peninsula is one of several ice-free areas along the coast of Antarctica in the summer, with a frigid and dry climate. The vegetation around Zhongshan Station is very sparse as a

result of the severe climatic conditions and exposure of the bedrock; the biological and chemical processes are very weak, and no true soil develops in this area (Zhu et al., 2008b; Bian et al., 2014). The atmospheric N_2O observation site ($69^\circ22'12''\text{S}$, $76^\circ21'49''\text{E}$, 18.5 m) was established on an outcropping rock, about 500 m to the northwest of Zhongshan Station.

1.2. Gas sampling and N_2O concentration determination

The air samples were collected using standard gas sampling flasks from 27 February, 2008 to 25 January, 2013. The flasks had been evacuated to -1.0×10^5 Pa in advance in the Atmospheric Chemistry Laboratory, China Meteorological Administration. The gas samples were randomly collected in the vacuum flasks. Two duplicate flasks of gas samples were collected each time, and their average concentration was used as the final result at the observation site. The sampling frequency was once a week. The N_2O concentrations were analyzed using an Agilent 6890 N Gas Chromatograph equipped with Electron Capture Detection (GC-ECD). Generally the N_2O in the gas samples was separated by the GC-ECD system with double columns for analysis and single column back-flushing. The temperatures for the detector and column were 395 and 75°C , respectively. A pre-column and a main-column (2 m, 80–100 mesh, Haye Sep. Q) were used with a methane-argon (5% : 95%) mixture as the carrier gas at a flow rate of 90 mL/min. The analysis time was about 10 min. The standard gas for the test was obtained from the World Meteorological Organization-Global Atmospheric Observation Network (WMO/GAW). The instrumental accuracy for the N_2O concentration measurement is better than $0.03\% \pm 0.09\%$. Details of the method can be found in the paper reported by Fang et al. (2011). Abnormal values were removed on the basis of the formula $|x_i - \bar{x}| > 3\sigma$, where x_i indicates the measured concentration, \bar{x} indicates the mean concentration and σ is the standard deviation of the monthly means. About 2% of the mean data were rejected based upon the above criterion.

1.3. Environmental parameters and other data sources

The data for environmental parameters, including air temperature, air humidity, wind speed and wind direction, were obtained from the Meteorological Observation Station at

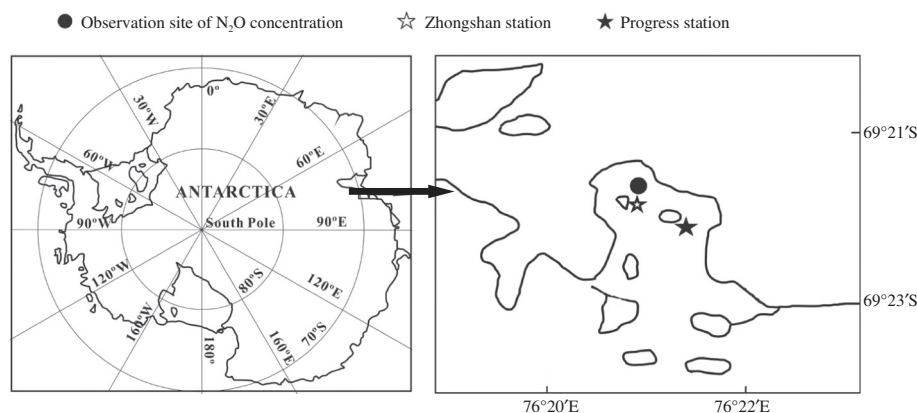


Fig. 1 – The observation site of atmospheric N_2O concentrations at Zhongshan Station, east Antarctica.

Zhongshan Station during the period of the observation. We compared our data with atmospheric N_2O concentrations from the World Data Centre for Greenhouse Gases (WDCGG) to further verify the reliability of the data. Typical observation sites with different latitudes were selected from the Northern Hemisphere to the Southern Hemisphere, including the observation sites Alert, Churchill, Jungfraujoch, Niwot Ridge, Ryori, Mauna Loa, Tutuila, cape Grim, Baring Head, Casey Station, Mawson, Arrival Heights and South Pole, and the information about these observation sites is summarized in Table 2 and Fig. 6. The N_2O concentration data at different observation sites were downloaded from the World Data Centre for Greenhouse Gases (<http://ds.data.jma.go.jp/gmd/wdcgg/cgi-bin/wdcgg/catalogue.cgi>) for comparison with the concentrations at Zhongshan Station.

1.4. Data analysis

All statistical analyses were performed with SPSS statistics 17.0 and Microsoft Excel for Windows 2007. Mean N_2O concentration, wind direction frequency, and mean annual and monthly N_2O concentrations at other sites around the world were analyzed using Microsoft Excel 2007. SPSS statistics 17.0 was used to draw statistical box figures for seasonal variations of N_2O concentrations. In all analyses where $p < 0.05$, the relationships were considered statistically significant. The map for global N_2O monitoring sites was processed by Photoshop CS6.

2. Results and discussion

2.1. Effects of wind on atmospheric N_2O concentrations

To test the potential influences of human activities from Zhongshan Station on background N_2O concentrations, wind direction frequency, mean wind speed and N_2O concentrations are shown in the rose figures (Fig. 2). The prevailing wind directions were persistently easterly and north-easterly with a prevalence frequency of 81.4%. Our observation site for N_2O concentrations was located upwind in the dominant wind direction to minimize the possible effects of human activities from the scientific stations. The N_2O concentrations in the north winds had the highest average, with a mean of 324.7 nL/L, followed by west winds (323.8 nL/L), southwest winds (323.8 nL/L), and north–north–east wind (323.5 nL/L), respectively, whereas the concentration under south–south–east wind was the lowest (320.3 nL/L). The concentrations in other wind directions were within the range of 322.3–322.9 nL/L. However, the wind frequency for the north, southwest, west and north–north–east only accounted for about 3.6%. Although possible influences of human activities from the scientific station might exist in these wind directions, they had no significant effects on atmospheric N_2O background concentrations. To further confirm the effects of wind directions on the N_2O concentration in spring, summer, autumn and winter, the frequency of wind direction and N_2O concentration in each season are also shown in Fig. 2b. Predominant wind directions were the east wind and northeast wind in spring and summer, whereas the east wind predominated in autumn and winter.

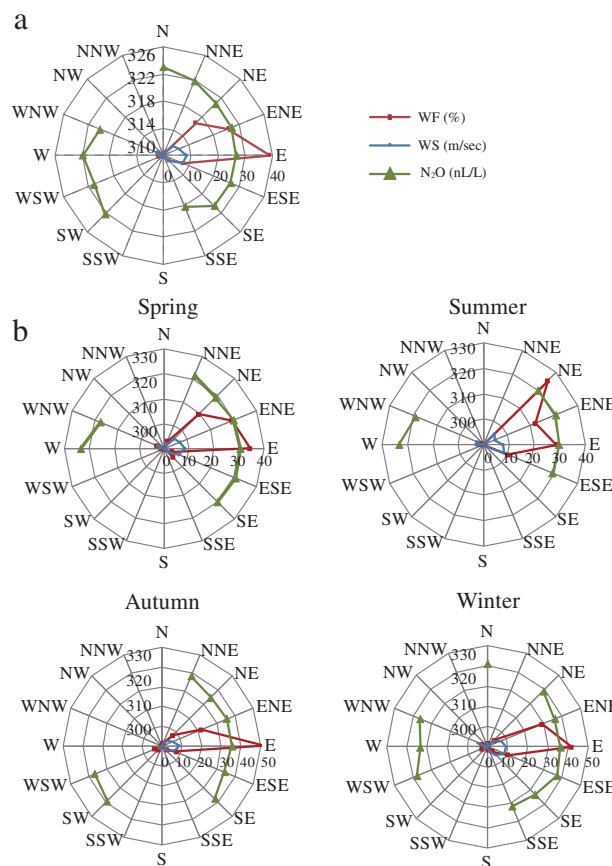


Fig. 2 – Rose figures representing mean N_2O concentration, wind direction frequency (WF), and mean wind speed (WS) at the observation site. (a) WF, WS, and N_2O concentrations under 16 wind directions during the period from 2008 to 2013; (b) WF, WS, and N_2O concentrations in four seasons under 16 wind directions.

N_2O concentration showed a slight increase in west wind conditions with a low frequency of 5%, and it could be increased by 5 nL/L, especially in spring and autumn. This might be associated with the seasonal changes of continental and marine air masses. The true reasons for these differences need to be further studied and discussed in the future.

In addition, weak winds and unstable wind speed are difficult conditions for the diffusion and mixing of the local pollutants. All the wind speed data from 2008 to 2013 were categorized into seven ranks (i.e., ≤ 1.5 m/sec, 1.5–3 m/sec, 3–6 m/sec, 6–10 m/sec, 10–15 m/sec, 15–20 m/sec and > 20 m/sec) from weak to strong winds. The mean N_2O concentrations under different wind speeds are shown in Fig. 3a. The mean N_2O concentrations under wind speeds of less than 1.5 and 15–20 m/sec were 1.8 and 1.5 nL/L higher than that under the wind speed of 1.5–15 m/sec, respectively. Little data for N_2O concentrations could be obtained for the wind speeds of less than 1.5 and 15–20 m/sec due to low frequency, and the reason for the higher N_2O concentrations observed needs to be further studied under these wind speeds. Overall the frequency for the wind speed of 1.5–15 m/sec was 96.4%,

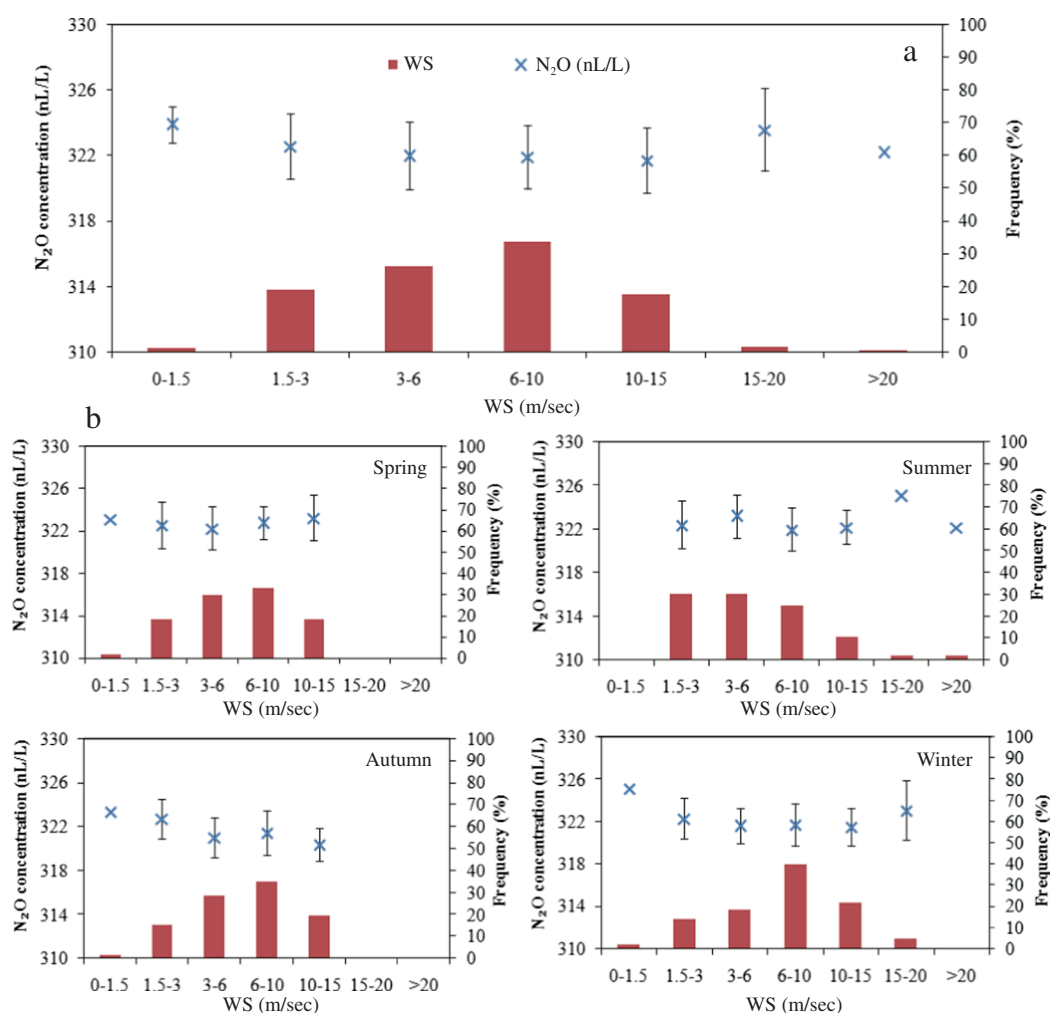


Fig. 3 – Relationships between atmospheric N₂O concentration and the frequency of different wind speeds at the observation site of Zhongshan Station. (a) N₂O concentrations and the frequency of different wind speeds during the period from 2008 to 2013; (b) N₂O concentrations and the frequency of different wind speeds in four seasons.

whereas low wind speed (≤ 1.5 m/sec) and strong wind speed (> 15 m/sec) had only 3.6% frequency. Therefore the effects of weak winds and the instability of wind speeds on annual N₂O concentrations could be ignored in our study area.

Fig. 3b displays the mean N₂O concentrations under the frequency of different wind speeds in four seasons. The N₂O concentrations for wind speed of ≤ 1.5 m/sec were about 1 nL/L higher than those under other ranks of wind speeds in autumn and winter, and the N₂O concentrations for wind speed of 15–20 m/sec were 2.5 nL/L higher than those under other wind speeds in summer, probably due to the anthropogenic pollution from Zhongshan Station. However, the N₂O concentrations under almost all the ranks of wind speeds showed consistent levels in spring. Although weak winds and unstable wind speeds are not sufficient to diffuse or mix the local pollutants, the weak and strong winds had very weak effects on the seasonal N₂O concentrations due to their extremely low frequency ($< 4\%$).

On the whole, the wind direction and wind speed had no statistically significant effects on atmospheric N₂O concentrations at our observation site during the four seasons. Thus the effects of disturbance by human activities from the scientific research station can be neglected.

2.2. Interannual variation of N₂O concentrations

Atmospheric N₂O concentrations showed an increasing trend from February 2008 to January 2013 at Zhongshan Station, east Antarctica (Fig. 4). Annual average N₂O concentrations increased from 320.5 ± 0.5 nL/L in 2008 to 324.8 ± 0.7 nL/L in 2012. The average growth rate was about 0.29% within linear fitting during these five years. The annual growth rates were 0.35%, 0.30%, 0.21% and 0.47% in 2008–2009, 2009–2010, 2010–2011 and 2011–2012, respectively (Table 1). In our study area, disturbance by human activities can be neglected because the observation site is located in the upwind direction from the scientific research station. Our observation data could reflect

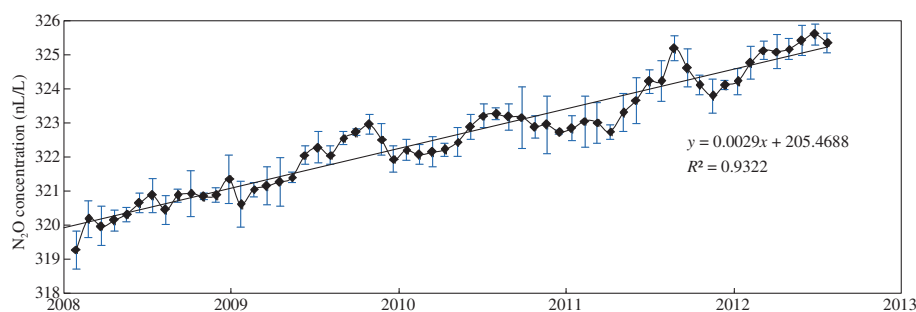


Fig. 4 – Changes of atmospheric N₂O concentrations at Zhongshan Station during the period from February 2008 to January 2013.

the increase in atmospheric N₂O background concentrations at the global scale, which might be related to the increase in anthropogenic N₂O emissions, including the increased use of nitrogen fertilizer in agricultural systems, industrial development and the increase in burning of fossil fuels (Denman et al., 2007; Syakila and Kroeze, 2011; Bouwman et al., 2013a, 2013b).

2.3. Seasonal variation of N₂O concentrations

As shown in Fig. 5, atmospheric N₂O concentrations showed a strong seasonal fluctuation during these five years. The concentrations appeared to follow a downtrend from spring to autumn, and then increased in winter. Generally the highest concentrations occurred in spring. This trend was mostly consistent with that at the observation site of Mawson Station, and a similar result also appeared at the site of Casey Station as reported by Allen et al. (2007). In our study area, predominant wind directions were from the east and north-east in spring, summer, autumn and winter, indicating that the wind directions had negligible effects on atmospheric N₂O concentrations during these four seasons.

Our observation site is very close to the Southern Ocean, and the seasonal variation of atmospheric N₂O concentrations might be effected by ocean currents and seasonal melting sea ice (Zhu et al., 2003a; Zhan and Chen, 2007). In addition, stratospheric air contained relatively low N₂O concentrations compared to tropospheric air. The low N₂O air transport from

the stratosphere to the troposphere might have a great influence on N₂O concentrations due to the seasonal atmospheric circulation (Thompson et al., 2014). Holton (1990) indicated that the downward flux from the stratospheric atmosphere would dilute the atmospheric N₂O concentration in the troposphere. Generally the transport from the stratosphere to the troposphere occurs mainly in winter, with the weakest in August to October in the Northern Hemisphere, whereas in the Southern Hemisphere the air transport from the stratosphere to the troposphere is stronger in autumn than in spring and winter (Shu et al., 2009). This phenomenon could be used to explain our result that observed N₂O concentrations in spring and winter were slightly higher than those in summer and autumn at the observation site.

On the other hand, N₂O is mainly produced by nitrification and denitrification in soil systems under the effects of microorganisms. The measurements of N₂O fluxes from tundra ecosystems in maritime Antarctica and in the Arctic have proved that N₂O emissions had a close correlation with soil temperature, and high N₂O emissions generally occurred during the cycles of soil freezing and thawing (Teepe et al., 2001; Schürmann et al., 2002; Huttunen et al., 2003; Zhu et al., 2009, 2013). Slightly enhanced N₂O concentrations in spring might be related to soil freezing and thawing cycles in the global ecosystems. On the whole the seasonal change of N₂O concentration is very complicated and affected by atmospheric circulation, climate change and soil environment change, etc.

Table 1 – Average N₂O concentrations (nL/L) at the observation site of Zhongshan Station, east Antarctica in the four seasons during the period from 2008 to 2012.

Year	Spring	Summer	Autumn	Winter
2008	320.8 ± 0.4	320.4 ± 0.0	320.1 ± 0.5	320.6 ± 0.3
2009	322.3 ± 0.4	321.0 ± 0.4	321.0 ± 0.6	321.6 ± 0.6
2010	323.1 ± 0.4	322.7 ± 0.3	322.1 ± 0.4	322.3 ± 0.5
2011	323.7 ± 0.6	323.1 ± 0.5	322.9 ± 0.5	323.0 ± 0.5
2012	325.3 ± 0.4	324.6 ± 0.6	324.0 ± 0.5	324.8 ± 0.5
2008–2009	0.46%	0.18%	0.26%	0.30%
2009–2010	0.26%	0.55%	0.35%	0.22%
2010–2011	0.18%	0.10%	0.24%	0.21%
2011–2012	0.48%	0.47%	0.36%	0.55%
Mean growth rate	0.34%	0.33%	0.31%	0.32%

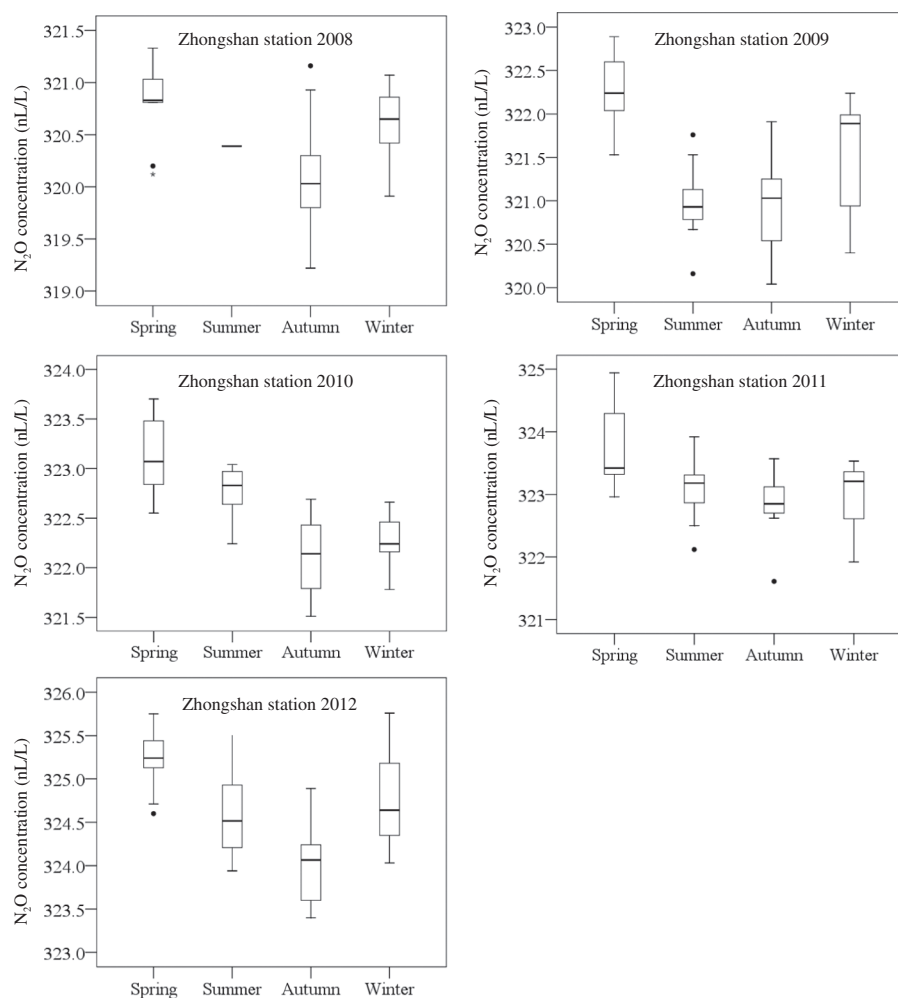


Fig. 5 – Seasonal variations of atmospheric N₂O concentrations at the observation site of Zhongshan Station.

As listed in Table 1, the N₂O concentrations in all the seasons showed an increasing annual trend from 2008 to 2012, with the highest growth rate of 0.34% in spring and the lowest of 0.31% in autumn. The growth rates were 0.33% and 0.32% in summer and winter, respectively. The seasonal N₂O growth rate reached the maximum at the observation site in 2012.

2.4. Comparisons with other observation sites

In order to further justify the reliability of N₂O concentrations, the data at our observation site were compared with those at other global sites from the World Data Centre for Greenhouse Gases (WDCGG). Fig. 6 shows the changes of N₂O concentrations at main global observation sites during the period from 2008 to 2012. The N₂O concentrations at Zhongshan Station (322.6 ± 0.7 nL/L) were quite close to the levels in the Southern Hemisphere (322.7 ± 0.7 nL/L) (Table 2), furthermore confirming that N₂O concentrations at our observation site are reliable, and can be considered representative of the background characteristics of global atmospheric N₂O levels. The overall atmospheric N₂O concentration in the Northern

Hemisphere (323.7 ± 0.2 nL/L) was about 1 nL/L higher than that in the Southern Hemisphere (322.7 ± 0.3 nL/L) from 2008 to 2012. The interannual growth rate of N₂O concentrations during these five years was 0.28% and 0.24% in the Northern Hemisphere and in the Southern Hemisphere, respectively. For example, the N₂O concentration in the Northern Hemisphere (325.5 ± 0.5 nL/L) was slightly higher than that in the Southern Hemisphere (324.1 ± 0.6 nL/L) in 2012. More N₂O emission sources, especially human sources, are concentrated in the Northern Hemisphere. Therefore the difference in the atmospheric N₂O background concentration between the North and South Hemispheres might be related to the emission sources from human activities, including farming and the burning of fossil fuels.

3. Summary

The atmospheric N₂O concentration at the observation site of Zhongshan Station continuously increased at a growth rate of about 0.29% from February 2008 to January 2013, and the annual average N₂O concentration reached 324.8 ± 0.7 nL/L in

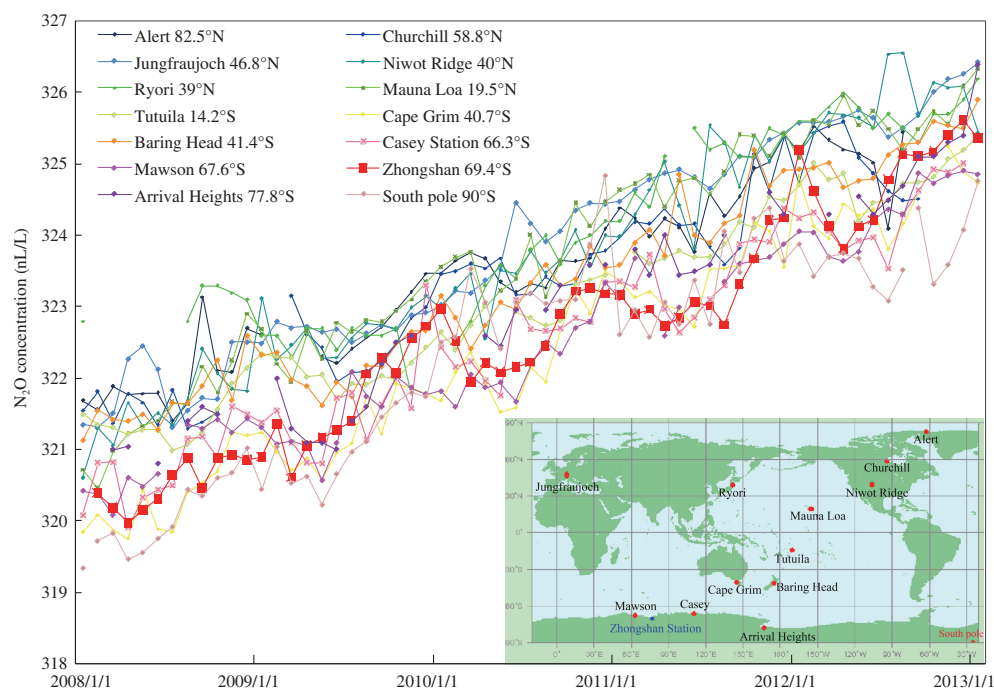


Fig. 6 – Comparisons between atmospheric N_2O concentrations at the observation site of Zhongshan Station and other global observation sites.

2012. This trend might be associated with the increase in the use of nitrogen fertilizer in agricultural systems, industrial development, the burning of fossil fuels, changes in land use and climate changes. The N_2O concentrations had an obvious seasonal fluctuation, and the concentrations in spring and winter were higher than those in summer and autumn. Overall the N_2O concentrations at our observation site were quite close to the levels in the Southern Hemisphere, but lower than those in the Northern Hemisphere. Therefore our result could be considered representative of the atmospheric N_2O background level at the global scale. This study provided

valuable data on atmospheric N_2O concentrations in east Antarctica, which is important to study on the relationships between N_2O emissions and climate change.

Acknowledgments

This work was supported by the Program of China Polar Environment Investigation and Assessment (No. CHINARE 2011–2015), and the National Natural Science Foundation of China (Nos. 41176171, 41576181). The authors appreciate the

Table 2 – Annual average N_2O concentration and its annual growth rate at the typical observation sites.

Station	Latitude and longitude	N ₂ O mean concentration (nL/L)					Mean	Growth rate (%)			
		2008	2009	2010	2011	2012		2008–2009	2009–2010	2010–2011	2011–2012
Alert	82.5°N 62.5°W	321.8	322.6	323.5	324.3	325.1	323.5	0.26	0.27	0.25	0.24
Churchill	56.6°N 94.1°W	321.5	322.4	323.5	324.2	325.0	323.3	0.28	0.32	0.24	0.24
Jungfraujoch	46.5°N 8.0°E	321.9	322.7	323.9	323.9	325.7	323.8	0.26	0.36	0.33	0.25
Niwot Ridge	40.0°N 105.5°W	321.6	322.7	323.5	324.8	325.9	323.7	0.35	0.25	0.39	0.35
Ryori	39.0°N 141.8°W	323.1	322.6	323.4	325.0	325.7	324.0	−0.15	0.25	0.50	0.20
Maunna Loa	19.5°N 155.6°W	321.5	322.7	323.7	324.9	325.6	323.7	0.36	0.31	0.38	0.21
NH mean		321.9	322.6	323.6	324.7	325.5	323.7	0.23	0.29	0.35	0.25
Tutuila	14.2°S 170.6°W	321.4	322.1	322.9	324.0	324.8	323.0	0.22	0.26	0.31	0.25
Cape Grim	40.7°S 144.7°E	320.4	321.3	322.3	323.4	324.4	323.3	0.29	0.31	0.36	0.30
Baring Head	41.4°S 174.9°E	321.6	322.1	323.2	324.1	325.1	323.2	0.17	0.32	0.28	0.30
Casey	66.3°S 110.5°E	320.7	322.8	322.5	323.3	324.3	322.7	0.66	−0.11	0.26	0.30
Mawson	67.6°S 62.9°E	321.5	322.2	323.6	324.3	320.9	322.5	0.24	0.41	0.24	−1.05
Zhongshan	69.4°S 76.4°E	320.5	321.6	322.6	323.3	324.8	322.6	0.35	0.30	0.21	0.47
Arrival Heights	77.8°S 166.7°E	321.2	321.7	323.0	323.8	324.8	322.9	0.14	0.41	0.27	0.29
South pole	90.0°S 24.8°E	320.1	321.1	323.1	323.4	323.6	322.3	0.29	0.63	0.09	0.07
SH mean		320.9	321.9	322.9	323.7	324.1	322.7	0.29	0.32	0.25	0.12

assistance of all the staff who lived through the winter at Zhongshan Station during the data collection. We also thank the Polar Office of the National Ocean Bureau of China for the support and assistance.

REFERENCES

- Allen, D.E., Dalal, R.C., Rennenberg, H., Meyer, R.L., Reeves, S., Schmidt, S., 2007. Spatial and temporal variation of nitrous oxide and methane flux between subtropical mangrove sediments and the atmosphere. *Soil Biol. Biochem.* 39 (2), 622–631.
- Artaxo, P., Bernsten, T., Betts, R., Fahey, D.W., Haywood, J., Lean, J., et al., 2007. Changes in atmospheric constituents and in radiative forcing. *Climate change 2007: The physical science basis*. Cambridge University Press, Cambridge, UK and New York, NY, USA.
- Bian, L.G., Tang, J., Lai, X., Wang, Y.T., Liu, L.X., Zhou, L.X., 2014. Monitoring carbon monoxide during 2008–2010 at Zhongshan station, Antarctica. *Acta Sci. Circumst.* 34 (2), 310–317.
- Bouwman, A.F., Beusen, A.H.W., Griffioen, J., Van Groenigen, J.W., Hefting, M.M., Oenema, O., et al., 2013a. Global trends and uncertainties in terrestrial denitrification and N₂O emissions. *Philos. Trans. R. Soc. B* 368 (1621), 20130112. <http://dx.doi.org/10.1098/rstb.2013.0112>.
- Bouwman, L., Goldewijk, K.K., Van Der Hoek, K.W., Beusen, A.H.W., Van Vuuren, D.P., Willems, J., et al., 2013b. Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period. *Proc. Natl. Acad. Sci. U. S. A.* 110 (52), 20882–20887.
- Denman, K.L., Brasseur, G., Chidthaisong, A., Ciais, P., Cox, P.M., Dickinson, R.E., et al., 2007. Couplings between changes in the climate system and biogeochemistry. *Climate change 2007: The physical science basis*. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 499–587.
- Fang, S.X., Zhou, L.X., Wang, W., Zhang, F., Yao, B., Xu, L., et al., 2011. Single standard calibration method by GC-ECD system on the background N₂O measurement. *Environ. Chem.* 30 (5), 1030–1033.
- Holton, J.R., 1990. On the global exchange of mass between the stratosphere and troposphere. *J. Atmos. Sci.* 47 (3), 392–395.
- Huttunen, J.T., Juutinen, S., Alm, J., Larmola, T., Hammar, T., Silvola, J., et al., 2003. Nitrous oxide flux to the atmosphere from the littoral zone of a boreal lake. *J. Geophys. Res.* 108 (D14), 4421.
- IPCC, 2013. *Climate change 2013: The physical science basis*. Cambridge University Press, New York, USA.
- Jungkunst, H.F., Fiedler, S., 2007. Latitudinal differentiated water table control of carbon dioxide, methane and nitrous oxide fluxes from hydromorphic soils: Feedbacks to climate change. *Glob. Chang. Biol.* 13 (12), 2668–2683.
- Khalil, M.A.K., Rasmussen, R.A., Shearer, M.J., 2002. Atmospheric nitrous oxide: Patterns of global change during recent decades and centuries. *Chemosphere* 47 (8), 807–821.
- Mosier, A.R., 1998. Soil processes and global change. *Biol. Fertil. Soils* 27 (3), 221–229.
- Priscu, J.C., 1997. The biogeochemistry of nitrous oxide in permanently ice-covered lakes of the McMurdo Dry Valleys, Antarctica. *Glob. Chang. Biol.* 3 (4), 301–315.
- Regina, K., Syvasalo, E., Hannukkala, A., Esala, M., 2004. Fluxes of N₂O from farmed peat soils in Finland. *Eur. J. Soil Sci.* 55 (3), 591–599.
- Schürmann, A., Mohn, J., Bachofen, R., 2002. N₂O emissions from snow-covered soils in the Swiss Alps. *Tellus B* 54 (2), 134–142.
- Shu, J.Z., Tian, W.S., Xie, F., 2009. Effects of atmospheric greenhouse gases and sea surface temperature changes on the transport process of the stratosphere and troposphere. *Proceedings of the 26th annual meteorological conference on atmospheric components, climates and environmental changes in China*, p. 238.
- Skiba, U., Fowler, D., Smith, K., 1994. Emissions of NO and N₂O from soils. *Environ. Monit. Assess.* 31 (1–2), 153–158.
- Sun, L.G., Xie, Z.Q., Zhao, J.L., Xing, G.X., Shi, S.L., Du, L.J., 2000. Monitoring the concentration of N₂O in the Fildes Peninsula, maritime Antarctica. *Chin. Sci. Bull.* 45 (21), 2000–2004.
- Sun, L.G., Zhu, R.B., Yin, X.B., Zhao, J.L., Xing, G.X., Shi, S.L., et al., 2001. Contrast between nitrous oxide concentrations in Antarctic Fildes Peninsula in 1999 and 2000 summers. *Chin. J. Polar Res.* 13 (2), 83–90.
- Syakila, A., Kroeze, C., 2011. The global nitrous oxide budget revisited. *Greenh. Gas Meas. Manag.* 1 (1), 17–26.
- Takakai, F., Morishita, T., Hashidoko, Y., Darung, U., Kuramochi, K., Dohong, S., et al., 2006. Effects of agricultural land-use change and forest fire on N₂O emission from tropical peatlands, Central Kalimantan, Indonesia. *Soil Sci. Plant Nutr.* 52 (5), 662–674.
- Teepe, R., Brumme, R., Beese, F., 2001. Nitrous oxide emissions from soil during freezing and thawing periods. *Soil Biol. Biochem.* 33 (9), 1269–1275.
- Thompson, R.L., Chevallier, F., Crotwell, A.M., Dutton, G., Langenfelds, R.L., Prinn, R.G., et al., 2014. Nitrous oxide emissions 1999 to 2009 from a global atmospheric inversion. *Atmos. Chem. Phys.* 14 (4), 1801–1817.
- Xing, G.X., Zhu, Z.L., 1997. Preliminary studies on N₂O emission fluxes from upland soils and paddy soils in China. *Nutr. Cycl. Agroecosyst.* 49 (1–3), 17–22.
- Zhan, L.Y., Chen, L.Q., 2007. Distributions of nitrous oxide mixing ratios in the atmosphere over the southern ocean. *Chin. J. Polar Res.* 19 (1), 49–60.
- Zhu, R.B., Sun, L.G., Liu, X.D., 2003a. Atmospheric nitrous oxide observations above the oceanic surface during CHINARE-18. *Prog. Nat. Sci.* 13 (8), 615–619.
- Zhu, R.B., Sun, L.G., Xie, Z.Q., Zhao, J.L., 2003b. Atmospheric nitrous oxide observations above the oceanic surface during the first Chinese Arctic Research Expedition. *Chin. J. Polym. Sci.* 14 (2), 138–148.
- Zhu, R.B., Liu, Y.S., Ma, E.D., Sun, J.J., Xu, H., Sun, L.G., 2009. Greenhouse gas emissions from penguin guanos and ornithogenic soils in coastal Antarctica: Effects of freezing–thawing cycles. *Atmos. Environ.* 43 (14), 2336–2347.
- Zhu, R.B., Liu, Y.S., Ma, J., Xu, H., Sun, L.G., 2008a. Nitrous oxide flux to the atmosphere from two coastal tundra wetlands in eastern Antarctica. *Atmos. Environ.* 42 (10), 2437–2447.
- Zhu, R.B., Liu, Y.S., Xu, H., Ma, D.W., Jiang, S., 2013. Marine animals significantly increase tundra N₂O and CH₄ emissions in maritime Antarctica. *J. Geophys. Res. Biogeosci.* 118 (4), 1773–1792. <http://dx.doi.org/10.1002/2013JG002398>.
- Zhu, R.B., Liu, Y.S., Xu, H., Ma, J., Sun, L.G., 2008b. Temporal and spatial variations of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ for atmospheric N₂O above the oceanic surface from Shanghai to Antarctica. *Sci. China Ser. D Earth Sci.* 51 (6), 899–910.
- Zhu, R.B., Sun, L.G., Ding, W.X., 2005. Nitrous oxide emissions from tundra soil and snowpack in the maritime Antarctic. *Chemosphere* 59 (11), 1667–1675.