

Short communication

Background-like nitrate in desert air

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HIGHLIGHTS

- Background-like nitrate existed in desert air.
- NO_x from desert soil and its conversion were likely responsible for the nitrate.
- Reactive nitrogen from deserts is not insignificant in the global nitrogen cycle.

ARTICLE INFO

Article history:

Received 9 July 2013

Received in revised form

8 November 2013

Accepted 17 November 2013

Keywords:

Nitrogen cycle

Reactive nitrogen

Soil emission

NO_x

Taklimakan desert

ABSTRACT

The atmospheric nitrogen cycle is a key process driving the earth's environmental evolution. Current model studies require knowledge of NO_x soil emissions from various land types, but desert emissions remain unquantified or are not addressed with high confidence. Our measurements at two observatories in Taklimakan desert during a dust episode showed an approximately stable and dust-independent nitrate in the air. Its concentration estimated from PM_{2.5}, PM₁₀ and TSP samples under non-dust, floating dust and dust storm conditions was $3.81 \pm 1.24 \mu\text{g m}^{-3}$, $2.95 \pm 0.69 \mu\text{g m}^{-3}$, $4.99 \pm 1.71 \mu\text{g m}^{-3}$, respectively, despite the more-than-one-order difference of dust loading. This concentration was much larger than that in remote marine and tropical forest air. Comprehensive investigation revealed a similar presence of nitrate in other desert air. The nitrate was hypothesized to be the consequence of the conversion of NO_x released from desert soils. These results indicate a background-like nitrate and active reactions of nitrogen compounds in desert air.

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1. Introduction

Reactive nitrogen compounds are a group of key chemical components in the atmosphere, governing atmospheric reactions in both the troposphere and the stratosphere (Crutzen, 1979) and acting as links in global biogeochemical cycles (Galloway et al., 2004). They are present in the atmosphere mainly in the form of nitrogen oxide (NO_x = NO + NO₂), as a consequence of both natural and anthropogenic processes (Logan, 1983). NO_x can be converted to other species via homogeneous and heterogeneous reactions in the atmosphere. Nitrate (including nitric acid) is one of the products of the reactions. Its dry and wet depositions to the surface are the major removal pathways of reactive nitrogen compounds from the air. On the other hand, nitrate is usually unstable under normal atmospheric conditions, can be converted back to NO_x through

photolysis or reactions with OH radicals and is actually one of the intermediate products in a series of reversible reactions involving NO_x. Its occurrence in the air is determined by the kinetic equilibrium of the reactions and closely dependent on the emission, transformation and deposition of the reactive nitrogen compounds (Seinfeld and Pandis, 2006).

High concentration of nitrate was observed in polluted urban atmosphere and attributed to large NO_x emission from anthropogenic activities associated with biomass burning and fossil fuel combustion (Blanchard and Tanenbaum, 2003; He et al., 2001). In the remote marine atmosphere, nitrate concentration was very small, for instance, $0.11 \pm 0.12 \mu\text{g m}^{-3}$ in the tropical south Pacific (Savoie et al., 1989). In continental areas, surface soil was recognized as one of the major natural sources of atmospheric NO_x (Galbally and Roy, 1978; Davidson and Kinglerlee, 1997).

Desert soil hosts the premier natural nitrate minerals on the earth (Ericksen, 1983; Walvoord et al., 2003). Nitrate can be reduced to NO_x through microbiological denitrification (Bowden, 1986) and thermal decomposition (Stern, 2001). Therefore,

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reactions associated with NO_x in desert air should be more vigorous than in the air of other remote areas. So far the atmospheric cycle of reactive nitrogen in desert air has rarely been investigated and the contribution of soil NO_x to the atmospheric reactive nitrogen is unclear. Although numerical simulations concerning the budget of atmospheric reactive nitrogen always encode the emission from surface, NO_x from desert soil was not considered in most models because of the lack of knowledge and hypothesized low concentration (Penner et al., 1991; Yan et al., 2005; Yienger and Levy, 1995). A few recent models considered the emission of reactive nitrogen compounds from desert soil (e.g. Steinkamp and Lawrence, 2011).

We analyzed aerosol samples of total suspension particles (described as TSP in the following descriptions), particles of aerodynamic diameter $<10 \mu\text{m}$ (as PM_{10}) and particles of aerodynamic diameter $<2.5 \mu\text{m}$ (as $\text{PM}_{2.5}$) which were collected at two sites in Taklimakan desert when a dust storm occurred. It was found that the mass concentration of nitrate in the air estimated from the differently sized samples was approximately constant under remarkably different atmospheric dust conditions. Since there was no demonstrable influence of anthropogenic activities on the air, the nitrate encountered in this study was considered more likely a background species in gas phase in desert air. Here, we report the results and discuss the processes responsible for the nitrate existence and the potential importance of the results.

2. Experiments

The observation was carried out in Taklimakan desert during a dust episode in April 2008. Samples were collected at the Aksu observatory ($40^\circ 37' \text{N}$, $80^\circ 45' \text{E}$; 1028 m a.s.l., located at the north edge of the desert) between April 12th and April 20th and at the Cele observatory ($37^\circ 01' \text{N}$, $80^\circ 44' \text{E}$; 1380 m a.s.l., located at the south edge) between April 22nd and April 26th. According to the dust occurrence, the whole observation period was divided into non-dust period (between April 12th and April 18th), dust storm period (between April 18th and April 20th) and floating dust period (between April 21st and April 26th). Three mini-volume samplers (Airmetrics, Oregon, USA) were used to simultaneously collect samples of TSP, PM_{10} and $\text{PM}_{2.5}$. Aerosol particles were collected onto 47-mm quartz microfiber filters (Whatman QM/A, Maidstone, UK) and analyzed for the major anions and cations, including SO_4^{2-} , NO_3^- , Cl^- , Na^+ , K^+ , NH_4^+ , Ca^{2+} and Mg^{2+} using an ion chromatography (Dionex DX-600). In total, 3 sets of TSP, PM_{10} and $\text{PM}_{2.5}$ were obtained in the non-dust period, 3 sets in the dust storm period, and 5 sets in the floating dust period. It should be pointed out that the suspended particulate matter at the two observatories in all periods, including the non-dust period, was always dominated by mineral dust. Details of the observatories and the sample collection and analysis were described in Wu et al. (2012).

For quality control of the sample collection and analysis, we also analyzed field blank and laboratory blank filters. Two blank filters were prepared after the sample collection at Aksu observatory on April 20th. They were randomly selected from the same batch of filters used for the sample collection and were mounted into the samplers. After about half an hour without turning on the vacuum pumps, they were removed from the samplers and stored similarly to the field samples. A filter that did not undergo transport and handling in the field was used as a laboratory blank. These filters were analyzed in the same procedure as the sample filters. Nitrate was not detected from the field blank filters and that from the laboratory blank filter was lower than the detection limit. Therefore, nitrate we observed from the sample filters should not be derived from contamination.

3. Results and discussion

3.1. Nitrate concentration

The mass concentrations of nitrate, $\text{PM}_{2.5}$, PM_{10} and TSP under different dust conditions are summarized in Fig. 1. Dust loading in the air largely varied during the whole period. The average mass concentrations of TSP, PM_{10} or $\text{PM}_{2.5}$ during the dust storm period were about 9–14 times of those during the non-dust period and 4–6 times of those during the floating dust period. Dust loading of $\text{PM}_{2.5}$ ($83\text{--}1340 \mu\text{g m}^{-3}$) accounted for less than 50% of that of TSP ($249\text{--}4266 \mu\text{g m}^{-3}$) indicating dust in coarse fractions (particles $>2.5 \mu\text{m}$ in diameter) dominated the dust loading. The significant difference of dust loading between PM_{10} ($173\text{--}2836 \mu\text{g m}^{-3}$) and TSP indicated the substantial contribution of dust particles larger than $10 \mu\text{m}$.

Nitrate concentration did not show a significant difference under different dust conditions. From differently sized particulate matter ($\text{PM}_{2.5}$, PM_{10} and TSP) samples, it was $3.81 \pm 1.24 \mu\text{g m}^{-3}$ under non-dust condition, $2.95 \pm 0.69 \mu\text{g m}^{-3}$ under floating dust condition, and $4.99 \pm 1.71 \mu\text{g m}^{-3}$ under dust storm condition. The average concentration of nitrate from TSP, PM_{10} or $\text{PM}_{2.5}$ during the dust storm periods was 1.2–1.4 times of that during the non-dust period and 1.5–1.8 times of that during the floating dust period despite the more-than-one-order difference of dust loading in differently sized samples. Under the same dust conditions, nitrate concentration from $\text{PM}_{2.5}$ was about 45%–90% of that from TSP and the concentration from PM_{10} was very close to that from TSP. These results indicate the independence of nitrate from dust loading, i.e. the presence of dust-independent nitrate, in the desert air and its approximately constant concentration in contrast to the large variation of dust loading.

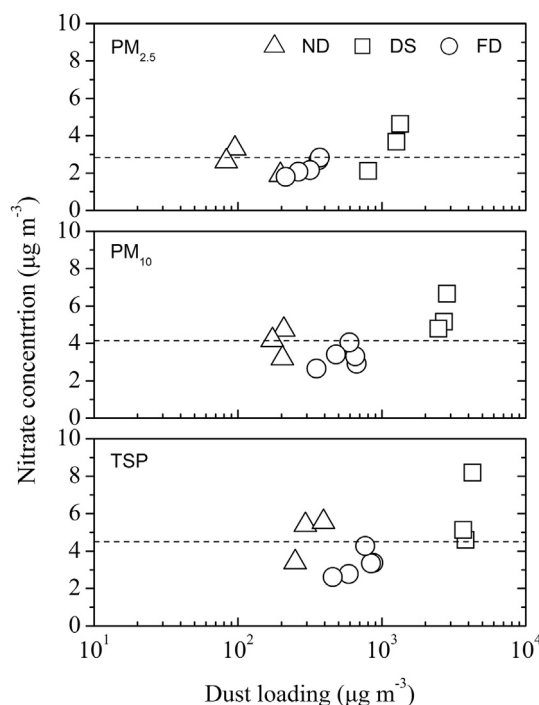


Fig. 1. Concentration of nitrate vs that of $\text{PM}_{2.5}$, PM_{10} and TSP under non-dust (ND), dust-storm (DS) and floating dust (FD) conditions. The dash lines show the mean values of nitrate concentration.

3.2. Original form and possible source of the nitrate

The most common form of nitrate in atmospheric aerosol particles is ammonium nitrate (NH_4NO_3). However, our analysis showed that ammonium concentration in all samples was lower than the detection limit (Wu et al., 2012). The nitrate we observed was unlikely in the form of NH_4NO_3 .

It has been reported that surface dust in Taklimakan desert contains a very small amount of nitrate (Nishikawa et al., 2000). But the nitrate we encountered was not expected to be dominated by that from surface soil. If the content were dominated by soil-derived nitrate, the content of nitrate in the samples should have been proportional to the dust loading, which was against the result that the nitrate was independent from the dust loading. In addition, our analysis of deposited dust which was collected on the roofs of buildings close to the Cele observatory revealed a nitrate content of 0.003–0.015%, corresponding to a nitrate concentration of 0.11–0.66 $\mu\text{g m}^{-3}$ regarding to the maximum TSP in our samples.

Nitrate in dust samples in the downstream areas of the Asian continent has been frequently attributed to the production via heterogeneous reactions of nitrogen oxides (including HNO_3) on the surface of mineral dust particles (Hwang and Ro, 2006; Song and Carmichael, 2001; Sullivan et al., 2007; Zhang, 2003). The reactants in the surface of dust particles are aqueous carbonates, such as dissolved calcium and magnesium carbonates. Nitrates in this form should be correlated with Ca^{2+} , Mg^{2+} or dust loading, and be enriched in coarse fraction (Song and Carmichael, 2001). This is inconsistent with the present result that the nitrate was not correlated with the dust loading. In addition, heterogeneous formation of nitrate on pre-existing dust particles is not efficient in dry dusty air because liquid water is one key medium for the relevant reactions (Vlasenko et al., 2006; Zhang and Iwasaka, 1999). Taklimakan desert is located in the hinterland of the Asian continent with an extreme continental-arid climate. The air was very dry and the relative humidity rarely exceeded 30% during the sampling periods. It was hard to account for the nitrate by NO_x -to-nitrate conversion on the surface of dust particles.

We suggest the absorption of nitric acid by the dust-loading sampling filters is responsible for the presence of the nitrate in the samples. Collecting particulate matter with quartz filters could trap nitric acid onto the filters if there was nitric acid in the air (Appel et al., 1984). Turpin et al. (1997) investigated the gas-particle partitioning of nitrate in hot desert air with a thermodynamic equilibrium model and pointed out that the nitrate should entirely be in the form of nitric acid. We consider that there was a stable content of nitric acid in the desert air when we collected the samples and the nitric acid was produced via gas-phase reactions of NO_x which was emitted from the desert surface soil.

Gaseous emission from soil is the primary export of nitrogen in arid lands (Feig et al., 2008). Deposits of nitrate were frequently observed in desert soil and attributed to the atmospheric deposition in geological time (Böhlke et al., 1997; Michalski et al., 2004; Qin et al., 2012). A recent study revealed that abiotic gas formation caused by thermal decomposition of organic and/or inorganic nitrogen was a key manner of nitrogen release from desert soil (McCalley and Sparks, 2009). Nitrification and denitrification by microbial activities are the major pathways of NO_x production at the soil surface, but the two processes must be very weak due to the low soil moisture in desert areas (Hartley and Schlesinger, 2000; Peterjohn and Schlesinger, 1991).

Nitrate deposits have been confirmed in some areas of Taklimakan desert. For example, Zhang et al. (2000) found widely-distributed nitrate in the soil of the Turpan-hami area in the northeast part of the desert. The estimated total nitrate resource there (2.6×10^9 t) was equivalent to that of Atacama Desert (Li et al.,

2010). Therefore, it is very likely that there were nitrate pools under the surface soil in the desert and they supplied the precursor NO_x responsible for the nitrate we observed. This hypothesis can explain the presence of the nitrate with the fact of no demonstrable influence of fossil fuel combustion and biomass burning on the air during the sampling period. Price et al. (1997a, 1997b) pointed out that production via lightning and stratospheric input could result in a remarkable increase of NO_x in the troposphere. But this potential must be very small because of the rare lightning at the desert area (Sayit, 2012).

It should be noted that nitric acid in the air flow in the samplers could not be completely absorbed by soil particle-loading quartz filters during particle collection (Appel et al., 1984). The total nitric acid in the atmosphere might be larger than that we detected. This may explain the small increase of nitrate loading with the large increase of dust loading in the samples (Fig. 1) because remarkable increase of dust loading on filters would have increased filter's resistance, which should have resulted in an increase of the trapping efficiency of acidic materials by the filters.

3.3. Comparison and implication

To the extent of our knowledge, there are three published papers in which nitrate of airborne particles in Taklimakan desert was described. Nishikawa et al. (2000) reported a nitrate concentration of 5 $\mu\text{g m}^{-3}$ in aerosols of Hotan and mentioned its large uncertainty. Yabuki et al. (2005) measured water-soluble components in aerosols collected by the use of low-volume Andersen samplers at Aksu from March 2001 to April 2004 and showed that nitrate concentration in TSP during two dust periods (1 March 2002 and 13 April 2002) was 2.27 $\mu\text{g m}^{-3}$ and 1.68 $\mu\text{g m}^{-3}$, respectively. Huang et al. (2010) reported the average nitrate concentration was 2.26 $\mu\text{g m}^{-3}$ and was not very different between dust and non-dust storm periods at Tazhong during a dust episode when there was less anthropogenic pollution. Sources and mechanisms potentially responsible for the nitrate presence were not confidently elucidated in those studies.

There are a limited number of publications in which measurements of nitrate in aerosols at other desert areas under dust or non-dust conditions were mentioned and almost all of them reported a nitrate concentration in the order similar to that of our results. Table 1 illustrates the available data. For example, the average

Table 1

Inter-comparison of nitrate loading in desert, remote marine and tropical forest air under natural conditions.

Site	Sample	Nitrate ($\mu\text{g m}^{-3}$)	Reference
Desert (dust condition)			
Aksu and Cele/Taklimakan desert	TSP	2.62–8.18	This study
	PM ₁₀	2.65–6.65	
	PM _{2.5}	1.80–4.64	
Aksu/Taklimakan desert	TSP	1.68–2.27	Yabuki et al. (2005)
Hotan/Taklimakan desert	TSP	~5	Nishikawa et al. (2000)
Tazhong/Taklimakan desert	PM _{2.5}	2.26	Huang et al. (2010)
Tongyu/Horqin sandland	PM _{2.5}	4.6 ± 2.5	Shen et al. (2011)
Meadview/Mojave Desert	TSP	2.17–2.23	Turpin et al. (1997)
Remote ocean			
Equatorial Pacific	TSP	0.16–0.17	Savoie et al. (1989)
Tropical South Pacific	TSP	0.11 ± 0.01	Savoie et al. (1989)
Pristine tropical forest			
Guyana	TSP	0.013	Gregory et al. (1986)
Amazon basin	TSP	0.43 ± 0.18	Talbot et al. (1988)
		(July)	
		0.39 ± 0.05	
		(August)	

nitrate concentration in dust periods was $4.6 \pm 2.5 \mu\text{g m}^{-3}$ at Horqin sandland, located at 3000–4000 km east of Taklimakan desert (Shen et al., 2011). Turpin et al. (1997) observed $2.17\text{--}2.23 \mu\text{g m}^{-3}$ nitrate entirely in gas phase in the air at Mojave Desert in the western United States during summer time when there were occasional dust storms and few anthropogenic activities.

Therefore, there is very likely background nitrate in the form of nitric gas in desert surface air and its concentration is about several micron grams per cubic meters ($2\text{--}8 \mu\text{g m}^{-3}$, to the extent of published literature). This concentration is actually much larger (tens of times) than the concentration of nitrate in the remote marine atmosphere ($0.11\text{--}0.17 \mu\text{g m}^{-3}$). It is also larger than the nitrate concentration in the pristine air of tropical forests ($0.013\text{--}0.43 \mu\text{g m}^{-3}$) (Table 1). Deserts, occupying approximately one-fourth of land areas, are about twice the size of tropical forest areas and one-tenth of marine areas on the earth. The net input of reactive nitrogen from desert surface air to the upper layers such as lower free troposphere could exceed that from tropical forests and is likely comparable to that from the ocean. In this regard, the emission of nitrogen compounds from deserts has to be encoded into numerical models in order to correctly assess the nitrogen cycles and its subsequent roles in the development and evolution of the earth environment.

4. Conclusions

Background-like nitrate which was supposed to be present in the form of nitric acid due to gas-phase conversions of NO_x from surface soils was found in the air of Taklimakan desert. Its concentration was several $\mu\text{g m}^{-3}$, which was larger than that in the remote marine air and in the pristine tropical forest air by one to two orders. Similar nitrate presence was also confirmed in other desert areas from the data of previous studies. It is likely there were active reactions associated with nitrogen compounds leading to the substantial nitrate in desert air. Since deserts occupy a large fraction of land areas on the earth, the emission of nitrogen compounds from surface soils in deserts could affect the local nitrogen mass balance and the global atmospheric nitrogen budget. Thus the nitrogen chemistry in desert air and its roles in global nitrogen cycles deserve further investigation in both field measurements and model studies.

Acknowledgments

This work was supported by the NNSF of China (Grant No. 40872211) and the West Light Foundation of CAS. It was also partly funded by the joint research program of JSPS and NSFC. The authors thank Prof. Chengyi Zhao, Prof. Jiaqiang Lei and the staff of Aksu and Cele observatories of CAS for their support of this study, and Prof. Jay Melton for his revision of words and grammar.

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