



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.

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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.

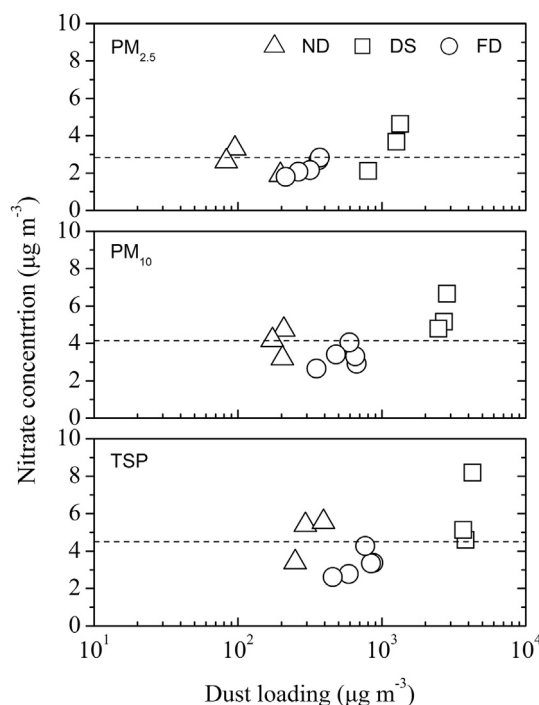


Fig. 1. Concentration of nitrate vs that of $\text{PM}_{2.5}$, PM_{10} and TSP under non-dust (ND), dust-storm (DS) and floating dust (FD) conditions. The dash lines show the mean values of nitrate concentration.

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