



Multi-year record of atmospheric and snow surface nitrate in the central Antarctic plateau



R. Traversi ^{a,*}, S. Becagli ^a, M. Brogioni ^b, L. Caiazzo ^a, V. Ciardini ^c, F. Giardi ^a, M. Legrand ^d, G. Macelloni ^b, B. Petkov ^e, S. Preunkert ^d, C. Scarchilli ^c, M. Severi ^a, V. Vitale ^e, R. Udisti ^{a,e}

^a Dept. of Chemistry "Ugo Schiff", University of Florence, Sesto F.no, Florence, Italy

^b IFAC-CNR, Sesto F.no, Florence, Italy

^c ENEA, Laboratory for Observations and Analyses of the earth and Climate (SSPT-PROTER-OAC), Rome

^d Université Grenoble Alpes, CNRS, Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), Grenoble, France

^e ISAC-CNR, Bologna, Italy

HIGHLIGHTS

- Continuous nitrate records in aerosol and snow obtained at Dome C for multiple years.
- One-month lag of nitrate summer maxima in snow with respect to aerosol was observed.
- Gaseous phase was found to be the most relevant contribution to nitrate budget at Dome C.
- Synoptic analysis of a major nitrate event showed a stratosphere-troposphere exchange.
- Uptake of atmospheric HNO₃ and bottom-up processes can explain snow summer maxima.

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ABSTRACT

Continuous all year-round samplings of atmospheric aerosol and surface snow at high (daily to 4-day) resolution were carried out at Dome C since 2004–05 to 2013 and nitrate records are here presented.

Basing on a larger statistical data set than previous studies, results confirm that nitrate seasonal pattern is characterized by maxima during austral summer for both aerosol and surface snow, occurring in-phase with solar UV irradiance. This temporal pattern is likely due to a combination of nitrate sources and post-depositional processes whose intensity usually enhances during the summer. Moreover, it should be noted that a case study of the synoptic conditions, which took place during a major nitrate event, showed the occurrence of a stratosphere-troposphere exchange.

The sampling of both matrices at the same time with high resolution allowed the detection of an about one-month long recurring lag of summer maxima in snow with respect to aerosol. This result can be explained by deposition and post-deposition processes occurring at the atmosphere-snow interface, such as a net uptake of gaseous nitric acid and a replenishment of the uppermost surface layers driven by a larger temperature gradient in summer. This hypothesis was preliminarily tested by a comparison with surface layers temperature data in the 2012–13 period.

The analysis of the relationship between the nitrate concentration in the gas phase and total nitrate obtained at Dome C (2012–13) showed the major role of gaseous HNO₃ to the total nitrate budget suggesting the need to further investigate the gas-to-particle conversion processes.

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1. Introduction

Nitrate is the end product of the oxidation of atmospheric nitrogen oxides (NO_x = NO + NO₂) and one of the most abundant

ions present in polar ice and snow, mainly as nitric acid in present-climate conditions (Legrand, 1987).

Chemical stratigraphies of nitrate from snow and ice layers have the potential to provide records of past changes in atmospheric composition (Legrand and Mayewski, 1997) including atmospheric NO_x cycling and oxidative capacity (De Angelis et al., 1984; Delmas et al., 1982; Legrand and Kirchner, 1990; Wolff, 1995), as well as past

* Corresponding author.

solar activity (Traversi et al., 2012) or major variations in Earth's magnetic field (Traversi et al., 2016).

Nevertheless, such a potential is based on the assumption that chemical concentrations in the air, snow, firn and ice cores are correlated. Hence, the knowledge of the link between atmosphere and snow composition at the time of deposition is basic to reconstruct the past climate and the past atmospheric chemical composition. The extent of such knowledge depends on whether the species of interest are gaseous or in the condensed phase, and if they are reversibly and/or irreversibly deposited to snow (Santachiara et al., 2016).

The relationship among the relative chemical composition at the various interfaces (air-snow, snow-firn, and firn-ice) is site-specific and, for each site, it depends on the meteorological parameters and on production mechanisms of the precipitation (Davidson et al., 1985; Dibb and Jaffrezo, 1997; Dick, 1990; Silvente and Legrand, 1993). Thus, in order to achieve a more complete overview of the transfer functions of chemical species interacting with snow and ice, additional field and laboratory experiments are both required.

From the point of view of the field work, different approaches have been followed, that involve the sampling of both atmospheric aerosol and snow cover, in polar and temperate glaciers. In some cases, fresh snow and aerosol were sampled independently (e.g. Shrestha et al., 2002; Ming et al., 2007) while in a number of other cases, simultaneous samplings of aerosol and snow were carried out both in the Arctic (Björkman et al., 2014; Davidson et al., 1985; Ianniello et al., 2016) and Antarctica (Dick and Peel, 1985; Erbland et al., 2013; Frey et al., 2009; Udisti et al., 2004; Wolff et al., 1998).

The results obtained by comparing the chemical composition of aerosol and fresh snow, even when collected at the same time, have frequently been contradictory, i.e. concentrations in snow and in air may be or may be not related (Davidson et al., 1985, 1993; Rahn and McCaffrey, 1979; Udisti et al., 2004; Wolff et al., 1998).

The discrepancy between the chemical composition of atmospheric aerosol and snow samples collected during overlapping time periods is difficult to solve due to both sampling features (thickness of the sampled snow layers, which is variable between top mm to top tenths of cm; different temporal resolution of aerosol and snow, scarcely representative aerosol samplings along the year-round period) and the possible influence of scavenging and post-depositional processes (re-emission of volatile HNO₃, UV photolysis, sublimation; Grannas et al., 2007, and references therein).

In order to provide a contribution to the understanding of dominant sources and sinks of nitrate, as well on their air-to-snow exchange in the Antarctic plateau, we present here nitrate records in atmospheric aerosol and surface snow sampled at the same time at Dome C along 8 years (nine years for the whole aerosol record), at high resolution, all year-round. It is the first time that such long and highly resolved records are achieved from continental Antarctica, where continuous and long-term atmospheric and snow samplings are particularly difficult due to the extreme meteorological conditions and, at the same time, the need of extra-care in avoiding contamination due to the low level of ion concentrations.

As far as Dome C is concerned, beside the work previously presented by Traversi et al. (2014), covering the 2005–2008 time period, the longest record of atmospheric nitrate is reported by Erbland et al. (2013) and Frey et al. (2009). In both cases, all year-round atmospheric (both particulate and gas-phase) samples were collected at Dome C (January 2007–2008 and January 2009–2010, respectively) by using a high-volume air sampler with different time resolution ranging between 5 and 7 days (Erbland et al., 2013) and 2 weeks (Frey et al., 2009). In particular, Erbland et al. (2013) carried out contemporaneous air and surface snow samplings all along the studied year, using a similar approach to the

one here presented.

Concerning central Antarctica, the longest record available to date is still that discussed by Weller and Wagenbach (2007) reporting the record of nitrate and other chemical species from Kohnen Station (Dronning Maud Land) along an almost 2-year long period (February 2003–December 2005), at 7 day- and 15 day-resolution. All the other available aerosol chemical data sets from Antarctic plateau areas are related only to the summer period i.e. November to February (Arimoto et al., 2004, 2008; Huey et al., 2004).

The multi-year record here presented enlarges significantly the data base of aerosol and snow chemical composition in the central Antarctic Plateau, allowing the more reliable detection of recurring seasonal patterns and providing a statistically more significant estimation of nitrate levels both in aerosol and surface snow in different times of the year. In order to document the total nitrate content in the atmosphere, gaseous and particulate nitrate were year-round sampled in 2012 and 2013 by deploying denuder tubes equipped with a back-up aerosol filter. Physical variables such as daily solar irradiance flux, snow surface temperature and meteorological parameters were examined for the longest time period overlapping with our aerosol and snow samplings, with the aim of providing hints on the major sources and atmosphere-to-snow processes controlling ground-level nitrate concentration at Dome C.

2. Experimental

2.1. Sampling and direct measurements

2.1.1. Aerosol and gas-phase

All year-round aerosol samplings have been carried out with various low volume sampling devices starting from December of 2004 at Concordia Station (Dome C, central Antarctica, 75° 06' S, 123° 20' E, 3220 m above sea level), located in the inner Antarctic Plateau, 1100 km away from the nearest coast.

The sampling site is located at the edge of the 'clean area', roughly 700 m away from the Italian-French permanent base and was upwind with respect to the dominant wind direction (south-west) in order to minimize contamination risks. Meteorological data provided by the Italian AWS station were carefully evaluated in a previous work (Traversi et al., 2014) for full four years (2005–2008) in order to verify the conditions of minimum or negligible contamination. Such analysis confirmed that the sampling station is properly placed up wind with respect to the Concordia base and that the occurrence of wind blowing from the opposite side is one order of magnitude lower and then negligible in affecting nitrate atmospheric concentration in the Dome C area on a seasonal and annual base, and also on a daily base for major nitrate events.

Here we present the data related to PM₁₀ and multi-stage impactor, accomplished with a daily (PM₁₀) and 3–4 days (4-stage impactor) temporal resolution, covering the time period December 2004–November 2013 for PM₁₀ and January 2006–November 2013 for the multi-stage impactor.

For PM₁₀ and the back-up stage of multi-stage impactor samplings, a Teflon (PTFE) membrane (with polymethylpentene support ring) and a polycarbonate membrane was chosen respectively, on the basis of the lowest blanks of the main and trace ionic components determined along with nitrate by Ion Chromatography (Becagli et al., 2012; Udisti et al., 2012). Moreover, the selected PTFE filter is certified to have a sampling efficiency higher than 99.5% for particles of 0.3 μm size. Further sampling details can be found in Becagli et al. (2012) and Traversi et al. (2014).

The gas-particulate nitrate partitioning was investigated at

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