EGU21-9425, updated on 02 Dec 2022
https://doi.org/10.5194/egusphere-egu21-9425
EGU General Assembly 2021

Innovative approach for new estimation of NO_x snow-source on the Antarctic Plateau

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Previous Antarctic summer campaigns have shown unexpectedly high levels of oxidants in the continental interior as well as at coastal regions, with atmospheric hydroxyl radical (OH) concentrations up to 4 x 10^6 cm^{-3}. It is now well established that such high reactivity of the summer Antarctic boundary layer results in part from the emissions of nitrogen oxides (NO_x ▯ NO + NO_2) produced during the photo-denitrification of the snowpack. Despite the numerous observations collected at various sites during previous campaigns such as ISCAT 1998, 2000, ANTCI, NITE-DC and OPALE, a robust quantification of the NO_x emissions on a continental scale over Antarctica is still lacking. Only NO emissions were measured during ISCAT and the ratio NO_2:NO was measured during NITE-DC and OPALE using indirect NO_2 measurements. This leaves significant uncertainties on the snow-air-radiation interaction. To overcome this crucial lack of information, direct NO_2 measurements are needed to estimate the NO_x flux emissions with reduced uncertainties.

For the first time, new developed optical instruments based on the IBB-CEAS technique and allowing direct measurement of NO_2 with detection limit of 10 x 10^{-12} mol mol^{-1}, (1σ), (Barbero et al., 2020) were deployed on the field during the 2019–2020 summer campaign at Dome C (75°06'S, 123°20'E, 3233m a.s.l). They were coupled with new designed dynamic flux chamber experiments. Snows of different ages ranging from newly formed drift snow to 16-20 year-old firn were sampled. Unexpectedly, the same daily average photolysis constant rate of (2.18 ± 0.38) x 10^{-8} s^{-1} (1σ) was estimated for the different type of snow samples, suggesting that the photolabile nitrate behaves as a single-family source with common photochemical properties. Daily summer NO_x fluxes were estimated to be (4.4 ± 2.3) x 10^7 molec cm^{-2} s^{-1}, 10 to 70 times less than what has been estimated in previous studies at Dome C and with uncertainties reduced by a factor up to 30. Using these results, we extrapolated an annual continental snow source NO_x budget of 0.025 ± 0.013 Tg.N y^{-1}, more than three times the N-budget of the stratospheric denitrification estimated to be 0.008 ± 0.003 Tg.N y^{-1} for Antarctica (Savarino et al., 2007), making the snowpack source a rather significant source in Antarctica. This innovative approach for the parameterization of nitrate photolysis using flux chamber experiments could significantly improve future global atmospheric models.