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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fluxes of nitrates between snow surfaces and the atmosphere in the European high arctic" *by* H. J. Beine et al.

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Measurements of boundary layer NOy in polar regions have revealed some surprising properties. Much higher levels of NOx and HONO have been observed than were anticipated in the air above snow surfaces, apparently due to emission from the snow-pack into an often very shallow stable boundary layer. It is believed that this is due to photochemical conversion of nitrate ions in the snow-pack and hence the origin and concentration of nitrate in snow is of great interest. The current paper is addressing this issue, reporting measurements of nitrate fluxes into and out of the snow-pack in the spring of 2001 at Ny Alesund, Svalbard. The flux of gaseous HNO3 and particle nitrate was measured using a denuder technique, and the nitrate content in the snow-pack as well as physical characteristics of the snow are described.

The main assertion from this study is that flux of HNO3 to or from the snow-pack is determined by whether it is acidic or alkaline. Acidic snow-packs appear to emit, alkaline snow-packs do not. It is suggested that the latter observation is due to the fact that the NO3- ions are associated with cations such as Na+ from seasalt or Ca2+ of crustal origin, which makes them not readily available for physical exchange or photochemical conversion into NOx and/or HONO. In addition, the alkalinity appears to favor deposition flux of HNO3. However, the contribution of HNO3 flux to the total nitrate content of the snow pack appears to be small; most is estimated to originate from deposition scavenging of the snow itself. The conclusions are reached from a comparison of similar measurements made a year earlier by the same authors at Alert, Nunavut, where the snow was generally acidic, in contrast to the Ny Alesund snow.

Unfortunately data on NOx are absent. This necessitates the assumption that NOx flux is negligible based on the absence of a measurable flux of HONO (roughly equal flux of NOx and HONO was observed from the acidic snow in Alert), but, as pointed out by Wolff in his comments on this paper, the validity of this assumption is debatable. It is shown in the paper that nitrate in snow constitutes a potentially large reservoir for atmospheric NOy, but a large contribution to atmospheric NOy would go undetected in the snow composition itself. Hence a less ambiguous determination of whether there is an NOx flux or not out of alkaline snow remains highly desirable.

The conclusions on the properties of alkaline snow should not come as a big surprise; after all, the whole idea of denuder sampling for HNO3 is based on this principle. Nevertheless, it is an important result in that it reminds us that not all (dirty) snow packs should be expected to emit NOy upon solar irradiation. Other interesting implications are mentioned by Wolff. Overall this is a nice paper, well worth publishing, but I agree with Wolff on the need to downplay the statements on the absence of NOx emission.

Additional comments:

* the paper often talks about Şwet depositionŤ. This seems to me a confusing term in

ACPD

3, S68–S70, 2003

the context of snow deposition. I suggest to use the term Şprecipitation scavengingŤ

* the discussion on page 85 about why the snow composition was quite variable is rather confusing and not always consistent. To me the most obvious and likely reason is simply mixing of different snows due to the windy conditions

* on the bottom of page 85 it is mentioned that the windpacked sublayer that formed on May 4 was accompanied by an increase in the nitrate concentration. This seems an interesting observation and I recommend some discussion on how this could have come about

* bottom page 86: in view of the discussions on page 85 (line 8-9), it seems unwarranted to assume that the nitrate content of the surface snow was representative for that in falling snow. Failing measurements of falling snow, at least this should be mentioned

* page 90: if the snow on April 29 was initially acidic, would you not have expected to see a flux of NOy out of the snow (positive in the terminology of the paper), at least on the first day or so after the snow fall?

* page 92 (line 24): the fact that pH appears to influence the NOy emission does not confirm, but at most supports the suggestions by Dominé and Shepson

* while all the different units appear correct as far as I can see, their amount is staggering which makes reading and comprehension sometimes quite difficult. I recommend the addition of a table of units and conversion factors.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 75, 2003.