Reviewer M. LEGRAND

The authors present the records of nitrate and ammonium extracted from an ice core spanning the 1220-2009 time period drilled at Lomonosovfonna (Svalbard) in the Eurasian Arctic. Having discarding a possible influence of melting, the authors discussed these ice Svalbard ice records in terms of past changes of atmospheric chemistry in response to changes of natural sources and growth of man made emissions in the northern hemisphere. During the 20th century both records seems to be influenced by anthropogenic pollution from Eurasia. At decadal scale, the pre-industrial nitrate level is highly correlated with the methanesulfonate (MSA) one, which is proposed to reflect a fertilising effect (nitrogen input to the ocean triggering the growth of DMS-producing phytoplankton). Eurasia was assumed to be the main source area for pre-industrial nitrate, whereas for ammonium, biogenic ammonia emissions from Siberian boreal forests were identified as the dominant pre-industrial source.

The short atmospheric lifetime of species like ammonium, nitrate, sulphate, or MSA motivates to increase efforts to gain ice core records at many places in the world, and in that sense the present work is useful to report. However, as it stands the manuscript suffers from two major weaknesses.

1. As for any ice core extracted from Svalbard, the large presence of melted snow layers rise the question to what extend the chemical ice core signals can be here safety related to atmospheric chemistry change. This crucial point needs to be addressed furthermore in the manuscript. As it stands, it is claimed in the abstract and the conclusion that this question is discussed in the paper but in fact it is only indirectly discussed when the common feature of nitrate and MSA is discussed in section 3.1 (see my other comments below). Since the effect of melt would differ from one chemical species to another one, I strongly suggest addressing more carefully this point as follows:
Put your Figures S1 and S2 (only available in the supplementary material) in the main text, for S1 please report not only ^{18}O and sodium but also melt, nitrate, ammonium, and MSA.
Please explain how your vertical lines (annual layer counting) were identified (at the first glance on the basis of ^{18}O but did the sodium profile really useful?).
Where are located other ions compared to sodium at such a seasonal scale? Did the delocalization due to melt differs from MSA to nitrate?
Could you calculate the ionic balance to evaluate the acidic character of snow layers and melted snow layers that may have influenced the remobilisation? Please comment.

2. The discussion of data in terms of sources (natural and anthropogenic) is rather vague, often based on comparison with other smoothed records extracted at other places in the northern hemisphere to identify sources or source regions. What are missed in the manuscript, that may help the reader to follow the comparison with various records (Altai, Alps, Greenland), are air mass back trajectories calculated for winter and summer at your site using HYSPLIT and the NCEP reanalysis for instance. Such analyses were already done by Eichler (GRL, 2009) for Altai in Siberia, by Kahl (JGR, 1997) for Summit in central Greenland or Fagerli (JGR, 2007) for the Alps (here with the EMEP transport-chemistry model). They need to be done for

Svalbard as well. Such information would then strengthen (or not) your argument based on correlations between records that may be sometimes coincidental.

Kahl, J. D. W., Martinez, D. A., Kuhns, H., Davidson, C. I., Jaffrezo, J. L., and Harris, J. M.: Air mass trajectories to Summit, Greenland: A 44-year climatology and some episodic events, J. Geophys. Res. Oceans, 102, 26861–26875, doi:10.1029/97jc00296, 1997.

Fagerli, H., M. Legrand, S. Preunkert, V. Vestreng, D. Simpson, and M. Cerqueira, Modeling historical long-term trends of sulfate, ammonium, and elemental carbon over Europe: A comparison with ice core records in the Alps, J. Geophys. Res., 112, D23S13, doi:10.1029/2006JD008044, 2007.

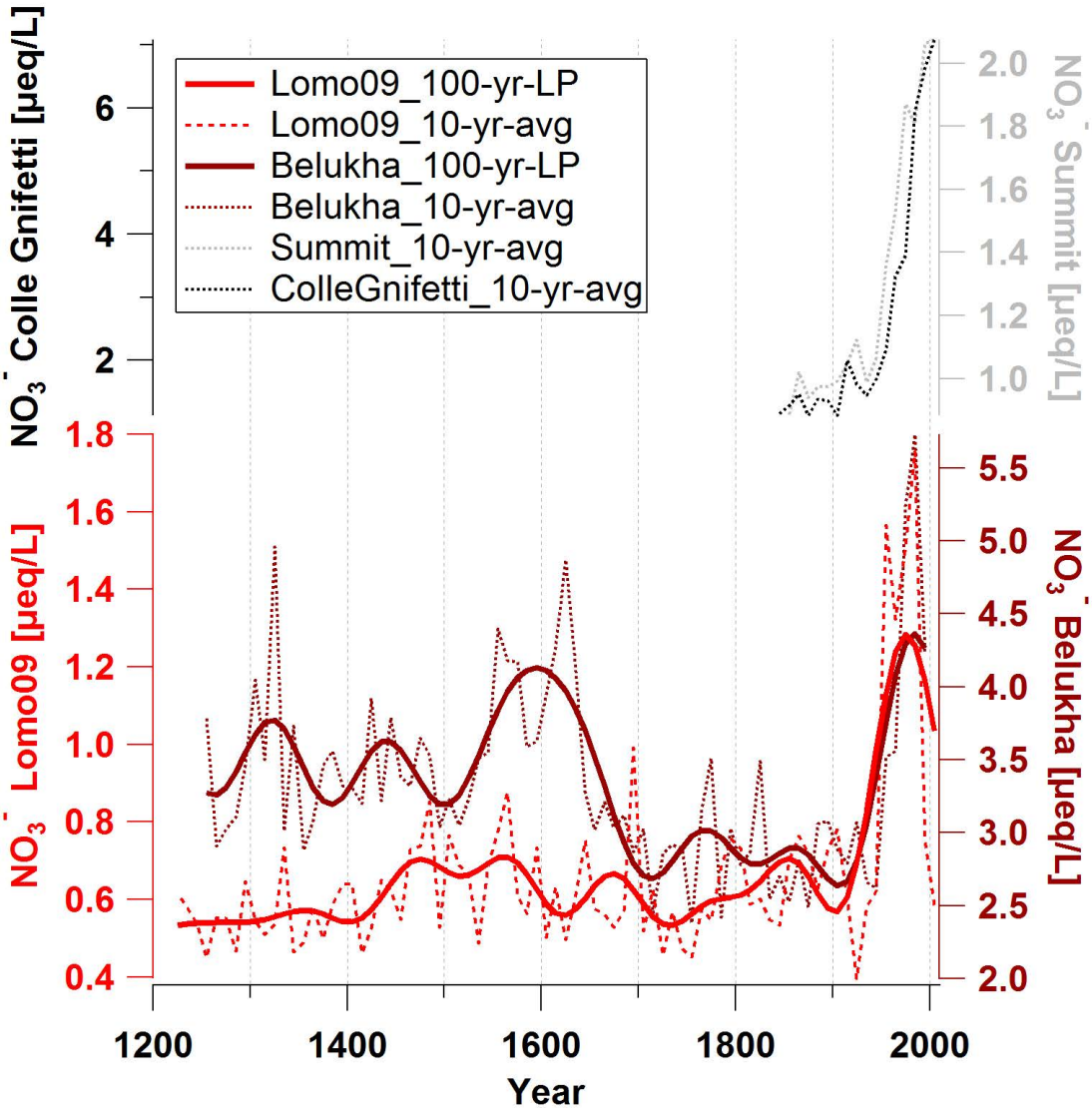
Sometimes your conclusions drawn when comparing different records are a bit subjective. I here take the example of the nitrate change and your comparison with Altai, the Alps and Greenland. My conclusion is very different from your. When discussing source regions of concern for anthropogenic NOx emissions you pointed out the similarity between your record and the one from Altai indicating that both records show a strong anthropogenic trend followed by a decrease after 1980 that contrasts with Alpine and Greenland records showing persisting high values after 1980. From that you concluded that the main source region for anthropogenic emissions at your site is Eurasia. First, it has be recognized that Greenland ice archives anthropogenic emissions from North America and Eurasia (polluted air masses from these two regions being advected in winter in the Arctic basin and transported over Greenland and lower latitudes in spring). Note that for Greenland you can also report in your Figure 4, in addition to Geng et al. (2014) the record from Savarino and Legrand (which, as your ice core, extend back to 1200 AD). For the Alps, the main source region is western Europe (see Preunkert et al., 2003 or Preunkert and Legrand, 2013, for nitrate records).
Savarino, J., and M. Legrand, High northern latitude forest fires and vegetation emissions over the last millenium inferred from the chemistry of a central Greenland ice core, J. Geophys. Res., 103, 8267-8279, 1998.

Preunkert, S., and M. Legrand, Towards a quasi-complete reconstruction of past atmospheric aerosol load and composition (organic and inorganic) over Europe since 1920 inferred from Alpine ice cores, Clim. Past, 9, 1403-1416, doi:10.5194/cp-9-1403-2013, 2013.

Preunkert, S., Wagenbach, D., and M. Legrand, A seasonally resolved Alpine ice core Record of Nitrate: Comparison with Anthropogenic Inventories and estimation of Pre-Industrial Emissions of NO from Europe, J. Geophys. Res., 108, D21, 4681, doi : 10.1029/2003JD003475, 2003.

Second, comparing Altai and your record my conclusions are different from you: whereas your nitrate level drops after 1980, this change is far less pronounced in the Altai record. I think you may have difficulties to fit your recent nitrate decrease with recent change of nitrogen oxide emissions. Furthermore, the Altai record clearly shows that anthropogenic emissions do not dominate preindustrial sources, and since a large variability is obvious for these natural sources in your Figure 4 (reported below), you cannot use the small decrease seen after 1980 as a sign of decreasing anthropogenic emissions there. In fact, except after 1980 your record is more similar

to the Greenland one with a rather low preindustrial level compared to the strong post 1940 increase at the opposite to the record at Altai where clearly natural sources can be as high as anthropogenic emissions.



Section 3.1: Whereas I found the idea to explain positive correlation between nitrate and MSA innovative and interesting, I would suggest to be more careful in your conclusion drawn by examining your 3 hypothesis (line 23-25, page 24678). I am not sure that you can discard an effect of melt as you did it, based on examination of correlation since the redistribution of nitrate may be different from the one of MSA.
Please also recognize that the good correlation between nitrate and MSA works for the periods around 1480, 1560, and 1680 but fails for the peak of MSA seen around 1350 (see Figure 5).
In discussing MSA, I always like to see the sulphate record (especially during the pre-industrial time). I am sure that the record is available since you used it for the dating purpose. It would have been also interesting as additional (back-up)

information to show the sulfate record when comparing anthropogenic emissions as recorded in Greenland and Altai.

Section 3.2: same comments as for nitrate: when comparing Altai and Svalbard: Fig 6 first indicates me that Svalbard and Greenland pre-industrial ammonium level are similar but are both one order of magnitude lower than at Altai, clearly pointing out the importance of continental biospheric emissions in Siberia compared to Greenland and Svalbard located far away from continental emissions.

Minor points:

Page 24668, line 23: I don't think that you can claim that PAN is efficiently wet deposited (PAN is not very soluble in acidic water).

Page 24668, line 25: Replace "in general" by "at a global scale"

Page 24670, line 16-18: I agree with your statement for nitrate but I don't see how possible is a migration of ammonium in a cold archives (without melting). At the opposite I am surprised that you don't mention previous studies having shown migration of MSA in snow and ice.

Page 24672, line 7: Please note that nitrate is not at all totally present in the aerosol phase.

Page 24668, line 24: What do you mean by "Values were not blank corrected" ? Either skips it or explains how blank are done, report if they are significant or not.