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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Partitioning of reactive nitrogen (NO_y) and dependence on meteorological conditions in the lower free troposphere" *by* C. Zellweger et al.

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Response to Referee #3

We would like to thank Referee #3 for his interest in our paper and his suggestions. The concerns he raised are addressed below.

Referee #3 mentions that he is not sure about the main focus of the paper. We feel that a two-fold focus is clearly visible, both from the length of coverage in the paper and from the abstract. The first focus of the paper is in fact to demonstrate that filters (in our case mostly based on meteorological parameters) are of crucial importance for the interpretation of any data observed at ground based measurement sites. The second focus is the presentation of a dataset, which is especially within the context of these filters highly valuable, since the paper presents a climatology of the measured

parameters for the total observation time and for specific meteorological conditions.

Referee #3 also mentions that the statement "This is the first data series on nitrogen oxides levels in the free troposphere over Central Europe" is not appropriate. He however acknowledges that this work represents "the first published results of ground based measurements of reactive nitrogen partitioning over a longer time period Europe". We agree that there are indeed a few publications on short-term measurements, however, they are not as numerous as one could conclude from the referee's statement (see below). We agree also that comparisons with other sites in Europe would be valuable. However, to our knowledge (and also to the knowledge of referee #3) no data from comparable ground based sites in Europe are published.

Referee #3 suggests in particular a comparison with the German station Hohenpeißenberg (985 m a.s.l.). Again, to our knowledge, NOy and speciated data from this station is not yet published. NOy measurements without further speciation except for NO and NO2 have been performed at the Hohenpeißenberg station since 1999, and hourly data are available from the World Data Centre for Greenhouse Gases [http://gaw.kishou.go.jp/wdcgg.html]. These data demonstrate that the two sites are not easily comparable, especially when the difference in altitude of 2595 m between the sites is considered. During 1999, NOy averaged 3.14 ppbv at the Hohenpeißenberg station, while a yearly average of only 0.88 ppbv was observed at the Jungfraujoch (JFJ). This suggests that the Hohenpeißenberg station is by far more often influenced by polluted air masses than the JFJ. Still, we will be happy to include these numbers as unpublished data in the revised paper with an appropriate citation, if the data producer is willing to provide the data. We can offer the same for unpublished data from other mountain stations.

Referee #3 also suggests that the JFJ time series should be compared with aircraft measurements performed over Europe. These measurements often cover only a few hours and were performed at different altitude levels in the troposphere. Nevertheless, we will be happy to include results of aircraft measurement campaigns in a revised

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manuscript. A few are mentioned in the following: Hering et al. [1998] described aircraft NOy measurements performed over the Swiss plateau during the POLLUMET (POLLUtion and METeorology) study in summer 1993. NOy concentrations ranged from 1.12 to 21.52 ppb. The flight altitude was significantly lower (500 to 1300 m a.s.l.) compared to the JFJ. However, this data may also be representative for air masses reaching the JFJ during thermally induced vertical transport (average NOy 0.99 ppbv) when dilution with undisturbed FT air during upward transport is considered. Other aircraft measurement campaigns were performed in the upper troposphere and are therefore also not easily comparable to JFJ data. For example, Ziereis et al. [2000] reported an average NOy mixing ratio of 0.24 ppbv measured during the POLINAT 2 campaign at the 8000 m level between 49°N and 57°N over the North Atlantic between 19 September and 25 October 1997. During undisturbed FT conditions, NOy averaged 0.37 ppbv (median 0.20 ppbv) at the JFJ for the above time period. Thus, the JFJ concentrations are relatively well comparable with the values observed during POLINAT 2. The JFJ data also compares well with the median NOy concentration of about 0.40 ppbv observed in continental-influenced air masses over the west Pacific throughout the troposphere without any apparent altitude dependence [Kondo et al., 1996].

Referee #3 also addresses the point that the identification of source regions would be interesting. We agree that this is in fact an interesting aspect but beyond the scope of this paper. However, the location of the JFJ in the middle of Europe and the high dependence of air pollutant concentrations on meteorological conditions should allow for the identification of source regions within Europe, and research in this direction is ongoing.

A last concern of Referee #3 is the relatively low values for HNO3. The loss of HNO3 in the inlet is addressed in section 2.2., where the total NOy measurements are compared between the University of East Anglia and EMPA. However, no further intercomparisons with other techniques were performed. It is also apparent from Figure 11 that HNO3 is still a significant fraction of total NOy, especially during undisturbed FT

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conditions. Furthermore, particulate nitrate (which includes HNO3 absorbed on particulate matter) can be an important fraction of total NOy depending on meteorological conditions.

References

Hering, A.M., J. Staehelin, U. Baltensperger, A.S.H. Prévôt, G.L. Kok, R.D. Schillawski, and A. Waldvogel, Airborne measurements of atmospheric aerosol particles and trace gases during photosmog episodes over the Swiss Plateau and the southern pre-alpine region, Atmos. Environ., 32, 3381-3392, 1998.

Kondo, Y., H. Ziereis, M. Koike, S. Kawakami, G.L. Gregory, G.W. Sachse, H.B. Singh, D.D. Davis, and J.T. Merrill, Reactive nitrogen over the Pacific Ocean during PEM-West A, J. Geophys. Res., 101, 1809-1828, 1996.

Ziereis, H., H. Schlager, P. Schlute, P.F.J. van Velthoven, and F. Slemr, Distribution of NO, NOx, and NOy in the upper troposphere and lower stratosphere between 28° and 61°N during POLINAT 2, J. Geophys. Res., 105, 3653-3664, 2000.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 2259, 2002.