Atmos. Chem. Phys. Discuss., 11, C9390–C9392, 2011
www.atmos-chem-phys-discuss.net/11/C9390/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term in situ measurements of NO_x and NO_y at Jungfraujoch 1998–2009: time series analysis and evaluation" *by* S. Pandey Deolal et al.

Anonymous Referee #1

Received and published: 27 September 2011

The paper of Pandey Deolal describes more than 10 years of continuous in-situ measurements of NOx and NOy at the Jungfraujoch. Based on this data set, a trend analysis is performed and diurnal cycles are discussed. The data set itself is unique and deserves publication, but in order to deduce reliable trends more emphasis should be paid to the evaluation of the data quality in particular for NOy. Although an intercomparison with a second NOy measurement device is performed, the results are rather inconclusive. Therefore, I recommend that the paper is revised before finally published in ACP. In particular the following questions/criticism should be addressed:

The quality of the NOy measurement depends strongly on its capability to account for

HNO3, which is affected by sampling line losses and the conversion efficiency of the NOy converter itself. The authors claim to address the line loss problematic by comparing their NOy measurement to a second measurement using an externally mounted converter. The comparison showed rather good agreement in the October and December, while later comparisons yielded very poor results. The authors argue that most likely NOx emission from a snow covered inlet tip of the external converter is responsible for the strong deviation in January/February. In the experimental section 2.2 the authors state, that the inlet line (20 cm Teflon tubing) is heated to 25°C an protected against snow and rain by a Teflon hood. So how does the inlet accumulate snow, and why only in January and February? Was there no snow fall in December? One way to address this last question would be to extend Figure 13 to the whole period. Additionally, comparing Fig. 10 and 11 it seems that the agreement between the two NOy measurements is good for high NOx/NOy ratios (e.g. most of October and December) and poor for low NOx/NOy ratios (e.g. Jan. 12 – 25).

The authors concede that the conversion efficiency for HNO3 might differ from NO2 and that a degradation of the conversion efficiency could be more severe for HNO3. Nevertheless, all information on the quality assurance for the EMPA instrument is very limited, actually it is restricted to one sentence, indicating that the conversion efficiency of the gold EMPA converter mostly ranged between 95 and 100 %, based on NO2 measurements. I consider this as insufficient, in particular if the authors aim to address potential trends. More information (e.g. a time series) of conversion efficiencies for NO2 AND HNO3 is mandatory to judge on the data quality and exclude instrument artifacts as a potential cause for long term trends as presented in Fig.4. Also, the authors should provide details about maintenance procedures for the EMPA converter (procedure and frequency of converter cleaning). Some of those details (time series of conversion for NO2 and information on cleaning) are only provided for the ETH measurements. Without a better characterization of the EMPA NOy data quality I consider the conclusion drawn by this paper (at least for the trend analysis) as highly superficial.

Specific Questions:

In the time series of NOy it is shown that the year 2003 is exceptional, which is explained by the authors as being due to the 2003 summer heat wave, causing high pollution levels over Europe, but Figure 7 indicates that the NOy enhancement reach far into the fall and winter of this year. I have a hard time to believe that these are remnants from the 2003 summer pollution. Additionally, the authors should extend Figure 4 by inclusion of a NOx/NOy trace. It seems that there is a strong trend for younger air masses (higher NOx/NOy) after 2003, which might indicate a shift in local pollution or a trend in the instrument performance (e.g. decreasing HNO3 conversion).

NOx/NOy ratios would also be helpful in the discussion of the seasonal variation. Convective transport as a cause of higher NOy in spring and summer should be characterized by higher NOx/NOy ratios, but Figure 5 (small panel) seems to indicate quite the opposite, with lowest (oldest, most processed air) NOx/NOy ratios in summer. This is in contradiction to the statement on page 2186, line 1, that only in summer the lifetime of NOx is long enough that rapid upward transport to JFJ can generate large NOx pollution events giving rise to large NOy enhancements.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21835, 2011.