

## ***Interactive comment on “Analysis of elevated spring-time levels of Peroxy Acetyl Nitrate (PAN) at the High Alpine research sites Jungfraujoch and Zugspitze” by S. Pandey Deolal et al.***

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We thank reviewer #1 for his/her careful review and the useful comments and suggestions, following our replies (in the in supplementary pdf file the comments of the reviewer are written in bold italics)

Reviewer's comment: Summary: This paper presents a detailed meteorological analysis of PAN data from the Jungfraujoch and Zugspitze sites during May 2008. The study presents the PAN data alongside several other trace gases, and segregates the data by meteorological patterns (clusters). The paper presents and adequately explains a

C6899

very thorough analysis of 1 month of data. This is not a groundbreaking piece of work, but it is certainly suitable for publication in ACP with relatively minor changes.

Reviewer's Specific Comments: Reviewer's comment: Figure 2 Discussion, pg 12739: Do the authors have any insight on why there appears to be larger interannual variability at the lower sites?

Reply: The sites of the right panel are located in the planetary boundary layer (PBL) including different types such as suburban, rural and low-level alpine. The interannual PAN variability is attributable (assuming similar emission strengths of the precursor) to the interannual meteorological variability: Due to photochemistry PAN concentrations increase in warm periods in summer (during high pressure condition) intermitted by “dilution” as caused by convection and frontal systems. The lack of such dilution of the polluted PBL might explain why PAN in the “heat wave” summer 2003 is very high at Hohenpeissenberg. The shaded area includes PAN concentrations of the different sites contributing to the rather large spread. The high altitude sites (left panel) are intermittently exposed to air reaching the sites from the European PBL and free tropospheric air. PAN measurements at Jungfraujoch show very large variability on hourly time scale (see Fig. 4 of Pandey Deolal et al., 2013) but it appears that frequency of air parcels with low and high PAN concentrations on monthly balances in a way that interannual variability becomes smaller.

Reviewer's comment: Why have the authors chosen to not shade the area including 2005 (JFJ) and 2003 (ZUG)? If there is not a suspected problem with the data, it is very interesting to acknowledge and understand the interannual variability in PAN. There are very few measurements of PAN globally that can offer any information on interannual variability of either production or venting to the free troposphere.

Reply: The measurements at JFJ of 2005 were excluded because they only contain measurements of short campaigns not necessarily representative for complete monthly mean values. No PAN observations were available for Zugspitze in 2003 and there are

C6900

no such data shown in Figure 2.

Reviewer's comment: Pg 12744, Lines 2-3: PAN could also decompose during transit.

Reply: Indeed, this is true, however, we only wanted to make a simple rough estimate and we do not have adequate information to take into account decomposition during transit which also depends on chemistry.

Reviewer's comment: Figures: Please add the cluster descriptors (i.e. "westerly advection" to all the plots and captions. It really slows down the reader to have to constantly refer to the text and remember your color schemes.

Reply: We will revise the figures accordingly.

Reviewer's comment: Overall Comment: This is my largest concern. The authors should attempt to determine how representative May 2008 is compared to the other years of data. I have confidence that the elevated PAN observed at JFJ during May 2008 was the result of recent boundary layer production; but in reality, this analysis is only based on approximately 10 days of data. Is this weather pattern common in spring? Does it occur frequently in other years?

Reply: Indeed, the representativeness of the results ("case study") of the described mechanism for other years is an important question. Replying to the question we refer to the study of Pandey Deolal et al. (2013), in which long-range backward trajectory analysis (performed by LAGRANTO using ERA interim wind fields) together with chemical filters was applied to the PAN measurements at JFJ covering the years 1997, 1998, 2008, 2009 and 2010. The study showed that high spring PAN values at JFJ can be (tentatively) attributed to European PBL influence. Since the underlying trajectory model was based on coarse resolution meteorology and did not represent turbulent and convective transport, the obtained results of the Pandey Deolal et al. (2013) study only allow a qualitative conclusion and, hence, motivated the current study where a more state-of-the-art transport model was used. In order to further explore the

C6901

representativeness of the weather conditions encountered in 2008 we compared the transport clusters obtained in our study with a long term weather type classification. The Alpine Weather Statistic (AWS) is a weather classification that was developed to characterise the weather situation at a given time over the Swiss domain (MeteoSwiss, 1985). The AWS was previously used to analyse PBL transport to JFJ (Henne et al., 2005). The AWS types "convective-indifferent" and "convective-anticyclonic" were identified as weather types for which PBL transport to JFJ was likely during the spring and summer months. Our JFJ cluster 3 largely corresponds with the AWS weather sub-types "convective-anticyclonic flat pressure" and "convective-indifferent easterly advection". The frequency of these two weather types for the years 2001 to 2010 and the months April and May was relatively large in 2008 (>15 days) but comparable to other years (2001, 2004, 2005; > 15 days). When looking at the frequency of all "convective-anticyclonic" and "convective-indifferent" weather types, which are likely to allow PBL transport to JFJ, the frequency in 2008 (30 days) was only slightly larger than the average frequency for all years (27 days). Hence, our conclusion on the representativeness of our 2008 case study is twofold. On the one hand, the occurrence of strong PBL influence during easterly flow in May 2008 was exceptional in its persistence and continuation for about 10 days. On the other hand, the frequency of weather types with likely PBL transport towards JFJ was not larger in 2008 than in other years. Therefore, we are convinced that our findings concerning the origin of the pronounced spring time PAN maximum at JFJ are not restricted to the analysed year but can be interpreted in a more general way. We will add this discussion in the revised manuscript. The selection of May 2008 is justified by the following arguments: During May 2008 JFJ experienced some of the largest hourly PAN mixing ratios ever recorded at JFJ (Pandey Deolal et al., 2013) and also the monthly mean PAN was among the largest on record (see Fig. 2 – this is even more evident in Fig. 2 of the revised manuscript in which PAN data of JFJ of the years 2009 and 2010 are included). PAN at ZSF was comparable to other years. Hence, May 2008 was selected for a more detailed analysis as the variability at the sites can help identifying the potential origin of air masses and

C6902

meteorological processes involved.

Reviewer's comment: Can the analysis be quickly re-run for another year, and quickly compared to see if that also resulted in elevated PAN?

Reply: Unfortunately, the analysis cannot be easily compared with other years because the extension not only requires the calculation of the FLEXPART model (such data for other years would be available) but also requires the many other steps of the data analysis which is not a simple task and therefore out of the scope of the paper.

References:

Henne, S., M. Furger, and A. S. H. Prévôt, 2005: Climatology of mountain venting induced moisture layers in the lee of the Alps. *J. Appl. Meteorol.*, 44, 620-633. MeteoSwiss, 1985: Alpenwetterstatistik Witterungskalender, Beschreibung der einzelnen Parameter. Pandey Deolal, S., J. Staehelin, D. Brunner, J. Cui, M. Steinbacher, C. Zellweger, S. Henne, and M. K. Vollmer, 2013: Transport of PAN and NO<sub>y</sub> from different source regions to the Swiss high alpine site Jungfraujoch. *Atmos. Environ.*, 64, 103-115. Wanner, H., E. Salvisberg, R. Rickli, and M. Schuepp, 1998: 50 years of Alpine Weather Statistics (AWS). *Meteorol. Z.*, 7, 99-111.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/14/C6899/2014/acpd-14-C6899-2014-supplement.pdf>

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 12727, 2014.