Atmos. Chem. Phys. Discuss., 14, C4082–C4084, 2014
www.atmos-chem-phys-discuss.net/14/C4082/2014/

Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #3

Received and published: 24 June 2014

The manuscript presents a detailed analysis of meteorological conditions associated with PAN concentrations measured at two high Alpine research sites (Jungfraujoch, Switzerland and Zugspitze, Germany) during May of 2008. The FLEXPART Lagrangian Particle Dispersion Model is used to identify transport regimes and the PAN measurements are interpreted in light of the 4 or 5 dominant airmass transport pathways. The measurements, co-located with other chemical and meteorological data are of high value to the atmospheric chemistry community, though it seems they have already been published, and the major contribution of this paper is to extend their interpretation to a detailed analysis for one month. My concerns center mainly around

the interpretation of the findings, and if these can be addressed, the paper should be suitable for ACP.

General

The use of FLEXPART, which considers turbulent and convective processes, is an advance beyond the earlier work published by the authors which relied on back trajectory analysis. The time scale considered (10 days), however, may be far too short to enable tracing emissions that contribute to raising background PAN concentrations since the PAN lifetime can be at least a few times longer. While the authors find compelling evidence for a European boundary layer influence on the highest observed PAN levels at these sites, the caveat noted in the conclusions section, that a "spring-time increase in the hemispheric PAN background cannot be ruled out from the current analysis" is very important. This limitation of the chosen analysis methods should be emphasized earlier in the paper. For example, P12737-12738 states there is no significant intercontinental transport during this period, could be qualified with 'rapid' since transport longer than 10 days is neglected.

Additional discussion should be included regarding the extent to which PAN to CO ratios can be cleanly interpreted as described on P12743-44 in terms of giving differences in production chemistry. How valid is the assumption of a constant CO or PAN background? From Figure 2 of Fischer et al. 2014, there appears to be spatial and vertical variability in the hemispheric background (ACP, 2679-2698; the first author here is a co-author yet this paper isn't referenced here). Is there not a strong signature of transport history as well? For example, the interpretation seems to assume a fixed ratio in the lifetimes for CO and PAN, but are their lifetimes affected similarly in a warm, moist air mass?

To my eye, the clusters for Jungfraujoch and Zugspitze could better align in Figures 4 and 5 as: JFJ 1 with ZSF 2; JFJ 2 with ZSF 3; JFJ 3 with ZSF 4 and JFJ 4 with ZSF 5. Perhaps a simple pattern correlation would confirm that the initial aligning of

ACPD

14, C4082–C4084, 2014

clusters between JFJ and ZSF is actually strongest? Or that the synoptic conditions are consistent with different transport pathways aloft vs. within PBL? There seems to be a lot of overlap with ZSF 1 and 5. Some discussion as to what unique information is retained in each of these clusters(ZSF 1 versus 5), and why the transport patterns on given days seem different at the two sites would help to clarify.

Specific

P12746 L22 Is this one stratospheric influence event responsible for the correlation, and if so, perhaps best to exclude it?

Section 4.2.3 articulates a goal of using the footprint cluster analysis to examine contributions from the free troposphere. From Figure 5, it seems that there are strong influences from the middle free-troposphere in addition to the European PBL. On P12747, the free tropospheric origin is dismissed as not dominating but why couldn't there be mixing between the PBL and free tropospheric air masses, and thus a combination of transported (longer-lived) PAN and freshly formed PAN?

P12749 In Cluster 5, are there really not NOx sources to the South? Couldn't an equally likely interpretation be that temperatures are too warm in these air masses for PAN to be stable?

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