

## ***Interactive comment on “Decoupling peroxyacetyl nitrate from ozone in Chinese outflows observed at Gosan Climate Observatory” by Jihyun Han et al.***

**Anonymous Referee #1**

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A review of "Decoupling peroxyacetyl nitrate from ozone in Chinese outflows observed at Gosan Climate Observatory" by Han et al. submitted to ACP

General comments:

The paper is based on continuous observations of PAN and other air pollutants at Gosan Observatory on Jeju Island in Korea. First, the authors described time series of these species, analyzed diurnal cycles, and then focused on four characteristic episodes, followed by detailed correlative analysis of PAN to other species, in particular O<sub>3</sub> and/or PM<sub>10</sub> and PM<sub>2.5</sub> components. The authors found better correlation of PAN with PM<sub>10</sub> than with O<sub>3</sub> in pollution plumes transported from China, while the correlation with O<sub>3</sub> is also reasonably good. The authors suggested this happened in biomass

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burning plumes. Overall, the paper is well organized and written. This is an interesting data and new results, and I enjoyed reading the paper. This paper would be a nice piece of work contributing to the community. However, the paper needs some clarifications and a bit more details in the analysis. I have several comments and suggestions for the authors to put into the revised paper.

Major comments:

The authors highlighted PM<sub>10</sub>. I wonder why PM<sub>10</sub> not PM<sub>2.5</sub>, as I would expect the same or even better correlation with PM<sub>2.5</sub>. Is this just because the authors had the PM<sub>10</sub> mass concentration data, not PM<sub>2.5</sub> mass concentration data? If so, the explicit statement of PM<sub>10</sub> might be misleading, as the readers would think that PAN was specifically correlated with larger particles. Otherwise, the authors can simply mention aerosols without size information. The authors need clarifications on this point.

The authors finding of good correlation of PAN to aerosols is interesting and this is obvious from the data, but on the other hand the correlation of PAN to O<sub>3</sub> is also good. So, I think decoupling of PAN from O<sub>3</sub> is a bit too strong statement, and furthermore, PAN as a potential indicator of overall aerosol formation aged air masses impacted by biomass burning (so I guess smaller particles like PM<sub>2.5</sub>) also sounds too strong, with only the observed good correlative behaviors. The authors would need to put more analysis or interpretation on the underlying mechanisms to make this statement more robust.

Specific comments:

Abstract: PM<sub>10</sub> and PM<sub>2.5</sub> OC and EC... Does it mean PM<sub>10</sub> mass concentration, OC and EC in PM<sub>2.5</sub>? Please clarify.

In Introduction (Page 3) and Results (Page 6), the authors mentioned previous measurements. However, some important references are missing for both urban/suburban and remote sites in East Asia, and I would note below-referenced work made in Japan.

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As far as I read the papers, Tanimoto et al. JGR (1999) reported PAN measurements in Tokyo to be 0.6 ppb in November, and Tanimoto et al. JGR (2002) reported approx. 0.5 ppb in spring at Rishiri Island in northern Japan.

Page 6, Line 138-140: what about correlation to PM2.5?

Page 7, Line 147-149: PAN is not always coupled with O<sub>3</sub>. . . Again, the authors used the word “decoupling” and implied PAN and O<sub>3</sub> are not correlated at all, but in fact they are reasonably correlated and the PAN-aerosol correlation was just better than PAN-O<sub>3</sub>. Do you consider to rephrase the statement?

Page 8, Line 167-168: elevated concentrations of O<sub>3</sub> and/or PAN at night. This is a bit ambiguous. From Figure 3, I can see O<sub>3</sub> is elevated but PAN is not. Please clarify or justify.

In Sections 4.2 and 4.3, the authors discuss fast and slow transport, respectively. I would suggest to put some estimates on time-scales (how fast and how slow these transport episodes were). In Page 8, Line 181-185, the authors mentioned PAN and O<sub>3</sub> formation in the outflow during transport. However, Tanimoto et al. SOLA (2008) paper reported negligible O<sub>3</sub> enhancement in fast transported plumes from biomass burning in Siberia.

Page 12, Line 266–: The paragraph on CAM-chem can be moved to the Experimental Section.

All figures: I can see the x-axis is local time, but please clarify.

Figure 4: What about adding the same figure for PM10?

Figure 6: These figures are highlight of this paper. However, I see sub-figures d and f are somewhat duplicating and I would delete these.

References:

Tanimoto, H., J. Hirokawa, Y. Kajii, and H. Akimoto, A new measurement technique of

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peroxyacetyl nitrate at parts per trillion by volume levels: Gas chromatography/negative ion chemical ionization mass spectrometry, *J. Geophys. Res.*, 104(17), 21,343-21,354, 1999.

Tanimoto, H., H. Furutani, S. Kato, J. Matsumoto, Y. Makide, and H. Akimoto, Seasonal cycles of ozone and oxidized nitrogen species in northeast Asia, 1, Impact of regional climatology and photochemistry observed during RISOTTO 1999-2000, *J. Geophys. Res.*, 107(D24), 4747, doi:10.1029/2001JD001496, 2002.

Tanimoto, H., K. Matsumoto, and M. Uematsu, Ozone–CO correlations in Siberian wildfire plumes observed at Rishiri Island, *SOLA*, 4, 65-68, doi:10.2151/sola.2008-017, 2008.

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