

## ***Interactive comment on “Variations of surface ozone at leodo Ocean Research Station in the East China Sea and influence of Asian outflows” by J. Han et al.***

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Thank you for your constructive comments for the manuscript acp-2015-338. The response for each comment is given below and manuscript was revised accordingly.

General Comments This manuscript describes the ozone concentration levels at leodo Station, located in the middle of Yellow Sea/East China Sea region. Temporal variations including diurnal, seasonal, and interannual variations, and the dependencies on the air mass origins were discussed. A major conclusion, as expected, is that the Chinese emissions strongly influenced. The multi-year ozone concentration data shown

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here for the first time are regionally representative and thus are important for evaluation of regional/global model simulations. The contents are suitable for ACP and logically sound. However, in certain parts, especially the interpretation of the leveling-off trend after 2009, attributed simply to stagnation of NO<sub>x</sub> emission in China, needs more consideration. After some more clarification on the following specific points, the manuscript is recommended for publication.

Specific points: 1. It is meaningful to compare the annual or monthly averages with those at Cheju (33°18'N, 126°10'E, EANET data, [http://www.eanet.asia/jpn/docea\\_f.html](http://www.eanet.asia/jpn/docea_f.html)) and at Fukue Island (32.75N, 128.68E, Kanaya et al., AAQR 2015, Appendix), both of which are located in the Yellow Sea/East China Sea. Comparison to data at Oki Island (36°17'N, 133°11'E, EANET), experiencing more aged air, may also be important. The comparison must be more important than those shown in Figure 3, where data at more distant locations were only used. In relation, in line 26 of page 6751, the statement that the ozone concentrations decreased with increased distance from China may not be valid when including the nearby stations, given that the ozone production continues for several days and maximum concentration could occur at somewhat distant locations (e.g., Oki Island) with more aging.

We replaced Trinidad Head data with Gosan data for comparison (shown below). As Fukue data are available since 2009, they were not integrated into Figure 3 for direct comparison with other measurements. However, it is particularly interesting to see Fukue data with the primary peak in April and the second peak in October (Figure 2 in Kanaya et al., 2015). It is what we observed at IORS and was reported by Tanimoto et al. (2005). Therefore, these two studies were cited for explaining seasonal variation of IORS O<sub>3</sub>.

As you mentioned, the statement of O<sub>3</sub> concentrations decreasing with increasing distance from China is only valid when comparing remote sites in northeast Asia. Therefore, the statement was limited to "remote site" and the line 26 in page 6751 was mod-

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ified like, "In these remote sites, the level of averaged O<sub>3</sub> concentrations decreased with increased distance from China."

2. Abstract Page 16748, line 9. The concept of "fractional contribution" is difficult to understand. Can the authors simply state that different levels of ozone concentrations were found for six well distinguished air masses?

It was modified such as, "At IORS, six types of air masses were distinguished with different levels of O<sub>3</sub> concentrations by the cluster analysis of backward trajectories."

3. Page 16749, line 3. Photochemical loss of ozone (O<sub>1</sub>D + H<sub>2</sub>O etc) needs to be mentioned as well.

This part with the related (line 24 page 16748 ~ line 3 page 16749) was rewritten as follows.

Tropospheric O<sub>3</sub> is primarily transported from the stratosphere upon tropopause folding and produced by in situ photochemical reactions involving carbon monoxide (CO) and hydrocarbons in the presence of nitrogen oxides (NO<sub>x</sub>) (Brasseur et al., 1999). Ozone is also lost by photochemical reactions and deposition to the Earth's surface.

4. Page 16749, line 28. Which time is the reference for the increase by 5-7 ppb in spring 2006?

It was spring from 17 April to 15 May in 2006.

5. Page 16750, line 4. Change a semicolon to comma.

It was changed. 6. Page 16750, line 25. How good was the slope of the correlation?

It was 0.731. The IORS TEI gave higher values than those of lab TEI calibrated against primary standard. In addition, the average calibration coefficient of IORS TEI was  $1.01 \pm 0.13$  for the measurement period.

7. Page 16751, lines 12-13. Parrish et al. (2012) reported that the increase in ozone

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in Europe has slowed down or even decreases were found at some sites during 2000-2010. The authors should mention that such trend in the hemispheric baseline could also affect the trend at IORS.

The statement regarding O<sub>3</sub> hemispheric baseline was added to the text as follows.

The long-term trend of O<sub>3</sub> at IORS is consistent with recent findings of slowdown in the increase of O<sub>3</sub> concentrations observed in Japanese background stations at Mt. Happo and others (Parrish et al., 2012). This hemispheric baseline likely affects O<sub>3</sub> distributions at IORS.

8. Page 16751, line 22. Add latitude and longitude information for Trinidad Head, as the site is not included in Figure 1.

In the revision, Trinidad Head site was replaced with Gosan and discussion was changed/ added accordingly.

9. Page 16752, line 8. The stated ozone destruction is actually observed at Minamitorishima (Figure 3a). This should be mentioned here.

The relevant part of line 5-12 was rewritten as follows.

Among the five sites, O<sub>3</sub> concentration was reduced in the afternoon only at Minamitorishima, implying O<sub>3</sub> destruction (Fig. 3). Considering O<sub>3</sub> loss is generally observed under low NO<sub>x</sub> conditions in the remote marine boundary layer (MBL) (Ayers et al. 1996), these variations indicate that IORS including other remote sites in East Asia were influenced by continental outflows.

10. Page 16752, line 26. Better to rephrase "all measured species were divided into five seasons"

The relevant part of the line 25~28 was modified as follows.

To examine seasonal characteristics of O<sub>3</sub> distributions, all measured species were divided into five seasons: March–April, May–June (pre-monsoon period), July–August,

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September–November, and December–February.

11. Page 16753, line 14. Do the authors mean Mar-April by spring, specifically?

Yes. In the present study, seasons were divided into five, in which March-April was classified into spring and May-June into dry summer before summer monsoon. It is always difficult to group months into seasons, especially in the study region. In the revision, monthly characteristics were more clearly stated (Abstract, 3. Ozone variations, and Conclusion).

12. Page 16753, line 21. Levy II (in the reference list also)

Typo was corrected.

13. Page 16754, line 3. Five-day must be incorrect, as 40 h is mentioned later twice. Were the backward trajectories calculated for the whole period of the ozone observation?

It is a mistake. Thank you very much for correcting it. "Five-day" was erased.

14. Page 16754, line 23. What is the typical altitude of the trajectory for the six cases? Can the difference in altitude among the cases affect the ozone climatology analysis?

The altitude information of each trajectory was not given in the manuscript, for which average altitude is shown in the following figure. The altitude was higher for NW1 and NW2 clusters and lower for SE and SW marine air masses. It is what we commonly observed in northeast Asia.

As NW1 was the most frequent from late fall till spring, it could pick up some O<sub>3</sub> from the stratosphere. However, the NW1 used to pass through Beijing area and therefore, its contribution is likely to be insignificant.

15. Page 16755, line 16. NW1 was more frequent than N (page 16754, line 24).

It was meant to state that the occurrence of W and N clusters was steady through the

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year, which was different from the others being much dependent on season. Thus, the sentence was reworded such as, "These two trajectories were constantly observed through the year with relatively less seasonal variation at IORS (Fig. 9b)."

16. Page 16756, line 12. Interpretation of the cluster N as stagnant needs more explanation. From Figures 7 and 8, most of the trajectories passed over the Korean Peninsula. What is the influence from the emissions in the Peninsula? Why can the authors mention that "Chinese influence is implicit in N" in line 3, page 16757?

N is typically observed under anticyclone system, which used to develop in spring and fall and led to stagnant condition. In comparison with the average trajectory shown in Figure 7, the individual trajectories of N cluster are fairly well scattered over China and Japan as well as Korea. If they were coded by O<sub>3</sub> concentration, ozone concentration is evidently higher on trajectories from China (figure shown below). The stationary high accumulates emissions from nearby lands, contributing to O<sub>3</sub> buildup. As you recommended, more discussion was added to the revision as follows.

Page 16756 line 12~17

The cluster N was commonly observed before and after summer monsoon season, during which a stagnant condition often developed under the influence of migratory anticyclone systems. It used to park over the Yellow Sea, accumulating pollutants from nearby lands including China, Japan as well as Korea. In fact, the high concentrations of O<sub>3</sub> turned out to be associated with air trajectories from Chinese coastal regions. The model results of Zhao et al. (2009) also showed that the high concentration of O<sub>3</sub> can be expanded under a high pressure system in East Asia. The model results of Zhao et al. (2009) also showed that the high concentration of O<sub>3</sub> can be expanded under a high pressure system in East Asia.

Individual trajectories of N cluster coded by O<sub>3</sub> concentration, of which average trajectory is given in Figure 7.

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17. Page 16758, line 4. Actually Itahashi et al. (2014) showed that NO<sub>2</sub> over China again increased in 2010, while O<sub>3</sub> at IORS decreased in 2010. Some more careful statement is necessary here, and also in page 16751.

Ozone change is not straightforwardly related to increase or decrease in NO<sub>2</sub> concentration. Moreover, NO<sub>2</sub> concentrations seems to fluctuate since 2009.

In 2009, NO<sub>2</sub> concentration was decreased in spring as well as in winter. In 2010, however, NO<sub>2</sub> was considerably increased only during winter. Of the two O<sub>3</sub> peaks at IORS, the spring maximum tended to decrease, leading to decreasing trend from 2009. In addition, we found slight change in annual frequency of trajectories. The W, NW1, and N trajectories responsible for high O<sub>3</sub> concentration were decreased since 2009. In contrast, maritime air masses were more frequently observed since 2009. In conclusion (page 16758), these two factors were stated equally as plausible reasons for decrease in O<sub>3</sub> at IORS.

For the page 16751, the effect of hemispheric baseline was stated before discussing NO<sub>x</sub>, as recommended above (comment 7).

The long-term trend of O<sub>3</sub> at IORS is consistent with recent findings of slowdown in the increase of O<sub>3</sub> concentrations observed in Japanese background stations at Mt. Happo and others (Parrish et al., 2012). This hemispheric baseline likely affects O<sub>3</sub> distributions at IORS.

18. Figure 8. Is the unit ppb?

Yes, it is. The unit was given in the figure caption.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/15/C8171/2015/acpd-15-C8171-2015-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 16747, 2015.

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