Correspondence to Referee #1

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.

1. Abstract: The average ozone concentrations for all the period is mentioned, but this does not mean much, since this average is a result of a variety of factors. Throughout the paper, ozone concentrations are noted with the precision of 0.1 ppb, but this is not meaningful. I would suggest just 52 ppb instead of 51.8 ppb, for example.

The average for the entire period was removed from abstract. Also, ozone concentrations were given with two significant digits.

2. Figure 1: The words “Yellow Sea” and “East Sea” are superimposed in the map. However, I find no need to write these names with a scientific importance, and suggest removing from the figure.

Figure 1 was remade to show five measurement sites.

3. Section 3: Diurnal and seasonal variations are discussed with Figures 3 and 4. There are some errors and unclear phrases.
Specifically: Page 16752, Line 24: "monthly" variations should be "seasonal" variations

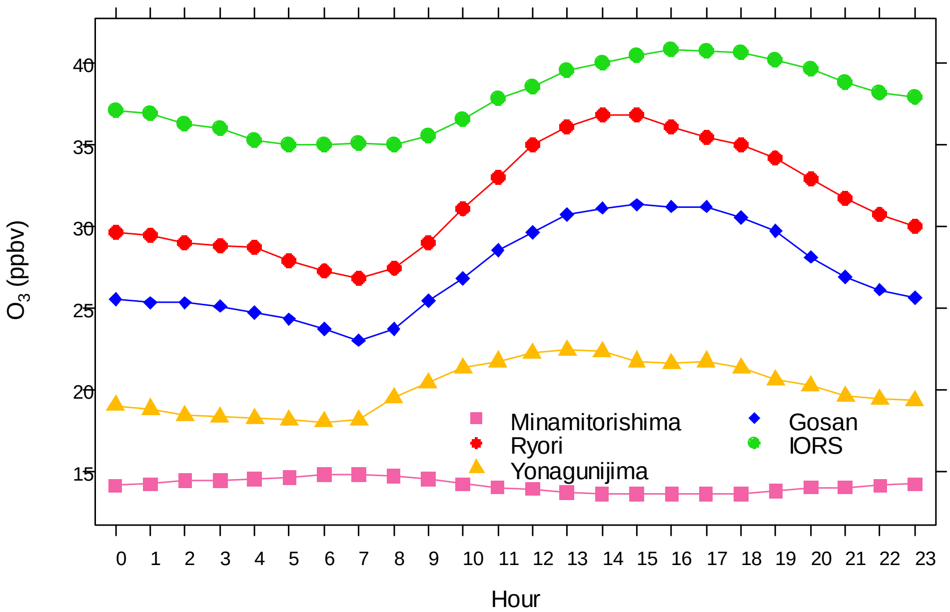
It is probably the page 16751 line 24. In Figure 3b, monthly means are presented for the five sites but they eventually represent seasonal variations. Thus, “monthly” was changed to “seasonal” as you recommended.

4. Page 16752, Line 9-10: Based on diurnal variations, the authors mention that ozone at IORS is influenced by Chinese outflow. This sentence is not logically sound, since diurnal variability is, in most cases, driven by local effects – emissions, chemistry, or meteorology. I would expect discussion of local effects. Also, the diurnal cycles (Figure 3a) are plotted for all the seasons. The appearance and magnitude of diurnal cycles depend on seasons - usually greater in summer than in winter. I would suggest showing seasonal cycles at IORS in four seasons first, and then compare (probably) summertime one with those at other sites.

Of course, the diurnal variation of ozone is driven by in-situ photochemistry. In remote regions with low NOx concentration, therefore, ozone is normally destroyed as what was observed in Minamitorishima. In IORS, however, ozone was found to be increased during the day through the year.

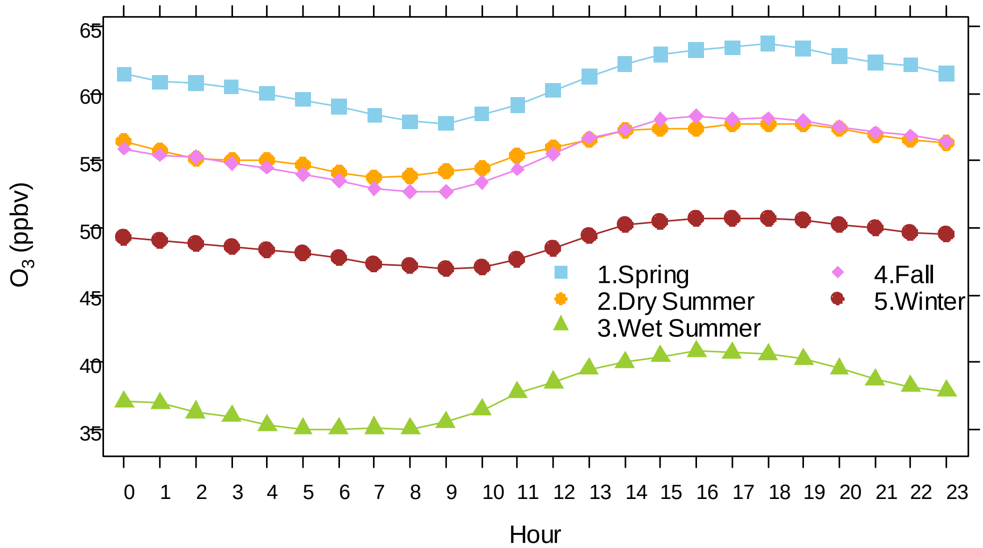
If comparing diurnal variations among seasons (Fig. 3a), the daytime buildup of ozone (the difference between the max. and min.) varied little: 6 ppbv in spring (Mar-Apr), 4 ppbv in dry summer (May-Jun), 6 ppbv in wet summer (Jul-Aug), 6 ppbv in fall (Sep-Nov), and 4 ppbv in winter (Dec-Feb). In contrast, the background concentrations were much different in seasons with being the highest in spring and the lowest in wet summer. The seasonal average was 61 ppbv in spring, 56 ppbv in dry summer, 38 ppbv in wet summer, 56 ppbv in fall, and 49 ppbv in winter. These results imply that ozone concentrations were greatly dependent on air masses, reflecting the degree of impact by continental outflows, which determined the background level of ozone..

At IORS, the daily buildup relative to the mean was 16 % in wet summer and was less than 10 % in the rest seasons. It was the highest during July-August, indicating the local effect as you said. This summertime buildup of 6 ppbv was higher than that of Yonagunijima (3 ppbv) and lower than that of Gosan (8 ppbv) and Ryori (10 ppbv), which also reveals the proximity to the land. The following figure shows the diurnal variations of O_3 at five sites only during wet summer (July~August).



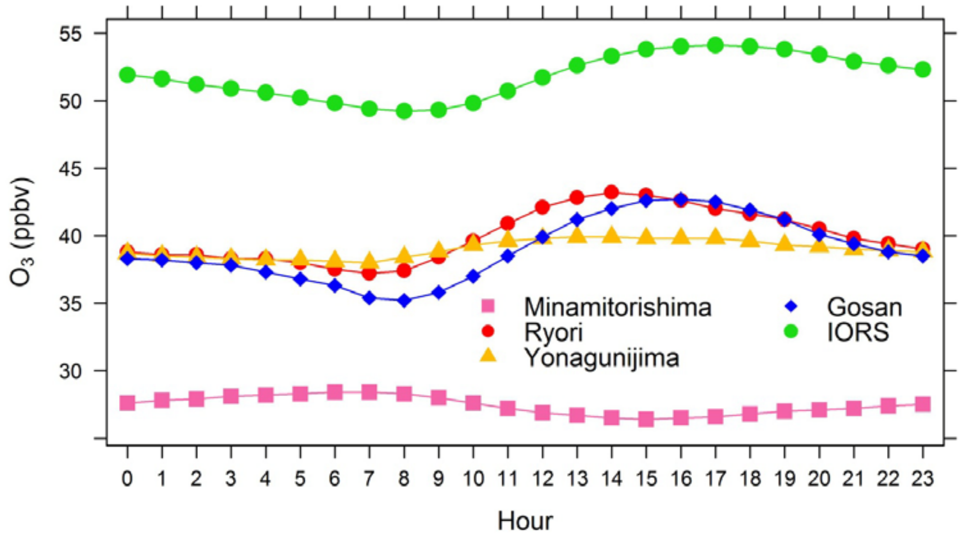
In the revised manuscript, Gosan data were replaced with those of Trinidad Head. Accordingly, text and Figure 3 were revised.

a)



O3 (ppbv)
65
60
55
50
45
40
35
1.Spring
2.Dry Summer
3.Wet Summer
4.Fall
5.Winter
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
Hour

b)



O3 (ppbv)
55
50
45
40
35
30
Minamitorishima
Ryori
Yonagunijima
Gosan
IORS
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23
Hour

c)

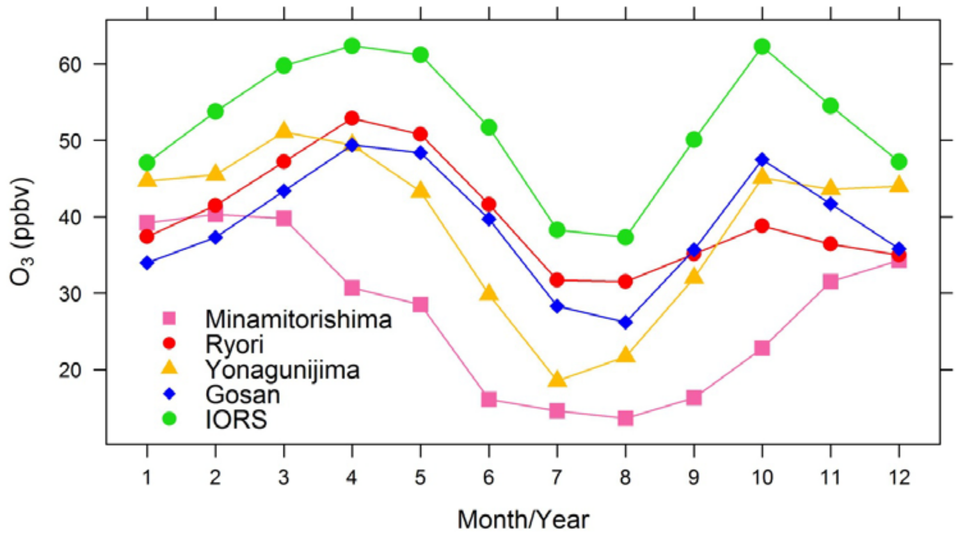


Figure 3. Comparison of diurnal and seasonal variations of O_3 concentrations at remote sites in the Northwest Pacific region including IORS, Gosan, Yonagunijima, Ryori, and Minamitorishima. All data were averaged for 8 years (2003–2010) and seasons were divide into spring (May-April), dry summer (May- June), wet summer (July-August), fall (September-November), and winter (December-February). a) diurnal variations of O_3 at IORS in different seasons, b) diurnal variations of O_3 at five sites, and b) monthly variations of O_3 at five sites.

5. Figure 3: The seasonal maximum is actually in autumn, not spring. Correct? There are both spring (higher) and autumn (lower) peaks observed at EANET stations, so this is consistent. A unique phenomenon is that the autumn peak is higher than the spring one here at IORS. Also, I would say that the spring peak is not April but "April-May". Latitudinal differences in the timing of the spring peak in this region is found in Tanimoto et al., GRL, 2005, so please look at it and add a bit more discussion.

At IORS, the monthly mean was the highest in April and October (62 ppbv), followed by May (61 ppbv). There is just 1 ppbv difference between April and May. Gosan showed similar tendency in their monthly distribution with the highest in April (49 ppbv), May (48 ppbv), and October (47 ppbv). In Figure 3c, the second peak is evident for these two sites. Tanimoto et al. (2005) found the second peak of O3 in October, which was more evident in their CTM (chemical transport model) results. As you mentioned, the data from IORS and Gosan may be good compliments for the EANET dataset, with which the characteristics of O3 in the northeast Asia would be better understood.

The relevant discussion (Page 16752 lines 13-20) was rewritten as follows.

At IORS, the monthly averaged O_3 concentrations were the highest in April and October (62 ppbv) and lowest in August (37 ppbv) (Fig. 3c). The O_3 concentrations remained high during March ~ May, resulting in a broad spring peak which was in contrast to a sharp fall peak. This is in accordance with a typical pattern that has been observed in other remote sites over Northeast Asia during the past decades (Chan et al., 2002; Jaffe et al., 1996; Kanaya et al., 2015; Kondo et al., 2008; Oltmans and Levy II, 1994; Tanimoto et al., 2005; Tanimoto et al., 2009; Watanabe et al., 2005; Weiss-Penzias et al., 2004). In particular, the second peak of O_3 was the most noticeable at IORS along with Gosan in October, of which tendency was observed in previous studies (Kanaya et al., 2015; Tanimoto et al. 2005).

Abstract was also rephrased, stating specific month, for which O3 was the lowest and highest.

6. Figure 3 again: I am not comfortable to see the plots of Trinidad Head, in particular for the diurnal cycles, since we can expect no link to IORS and other East Asian sites.
7. Figure 3 again: I think ozone data is available at Gosan site on Jeju Island. Can you compare IORS and Gosan?

For 6 and 7, the measurements of Trinidad Head were replaced with those of Gosan and the discussion was revised accordingly.

8. Page 16752, Line 23: Do you mean ozone is removed by rain? The solubility of ozone is not high, so my understanding is that ozone itself is not effectively removed by rain. Can you please clarify or elaborate more?

This part (Line 21-25) was rewritten for clarification as follows.

In summer, the study region is under influence of Asian monsoon system which brings moist air from the Pacific Ocean. Meteorological parameters including relative humidity, wind speed, and visibility indicate a clear shift in air mass from pre-monsoon to monsoon season (Fig. 4b). At IORS, O3 concentration was noticeably decreased during summer, even though temperature was high. Likewise, the O3 level of Gosan was reduced down to the minimum in summer, when the levels of precursors were the lowest with heavy rainfall.

9. Page 16754, Line 3-12: This paragraph should be moved to 2. Methodology section. "w.e.re" must be a typo.

The background information on trajectory analysis was moved to the end of the methodology section. Typo was corrected.

Correspondence to Referee #2

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.

1 Overall Comments

1. While the analysis of air mass trajectories was appropriate and interesting, I was disappointed to see very little mention of any other meteorological factors, despite their availability. Were the influences of the other meteorological observations taken at the IORS station (listed in the methodology section) examined along with wind speeds? What about weather conditions at other stations upwind? As it stands, only the effects of precipitation are directly described at all, and then only in a qualitative fashion. Whether or not other meteorological factors prove to be important here, I would expect at the very least some more substantial discussion of their significance (or lack thereof).

2. The figures in general show varying degrees of polish and clarity, and some of them are in need of attention. Unifying colors between figures, matching colorbar scales, and lining up axes within multipanel figures would go a long way towards improving their overall effectiveness (see specifics below).

2 Specific Comments

1. Abstract, page 16748, line 15: The phrase "of which extent was apt to be changed by" is awkward. I suggest "the extent of which was affected by".

It was modified like "of which extent was dependent on meteorological state".

2. Introduction, page 16749, lines 2-3: This sentence implies that deposition is the dominant sink of tropospheric ozone, which I do not believe is the case. Please support and clarify this statement.

This part is to give a general view of tropospheric O3, which is primarily transported from the stratosphere, recycled/produced through photochemical reactions, and some of it is deposited to the earth surface. The relevant part was rewritten as follows.

Tropospheric O3 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 (NOx) (Brasseur et al., 1999). Ozone is also lost by photochemical reactions and deposition to the Earth's surface.

3. Section 3, page 16752, line 9: I believe "IORS including other remote sites" should read "IORS and other remote sites".

It was corrected.

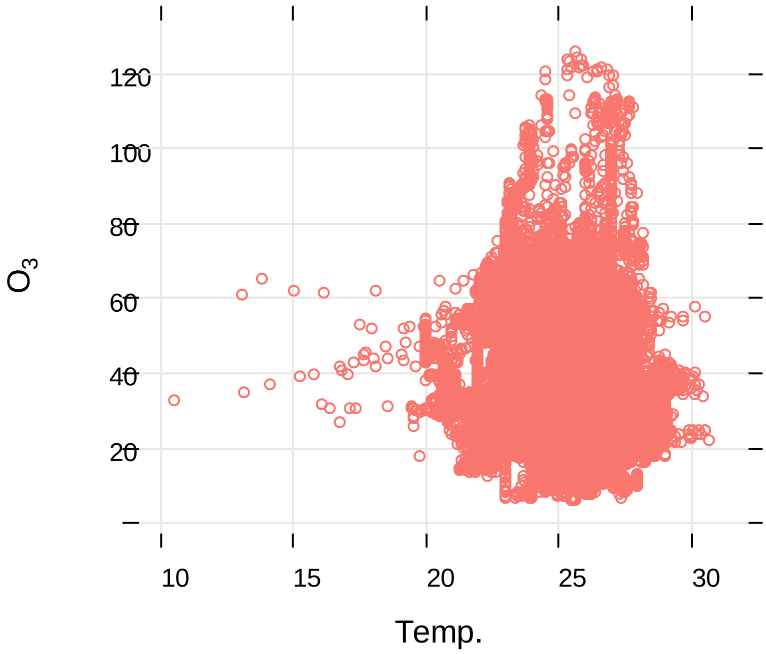
4. Section 3, page 16752, lines 13-20: There seems to be some redundancy here, with two pairs of sentences essentially saying the same thing: "highest O3 concentrations were mostly observed in spring with an apparent minimum in summer" and "this is a typical pattern" vs. "monthly averaged O3 concentrations were the highest in April and October and lowest in July" and "This accords with what has been observed". Consolidate or further differentiate the repeated statements.

This paragraph gets shortened as follows.

At IORS, the monthly averaged O_3 concentrations were the highest in April and October (62 ppbv) and lowest in August (37 ppbv) (Fig. 3c). The O_3 concentrations remained high during March ~ May, resulting in a broad spring peak which was in contrast to a sharp fall peak. This is in accordance with a typical pattern that has been observed in other remote sites over Northeast Asia during the past decades (Chan et al., 2002; Jaffe et al., 1996; Kanaya et al., 2015; Kondo et al., 2008; Oltmans and Levy II, 1994; Tanimoto et al., 2005; Tanimoto et al., 2009; Watanabe et al., 2005; Weiss-Penzias et al., 2004).

5. Section 3, page 16752, lines 22-23: Here there is a hint of temperature dependence, but no explanation of what the data show or whether the temperature/ozone relationship matches up with expectations. Is there a positive temperature correlation during clear days?

The lines 21-25 and relevant part was rewritten as follows.
In summer, the study region is under influence of Asian monsoon system which brings moist air from the Pacific Ocean. Meteorological parameters including relative humidity, wind speed, and visibility indicate a clear shift in air mass from pre-monsoon to monsoon season (Fig. 4b). At IORS, O3 concentration was noticeably decreased during summer, even though temperature was high. Likewise, the O3 level of Gosan was reduced down to the minimum in summer, when the levels of precursors were the lowest with heavy rainfall.
We have rainfall data only for the first couple of years and no criteria to tell if it was clear or not. The following figure presents all O3 measurements against temperature in July, where no correlation was found between the two.



6. Section 4.1, page 16754, line 3: Fix typo ("1500ma.s.l.w.e.re calculated").

This part (the first paragraph) was move to "2. Methodology" section. Typo was corrected.

7. Section 4.3, page 16756, lines 20-21: Please clarify the sentence "It is not certain for a higher and lower frequency of NW2 and W in 2004 than in other years." I am not sure what it means.

It was to state a big change in the frequency of the cluster NW2 and W from 2004 to 2005. Because it doesn't deliver any meaning, it was removed in the revised manuscript.

8. Figure 4b: I think including (essentially) 4 y-axes is a bit much. These would be much clearer separated out into 4 vertically-stacked panels.

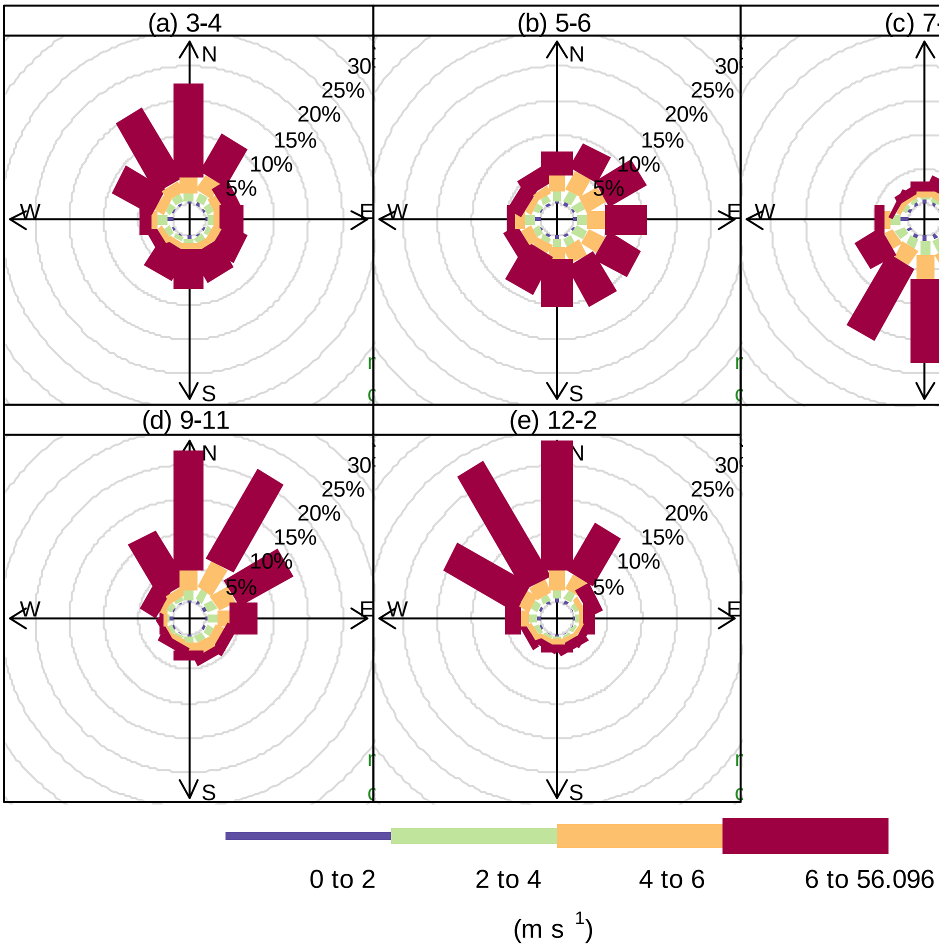
Figure 4b was modified.

9. Figure 5a-e: Colorbars are all of varying scale, reducing the effectiveness of seasonal comparison. I recommend unifying the scales under a single colorbar and (if possible) getting rid of any tiny, extraneous text that is cluttering up individual panels.

Figure 5a-e were remade with the same color scale.

10. Figure 5f: The meaning of the stacked bar plots is difficult to interpret as presented. I recommend turning them into a set of simple polar plots, such as those produced by the windRose function of the openair package.

Figure 5f was to provide a seasonal characteristic of winds in a simple way. Considering the importance of winds in this study, it was replaced with windrose plot shown below.



11. Figure 6: I was distracted by the unaligned axes in the panels of this figure. Clean up placement.

Figure 6 was remade, in which all plots have the same scale.

12. Figures 9 and 10: Unifying the color scheme used here with that of Figure 7 would greatly improve the clarity of all three, making it easier to flip back and forth between them.

Colors and symbols are all modified so that they represent the same trajectory in Figure 7, 9, and 10.

Correspondence to Referee #3

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

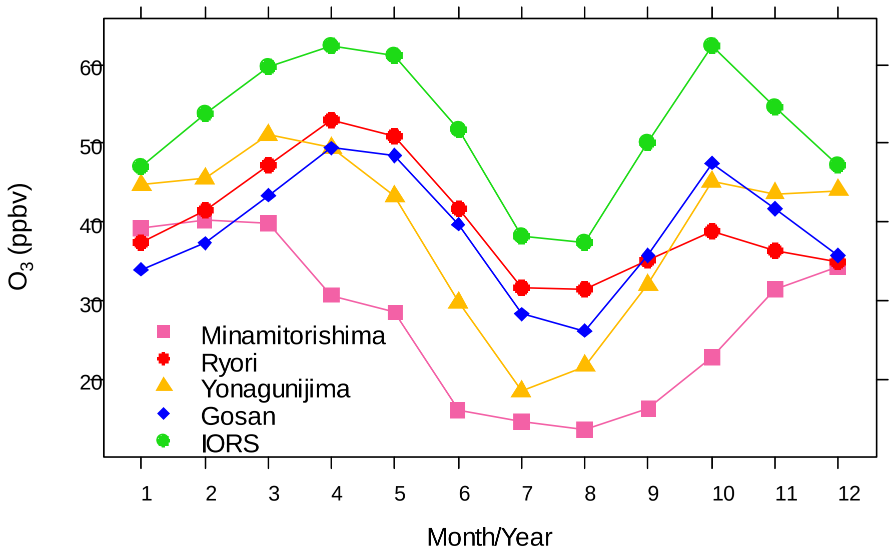
This manuscript describes the ozone concentration levels at Ieodo Station, located in the middle of Yellow Sea/East China Sea region. Temporal variations including diurnal, seasonal, and interannual variations, and the dependencies on the air mass origins were discussed. A major conclusion, as expected, is that the Chinese emissions strongly influenced. The multi-year ozone concentration data shown 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 NOx 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) 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 O3.

As you mentioned, the statement of O3 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 modified like, "In these remote sites, the level of averaged O3 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 O3 concentrations by the cluster analysis of backward trajectories."

3. Page 16749, line 3. Photochemical loss of ozone (O1D + H2O 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 O3 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 (NOx) (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 ± 0.13 for the measurement period.

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

The statement regarding O3 hemispheric baseline was added to the text as follows.

The long-term trend of O3 at IORS is consistent with recent findings of slowdown in the increase of O3 concentrations observed in Japanese background stations at Mt. Happo and others (Parrish et al., 2012). This hemispheric baseline likely affects O3 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, O3 concentration was reduced in the afternoon only at Minamitorishima, implying O3 destruction (Fig. 3). Considering O3 loss is generally observed under low NOx 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 O3 distributions, all measured species were divided into five seasons: March–April, May–June (pre-monsoon period), July–August, 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.

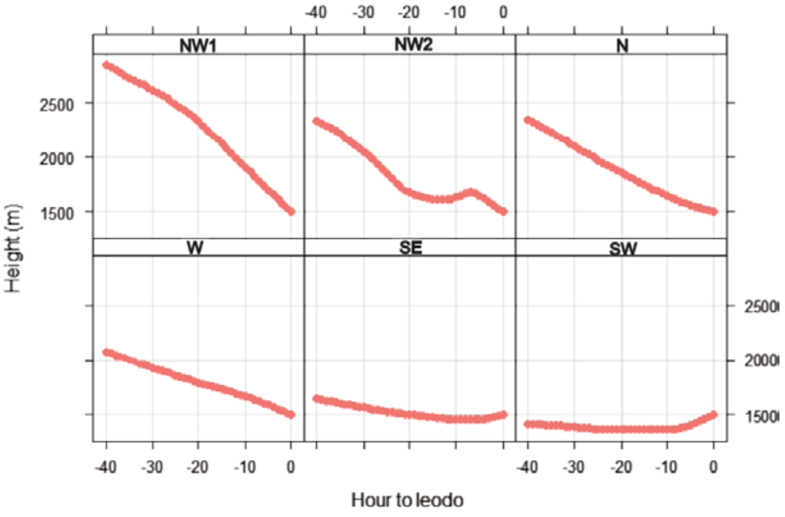
13. Page 16754, line 3. Five-day must be incorrect, as 40 h is mentioned later twice. Were the backward trajectories calculated for the whole period of the ozone observation?

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 O3 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).

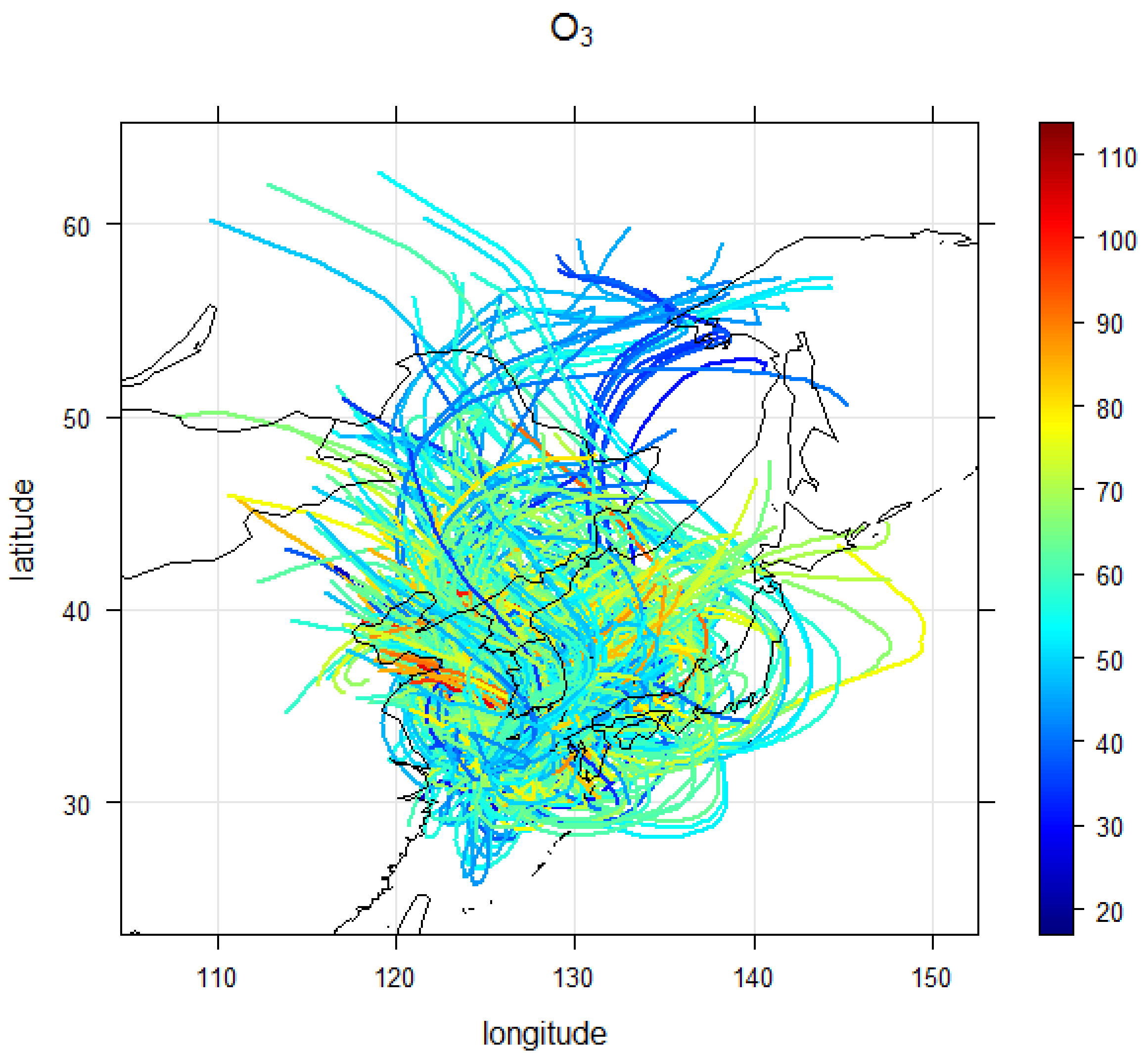
It was meant to state that the occurrence of W and N clusters was steady through the year, which was different from the others being much dependent on season. Thus, the sentence was reworded such as, "These two trajectories were constantly observed through the year with relatively less seasonal variation 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 scatted over China and Japan as well as Korea. If they were coded by O3 concentration, ozone concentration is evidently higher on trajectories from China (figure shown below). The stationary high accumulates emissions from nearby lands, contributing to O3 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 O3 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 O3 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 O3 can be expanded under a high pressure system in East Asia.



Individual trajectories of N cluster coded by O3 concentration, of which average trajectory is given in Figure 7.

17. Page 16758, line 4. Actually Itahashi et al. (2014) showed that NO2 over China again increased in 2010, while O3 at Ieodo 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 NO2 concentration. Moreover, NO2 concentrations seems to fluctuate since 2009.

In 2009, NO2 concentration was decreased in spring as well as in winter. In 2010, however, NO2 was considerably increased only during winter. Of the two O3 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 O3 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 O3 at IORS.

For the page 16751, the effect of hemispheric baseline was stated before discussing NOx, as recommended above (comment 7).

The long-term trend of O3 at IORS is consistent with recent findings of slowdown in the increase of O3 concentrations observed in Japanese background stations at Mt. Happo and others (Parrish et al., 2012). This hemispheric baseline likely affects O3 distributions at IORS.

18. Figure 8. Is the unit ppb?

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