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Title: Lidar detection of high concentrations of ozone and aerosol transported from Northeast Asia over Saga, Japan

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Iteration: Minor Revision

Reply to Co-Editor comments and Anonymous Referee #1 comments

Co-Editor Decision: Reconsider after minor revisions (Editor review) (01 Dec 2016) by Yugo Kanaya

Comments to the Author:

Dear Authors,

Thank you for submitting your revised manuscript. Re-evaluation was made by our two reviewers. One suggested accepted as is, and the other requested minor revision. I would appreciate you could further respond to the comments by the reviewer #2. Also, I hope if you could take into account the handling editor's comments below:

The authors wish to thank the Co-Editor and the reviewer #2 for helpful and thoughtful comments. Each comment is addressed individually below. The reference comments are written in black, and our responses are described in red.

1. Although the focus of the manuscript is successfully shifted to the aspect of transport analysis, the Introduction part remained focused on the technical aspects and the authors' works. It is better to add some more general description on O₃ and aerosols pollutions reaching this region in spring from the continent, citing literature (including those not using lidars), touching on what was studied before and what is yet to be analyzed (e.g., three dimensional features of ozone). This will highlight the value of this manuscript.

As suggested, we added the next sentences and 5 references:

Lines 45-50 and line 77 in the revised paper:

The aerosols transported from the East Asia to the western Japan were observed by lidar and their vertical distributions were reported (Iwasaka et al., 1988; Murayama et al., 2001; Hara et al., 2009). On the other hand the ozone pollutions from the Asia were mainly studied by the surface measurements (Akimoto et al., 1996; Yamaji et al., 2006). Continuous ozone vertical distributions by ozone DIAL are very useful for studying the transport process and the origin.

In this paper we report an event during which high concentrations of ozone and aerosols were observed almost simultaneously below an altitude of 1.5 km over Saga on 22 March 2015 by Mie lidar and ozone DIAL, which substantially impacted surface air quality. We also compared the observational results with those simulated by the models.

2. lines 150-151. Is an old-type ozone monitor based on wet chemistry is still under use at this site, where the PAN interference matters?

Ozone monitor is a UV photometer, and we revised as follows:

Lines 159-161:

Because the surface Ox was observed by an UV photometer, the contribution of other components such as peroxyacetylene nitrate (PAN) to oxidant concentrations was extremely low, and the oxidant volume mixing ratio was considered to be that of ozone.

3. line 181. Add base year of REAS2.1 emission inventory used for modeling.

We added “the REAS 2.1 emission inventory in 2007” in line 188.

4. lines 186 and 194. How is the 20% of systematic error related to the uncertainty of 12% in line 194?

We estimated as follows:

$$\text{uncertainty}^2 = (\text{systematic error})^2 + (\text{statistical error})^2$$

$$\text{the uncertainty: } (0.2^2 + 0.1^2)^{1/2} = 0.22$$

$$\text{the mean value of the uncertainty: } (0.07^2 + 0.1^2)^{1/2} = 0.12$$

5. lines 266-267. The skyradiometer could have different sight than the lidar. This is also a possible reason for the difference.

We added “The sky radiometer could have different sight than the Mie lidar. This might be also a possible reason for the difference.” in lines 276-277.

6. line 296. When the contribution from dust particles is 100%, those from others need to be 0%.

We replaced “(60–100%)”, “(40–60%)”, and “(30–40%)” in lines 306-307 with “(about 60–100%)”, “(about 40–60%)”, and “(about 30–40%)”, respectively, and also corrected line 310 in the same manner.

7. Figure 8(b). There are several regions with increased dust extinction coefficient. Where do the authors think are the source regions of dust? The maximum during 1800 20 March - 0000 21 March is best connected to the increase over Saga; however, the region is over the ocean or the Shandong Peninsula and thus is not likely the source region.

Meteorological fields (e.g., wind velocities and pressure levels) used in the simulation of dust extinction coefficient (i.e., MASINGAR mk-2) are not identical to those used for the backward trajectory (i.e., HYSPLIT). This could explain that there are several regions where dust extinction coefficients are increased. Please find the next figure of dust emission simulated by MASINGAR mk-2 on 19 March (in UTC).

Based on this figure, we added the following sentences in lines 323-325:

The MASINGAR mk-2 simulation suggested that the dust particles emitted during 18:00–24:00 UTC on 19 March around 40°N, 105°E were responsible for the dust storm captured by the Mie lidar observation.

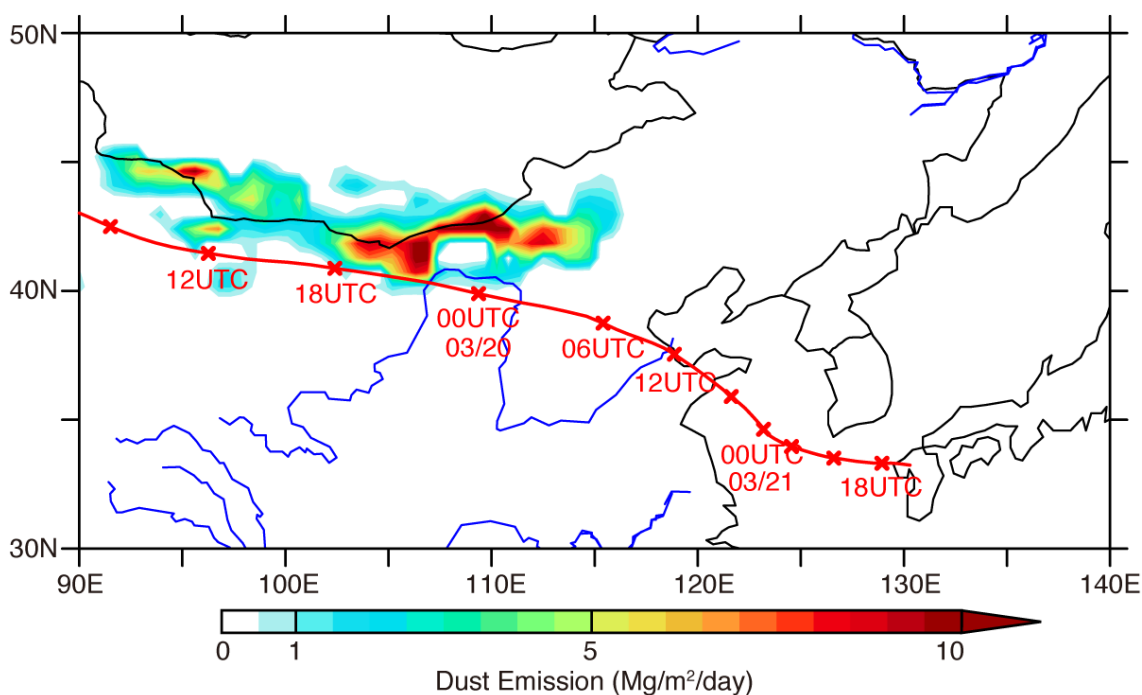


Figure S1, Daily averaged dust emission on 19 March. Red line shows the HYSPLIT-model backward trajectory from Saga at 1500 m above ground level (AGL) ending at 06:00JST on 22 May 2015 (identical to the backward trajectory shown in Figure 8).

Submitted on 30 Nov 2016

Anonymous Referee #2

In the revised version of the manuscript, a good effort has been done to improve the quality of the paper. Most of the corrections suggested were properly addressed. Before the acceptance of the paper, I recommend to address the following two minor points:

1) The altitudes detected from the lidar measurements (shown in Fig. 11) only in some cases correspond to the atmospheric boundary layer top. For numerous profiles (e.g. from 1200 JST of 20 March to 12 00 JST of 21 March), the method detects altitudes above 2 km that are not realistic boundary layer heights for this season. The algorithm applied to lidar measurements detects vertical gradients of aerosol backscattering, thus the top of aerosol layers that may not be the boundary layer top. A physical identification of the boundary layer top is done by recognizing the temperature inversion from radiosoundings as already

shown in the figure. My recommendation is to screen out all heights detected from lidar data that do not correspond to the boundary layer height shown by the radiosoundings. Also, the difference between residual layer and mixing layer does not seem to be clear in the current paper. Radiosounding can tell the location of both as below the mixing layer the profile of potential temperature is neutral above an unstable layer near the surface and the residual layer corresponds to a neutral profile above a stable layer.

As suggested, Mie lidar had a tendency to detect not realistic mixed layer heights in the nighttime. Therefore we estimated the top altitudes of the mixed layers from two hours after sunrise to two hours before sunset, and revised in lines 342-344 as follows:

“we show in Fig. 11 the time variations of the top altitudes of the ~~mixed atmospheric boundary~~ layers from two hours after sunrise to two hours before sunset during ~~from~~ 11:10 JST on 20 March ~~through~~ 14:33 JST on 31 March 2015”, and we replaced the old Fig.11 with the revised Fig.11.

And we deleted “However, Mie lidar had a tendency to detect the residual layers in the night and morning time, e.g., 21–22 March.” in lines 349-350 and “ above the residual layer” in line 353.

2) The lidar ratio for both dust and pollution aerosols is the same (50 sr). This is an approximation as shown by the authors themselves: dust seem to have lower lidar ratios than pollution aerosol. I recommend to include in the text of the manuscript the values given in the answers to the reviewers : “Their summaries are as follows: Sakai et al.(2003): Asian dust 47 ± 18 sr, Cattrall et al.(2005): Dust (spheroids) 42 ± 4 sr, SE Asia Pollution 58 ± 10 sr, Anderson et al.(2003): ACE-Asia Pollution (Fine-dominated, submicron portion) 50 ± 5 sr, Dust (Coarse-dominated, Dust-like chemistry, Supermicron portion) 46 ± 8 sr.” and then indicate that as a simplification, they use the same value for both species.

As suggested, the next sentences are added in lines 102-106:

Their summaries are as follows: Sakai et al. (2003): Asian dust 47 ± 18 sr, Cattrall et al. (2005): dust (spheroids) 42 ± 4 sr, South East Asia pollution 58 ± 10 sr, Anderson et al. (2003): ACE-Asia pollution (fine-dominated, submicron portion) 50 ± 5 sr, dust (coarse-dominated, dust-like chemistry, supermicron portion) 46 ± 8 sr. As a simplification, we used the same value for both species.