

Interactive comment on “Long-term change in the contributions of various source regions to surface ozone over Japan” by Tatsuya Nagashima et al.

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Received and published: 5 May 2017

Response to the comment of Referee #1 The authors greatly appreciate your critical reading of our manuscript and highly valuable suggestions and comments. Our responses to your comments are listed below. (Pages and lines are those in the track-changed manuscript)

(RC): Referee Comment / (AR): Author Response

Major comments:

(RC) The paper is thoroughly written, but reads a bit too descriptive and too technically detailed. I would encourage the authors to make a bit more effort in reorganizing the sentences and try to put focus on the major scientific findings in this work.

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(AR) Thank you for the overall comment. I read through the manuscript and checked the points which seems too descriptive and technically detailed, and deleted or modified them if I could. The list of changes in the manuscript is below.

- P2, L39
- P4, L138-140 / L145-147
- P5, L180 / L183-184 / L204 / 206
- P6, L250 / L256-257
- P7, L304 / L306-307
- P8, L322-323 / L326-328 / L360-361 / L364-365
- P9, L366 / L378-379 / L407
- P10, L447-449 / L457
- P11, L458 / L466-468 / L490-491
- P12, L511

(RC) Why up to 2005? and why surface ozone? Needs a bit of more sentences to justify these points.

(AR1) The model simulation should include the period starting from 1980s and covering after the year 2000 to a certain extent to cover the years reported to have increasing trend in the surface O₃ over Japan. Therefore, I selected the REAS v1.2 inventory because it was the only inventory data at the time of model calculation covering from 1980 to 2005 and focused on the whole East and Southeast Asian regions. The simulation period covering up to 2005 is mainly due to the temporal coverage of the emission inventory data used (REASv1.2). However, I'm strongly sure that I could obtain the basic understanding about the role of various source regions on the recent reported trend in surface O₃ over Japan even with the simulation up to 2005 in this study, although

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I know the anthropogenic emission of air pollutants in Asian region has been varied continuously even after 2005 and examining its impact on the air quality in Japan is also important. I added the following sentence to justify the reason more clearly (P4, L159-162).

"The end of simulation period (2005) was determined mainly due to the temporal coverage of the Asian emission data described below, however, this period sufficiently covered the years reported to have increasing trend in surface O₃ over Japan in the previous literatures."

(AR2) The long-term increasing trend of the surface O₃ in Japan during the last about 30 years despite of the continuous efforts to reduce the emission of O₃ precursors in Japan and the consequent high violation rate of national ambient air quality standard (AAQS) in Japan, almost all the ambient air monitoring sites has been failed to meet the AAQS for a long time, are the persistent issues for environmental administration in Japan, therefore I focused on the surface O₃. I added and modified the sentences stating the reason more clearly as follows (P2, L84 – P3, L92).

"In Japan, analysis of long-term observations by the ambient air quality monitoring network . . . until the present (Ohara and Sakata, 2003; . . . Akimoto et al., 2015). And the consequent high violation rate of national ambient air quality standard (AAQS) for surface O₃ (hourly mean concentration of 60 ppbv) has been the persistent issue in environmental administration for a long time, therefore, there is an urgent need to study the reason for the increasing trend and examine the countermeasures. One clue is that the simultaneous observations of O₃ precursors such as . . . inconsistent with the increasing trend of O₃ over Japan."

Specific comments:

(RC) Title: I would prefer to "Long-term changes in the source contribution to surface ozone in Japan", just for your consideration.

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(AR) Thank you for the suggestion. I also like the simple one. I change the title as you suggested.

(RC) Introduction, L49 - : The authors started mentioning the trends in Taiwan, China, and South Korea, but historically the trends over Japan were studied with ozone sondes or surface measurements prior to these areas. Hence, I would suggest the authors to start with Japanese trends then followed by recent reports in other countries.

(AR) Thank you for the suggestion. I changed the manuscript accordingly as follows.
(P2, L56-83)

"Japan experienced a rapid industrialization ahead of other Asian countries, and an increasing trend has been found in various observations of tropospheric O₃ ... until the mid-2000s (Tanimoto, 2009; Tanimoto et al., 2009; Parrish et al., 2012). During the recent decades, an increasing trend in tropospheric O₃ has also been observed at ... in tropospheric O₃ for other regions in the world (Cooper et al., 2014).

(RC) P8, L318, section 3.3: The authors phrased "IAVs" in several places in the paper. The authors use the term "IAVs" not to mention (single) year-to-(single) year variability but rather decadal changes or changes during several years and the next several years (Explanation of Figure 3, for example). In Figure 3 the observed IAVs are not necessarily correlated with the modeled IAVs, on (single) year-by-year basis. So, I would encourage the authors to come back to this point and rephrase where necessary.

(AR) I carefully checked all the "IAVs" and rephrased some of them which do not mean year-by-year variation but rather longer (e.g. decadal) temporal variation to "long-term variation" or "temporal variation". The following is the list of changes.

-P8, L334: "IVAs -> long-term variation"

-P11, L494 (title of the section 3.3): "IVAs -> temporal variations"

-P12, L513: "increase or the IAV -> temporal variation"

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-P12, L514: "IVA -> temporal variation"

-P12, L542: "IVAs -> temporal variations"

-P13, L572: "IVA -> temporal variation"

-P14, L627 / L631: "IVA -> temporal variation"

-Table 2: "IAV -> Var", "InterAnnual Variation (IAV) -> Temporal Variation (Var)" <in the caption>

(RC) P7, L315-317; 2.70 and 2.58 ppbv/decade are too precise. I would suggest 2.7 and 2.6. But are these precise at 5% risk level?

(AR) Yes. These trend values are precise at 5 % risk level. So, I'll keep them as is.

(RC) P9, L374: last decade needs to be more specific. 2000s?

(AR) It means the period from 1996 to 2005. I added the period in the manuscript as follows. (P9, L390)

"The contribution of domestic production had a large IAV and was larger in the last decade (1996-2005) than previously."

Best regards,

Tatsuya Nagashima

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/acp-2016-1087/acp-2016-1087-AC1-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-1087, 2017.

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