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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impacts of anthropogenic and natural sources on free tropospheric ozone over the Middle East" *by* Z. Jiang et al.

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This is a nice and original study making use of advanced modelling tools (e.g. the adjoint of GEOS-Chem) and previous assimilation results (Miyazaki et al. 2015) in order to better identify the causes for the observed seasonal cycle of tropospheric ozone over the Middle East. The study provides interesting conclusions regarding the importance of lightning as well as of transport patterns bringing South Asian air to the Middle East. In particular, it shows that the free tropospheric ozone summertime enhancement is less due to a transport of ozone from outside the area than an import of ozone precursors (most importantly NOx).

The article is clear and well written, and the methods are generally sound. I recommend the article for publication in ACP, if the authors address my only major comment (see below), related to the analysis of the role of transport using an idealized CO-like tracer:

In agreement with previous studies, Middle eastern O3 is shown to be NOx-limited. This questions the relevance of model results for a CO-like tracer with a lifetime of 30 days and only surface emissions, as opposed to NOx which has a lifetime lower than one day and is partially emitted in the free troposphere. In fact, although lightning emissions are much lower than anthropogenic emissions over the Indian subcontinent (see Fig. 1), both emission categories contribute about equally to middle eastern summertime O3 (Fig. 4a-b), which demonstrate very well the importance of the level at which NOx is emitted. This asks for more discussion, given that lightning and anthropogenic emissions have a different seasonality. I wonder whether the authors could conduct additional tests using a shorter-lived tracer and/or with a source located in the free troposphere.

Thank you for the valuable comments and suggestions! We expanded Figure 5 by releasing combustion CO emissions from surface (1-day lifetime, Figure 5g-5i) and middle free troposphere (7-day lifetime, Figure 5j-5l). The results suggest that free tropospheric NOx sources have larger impacts than surface sources on free tropospheric O3.

Minor comments:

Q1: p. 35524, l. 30: besides emissions and chemistry, transport processes are also poorly quantified.

Thank you for this suggestion! The text has been changed.

Q2: p. 35524, l. 33-35 "we use updated reactive nitrogen (NOx) emissions... to provide an improved estimate of O3 precursor emissions": awkward. The purpose of the study

is not to improve emission estimates. Please rephrase.

The statement has been modified to clarify the purpose of this study.

Q3: p. 35527, l. 5-7: were CO emissions not constrained as well? If not, a short justification might be needed.

Because of the limitation of the short horizontal localization length (with the cut-off radius of 1643 km) and the short data assimilation window (i.e., two hours) the influence of long-range transport processes cannot be sufficiently considered in the data assimilation framework of Miyazaki et al. (2015). Thus, the estimated surface CO emissions may have large uncertainty. Therefore, we did not use the optimised CO emissions in this work.

A short description was added at the end of this paragraph.

Q4: p. 35527, l. 12-13: I suppose that the ozone observations also indirectly constrained NOx emissions through photochemistry, a fact not really conveyed by this sentence.

A major advantage of the multispecies data assimilation is that observations of one species (for example, O3) can provide additional constraint on other species (for example NOx).

The description has been modified.

Q5: p. 35528, l. 4-5: The model performs indeed very well in summer and fall (Northern hemisphere), but less so in winter and spring. Could you comment? Were the CHASER-predicted O3 fields from Miyazaki et al. similar to those calculated by GEOS-Chem?

Based on our ongoing studies, we speculate that a possible reason is the seasonal change of O3 chemical environment due to the decrease of biogenic emissions in winter. The model may not provide a good description for O3 production in low VOC

condition.

We confirmed that a CHASER simulation using the same optimised surface NOx emissions has a negative bias against TES retrievals in the Northern extratropics in spring, as commonly found in the GEOS-Chem simulation. This common negative bias in the two models could be caused by similar model errors that may be related or unrelated to errors in surface NOx emissions. The agreement between the CHASER simulation and TES retrievals is found to be better in summer and fall than in spring, as also seen in the GEOS-Chem simulation. However, the spatial distribution and the magnitude of model bias were different between the two models to some extent. The better agreement in summer suggests that the both models realistically represent ozone photochemical productions, given the optimised surface NOx emisisons.

Q6: p. 35529, l. 9: insert "during the summer" after "O3 enhancement"

Changed.

Q7: p. 35530, l. 7: Asian lightning emissions appear to contribute about as much as anthropogenic NOx emissions from Asia to middle tropospheric O3 (Fig. 4a).

Contribution from lightning is really significant. The text has been changed.

Q8: p. 35530, l. 14-16: "... are not significant" is too strong (see Fig. 4). Maybe "less" or "much less" significant.

Thank you for this suggestion! Changed.

Technical comments:

Q9: p. 35526, l. 8: "will allow us" (drop the s)

Changed.

Q10: p. 35526, l. 19: insert a space before FT2000

Changed.

Q11: p. 35528, l. 4: "seasonality"

Changed.

Q12: p. 35528, l. 22: "the maritime continent" is a bit obscure if simply "ocean" is meant.

We use “the maritime continent” to emphasize on the ocean around Indonesia. The word “ocean” may be too general.

Q13: p. 35528, l. 24-25: drop "the" before "highest"

Changed.

Q14: p. 35529, l. 10: "troposphere"

Changed.

Q15: p. 35529, l. 16: I suppose what is meant here is "the rest of Asia", not the entire continent.

Thank you for pointing out this issue. Changed.

Q16: p. 35529, l. 18: "they were not able" (instead of "are")

Changed.

Q17: p. 35530, l. 27: "is produced" (instead of are)

Changed.

Q18: p. 35531, l. 4: "very small" (instead of "much small")

Changed.

Q19: p. 35533, l. 16: "observations"

Changed.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 35523, 2015.