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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Transpacific transport of ozone pollution and the effect of recentAsian emission increases on air quality in North America: an integratedanalysis using satellite, aircraft, ozonesonde, and surface observations" *by* et al.

et al.

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We thank the reviewer for the detailed and helpful comments and suggestions. We have addressed all the comments in our revised manuscript. Please see below for our response to each comment:

Comment: This paper gives a very nice overview of the import of the effect of Asian emissions on U.S. ozone levels using a number of measurement platforms and the GEOS-chem model. It makes a nice contribution to the literature and should be pub-

lished. The comments below should be addressed before the paper is published.

1) Simulated ozone off the west coast of N. America seems to underestimate the aircraft measurements by about 5 ppbv, which the paper attributes to an underestimate in the input of stratospheric ozone. This is probably a reasonable explanation, considering that the model uses the Synoz parameterization. Model underestimates of 10 ppbv are seen at Richland. However, GEOS-Chem seems to reproduce the ozone record at Mt. Batchelor (MBO) without any significant bias. The discrepancy between the model measurement comparison with the ozonesondes and aircraft measurements versus Mt Batchelor is not discussed. Presumably if the STE of ozone was increased in GEOSchem (to reconcile the model with the aircraft and ozonesonde measurements) this would significantly impact the simulation over MBO. I would guess as a result GEOSchem would significantly overestimate the ozone at MBO. This would seem to imply the estimated 9 ppbv contribution from Asia is too high. If the authors understand how to reconcile these set of measurements it would be helpful and would strengthen the paper. If not, this discrepancy should be stated and the strong conclusion that Asian pollution contributes 9 +/- 3 ppbv of ozone at MBO should be explicitly tempered by the fact that model underestimates ozone by 5-10 ppbv in other nearby locations.

Response: There is less stratospheric influence at MBO than at Trinidad Head and Richland. Weiss-Penzias et al. JGR (2006) found that the average ozone at MBO is significantly lower than the ozone profiles from Trinidad Head at 740 hPa (MBO altitude). The mean ozone concentrations at 740 hPa measured at Trinidad Head and Richland during INTEX-B were 60 ppbv and 62 ppbv, respectively, compared with 53 ppbv at MBO.

We now state in the text, "The mean observed ozone concentration at MBO during INTEX-B is 54 +/- 10 ppbv (mean +/- standard deviation), compared with 53 +/- 9 ppbv in the model. It is lower than the mean ozone observed at Trinidad Head and Richland at 2.7 km during INTEX-B (60 ppbv and 62 ppbv, respectively as shown in Fig. 5), because stratospheric influence at MBO is weaker (Weiss-Penzias et al., 2006)."

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Interactive Comment

Other Comments. 1) Page 8152, line 25: 495 Tg ozone y-1 should read approximately 495 Tg ozone y-1, as Synoz only constrains the ozone flux to the specified stratospheric input over a long-term time average. Interannual variability may occur, although its amplitude is constrained.

Response: We added "approximately" in the text as suggested.

Comment: 2) I found the discussion on emission changes rather confusing and had to reread it to make sure I understood what is going on. I think the following points would help to clarify the discussion and interpretation. 1) I think it would be helpful, if the authors would explicitly name the emission inventory that you are using, e.g. S2006(prime); (Then the various emission inventories could be explicitly named on the bottom of 8152). 2) Then, if I understand correctly, S2006(prime); is the same as S2006, but with the NOx emissions equal to twice those of S2000 over East Asia (20-50 N and 100- 150 E). It would help to explicitly state this on page 8154, as well as the countries over which you are doubling the anthropogenic emission inventory (i.e., at least over China, Japan and S. Korea). Then if I understand correctly you are attributing the factor of 2 emission increases over China with growth in emissions, but over Japan and S. Korea to low emission estimates in S2000. 3) Thus, only approximately 80

Response: Following the suggestions, we have clarified the discussion in the revised manuscript: (1). we included country names on page 8154, line 2 that "over eastern Asia (including China, Japan and Korea)"; (2). we replaced the last paragraph of Sect. 3 with "In what follows, we will interpret the doubling of anthropogenic NOx emissions in eastern Asia relative to S2000 as representing the actual 2000-2006 regional growth rate in emissions. This interpretation overestimates the actual growth by about 30% due to the apparent underestimation in S2000 for Japan and Korea. In any case, our standard simulation for 2006 includes our best estimate of East Asian emissions for that year constrained by the OMI data."

Comment: 3) Page 8154, line 23: Are the results of Jaegle and Wang consistent with

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your assumptions? Their estimates suggest a 30-50% emission increase is reasonable; the author’s emissions are 2-3x what they report. Can the authors clarify?

Response: They get a 30-50% increase, and we get a factor of 2. At least it is in the same direction. They used GOME, which has a much coarser resolution than OMI, and that might matter for Korea and Japan. There are enough factors that could contribute to this difference between top-down estimates that we would rather not elaborate.

Comment: 4) Page 8155, line 15: likely because of an OH overestimate. This seems a bit presumptuous. Certainly your emissions of CO are also rather uncertain. consistent with an OH overestimate would be a better way to phrase it.

Response: We have changed the text accordingly.

Comment: 5) Page 8156, line 5. You seem to be assuming an error in the measurements because NO/NO2 in Geos-Chem is in close agreement with the NASA Langley photochemical model. Is it not just as likely that the photochemical model is also missing some important process or input? Or does the 15% error in HO, HO2 in that NASA model lead you to believe this is unlikely. Please clarify.

Response: The observed and modeled NO, NO2 and the ratio of NO/NO2 are in agreement for the C-130 data set and for the DC-8 dataset below 6 km. Above those heights the model and observations deviate and the observed NO/NO2 ratios deviate from photostationary state. The overestimation of the DC-8 NO2 data grow beyond reasonable estimates of the uncertainty in the measurement zero offset above 8 km, increasing to approximately 30 pptv at 10 km and 40 pptv at 12 km. It is well established that the NO/NO2 photostationary state is accurately (to better than 30%) represented by photolysis of NO2, and the reactions of NO with O3, HO2ň and RO2. And the measurements described here strongly support that interpretation with the photostationary state expression is in agreement with the observations in the lower troposphere. We interpret the systematic differences between observations and the modeled NO/NO2

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as an indication that there is an altitude dependent error in some combination of the NO, NO2 or J-value measurements.

We now state, "Observations of NO and NO2 from both the DC-8 and the C-130 are in photostationary state and the absolute values are in agreement with the GEOS-Chem simulation at low altitude (below 6 km for the DC-8 and below 4 km for the C-130). At higher altitudes the NO/NO2 ratio from both platforms is inconsistent with the assumption of photostationary state. Differences for the C-130 data are within the uncertainty in the instrument zero offsets ($<$5 pptv). For the DC-8 the differences become larger than that can be explained as uncertainties in the measurements above 8 km. The GEOS-Chem model overestimates NO measurements from DC-8 by 50% at 10 km (60 vs. 40 pptv) and underestimates NO2 at the same altitude by a factor of 2 (20 vs. 40 pptv). By coincidence NOx is in agreement. If we attribute all of the error to one or the other measurement, than the GEOS-Chem model predicts either 50% too much NOx or 100% too little at 10 km."

Comment: 6) On the bottom of page 8156 you suggest the model error in ozone is due to an underestimate in STE. Yet, by filtering out stratospheric air one might think you have minimized this error. Please clarify?

Response: We now state, "The stratospheric filter does not exclude stratospheric influence within the troposphere, as mixing of stratospheric and tropospheric air masses causes the O3/CO ratio to drop rapidly below the filter threshold."

Comment: 7) Page 8157, line 20: Is the decreasing trend in CO during April and May also attributable to the increase in OH?

Response: Yes, it can also be a reason. We changed the sentence on page 8157 to "CO shows a decreasing trend from April to May over the Northeast Pacific due to the seasonal decline of biomass burning in Southeast Asia (Duncan et al., 2003) and the seasonal increase of OH concentrations".

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Comment: 8) Page 8157, line 26: was event 2 also seen at MBO?

Response: As indicated in Fig. 10 and 14, event 2 was also observed at MBO around May 10th. We changed the sentence on page 8157, line 26 to "Event 2 was observed from the DC-8 on 9 May as discussed in Sect. 5.2, and arrived at MBO around 10 May.".

Comment: 9) Page 8158, line 12. Is the correlation of 0.5 significant?

Response: Yes, we added on page 8158 that r is "significant with 95% confidence".

Comment: 10) Page 8158, line 23. This north-south split in the plume has also been noticed in transport to Hawaii (e.g., Hess and Vukicevic, JGR, 2003).

Response: The study by Hess and Vukicevic discussed a pollution plume transported to MLO and PAN decomposition during subsidence, but it does not describe the north-south split in the Northeast Pacific as we discuss here.

Comment: 11) Page 8160, line 4: The ozone production in the southern branch is relevant for impact on the United States. It might be better to say something like the direct impact. Much of the southern branch gets shunted into the subtropical boundary layer where it is subject to rapid photochemical destruction; it may be that slow subsequent ozone production from the Northern Branch might be indirectly important on longer temporal and spatial scales.

Response: Yes, we changed the wording so that "The ozone production in the southern branch is relevant for direct impact on the United States"

Comment: 12) Figures 11 and 12 show two maximums in ozone production, ozone and NOx. The maximum in the central Pacific was examined by Hess and Vukicevic (JGR, 2003) in relation to the MLO experiment on Hawaii. They also showed the importance of PAN decomposition in descending anticyclonic air and the subsequent ozone production.

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Response: We included the citation of Hess and Vukicevic (2003) on page 8169, line 3.

Comment: 13) The increase in ozone attributed to increases in Asian emissions of 3-5 ppbv is likely too high by 20% (assuming S2000 is 20% too low in their estimated NOx emissions over Japan and S. Korea). Thus, an estimate of 2.4-4 ppbv is probably more reasonable. This should probably be pointed out.

Response: The effect is nonlinear. We now state, "rising Asian emissions from 2000 to 2006 have increased ozone at MBO by 3 ppbv on average in April-May and up to 5 ppbv in events, although a small part of that increase could reflect the underestimate of emissions for Japan and Korea in the baseline S2000 inventory for 2000."

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8143, 2008.