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Interactive comment on "Decadal changes in global surface NO_x emissions from multi-constituent satellite data assimilation" *by* Kazuyuki Miyazaki et al.

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This is a very interesting assimilation study on global NOx emissions. It is innovative in that it uses not only NO_2 data to constrain the emissions, but also data for other related chemical compounds (O_3, CO, HNO_3). Furthermore, it uses NO_2 data from 3 different nadir sensors (OMI, SCIAMACHY, GOME-2) which have different overpass times. In most inverse modeling and assimilation studies for NOx, the data from only one satellite sensor are used (and data from other sensors are possibly used for evaluation).

General comments

1. I certainly appreciate the effort made by the authors to incorporate more data.

There is logic to it: more data should be better than just one dataset. It is argued (maybe a bit too emphatically) that non-NO_2 datasets improve the NOx emission estimation because they should lead to better estimation of the NOx lifetime in the model. In general, that might be true, but I wouldn't be so sure that it is automatically the case. I find that adding more data from different species might contribute to obscure the interpretation of the results, because the additional data come with their own limitations and uncertainties (including biases) which are not all well characterized. I am not fully convinced that authors understand perfectly the role of the different datasets in the assimilation. I wonder in particular to what extent the NOx emission updates are driven by the non-NO_2 datasets. For example, ozone is apparently biased low in the model. Increasing NOx emissions is naturally found to improve ozone. But ozone could be biased low due to other reasons (transport, deposition, NMVOC chemistry and emissions). So, is ozone improved for the good reasons? Who knows? Many other CTMs overestimate surface ozone. I encourage the authors to moderate their claims regarding the advantages of additional data.

That being said, I concur that assimilating non-NO_2 dataset should contribute to improve (somewhat) the NOx lifetime in the model, which is a good thing. But I would expect the authors to provide a more quantitative and systematic analysis of how the non-NO_2 datasets influence the assimilation results. I also encourage the authors to be more cautious in their discussion, to reflect the possible limitations and complications associated with the use of additional, non-NO_2 measurements.

2. Regarding the use of 3 different NO_2 sensors, it is obvious (and I think the authors know) that the diurnal cycle alone cannot explain entirely the difference between NO2 columns from e.g. GOME-2 and OMI. And even if it would, it is also obvious that the diurnal cycle of NOx emissions is only one among many different processes affecting the diurnal cycle of NO2 columns. This article presents a smart

but crude procedure to improve the match with the 3 sensors simultaneously in spite of their inconsistencies: additional control parameters are introduced which allow modifying the diurnal cycle of emissions at every model pixel. Unfortunately, the result is not much credible as it would imply much stronger rush hour emission peaks even in regions where mobile emissions (cars) are not the main NOx source category. Power plans, industries, etc. do not have peak activity around 8 AM. The most negative values of the Etc parameter (Fig. 13) are found in Inner Mongolia, which has only few cars but does have power plants. Even though the diurnal cycle adjustment serves its purpose, it is clearly artificial. The authors should provide a better explanation of why they choose this procedure. Maybe it is the only one which works since we don't really understand the reasons for the inconsistency between morning and afternoon sensors. More discussion is warranted.

3. Although the paper is already quite long, I would expect at least some comparisons with independent NO_2 measurements. The reader has no clue regarding how the model performs for vertical profiles of NO_2. Also, given the focus of the paper on the diurnal cycle, comparisons with ground-based remote sensing data data could be useful. Therefore, although this article is clearly interesting and the methodology appears generally sound, I recommend that the authors try to address those main comments, as well as the other comments listed below, before it can be published in ACP.

Other comments

- Page 5, 1st full paragraph: I find odd to apply a unique diurnal cycle to all emissions in a given region, e.g. the anthropogenic-type cycle over Europe, eastern China, Japan, North America. This is strange. Why not make a weighted average based on the fractional a priori contribution of anthropogenic, biomass burning

and soil emissions? The diurnal cycle in New York doesn't have to be the same as in Wyoming.

- Page 5, 2nd paragraph: What is the vertical LNOx profile parameterization?
- Page 8, 1st full paragraph: Apparently the retrievals of Boersma et al. (2011, 2004) are used. But then the section goes on mentioning reduced errors based on Maasakkers (2013) even though the retrieval of Maasakkers is not used. This is difficult to understand.
- Page 10, lines 9-10: The assimilation effectively corrected the NO_2 columns at the different overpass times. The complete diurnal cycle of NO_2 concentration is an entirely different story. The model performance could be checked by comparisons with ground-based remote sensing data.
- Page 10, line 13: Here and elsewhere, the observation errors for highly polluted cases are considered very large. But the relative errors there are often of the order of 25-35% which is generally less than at more remote locations. Of course during the winter, things are different due to large zenith angles, clouds, and snow.
- Page 10, line 17: Some explanation on why the model lifetime of NOx would be too short would be useful.
- Page 12, on trends: The total number of observations changes during the 10-year period. Also the different satellites provided data during different periods. Some comments on possible consequences for trend estimation are needed.
- Page 12, lines 12-14: After assimilation, the emission is higher in January than in July over the U.S. Therefore, it is very unlikely that soil emissions can explain the changes in seasonality.

- Page 12, lines 15-17: The emission factors are indeed uncertain, but so are also the biomass burnt estimates.
- Page 12, lines 17-18: Note that the year-to-year variations over South America are very large.
- Page 12, lines 30-34: the increases aren't highest just over cities and lowest over remote areas. The entire N-E China and the Guangzhou area show large increases. The Chengdu/Chongqing area (with emission decreases) is certainly not "remote". Over N-E China, given the model resolution, it is not possible to distinguish urban from rural areas. Furthermore, Inner Mongolia shows large increases.
- Page 14, 1st full paragraph: The results regarding the trends in Europe are difficult to understand. Could you compare with previous studies for Europe, e.g. Curier et al. (Remote Sens. Environ. 2014, doi:10.1016/j.rse.2014.03.032)? From Figure 3, the OMI observations indicate a positive NO_2 trend, whereas GOME-2 shows the opposite trend. Such difference cannot be due the diurnal cycle of NO_2. Apparently those instruments have drifts which can be interpreted as emission trends. Please comment on this.
- Page 14, line 23 "The summertime peak enhancement is obvious over remote regions": Could you substantiate that claim?
- Page 15, line 10: Couldn't this be verified with e.g. MODIS fire counts or other biomass burning proxies?
- Page 15, line 27: Although temperature has some effect, the shorter NOx lifetimes at tropical latitudes such as India are primarily due to higher photolysis rates and specific humidity.

- Page 16, line 3: Why would high resolution analysis be required? This shouldn't be so complicated. For example, biomass burning has a distinct seasonality which can be probed at coarse resolution.
- Page 16, end of section 4.2.6. The high temporal correlation between N. Africa and Central Africa is interesting. Would this be related to biomass burning or to soil emissions (or both)? Examination of MODIS fire counts could help, also possibly temperature data. Is this correlation also found in the NO_2-only assimilation?
- Page 19, first paragraph: Basically, the improved ozone is due to the general increase in NOx emissions over all regions, whereas the a priori model seems to have a negative bias in surface ozone. In the study of Travis et al. (ACPD, 2016, doi:10.5194/acp-2016-110), NOx emissions over the U.S. are found to be largely overestimated in comparisons with aircraft data.
- Page 19, last full paragraph: Could you also provide the global tropospheric chemical lifetime of methane (or methyl chloroform) in the model?
- Page 20, first sentence: "The inverse lifetime is expected to be proportional to the ratio of NOx to NO_2". It's the other way around. Increase the NOx to NO_2 ratio should increase the fraction of NO (which does not react with OH) and therefore decrease the sink of NOx, i.e. the inverse lifetime. The main effect of a NOx emission increase is (most often) increased OH levels and therefore shorter NOx lifetime. The point which is made in this paragraph is unclear.
- Page 20, lines 20-25: The large adjustments are first said to suggest a change in diurnal evolution of the emissions. Then they are said to suggest other possible causes related to the model or the retrievals. Correct, but then the first suggestion is not necessary. Values of Etc as negative as -0.6 or -0.8 are found at some locations, which are impossibly large. Large Etc should be found only in areas

where traffic is the dominant source. This does not appear to be the case (Fig. 13).

- Page 22, line 4: Is the given observed NO_2 concentration trend for OMI or for all sensors? The trend appears very different between GOME-2 and OMI.
- Page 22, line 6: Why would NO_2 have become more long-lived? Does OH show a negative trend in this region? If so, what are the causes for this trend? Note that the fraction of NO_2 to NOx is determined mostly by ozone and the photolysis rate of NO_2. A shift in NO_2:NOx emission ratio does not matter much except directly over emission areas (titration effect). The paragraph seems to imply that the NO_2:NOx emission ratio in the model has changed over the 10-year period. Is this true?

Technical corrections

- Page 1: "Forkert" should be "Folkert"
- p. 1 l. 6: "biased" should be "biases"
- p.1 l. 8: "the development" : do you mean the evolution?
- p. 2 l. 2: "traffic rush hours, economic activity..." those are not "source categories". Sentence is confusing, please rephrase.
- p. 2 l. 25: Kalam should be Kalman
- p. 2 l. 33: insert a hyphen between "multi" and "constituent" same line: replace advancement by advance or progress

- p. 3, l. 21-22: The sentence "The OH magnitude and gradient is the primary chemical pathway for propagating observational information..." does not make much sense. Rephrase or delete.
- p. 3 l. 31: Replace maybe "an EnKF technique" by "a variant of the EnKF technique"
- p. 4 l. 25: Explain "background spread"
- p. 4 l. 30: Isn't Yienger and Levy (1995) the correct reference for GEIA NOx? Please check.
- p. 6 lines 12, 16, 22: I think "Ets" should be "Etc"
- p. 6 l. 31: GOME-2 (not GOME-II)
- p. 10 l. 24: Here and at other instances, replace "c.f." by more standard phrasing (e.g. "see")
- p. 11 l. 12: the sentence seems to imply that the chemical lifetime of NOx might be underestimated, which is not what you mean here. Rephrase.
- p. 11 l. 20: I suppose you mean GFED 3 here, not EDGAR 4.2 (see section 2.1)
- p. 12 l. 9: "southern parts of the Eurasian continent" : don't you simply mean China? The seasonal variation over Southeast Asia does not show a summer maximum, so it does not fit into the point made in this sentence.
- p. 12 l. 18 "assumptions applied for the a priori emissions" I think you could be more specific (use of climatology after 2011)
- p. 12 l. 27: "The EDGAR v4 emissions are too low": that statement is too blunt for several reasons. Replace "EDGAR v4" by "our a priori inventory" (since EDGAR

for 2008 is used after 2008, and since soil emissions are not from EDGAR). Furthermore, add something like “Our assimilation indicates that…”.

- p. 12 l. 28: “too low by a factor of 0.6”: awkward. Should be too low by a factor of 1/0.6 (i.e. about 1.7)
- p.12 l. 29: emissions are maximum in June, not July.
- p. 13 l. 34: “in the reported mobile emissions”: why specifically in this source category?
- p. 14 l. 1: replace “reveal” by “show”
- p. 14 l. 2: replace “by” by “after”
- p. 14, l. 18: “around Atlanta (…) and Denver”: this seems to indicate that increments are found mostly over cities, which is not true. Consider replacing by “Southeast US and most of Western US”
- p. 14 l. 19: delete “over” after “around”
- p. 14 l. 20: Los Angeles
- p. 15 l. 9: I think the word “boreal” is superfluous here (and at many other instances in the text)
- p. 15 l. 20-21: “particularly strong increase around Delhi” but the changes over Delhi are lower than the regional average!
- p. 16 l. 5: Replace “by data assimilation” by “due to data assimilation”
- p. 16 l. 33: replace “reflection” by “reflecting” same line: replace “when” by “whereas”

- p. 19 l. 17: LNOx (instead of LNO)
- p. 19 l. 34: here and elsewhere in the manuscript, insert hyphen between “multiple” and “species”
- p. 20 l. 17: Replace “rom the three…” by “constrained by the three…”
- p. 21 l. 8: “using either model after data assimilation” : awkward, the model is used for data assimilation
- p. 32: Table 2 and elsewhere: Replace “Australis” by “Australia”
- p. 41 Figure 6: it is impossible to distinguish black and dark blue on the “South America” plot. Consider using other colors.
- p. 44, Figure 9: the title of the middle panels should be “A posteriori – A priori”. Same for the title of the right panels, the minus sign is missing.

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