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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Top-down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: assessing anthropogenic emissions of CO, NO_x and CO_2 and their impacts" *by* J. Brioude et al.

Anonymous Referee #3

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1 Overview

The manuscript by Brioude et al. uses aircraft data from the CALNEX 2010 campaign and an inert-tracer Lagrangian inversion scheme to constrain emissions of NO_x, CO and CO_2. The study considers a range of model configurations and presents detailed comparisons of their top-down emissions estimates to several bottom up inventories, as well as other recent works on emissions constraints coming from the CALNEX campaign. Further, they consider observations from a flight in 2002, and they use this to

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derive long-term emissions trends. The ability to use aircraft measurements to provide mesoscale constraints on emissions is well within the scope of ACP, and the conclusions regarding the emissions in the LA region are of value to the air quality community. The methods are fairly well tested, building upon several recent works by Brioude, although there are a few ways in which the results could be further tested. I would strongly encourage the authors to probe the robustness of their inversion at scales commensurate with those upon which the conclusions are based, and to formally assess the assumption of daytime NO_y as a conservative tracer whose variability is exclusively owing to errors in NO_x emissions from the previous 24 hrs. These issues, as well as some minor suggestions, are detailed below.

2 General comments

- The NO_x emissions constraints are based upon the assumption that daytime NO_y is a conservative tracer, whose variations can be exclusively ascribed to errors in NO_x emissions from the preceding day. This is a convolution of a few assumptions that warrant further consideration. That the effect of NO_x lasts only a day is addressed in a separate comment below. Now, consider the remaining assumption that NO_y is (a) conserved and (b) governed exclusively by NO_x emissions. Given that these assumptions are repeated from earlier work (Brioude et al., 2011), but not tested nor supported by literature citations there nor in the present manuscript, I strongly recommend that it be evaluated at this stage. The modeling effort would be minimal – simply perturb the NO_x emissions on one day in the LA Basin in WRF-Chem, and then track how NO_y changes.

- The validation effort is a bit limited. The reasons for restricting the comparisons to specific times and locations doesn't really seem justified given that the authors extrapolate their emissions constraints to much broader temporal (e.g., daily to

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annual average trends) and spatial scales (e.g., 31454.9) than they allow to be included in the validation tests using only aircraft data from 10 am to 6 pm LT between 200 and 700 m. If we are to believe the emissions constraints are valid at broad scales, then they should be evaluated at such scales as well. Are there other observations from CALNEX (e.g., surface monitoring) that could provide additional assessments of the top-down emissions estimates? Using the same observations that were used in the inversion itself is not as stringent of a test. Even testing the inversion results from one flight relative to observations from a different flight would not reveal some forms of systematic bias.

3 Specific comments

- 31444.1: This seems a bit misleading, as remote sensing can also sample pollutants at different distances downwind of a source. There have been many papers assessing plumes from power plants (e.g., Valin et al., AMT, 2011; Wang, Streets et al., ERL, 2010).
- 31446.6: Can the authors more quantitatively assess, rather than assert, the relative magnitudes of the sources of error?
- 31448.27: Is comparing surface fluxes estimated using 24 h and 48 h sufficient to quantify the error? Instead, aren't at least three tests necessary to show that the results are converging? Otherwise, we might just as easily conclude that each additional day back considered would change the result by an additional 5%. It also seems hard to rationalize the conclusions. For CO and CO_2, wouldn't it depend upon the meteorological conditions surrounding the individual flight? It just seems that in stagnant conditions, emissions within the domain from much further back than 24 h could have an influence.

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- 31449.6: An adjoint is unnecessary, or is FLEXPART equivalent to the adjoint of a Lagrangian model? I think it is really the latter, as time-reverse Lagrangian models are considered to be exactly that (e.g., Pillai et al., ACP, 2012, and references therein). So it is perhaps more precise to say "adjoint of WRF-Chem".

- Section 2.4: Unless I missed it, there doesn't appear to be much about the prior or observation error statistics. How are these determined? Are they assumed to be uncorrelated? Are observations randomly sampled to ensure this as in previous works by Brioude?

- Section 2.4: Restriction of the inversion to only consider sources for which their is already a significant emission could bias the inversion. It would not be possible to use this technique to infer a source that was not present in the initial inventory. This should be recognized, and if the authors can assume that the only errors in the inventories worth discovering are adjustments to the magnitudes of known sources, that should also be justified and explained further.

- 31454.15: The wording here is a bit odd. I think it may be clearer to say "modified the spatial distribution of the CO surface fluxes compared to the prior," because the prior itself has not actually changed. If the spatial distributions are shifting though, it does raise some concern about ruling out the possibility of a missing source, as mentioned in the previous comment.

- 31455.23: It would also be useful to compare these slopes to the a priori simulations, to see what the improvement has been following the inversion.

- 31462.9: Why is the correlation higher for the 12 km case than the 4 km case?

- 31457.9: Could the authors discuss a bit further the sectors contributing to CO_2 and why there is less of a weekend effect for these species relative to CO and NO_x?

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- Figure 5: Can the authors comment on the apparent increase in weekend emissions of CO_2 in the San Diego region?
- 31463.2: NH_3 is not a conservative tracer, so extension of these methods to this species are not clear.
- Abstract and discussion: mention forecast, but not sure if really mean forecast or reanalysis.

4 Editorial comments

- 31444.16: Conclusions and . . .
- throughout: italicize subscript x on NO_x
- 31448.1: cumulus scheme
- 31453.16: associated with
- 31453.21: to the NEI
- 31454.26: converted to daily average
- 31464.19: Middlebrook and Roya

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 31439, 2012.