Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-180-RC1, 2021

Comment on acp-2021-180

Anonymous Referee #2

Referee comment on "Spatially and temporally resolved measurements of NO_x fluxes by airborne eddy covariance over Greater London" by Adam R. Vaughan et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-180-RC1, 2021

General Comments:

The authors generate high-resolution, observationally based maps of NOx emission fluxes for the Greater London region using eddy covariance measurements from aircraft and environmental response functions that model flux-source driver relationships. The derived emission maps provide a direct comparison to the NAEI bottom-up inventory, which underestimates regional NOx. The study showcases a novel and compelling use of airborne eddy covariance measurements that are strengthened by source area modeling and the development of a machine learning based flux-driver model that enables upscaling beyond the immediate measurement footprint. However, the authors neglect several important details about the quality and uncertainty of the underlying eddy covariance NOx measurements. Showcasing some quality metrics would build confidence in the measurement system as detailed in the specific comments below. Furthermore, there is no systematic treatment of the flux measurement uncertainty. The authors state the typical random and systematic errors for the 400 m average, which are quite high (> 100%). How do such errors reduce on further averaging? How does the measurement uncertainty effect the evaluation of the NAEI NOx emissions? In general, the figures lack any depiction of associated uncertainties. A better emphasis on uncertainty would support the main conclusion that the data-generated maps are valid for this type of inventory evaluation.

Specific Comments:

- L69: What is the typical aircraft speed?
- L71: What times of day did flights occur? This information should be added to Table 1 to give a better sense of diel sampling.
- Section 2.2: Much of the detail in this section could probably be moved to a

supplement. Main points, including instrument accuracy and precision, should remain, but details on calibrations, dark counts, etc, read more like an instrument paper and distract from the main results.

- L114: Is the 3.3% correction referenced here to generate dry mixing ratios from wet? Or is this the correction due to quenching of the chemiluminescence reaction from water vapor? If the former, although the correction is small, the authors should still note that unless using dry mixing ratios to generate NOx fluxes, they may need to apply a Webb density correction to account for heat and water vapor. No mention of a density correction is made later on in the text.
- L133-134: Detection limits of 49 and 78 pptv for NO and NO2 seem unrealistically low for a 9 Hz integration time. Another reference from the paper using the same instrument (Lee et al, 2009) quotes a 2-sigma LOD for NO of 36 pptv at 1Hz. If this is true, adding the Allan-Werle plot to the supplement would be useful.
- L145: This is not a strictly correct error estimation. The individual uncertainties should be propagated through all the equations used to calculate NO/NO2 mixing ratio.
- L149-150: Which species were measured by the PTR-MS and Picarro? Were they used in this analysis at all?
- Section 2.2.3: Was there any treatment or consideration of a vertical flux divergence? This is an important point that the authors should address.
- L225-226: "whereas time-frequency EC gives a flux measurement every 400 m along the transect using a 4000 m moving window…" How is the 4000 m moving window applied to the 400 m CWT fluxes? Are the measurements overlapping? This wording is unclear.
- L245: One of the advantages of the CWT method is that by decomposing the signal into the time domains, the strict criteria for stationarity is not necessary.
- L250: It's not explicitly stated, but was data with $u^*<0.15$ m/s discarded?
- Section 3.1: Some additional quality metrics for the NOx fluxes would strengthen the results. Examples include a lag-covariance plot, a CWT cross-scalogram, wind and scalar power spectra and co-spectra. Such figures could be in the supplement but would build confidence in the application of the eddy covariance technique to NOx fluxes and would give a visual idea of signal-to-noise.
- L299-300: The point-by-point errors are significant. How do errors reduce when averaging? Figure 5 shows the standard deviation in shading, but it would be helpful to get a sense of the error when averaging all transects of a given type together. These uncertainties should be reported, even if not displayed in the figure.
- L334-335: In general, not much explanation is given in the manuscript about when sampling occurred. At what times are the measurements acquired? How is averaging performed across transects sampled at multiple times of day? Doesn't this dampen any diel variability? How is the timing compared to the emissions inventory? Are you comparing emissions for each transect time independently or to a mean? It would be helpful to add some clarity on these points in the text.
- Figure 6: What does the shading represent? Is it the uncertainty of the measured/inventory fluxes? Also, please elaborate further on the GAM. There is not much description in the text. Are the NAEI estimates here generated from the flux footprint?
- L410: The description of Figure 8 is unclear, particularly the phrase "median average". Please elaborate on what is being depicted.
- Figure 8: Should specify that these are NOx emission rates. What is the uncertainty in the flux projection? It would be helpful to see a map depicting the associated uncertainties. The standard deviation only indicates the variability between different legs but does not help quantify how any uncertainty in the measurement propagates into the ERF model.
- L431: It would be helpful to show a figure depicting the comparisons between ERF-reproduced and measured fluxes in the supplement to get a visual idea of how robust the ERF technique is.

Technical Corrections:

- Figure 1: Lat/lon coordinates for each transect do not align with those listed in Table 1.
- Table 1: Add typical transect altitude or range of altitudes. Add time of day each transect was sampled.
- Table 2 is incorrectly labeled as Table 1.
- Table 3: Label column "ratio" with something more descriptive.
- Figure 6: Describe what shading indicates in the caption.
- Figure 7: Identify blue and black curves in the caption.
- Figures 8 & 9: Specify NOx with units or in captions