

Comment on acp-2021-982

Anonymous Referee #2

Referee comment on "Eddy covariance measurements highlight sources of nitrogen oxide emissions missing from inventories for central London" by Will S. Drysdale et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-982-RC2>, 2022

The authors present eddy covariance measurements of NOx fluxes over London and interpret the results in terms of predominant sources and bottom-up predictions. The topic is of relevance to ACP and in general the quality of analysis is appropriate. I list some comments below that in my view should be addressed prior to acceptance.

Main comments

Section 2.2. Here the authors derive a 23-62% correction due to vertical flux divergence but then don't apply it as too uncertain due to issues with the model mixing height. What helps put the study on more solid footing is that this correction would only increase the measured fluxes, and the authors are already inferring an inventory underestimate. However a couple aspects of this are somewhat surprising, at least to me, and merit more discussion.

- The first is the size of the effect, and here it would help to give some information on the modeled boundary layer height values. Based on Eq 1, 23-62% corrections require heights of only be 500-1000m. If I'm reading it correctly, the observational comparison later in this section indicates that the modeled boundary layer heights are biased substantially low, so that the flux divergence influence as estimated is too big.
- I believe application of Eq 1 implies that the measurement height is always out of the constant flux layer, and further that there isn't actually a constant flux layer at all ... i.e. the correction begins at the surface rather than at the top of the surface layer. Some more physical justification is needed of its applicability.
- Another surprising aspect is the inconsistency between the entrainment-based correction approach (23-62%) and the single-point correction approach (0%). The authors attribute the latter to "attenuation of the concentration enrichments at this measurement height, rather than the lack of stored flux" but it is not clear to me how physically plausible this actually is. Some more information / discussion is needed.

2.2.1.3 The authors derive a 5% spectral correction due to high-frequency sampling losses. However there was large variability in sampling flow (3-30 L/min) and turbulent characteristics (Re 120-2300). Does the degree of high-frequency loss vary significantly across these conditions? Is it really only 5% for the low-flow / laminar periods?

Sampling took place on a 13m mast atop a 177m building. Surrounding buildings are all quite a lot shorter but to what degree might the BT Tower itself disrupt the sampled flow field in a way that would bias the fluxes?

2.2 I appreciate the discussion of eddy covariance QA/QC. It would also be helpful to show ogives and to give information about the range of sampling lag times.

2.3.1 please state the temporal resolution of NAEI in this section. Is it annual?

2.3.2 what is the spatial resolution of LAEI? Also 1km2?

L252, "Day of week and month of year factors were still applied". Unless I missed it, this is the first mention of temporal variability in the inventories. Such factors need to be more carefully described in the corresponding methods sections.

Conclusions. It seems that pinning down the uncertainty due to entrainment / storage should be an important priority for future work, as this term is similar in magnitude to the inventory bias inferred. Are there plans along these lines that can be mentioned in the conclusions?

Conclusions. The reader is left a bit unsatisfied by the lack of take home messages. The authors might consider discussing some implications of their findings; e.g. what do the derived emission errors mean for AQ predictions?

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Technical/editorial comments

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some typos and grammatical issues throughout; please correct.

Lines 25-35, the narrative here as written is confusing and hard to follow.

67, "hitherto unreported sources" implies missing sources when it seems the problem is mainly underestimation of known sources. Perhaps "underreported"

Figures are not referred to in order

Table 1, I'm confused here because some directions the sectors add up to <<100% and others add to >100%. Please clarify what is going on.