Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2020-81-RC1, 2020

Atmospheric Measurement Techniques Discussions

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Interactive comment

Interactive comment on “The quantification of NO_x and SO_2 point source emission flux errors of mobile DOAS on the basis of the Gaussian dispersion model: A simulation study” *by* Yeyuan Huang et al.

Anonymous Referee #2

Received and published: 14 May 2020

Review of “The quantification of NOx and SO2 point source emission flux errors of mobile DOAS on the basis of the Gaussian dispersion model: A simulation study“ by Yeyuan Huan et al.

In this manuscript, the authors report on an extensive sensitivity study on car-DOAS measurements of NO2 and SO2 for the estimation of fluxes of point sources. Pollutant plumes are modelled with a Gaussian dispersion model, zenith-sky observations of these fields from a moving platform are simulated and emission fluxes computed from these simulated measurements. The simulations are executed for a range of emission

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strengths, wind speeds, measurement distances and sampling ratios covering a large part of the possible parameter space. Based on these simulations, uncertainties of the retrieved fluxes are estimated and discussed in detail, leading to recommendations for optimized measurement conditions and measurement approaches for flux measurements using car-DOAS observations.

The topic of this study fits well into the scope of AMT as it provides detailed uncertainty estimates for a frequently used atmospheric measurement method. The results are new and interesting for the community and will prove useful for the design and interpretation of future emission flux measurements using car-DOAS observations. The manuscript is well organised and overall clearly written. I have however a number of concerns which the authors need to address before it can be accepted, and therefore recommend the manuscript for publication in AMT only after major revisions.

Major comments

- Two of the key error sources the authors discuss are wind direction and wind speed uncertainties. While I agree that wind speed introduces important uncertainties in flux estimates, I think that wind direction at such relatively short distance from a point source can be estimated with reasonable accuracy just from geometry. I strongly disagree with the way the authors have estimated wind uncertainties: They use a statistical evaluation of wind measurements at one site and interpret the standard deviations they find in these measurements as uncertainties which they then parametrise as a function of wind speed. However, this is not the quantity of interest for flux measurements, where wind speed and direction is usually taken from a close-by measurement or a model. I think either the authors need to explain why I am wrong, and this is after all a good representation of the uncertainty of the wind information usually employed in car-DOAS measurements, or they should use another, maybe simpler representation of the wind field uncertainty.

- One important discussion point in the manuscript is the sampling error. Unfortunately, even after reading this part several times and checking the referenced papers for an explanation of what exactly this sampling uncertainty is, I was not able to understand it. In an ideal world, where the sky brightness does not change, the instrument has no dead time between measurements, and there are no non-linear effects in the DOAS evaluation, the retrieved VCDs are the mean VCDs over the distance travelled during the integration time. If the wind field is homogeneous as I assume is the case in this study, then I fail to see where the "sampling error" comes into play. I think the authors need to provide a better explanation of what exactly they are referring to with this term.

- One relevant point of the manuscript is the discussion of the NO2/NOx ratio on the measurement uncertainty. Maybe I overlooked this information, but it was not clear to me how this uncertainty was computed – did the authors just assume that no correction is applied in the retrievals, so the error is defined as the difference between an assumed steady state ratio and the real NO2/NOx ratio? Or is the uncertainty only increasing because the NO2 column is decreasing as one approaches the stack? Please clarify in the manuscript.

- I realize that this may be bordering onto a philosophical discussion, but I do not agree with the distinction the authors make between SCD uncertainty and undetectable SCD. In my view, this is two aspects of the same thing as the signal from the "undetectable SCD" is not missing, but just hidden in the noise. If the integration time is increased or more transects through the plume are averaged, then the "undetectable SCD" is reduced. The separation of these two effects may be illustrative to explain why measurements should not be done in the far field of the plume, but it is in my opinion not correct to claim that repeating measurements does not decrease the "undetectable SCD" as is stated in the manuscript. In general, I think that adding this as an additional error term is not mathematically correct.

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- The authors took a "top-down" approach for this manuscript, showing simulation results and then explaining them. While I understand that this is how science often works, it does not necessarily help in making the manuscript focused and revealing the underlying physical effects. In my opinion, a "bottom-up" approach explaining the key effects (such as dependence of VCD on wind speed, NOx/NO2 ratio on time since emission, sampling error on measurement distance) and then illustrating it in the simulation results.
- Overall, the manuscript would benefit from detailed proof reading. In some parts, in particular the description of the plume modelling, it is difficult to follow because of the use of English. In response to a suggestion from the quick-look review, the authors have in part introduced "air parcel" where they used "plume" before, but that did unfortunately not help to clarify this section. I hope that this section can be made clearer. Also, the use of "ambient SCD" is confusing and should be replaced by another formulation, maybe simply SCD.

Detailed comments

1. Abstract: Too long, please just summarize the main points instead of giving a detailed account of the study.
2. Somewhere you should have a brief discussion of those aspects of the measurements which also lead to errors, but are not treated in this study, for example stratospheric correction, uncertainty in background measurement, non-Gaussian behaviour of plume, vertical wind shear, . . .
3. Section 2: I think it would be good to have a brief description of flux derivation with car-DOAS measurements first to remind all readers of how this is done and what the relevant quantities are. Later in this section, one could then refer to this introduction.

4. Section 2.1: The wording here is in parts confusing – “emission flux simulation” should be “simulation of emission flux measurement” as it is not the emission flux itself which is simulated.

5. Table 2: I’d suggest to remove those cases which are not use in this work (D, E, F)

6. Lines 175 – 183: I’m confused by this part which suggests that some temporal variations of the wind field need to be taken into account. However, as far as I understand the simulations, this is not the case. Please clarify and if this is not used in the simulations, please remove it.

7. Section 2.2.2: Please re-read carefully and make clear where you talk about the whole plume, a transect of the plume or an air parcel within the plume. For example, in line 200 you write “[O3] is the mean concentration in the plume at time t” but in a static model as I assume you have, there are no changes with time. I assume in this case you are talking about the [O3] in an individual air parcel moving through the plume. In line 208, you write “For simplicity, we assumed that the O3 concentration within the air parcel of the plume is the same everywhere“ but I assume that you mean that [O3] is the same on a transect of the plume.

8. Section 2.2.2: In my opinion, additional assumptions were made in this section which should be mentioned, in particular that no NOx is present in ambient air as otherwise this would be mixed into the plume and more importantly, that no O3 is consumed in the reaction with NO (which is clearly not correct). Please add this to the discussion.

9. Equation 12: I think it would be good to explicitly show the dependence of R_{NOx} on (x) here.

10. Equation 13: Please introduce ΔF and l. Do you assume that there is no NO2 or SO2 outside the plume?

11. Equation 14: For clarity, ΔF and s should also have an index j here.

12. Equation 16: Why do you change notation here for the inner product? Why is the index now i and no longer j?

13. Equation 18: In order to be able to write the total error in this way, you need to assume that the errors are random, have a Gaussian distribution and are independent of each other. Is that a reasonable assumption?

14. Equation 18: Why is Q used here instead of F?

15. Line 310: I agree that GPS errors tend to cancel, but this is not necessarily true for flux errors. If one distance is too long, and this happens to be inside the plume, while the next distance is too short but is already outside the plume, the flux will be overestimated in spite of the fact that the sum of the two distances has only a small error.

16. Section 4.3.1.1: There is also the effect of the plume width decreasing with increasing wind speed which counteracts the effect of increasing “undetectable SCD”.

17. Line 498: I understand the idea of the authors that if NOx is in steady state, it is easy to compute the NO2/NOx ratio. However, at least in principle, one can estimate the NO2/NOx ratio from the ambient O3 concentration, the wind speed and the distance from the source as explained earlier in the manuscript.

18. Line 542: I don’t think that this estimate is really conservative as two assumption son the NO to NO2 conversion were made which both go into the direction of too fast conversion: 1) instantaneous mixing of O3 into the plume and 2) no consumption of O3 in the reaction with NO. In reality, the centre of the plume will have lower O3 than the outer parts and O3 levels will be generally lower than in the ambient because of the high NO concentration inside the plume.

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19. Line 554 and following: It is still not clear to me why the absolute retrieval error decreases at large distance. This is counterintuitive to me.

20. Figure 14: I do not understand why absolute flux retrieval errors are shown here – relative errors (not R2 but relative errors of the total flux) would be easier to understand.

21. Line 574: AS discussed above, I disagree with the statement that the undetectable flux cannot be reduced by multiple measurements.

22. Figure 19: As Figure 14 – why absolute instead of relative flux errors?

23. Line 701: Why do lower emission rates lead to variations in plume width? In relative units, this should not be the case.

24. Line 787: Which missing error source are you referring to?

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