

Dear Reviewer,

We have revised our manuscript based on your comments. We thank you very much for your insightful comments which we have used to greatly improve our manuscript. Below, please find our specific responses (in blue) along with each of your comments. Thank you so much for taking care of our submission.

Sincerely,

Jia Su

Comments to Authors:

1. 65-67: This statement on satellite observations is not correct. With the recent high spatial resolution observations of NO₂ by TROPOMI on Sentinel-5P since 2017, with 3.5 x 5.5. km² pixels, plumes of NO₂ by cities, power plants, and even ships can be tracked. Appropriate references to the novel TROPOMI NO₂ observations should be given, for example:

- Lorente, A., Boersma, K.F., Eskes, H.J. et al., Quantification of nitrogen oxides emissions from build-up of pollution over Paris with TROPOMI. Sci Rep 9, 20033 (2019).
<https://doi.org/10.1038/s41598-019-56428-5>

- Georgoulas et al., Detection of NO₂ pollution plumes from individual ships with the TROPOMI/S5P satellite sensor, Environ. Res. Lett. 15, 124037, 2020

We modified our statement on satellite observations according to reviewer's comments and added above two references for the manuscript.

2. 74: a reference to an NO₂ sonde system should be given: Sluis, W. W., Allaart, M. A. F., Pijters, A. J. M., and Gast, L. F. L.: The development of a nitrogen dioxide sonde, Atmos. Meas. Tech., 3, 1753–1762, <https://doi.org/10.5194/amt-3-1753-2010>, 2010.

We added the reference for the manuscript.

3. Section 2:

Here consistency is needed, and proper introduction of formulae.

- 1. 79: 24x7 operation
- 1. 85: absorption > absorbing

- l. 87: quant
- l. 82 ff: subscripts that are words, like “on” and “off”, should be in upright font.
- Please give the units of all quantities used: λ , β , α , etc. etc.

We modified them according to reviewer’s comments.

- Eqs. 1-3:
 - Are these relations only valid for an upright pointing lidar? **NO, they are valid for all lidars.**
 - Both capital Z and small z are used as variables. Are they the same height variable?? This consistency question holds throughout the paper. **Both capital Z and z are height variable. The capital Z is the height of lidar signal, while z is height variable of integral formula.**
 - Please give the units of X, C1, C2, C3, σ , N, etc. **We added them in our manuscript.**

l. 124 and other places: acronyms like AED should be in upright font; only symbols are in slant font. **We modified them according to reviewer’s comments.**

- l. 116: derivatives w.r.t. which variable? **We modified it. “The molecular density of the gas of interest can be obtained using Eqs. (1), (2) and (3).”**
- What does the D mean in AED, MED, OAD ? **D means difference.**
 - l. 129: unclear sentence. The text of the method description should be clarified. **We modified it. “For correction of AED and B, we need accurate aerosol measurements. However, accurate aerosol measurements are not easily to be obtained. From the above NO₂ retrieval relative equation, AED are determined by the three wavelengths, so how to choose the three wavelengths is very critical to reduce AED and improve the NO₂ retrievals accuracy.”**

4. In comparison to Eqs. 4-9, how does the two-wavelength DIAL NO₂ retrieval equation look like? This is relevant for the comparisons shown later on. **We added the two-wavelength equations as supplements.**

5. The description of the A, B, C methods in Sect. 2 should be improved:

- l. 135-136: These methods A and B have not been introduced yet. Please give a name for the methods: increasing absorption, decreasing absorption, and maximum in absorption **We used suggested increasing absorption, decreasing absorption, and maximum absorption as names for methods A, B and C.**

- l. 137: missing reference. Or should it be Liang? **We added the missing reference.**

- l. 142-143: please compare to the two-wavelength DIAL equation. Please explain why eq. 12 is better. **We added explanation for it.**

- Eq. 12: is the +-sign the most important difference between the three methods? **We added them according to reviewer's comments.**

- l. 152: please first introduce the derivation of Eq. 13. **We revised it.**

- l. 153: what does K represent? what is the relation to the earlier equations? **K is part of Eq.(7) which is related to three wavelengths.**

- l. 156: please give some examples for the three wavelengths driven by K. **We added some examples in manuscript.**

- Eq. 13: please first show the equation, then discuss it. **We revised it according to reviewer's comments.**

6. Sect. 3:

- l. 169: what about the wavelengths below 400 nm? **NO₂ have strong absorption between 420 nm and 450 nm, so we selected the wavelengths above 400 nm.**

- l. 172: what about the relative weight of the two rules/criteria?

- l. 177: how does the two-wavelength NO₂ retrieval equation looks like? **We added the two-wavelength equations as supplements.**

- l. 182: please give the physical unit of the lidar ratio. **The lidar ratio is the ratio of aerosol extinction coefficients to aerosol backscattering coefficients.**

- l. 190: which HITRAN version? **The version is 1.1.2.0**

- l. 191: e in italics **We revised it.**

- l. 193-194: this line colour code information belongs in the figure caption. **We revised it.**

- l. 200: far less > much smaller **We used "much smaller" instead of "far less". .**

- l. 223: This is a strange order of this section: first instrument description, then simulation, and then again instrument part. Please restructure section 3 into two subsections: (1) instrument description, (2) simulation of the retrieval. **We adjusted order for the section in our manuscript.**

- l. 239: missing words at the end of this sentence ? **Yes, we revised it.**

(7) Sect. 4:

This section on error analysis requires drastic improvement: clarification, better introduction of equations, consistency with the rest of the paper, correction of grammar and typo's.

- l. 242: ... from standard uncertainty: please explain., **Leblanc introduced standard uncertainties for Lidar (Eq.(2) in his reference)**
- Eq. 14: what is the unit of U? **The unit of U is “%”.**
- l. 246: and not discussed in this work: unclear. **We added relative uncertainties for Lidar signal noise.**
- l. 249: what is the uncertainty function u? please give reference. **u are standard uncertainties. Capital U are relative standard uncertainties. And we added a reference for it.**
- l. 249: how are these equations 15-18 derived? please first show the equations, and then explain the variables in them. **We revised it.**
- l. 250: in Section 2 the subscript ‘a’ means aerosols, and ‘m’ means molecules. It is very confusing that here ‘a’ means air. Please be consistent. **We used “m” instead of “a”.**
- l. 260 ff: 8-hour or eight-hour: be consistent **We revised it.**
- l. 272: Fernald’s method: give reference. **We added a reference for Fernald’s method.**

8. Sect. 5:

- What is vertical resolution of the measured NO₂ profiles? **The vertical resolution of measured NO₂ profiles is 100 m.**
- l. 327-328: ... good agreement between the OMI ... > with the OMI satellite measurements **We revised it.**
- Fig. 12: what is the reason of the still large differences between measurement and model ?

Matthew S. Johnson (co-author) did a lot work about comparing NO₂ profiles from aircraft measurements and WRF-Chem model, and said NO₂ profiles from WRF-Chem model are generally a little lower than real NO₂ profiles.

Figures

- Captions should be self-explanatory! **We revised it.**
- Fig. 2: unclear alignment of the wavelengths in the legend. What is the source of this NO₂ absorption cross-section spectrum? Caption: strong absorption cross-section spectrum ...

We downloaded absorption data of NO₂ from Hitran and plotted it using Matlab.

We revised alignment of the wavelength in the legend.

- Fig. 3: Are this figure and table taken from the manufacturer's brochure? It should be a new figure for this manuscript, otherwise there is a copyright issue. **Yes. Thanks! We redo them.**
- Fig. 5: Please indicate in the legends whether it is 2- or 3-wavelength DIAL. Explain the quantities in the caption. What is meant with the x-axis label difference of ... ? between what and what? **We revised it according to reviewer's comments.**
- Please combine figures 8, 9 and 10. Only e is varying. Explain the legends in the caption. **We revised it according to reviewer's comments.**
- Fig. 11, caption: 'except U_s': what does this mean?? explain what e is. what is TU? explain the legends. **We revised it according to reviewer's comments.**
- Fig. 12: explain the black error bars in b and d. **We added an explanation for them.**

Textual

There are several typos and grammatical mistakes. Please carefully check the English language throughout the manuscript.

Often, the article is missing, e.g. on l. 36: ... from the WRF-Chem model....

The singular/plural should be checked, e.g. l. 42: ... the main emission sources ...

Check typography of the references.

We revised it according to reviewer's comments.