

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. A track changes version of the manuscript shows all changes marked with red.

Thank you so much for taking care of our submission.

Sincerely,

Jia Su

Comments to Authors:

1. In section 2 it would be useful to cite a few examples of prior works on three-wavelength (or dual-DIAL) lidar analysis. (There are many available, and previous work is suggested in lines 135ff, but that citation is missing/mislabeled in the references section.)

We added a few examples of prior works on three-wavelength lidar analysis from line 137 to line 141 and the missing reference at line 463.

“Wang used three wavelengths corresponding to the strong, medium and weak absorption of O₃ to obtain an accurate stratospheric ozone profile in the presence of volcanic aerosols. Liu used three wavelengths of 448.10nm, 447.20nm and 446.60 nm corresponding to the strong, medium and weak absorption of NO₂ to retrieve NO₂.”

2. In equation 5, it would be helpful to label this term as NAD since it is described as this in the text. (e.g. “NAD = $\Delta\sigma N$ = ...”)

The $\Delta\sigma N$ is the absorption cross section difference for NO₂. In general, σ is expressed as cross section, so we think $\Delta\sigma N$ is better than the expression of NAD.

3. In equation 7, it would be useful to include the term K that will be used later (e.g. “AED = ... = $K\alpha\alpha(Z)$ ”)

We revised it at line 121 according to reviewer’s comments.

4. Since the three-wavelength results are compared against the two-wavelength results throughout this paper, it would be useful to readers to include the two-wavelength equations that correspond to equations 4 through 9.

We added the two-wavelength equations as supplements.

5. In section 2a, it would be useful to note that DIAL systems for other atmospheric gases like ozone, it is only practical to use wavelength selection Method B because of the shape of the ozone absorption spectrum (lacking narrow peaks). The shape of the absorption spectrum of NO₂ allows for an especially favorable three-wavelength analysis using Method C because it is possible to choose the points spanning over the peak as shown in figures 1 and 2.

We added it from line 147 to line 149 according to reviewer's comments.

“However, for DIAL systems to measure other atmospheric gases like ozone, it is only practical to use wavelength selection Method B because of the shape of the ozone absorption spectrum (lacking narrow peaks).”

6. In sections 2 and 3, it is mentioned that the wavelengths are optimized according to the rules a. (maximize NAD) and b. (minimize AED), but this is a multivariate optimization. It would be useful to provide more detail of the optimization process and how the authors arrived at the final wavelengths.

Our two rules are increasing absorption cross section difference of NO₂ and decreasing AED. From Eq.12, AED can be determined by the value of K. We can choose the appropriate three wavelengths to make the value of K equal or close to 0, the value of AED will be equal or close to 0. We added explanation for it from line 179 to line 180. We simulated B using three-wavelength Dial technique and two-wavelength Dial technique, and found that three-wavelength Dial technique can decrease the value of B.

“Combining the OPO laser energy outputs, NO₂ absorption spectral and two three-wavelength chosen rules, 438 nm, 439.5 nm and 441 nm shown in Fig. 2 result in the wavelengths of HU three-wavelength DIAL system because $\Delta\sigma_N$ of the three-wavelength pair is more than other three-wavelength pairs in NO₂ strong absorption spectral zone and the K value of the three-wavelength is close to 0.”

7. There are two sections of text (lines 163ff and lines 223ff) that describe the lidar hardware and should be combined.

We revised it at line 229 according to reviewer's comments.

8. In section 4, Disregarding the uncertainty introduced by the lidar signals, U_s , should not be taken lightly. In particular, because this term is the result of taking a derivative of (logs of ratios of) signals, it can be very susceptible to noise in the raw signals. The modelled analysis in this paper uses relatively noise-free aerosol and ozone profiles (figure 4) which reduces this issue and facilitates evaluation of the optimizations presented. It is noted that the lidar signals in section 4 were integrated for 2 minutes to reduce the signal noise, and as a result, the resulting NO₂ profiles are relatively smooth. However, there should be some discussion of this noise source and its contribution to the resulting NO₂ profile uncertainty. This provides readers with an estimation of the relative contribution of signal noise which ultimately depends on lidar specifications (e.g. power and aperture) as well as on temporal and spatial resolution (i.e. averaging).

We added an equation (line 263) and analysis (between line 287 and line 295) for uncertainties owing to noise of three-wavelength lidar signals.

$$U_s(z) = \frac{u \left\{ \frac{1}{2} \times \frac{d}{dz} \left[\ln \frac{X(\lambda_1, Z)(\lambda_3, Z)}{X(\lambda_2, Z)^2} \right] \right\}}{N_N(Z) \Delta \sigma_N} =$$

$$\frac{1}{2} \times \frac{\sqrt{\left\{ \frac{d \left\{ \frac{d}{dz} \left[\ln \frac{X(\lambda_1, Z)(\lambda_3, Z)}{X(\lambda_2, Z)^2} \right] \right\}}{d[X(\lambda_1, Z)]} \times u[X(\lambda_1, Z)] \right\}^2 + \left\{ \frac{d \left\{ \frac{d}{dz} \left[\ln \frac{X(\lambda_1, Z)(\lambda_3, Z)}{X(\lambda_2, Z)^2} \right] \right\}}{d[X(\lambda_2, Z)]} \times u[X(\lambda_2, Z)] \right\}^2 + \left\{ \frac{d \left\{ \frac{d}{dz} \left[\ln \frac{X(\lambda_1, Z)(\lambda_3, Z)}{X(\lambda_2, Z)^2} \right] \right\}}{d[X(\lambda_3, Z)]} \times u[X(\lambda_3, Z)] \right\}^2}}{N_N(Z) \Delta \sigma_N} \quad (18)$$

From Eq. (18), U_s is determined by uncertainties of three-wavelength lidar signals. The uncertainties of lidar signals with average integration time of 1 minute and 2 minutes are derived from Poisson statistics associated with the probability of detection of a repeated random event [Measures, 1984; Leblanc et al., 2016]. NO₂ number density relative uncertainty owing to the noise of lidar signals with average integration time of 1 minute and 2 minutes are obtained shown in Fig11. We can see U_s using two-wavelength DIAL technique is smaller than using three-wavelength DIAL technique. With increase of average integration time from 1 minute to 2 minutes, U_s can be effectively reduced.

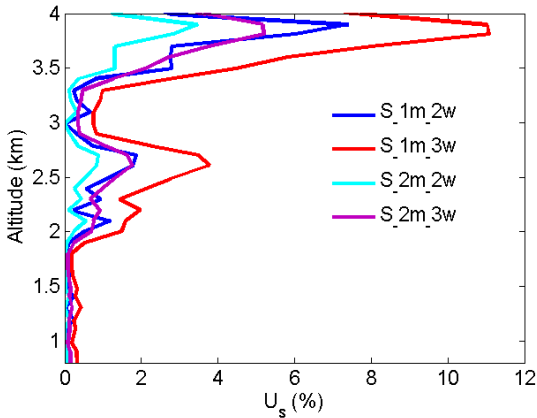


Fig.11 NO₂ number density relative uncertainty owing to the noise of signals with average of 1 minute and 2 minutes.

9. In section 4, the notation of “Na” and “σa” for number density of air and Rayleigh (air) scattering cross section, respectively, might be less confusing as “Nm” and “σm” to be consistent with the rest of the text where “m” denotes molecular terms and “a” denotes aerosol terms.

We revised them from line 254 to line 256 according to reviewer’s comments.

10. In section 4 (line 272), a reference to Fernald’s paper describing the lidar inversion procedure should be provided.

We added a reference to Fernald’s paper at line 430 according to reviewer’s comments.

“Fernald, F. G., Herman, B. M., and Reagan, J. A.: Determination of aerosol height distributions by lidar, *Journal of Applied meteorology.*, 11(3), 482-489, 1972”

10. In section 5 (line326), the vertical resolution of the WRF-Chem results should be provided since the comparison with the lidar will be in this dimension.

We added it at line 343 according to reviewer’s comments.

“As a first-order assessment of the HU lidar NO₂ profiles, we compare the retrieval results to simulated data from the Weather Research and Forecasting Chemistry (WRF-Chem) model (Grell et al., 2005) at 12 km × 12 km spatial resolution and 200 m vertical resolution. “

11. Typographic errors:

Line 118 (equation 4), the numerator of the lidar signal term should be $X(\lambda_1, Z)X(\lambda_3, Z)$.

We revised it at line 118.

Lines 193-194, "...light blue lines are NAD." should be "...light blue lines are OAD."

We revised it at line 200.

Line 217 (figure 5 caption), Add "B" to the list of values described as shown on the graphs.

We added it at line 223.

Line 228 "NO2" should be "NO₂"

We revised it at line 234.

Line 239, "NO" should be "NO₂"

We revised it at line 245.

Line 257 (equation 18), "In" should be "ln"

We revised it at line 261.

Lines 271ff, Font used for "αα" is different than that used elsewhere (e.g. compare with line 109).

We revised it from line 265 to line 270.

Line 309, "Fig. 8" should be "Fig. 12"

We revised it at line 326.