Authors' Response to Reviewer #2's Comments

Dear Reviewer,

Thank you for your comments concerning our manuscript entitled "Tropospheric ozone sensing with a differential absorption lidar based on single CO_2 Raman cell" (MS NO: egusphere-2024-1853). Those comments are all valuable and very helpful for revising and improving our paper. We have studied the comments carefully and have made corrections which we hope will get your approval. Our point-to-point responses to the reviewer's comments are as flowing:

General comments:

The manuscript presents a novel setup of ozone lidar using CO_2 Raman tube and its application in several field measurements. The instrument seems to be very promising. The result of the comparison between lidar and vertical measurement was fantastic. Though there are plenty of sloppy writing problems and a few technique questions, current manuscript is worthy to publish. I would recommend a major revision after addressing the following comments.

Reply: Thanks for the comments.

Specific comments:

1. Line 50: 'Fuel laser'? Or dye laser?

Reply: Thanks for the comments. "fuel laser" have been amended as "dye laser" on line 48, 50, 52, 53.

2. Line 57: H_2 and D_2

Reply: Thanks for the comments. "H_2, D_2" has been amended as "H_2 and D_2".

3. Line 58: Lidar can not be deployed by lidar.

Reply: Thanks for the comments. "The atmospheric ozone profiling Lidar developed by National Oceanic and Atmospheric Administration (NOAA) deploys a scanning four-wavelength ultraviolet differential absorption lidar" has been amended as "National Oceanic and Atmospheric Administration (NOAA) deployed a scanning four-wavelength ultraviolet differential absorption lidar".

4. Line 61-65: Please rephrase this sentence.

Reply: Thanks for the comments. "This lidar measures tropospheric ozone and aerosols based on Raman shift wavelengths produced by D_2, H_2 Raman gases pumped by the frequency-quadrupled Nd:YAG lasers. There are two main problems in using the D_2, H_2 dual Raman cells: One is that the D_2, H_2 Raman cells share the frequency-quadrupled Nd:YAG laser and places higher requirements on the pump laser; the other is that the overlaps of the receiver field of view and laser beam divergences are different, which enlarge the blind area of the ozone detection, whereas the ozone lidar using the CO_2 single Raman cell can overcome these drawbacks." has been amended as "The lidar system measures tropospheric ozone and aerosols by utilizing the Raman shift wavelengths generated from D_2 and H_2 gases. However, there are two primary challenges associated with employing the D_2 and H_2 dual Raman cells: (1) Shared Laser Resource: The D_2 and H_2 Raman cells are both pumped by the same frequency-quadrupled Nd:YAG laser. This shared resources places increased demands on the pump laser's performance and stability. The lase must provide sufficient energy to effectively pump both Raman cells. (2) Receiver Field of View and Laser Divergence Overlap: The second challenge arises from the varying overlaps between the receiver's field of view and the divergences of the laser beams for the D_2 and H_2 Raman cells. These differences can result in a larger blind area during ozone detection. The blind area refers to the region where the lidar system is unable to accurately measure ozone concentrations due to the geometric constraints of the laser beam and the receiver's field of view. This can lead to incomplete or inaccurate data regarding the ozone levels in the troposphere. In contrast, an ozone lidar system utilizing a CO_2 single Raman cell has the potential to address these issues. The single Raman cell design simplifies the system by eliminating the need to manage two separate Raman cells, thereby reducing complexity and the need for Nd:YAG. Furthermore, the single Raman cell system may offer a more consistent overlap between the receiver's field of view and the laser beam, which can help to minimize the blind area and enhance the accuracy of ozone detection.".
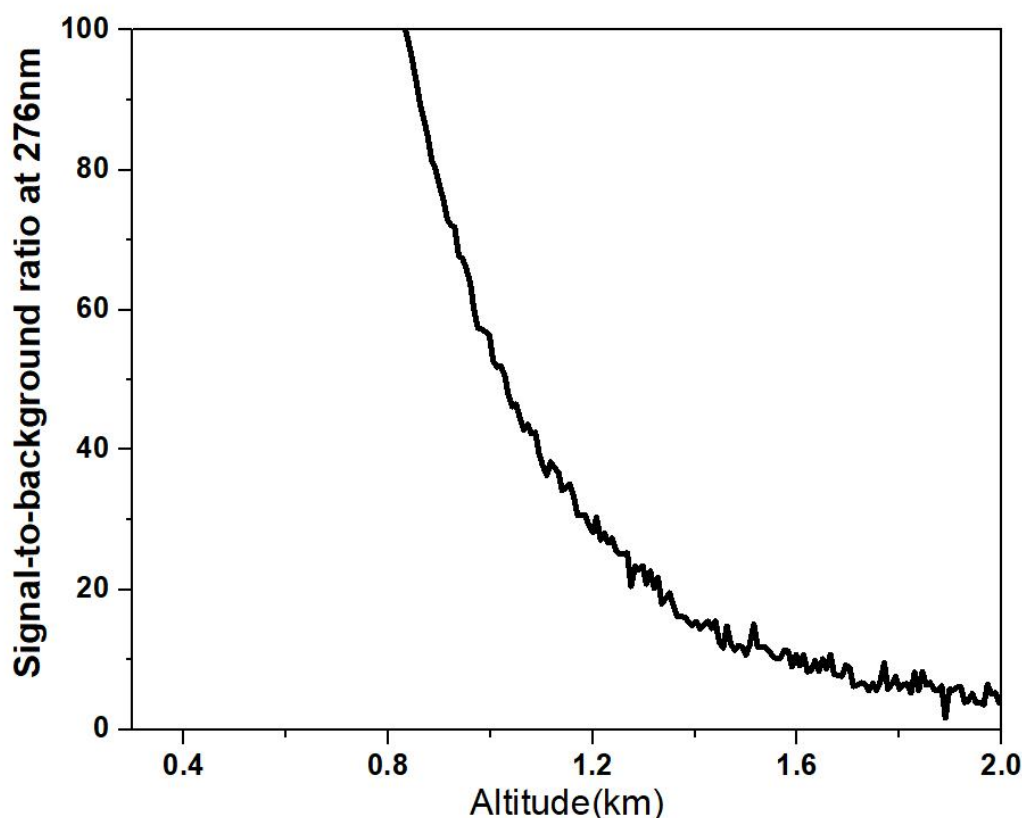
5. Line 108: Nd:YAG. The sentence also needs to be improved.

Reply: Thanks for the comments. "Nd: YAGA quad-frequency laser pumped single D2 Raman tube generates first-order stokes light and second-order stokes light, corresponding to the differential absorption wavelengths of 289 nm and 316 nm." has been amended as "The Nd:YAG quad-frequency laser, when used to pump a single D_2 Raman tube, generates both first-order and second-order Stokes light. These correspond to the differential absorption wavelengths of 289 nm and 316 nm, respectively.".

6. Line 118: Give the reason why 276nm laser can only be used under 600m.

Reply: Thanks for the comments. Due to the strong absorption of ozone at 276nm and the strong atmospheric backscatter at this wavelength, the detection height of the 276nm wavelength signal is limited. As shown in the figure, the signal-to-background ratio of the 276nm signal is greater than 100 below 600m, which meets the detection requirements with sufficient signal-to-noise ratio. Above 800m, it quickly drops

below 100. To accommodate different aerosol types and weather influences, and considering that aerosols are mainly distributed below a height of 600m, a height of 600m was adopted as the stitching height for the differential wavelength pair.

7. Line 127-129: SO$_2$ interference should be addressed in detail, here and in section 4

Reply: Thanks for the comments. The table below analyzes the extent of interference from NO$_2$ and SO$_2$ gases. The interference from NO$_2$ at the 276.2nm/287.2nm wavelength pair and the 287.2nm/299.1nm wavelength pair is 0.98% and 3.5% of the NO$_2$ concentration, respectively, which can be neglected. The impact of SO$_2$ on ozone is more significant, with impacts of 8.9% and 34.7% of the SO2 concentration at the two wavelength pairs. The typical environmental concentration of SO$_2$ is a few (ug/m³). If assessed at 10 ug/m³, its impact would be approximately 0.89 ug/m³ and 3.5ug/m³, which is relatively small compared to other sources of error and is therefore usually not considered. Another factor is that if the interference effect of SO$_2$ concentration is taken into account, it would also be necessary to measure the spatiotemporal distribution of SO$_2$ in real time, which would undoubtedly be very costly.

Wavelength pairs (nm)	SO$_2$(e^{-20}cm^2)	NO$_2$(e^{-20}cm^2)	O$_3$(e^{-20} cm^2)	NO$_2$ interference	SO2 interference
276.2/287.2	30	3.3	335.43	0.98% * NO$_2$ Concentration	8.9% * SO$_2$ concentration

287.2/299.1	52.8	5.4	152.21	3.5%*NO₂ concentration	34.7% * SO₂ concentration

8. Line 147: How about the rest power of the pumping laser? Does 266nm laser still exist in the output laser beam?

Reply: Thanks for the comments. The 266nm laser still exist in the output laser beam, with the residual single-pulse energy at 266nm being approximately 14mJ.

9. Line 166-167: Please rephrase this sentence. The reason to use JGS1 quartz should be mentioned.

Reply: Thanks for the comments. "JGS1 quartz material is adopted in the three sets of high-reflection flat concave mirrors." has been amended as "The three sets of high-reflection flat concave mirrors are constructed using JGS1 quartz material, which is chosen for its superior optical properties and resistance to laser damage, ensuring high reflectivity and durability in the system."

10. Line 199: What is the meaning of 4000 laser pulse? Total measurement time?

Reply: Thanks for the comments. 4000 laser pulses correspond to the raw data acquisition time. Given that the laser operates at a frequency of 10 Hz, 4000 laser pulses equate to a data acquisition period of 400 seconds.
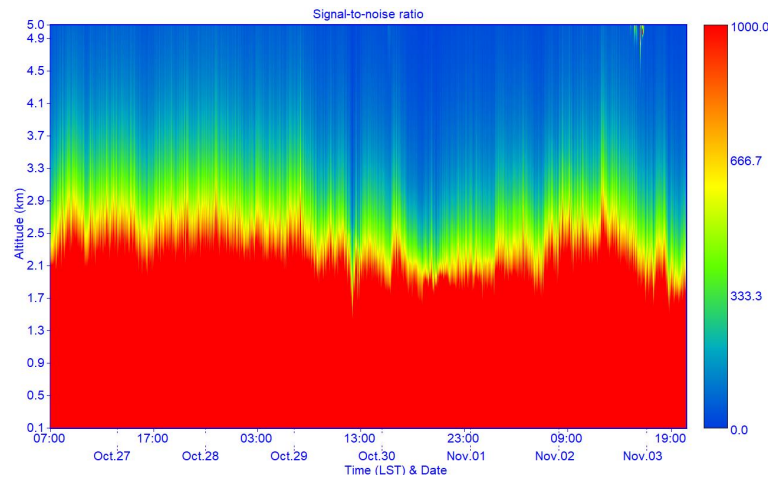
11. Line 204: How to determine the statistical error should use more serious statistics. Currently the error estimation method as well as clear conclusion of the estimation are missing. The same for section 4.2.

Reply: Thanks for the comments. The statistical error is

$$\frac{\delta(N(z))}{N(z)}$$
$$= \frac{1}{2\Delta\delta\Delta z N(z)} \sqrt{\frac{\delta P(\lambda_{on}, z)^2}{P(\lambda_{on}, z)^2} + \frac{\delta P(\lambda_{on}, z+\Delta z)^2}{P(\lambda_{on}, z+\Delta z)^2} + \frac{\delta P(\lambda_{off}, z)^2}{P(\lambda_{off}, z)^2} + \frac{\delta P(\lambda_{on}, z+\Delta z)^2}{P(\lambda_{on}, z+\Delta z)^2}}$$

The statistical error of ozone lidar data is inversely proportional to the absorption cross-section difference, the difference distance, the unknown gas concentration, and the SNR of the ozone data. The statistical error of the ozone lidar is related not only to the hardware of the device but can also be considered constant in the short term, aside from its dependence on atmospheric conditions and solar irradiance. Generally, due to the influence of solar irradiance, the signal-to-noise ratio (SNR) of daytime signals is typically lower than that of nighttime signals. During the observation period from October 26 to November 3, 2018, the SNR of the 299.1nm signal remained essentially stable as shown in the blow figure. Therefore, the statistical error of ozone at 11:00 to 12:00 on October 26, 2018, was used to analyze the performance of the ozone lidar. It

is important to note that an ozone lidar is an in situ measurement device that is closely related to atmospheric conditions, and its SNR can drop sharply during extreme weather conditions such as rain or fog, leading to a significant increase in statistical error.

12. Line 229: I can not find where the aerosol extinction coefficient is less than 0.3 km^{-1} in Fig.7.

Reply: Thanks for the comments. "0.3 km" has been amended as " 0.5km".

13. Line 236: 'times'?

Reply: Thanks for the comments. " Figure 8(a) shows the times of the aerosol correction term" has been amended as "Figure 8(a) shows the aerosol correction term"

14. Line 251: Something missing before 'therefore'.

Reply: Thanks for the comments."It indicated that the aerosol correction term changes rapidly in the process of aerosol transport and when the boundary layer aerosol concentration is high" has been amended as "The analysis indicates that the aerosol correction term exhibits rapid fluctuations during the transport of aerosols, particularly when the concentration of boundary layer aerosols is elevated."

15. Line 285: Fig.12 is not readable.

Reply: Thanks for the comments. We have redrawn fig.12.