Response to Referee #4:

The authors greatly appreciate the helpful comments of referee #4. In the following, we present our point-by-point responses to the Referee #4. The referees' comments are in blue italic and our responses are in black. We have made appropriate changes in the revised manuscript by taking the comments into account.

Main comments:

1. I find the quality of this work to be questionable, as will be revealed by several comments below. With this in mind, I don't really have a compelling reason to see this paper published. Had the authors succeeded in demonstrating the utility of the 1370 nm band with real data, I would be in favour of publication, but, as is, it could be that the lack of success is simply due to weaknesses in their method (see specific weaknesses below and also the neglect of forward scattering).

Responses: Thank you for your comment. The PWV results obtained by using the water vapor absorption band near 1370 nm with the real data. Figure 5 and Figure 11 in this revised manuscript shows the same conclusion, that is, for a dry atmosphere, the PWV obtained by using the water vapor absorption band near 940 nm (BAND1) is higher than that of the band near 1370 nm (BAND2). It can be seen from Fig. 11 that the PWV retrievals of BAND1 are closer to those of CE-318 sun photometer, which demonstrated that the algorithm is OK, because it is not easy to say which PWV obtained by using BAND1 and BAND2 is closer to the 'true' value, Therefore, we used the model to simulate some theoretical spectra for the inversion test. In the test results (Fig. 5), the PWV retrievals obtained by using BAND1 are also higher than those of BAND2, which is consistent with Fig. 11, while the PWV retrievals from BAND2 are closer to the theoretical inputs, which are assumed to the "True" value. Therefore, we proposed that for dry environment, we can try to introduce the stronger water vapor band around 1370 nm for PWV inversion when measurements are available.

Thank you for pointing out the problem of the forward scattering issue due to the large FOV of the EKO instruments (5°), we did neglect the possible impact in previous algorithm. As shown in Fig. 1, the Circumsolar radiation Ratio (CR) at infrared wavelengths (870 nm) is very small, which will not affect the water vapor retrieval. However, in the inversion of aerosol optical depth, especially for high AOD loading atmosphere and at shorter wavelengths, it is sure necessary to perform CSR correction. We have added CSR correction in the revised manuscript.

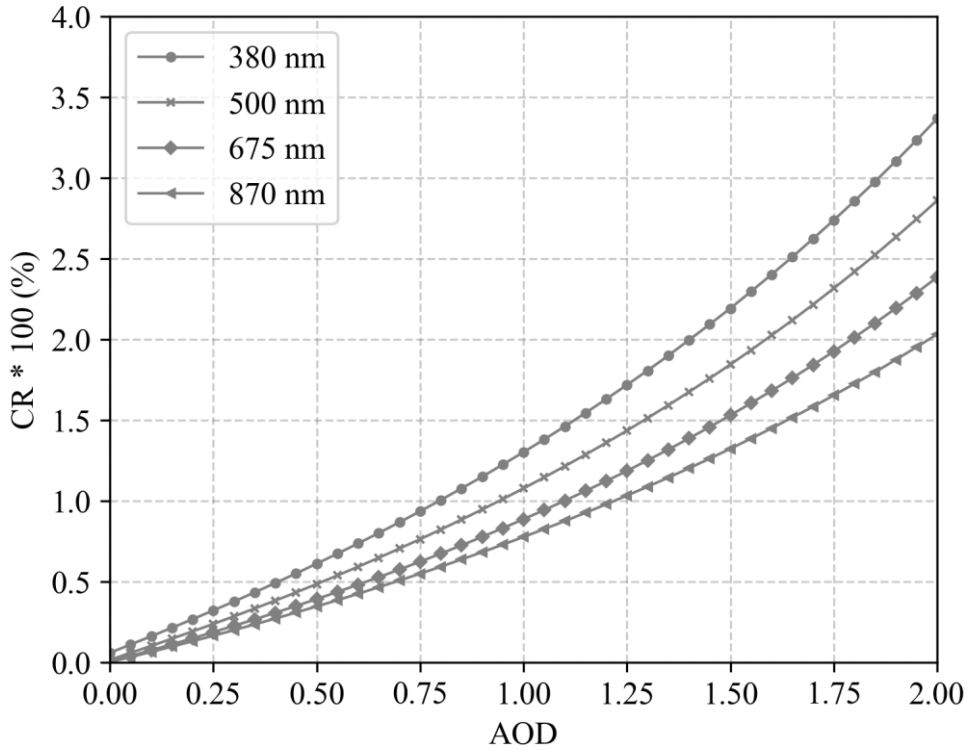

Figure 1. Simulations of CR * 100 (%) for SZA 30 ◦ with AOD from 0 to 2, at 380 nm, 500 nm, 675 nm and 870 nm, for 2020 annual average MERRA2 aerosols data in Beijing area for FOV between 1.2 ° and 5 °.

2. I agree with many of the comments raised previously by Reviewer 1 and find that Reviewer 2 was not thorough. Specifically, the point raised by Reviewer 1 about MS711 and MS712 being very different instruments is noteworthy. These instruments should be validated separately. Also, MODTRAN4.3 is used, which relies on HITRAN1996 (I believe). This is outdated. MODTRAN 5 and 6 are available and the

spectroscopic parameters for water vapor have changed. I know that the Brown et al., 2002 parameters are used in MODTRAN 5.2 for example:

"Brown, L. R., Toth, R. A., and Dulick, M.: Empirical line parameters of $H2^{16}O$ near 0.94 μm: Positions, intensities and air broadening coefficients, J. Mol. Spectrosc., 212, 57–82, 2002."

Responses: Thank you for your comment. We have accepted the suggestions and comments proposed by reviewer 1. MS711 and MS712 have the same full width at half maximum (FWHM) and wavelength accuracy, and the possible impact of the differences between the two instruments has not been considered in the relevant applications of this manuscript. We noticed that the current version of MODTRAN4.3 may be old, so the water vapor transmittances were calculated under the same conditions using MODTRAN5.2 and MODTRAN4.3. Figure 2 shows the results of water vapor transmittances calculated using two versions of MODTRAN model. Because a narrow band other than single wavelength is used, the water vapor transmittances calculated by MODTRAN4.3 and MODTRAN5.2 are almost the same.

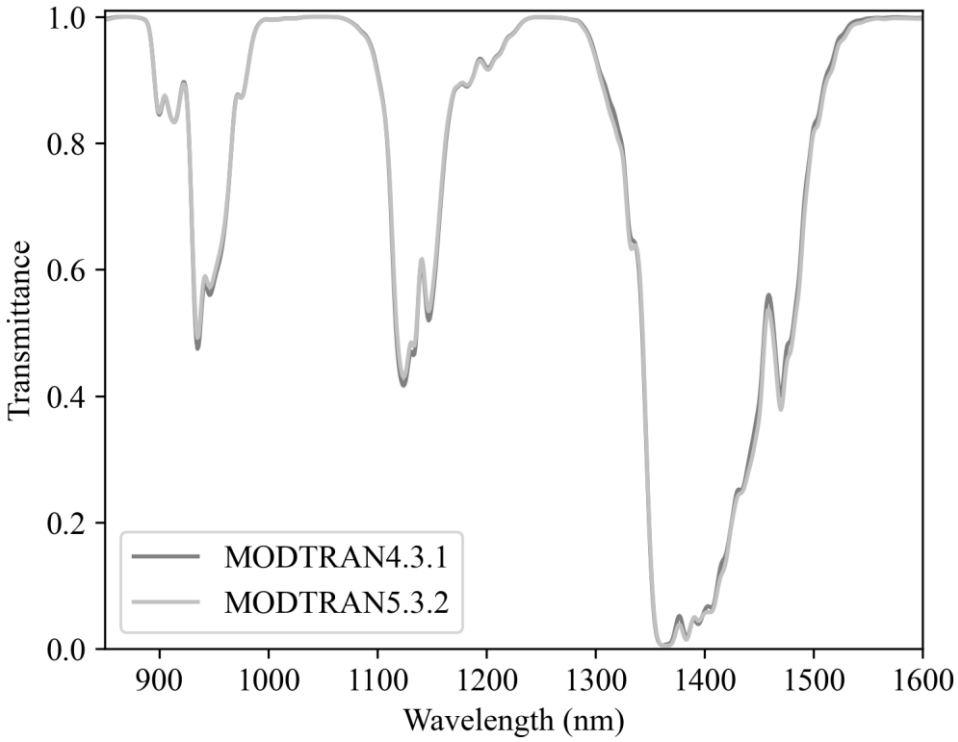

Figure. 2 Water vapor transmittances calculated by MODTRAN4.3 and MODTRAN5.2 under the same conditions, respectively (SZA=0 °, PWV=1.0 g*m^{-2}, FWHM=7 nm).

3. It seems that the main point of Fig. 3 was to show the absorption due to water vapour at ~940 nm is small when PWV is 0.5 cm, but the figure shows that absorption is quite strong. The comparison of the suitability of the 940 and 1370 nm bands for PWV retrieval is one of the main points of the paper. The authors appear to hypothesize that the stronger 1370 nm band would be more suitable for a dry atmosphere, but their results with real data do not make a strong case for this longer wavelength band.

Responses: Thank you for your comment. Figure 3 in the manuscript shows that the water vapor absorption at 940 nm decrease significantly with the decrease of PWV in the atmosphere, but the water vapor absorption near 1370 nm is still strong. Therefore, we suppose that the water vapor band near 1370 nm may be more suitable for drier atmosphere, which was proved with simulated retrievals by radiative transfer modelling.

4. 5% (relatively) uniform noise throughout the two bands is not realistic. A proper instrument model should be used since SNR will decrease at wavelengths for which the transmitted irradiance is low in the case of a grating spectrometer. This could be why the results are quite different between simulated data (Fig. 5) and real data (Fig. 10).

Responses: Thank you for your comment and sorry for the unclear sentences given in previous manuscript. In the inversion test, the noise imposed on the theoretical spectrum is not a uniform 5% noise, but a random noise within ±5% superimposed on each wavelength. The results in Fig. 5 and Fig. 11 (previous Fig. 10) are consistent, that is, for dry atmosphere, the PWV retrieved by using band near 940 nm are higher than those of the band near 1370 nm. As shown in in Fig. 5, the results of PWV obtained from band near 940 nm are also shown a relatively larger uncertainty due to the noise added in the simulated spectrum.

Specific comments

1. L17: "CE-318" -> "CE-318 sun photometer."

Response: Thank you for your comment. We have corrected it in the revised manuscript (line 21).

2. Delete "which shows that" and start a new sentence with "The two …"

Response: Thank you for your comment. We have revised it in the manuscript (line 21).

3. L30: I don't know if acronyms need to be defined in the main body of the paper to make it independent of the abstract.

Response: Thank you for your comment. We checked some articles that such acronyms are allowed.

4. L31 (and L41): "etc." should not be used like this. "and others" is preferable.

Response: Thank you for your comment. We have corrected it in the revised manuscript (line 32 and line 41).

5. L41: I suggest deleting "etc." here.

Response: Thank you for your comment. We have revised it in the manuscript (line 41).

6. L44: Define PHOTONS

Response: Thank you for your comment. We have included the full name of PHOTONS in the manuscript (line 45).

7. L44: CE-318" -> "the CE-318"

Response: Thank you for your comment. We have revised it in the manuscript (line 46).

8. L48: Delete "the"

Response: Thank you for your comment. We have revised it in the manuscript (line 50).

9. L51: This sentence does not give the spatial domain. Is it a global comparison? 4 references are provided at the end of the sentence but it is not clear which one contains the standard deviation of PWV retrieval differences and the overestimation of AOD.

Response: Thank you for your comment. We have described it in detail in the manuscript (lines 52-55). The PWV retrievals difference between PSR and CIMEL is quoted from Table 2 of Kazadzis et al. (2014). The AOD retrievals difference between PSR and CIMEL is quoted from Figure 3 of Kazadzis et al. (2018a) and also described in Kazadzis et al. (2014).

10. L52: End this long sentence after "CE-318".

Response: Thank you for your comment. We have revised it in the manuscript (line 55).

11. L52: The second half of this sentence "and the PWV given…" does not make sense. What is meant by "integration"? 940 nm is mentioned, so what are the "different wavelengths"?

Response: Thank you for your comment. Sorry for the error in our description, it has been revised in the manuscript (line 57-58).

12. L52: "single" -> "a single"

Response: Thank you for your comment. This has been corrected it in the revised manuscript.

13. L63: environment -> environments

Response: Thank you for your comment. We have corrected it in the revised manuscript (line 66).

14. L63: There should be a sentence stating whether and why 1370 nm would be less useful in humid environments.

Response: Thank you for your comment. We have added some explanations to the manuscript (lines 64-66).

15. L88: need -> needs

Response: Thank you for your comment. We have corrected it in the revised manuscript (line 90).

16. L100: Delete the two consecutive sentences starting with "The light grey"

Response: Thank you for your comment. We have corrected it in the revised manuscript (line 103).

17. L107: 0 ° solar -> 0 ° solar

Response: Thank you for your comment. We have revised it in the manuscript (line 106).

18. L113: atmosphere -> atmospheres

Response: Thank you for your comment. We have revised it in the manuscript (line 112).

19. L146 (Table 2): mode -> model

Response: Thank you for your comment. We have revised it in the manuscript (line 146).

20. L146 (Table 2): "Altitude" –> "Altitude of surface"

Response: Thank you for your comment. We have revised it in the manuscript (line 146).

21. L146 (Table 2): DISORT is not needed for modelling transmittance. DISORT is used when scattered light into the field of view is considered.

Response: Thank you for your comment. We have revised it in the manuscript (line 146).

22. L151: Give the update equation (i.e. how is the increase/decrease of PWV calculated from iteration to iteration). Is Chahine's method used?

Response: Thank you for your comment. We did not use Chahine's method, and the increase or decrease of PWV depends on whether Δ in Eq. 6 in the manuscript is positive or negative as described in lines 148-150.

23. L156: The PWV might be randomly generated but it the upper bound is clearly not 0.5 as shown in Fig. 5.

Response: Thank you for your comment. The PWV we input is a random number within 0-0.5 cm, but the curves where the transmittance reaches 0 are filtered out, which leads to the fact that the maximum value of PWV in the Fig.5 does not seem to reach 0.5 cm. We have made revisions in the manuscript (line 154).

24. L156: "0-0.5" -> "0 and 0.5"

Response: Thank you for your comment. We have revised it in the manuscript (line 154).

25. L156-157: Delete "and generating 1000 simulated spectral curves."

Response: Thank you for your comment. We have revised it in the manuscript (line 154).

26. L160: "in" -> "in a"

Response: Thank you for your comment. We have revised it in the manuscript (line 158).

27. L162 (Fig. 5 and Fig. 10): use grey font for the statistics for Band 1.

Response: Thank you for your comment. We have revised it in the manuscript.

28. L168: The transmittance due to Rayleigh scattering is approximate. Better formulations exist, but this might be OK since 340 nm seems to be the shortest wavelength used for AOD.

Response: Thank you for your comment. We will pay attention to using the updated Rayleigh scattering formula in our future work.

29. L171: "the wavelengths used for AOD inversion were carefully selected by using MODTRAN to calculate and filter the wavelengths corresponding to the transmittances greater than 0.999 that do not include Rayleigh scattering and continuous water vapor absorption" -> "the wavelength used for AOD inversion are those for which the MODTRAN transmittances excluding Rayleigh scattering and water vapor absorption are greater than 0.999."

Response: Thank you for your comment. We have revised it in the manuscript (lines 168-169).

30. L175: "as" –> "as described in"

Response: Thank you for your comment. We have revised it in the manuscript (line 172).

31. L177 (Fig. 6): 0.99 -> 0.999 (in legend)

Response: Thank you for your comment. We have revised it in the manuscript (line 175).

32. L184: on -> from

Response: Thank you for your comment. We have revised it in the manuscript (line 203).

33. L190 (Fig. 7 caption): Either here or preferably in Table 2, the authors should clearly state the average PWV for the "low" or "rich" water content. There is no mention of what 'rich' means quantitatively in this paper.

Response: Thank you for your comment. We have indicated in the abstract that a dry environment refers to PWV<0.5 cm, and other conditions are called rich in water vapor content.

34. L204: "Presumably due to the large calibration uncertainty…" This explanation is rather simple. Also, the first part of this sentence really belongs at the end of the previous sentence. These two extreme wavelengths have large AOD relative uncertainties for non-instrumental reasons. At 340 nm, aerosols contribute more weakly to the total extinction because of strong Rayleigh scattering and ozone absorption. At 1640 nm, aerosol extinction is weak, particularly when particle size is small (radii < 160 nm). This brings me to another point. Table 2 says there is no boundary layer aerosol used. So how is AOD retrieved? Show the true AOD at each wavelength in Fig. 8.

Response: Thank you for your comment. We have added more explanations in the revised manuscript (lines 221-225). The MODTRAN model settings in Table 2 are only used for PWV inversion. The inversion of AOD is based on Beer Lambert's law that described in sect. 3.3.

35. L225: The discussion here needs to be improved. The authors' statement should be clear that biases do not result from low signal-to-noise ratio.

Response: Thank you for your comment. We have revised it in the manuscript (lines 243-246).

36. L228: "with decreasing" -> "for low"

Response: Thank you for your comment. We have revised it in the manuscript (line 247).

37. L229 (Fig. 10): The 1370 nm band is worse in terms of slope, intercept, and R.

Response: Thank you for your comment. Figure 10 is Fig. 11 in the revised manuscript. Figure 11 is a linear fitting diagram of the PWV retrievals of the EKO instruments and the PWV retrievals of CE-318 sun photometer, and the PWV retrievals of CE-318 cannot be completely considered as the true value. Therefore, the worse fitting effect of the PWV retrievals in the band near 1370 nm and those of CE-

318 in Fig. 11 does not mean that the PWV inversion effect of 1370 nm in a dry environment is worse.

38. L235: "may be more accurate" is not supported by the evidence (Fig. 10). Speculative statements should be removed even if they are logically defensible.

Response: Thank you for your comment. As discussed in question 37 Fig. 11 can only show that the PWV retrievals of the band near 940 nm are larger than those near 1370 nm, but it cannot come to conclusion which one are more accurate. Therefore, we just introduce the 1370 nm band because the measurements are available, the results shown in Fig. 5 are consistent with Fig. 11, so we suppose 1370 nm could be introduced for PWV retrievals of dry atmosphere when measurements is available.

39. L247: Why would AOD provide "some assistance" for retrieval of other trace gases from direct sun measurements? L247: "other trace gases retrieval." -> "retrieval of abundances of other trace gases."

Response: Thank you for your comment. We removed such ambiguous statements.

40. L258. Start a new sentence: "The specific …"

Response: Thank you for your comment. We have revised it in the manuscript (line 275).

41. L293: Very questionable speculation…. should be removed unless it can be supported using real data.

Response: Thank you for your comment. We explained the reason for this speculation in the previous responses.

42. L296: are -> is

Response: Thank you for your comment. We have revised it in the manuscript (line 304).

43. L298: Start a new sentence after "wavelengths": "This will be considered in the future version.

Response: Thank you for your comment. We have included the CSR correction in this round of revisions and removed this statement in the revised manuscript.