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Atmospheric Measurement Techniques Discussions

Interactive comment on "A rapid method to derive horizontal distributions of trace gases and aerosols near the surface using multi-axis differential optical absorption spectroscopy" *by* Y. Wang et al.

Anonymous Referee #3

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General Comments:

This manuscript describes a simple, parameterizable, method for inversion of MAX-DOAS observations to near-surface mixing ratios of three important gaseous pollutants. The method is described, as is the instrument used to make the observations. The results of the method are compared to long-path DOAS observations and visibility monitors and relatively good agreement is found.

The manuscript contains valuable information, and particularly the comparison with

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in-situ observations is useful to the community. However, the manuscript, as written needs improvement before final publication. These general comments contain major issues to be resolved by the authors, and following that, some specific comments are given.

1) The manuscript is too long given its result and the language is difficult to read at times, making it confusing. This is a technique article that is describing a method that is similar to work others had done previously. Many of the later figures and text attempt to make a simple interpretation of the data on four days in terms of wind speed and direction, weekend effect, etc. While all this interpretation seems feasible, it is not relevant to the technique, which is the point of the manuscript. The manuscript would be a much more valuable contribution to the literature if the manuscript were more focused on the technique, and the comparison to LP-DOAS, which demonstrates the method's quality.

2) Somewhere in the manuscript, possibly around the middle of page 8133, where light path properties are discussed, it would be useful to mention how topography can affect the effective altitude above ground level of the light path. Hills in the distance may block a 1 degree elevation angle light path, as well as valleys or simply placement of the instrument on a hill could cause the direct light path to be higher above the ground surface. It should be noted that the light path as calculated by this method is not the exact distance over which photons may have traversed. Some photons that end up being detected at the 1 degree elevation angle will have traveled shorter or longer distances along this final light travel direction. Therefore, hills at further distance than the "effective light path" can still affect the observation. Additionally, the concept of the "sensitive altitude range" is much more of an approximate concept than an exact range over which the trace gas concentration is averaged. Therefore, Figure 14 is really not very useful. In fact, a secondary vertical axis could be placed on Figure 13 to give the same information. However, one might even eliminate both figures and simply indicate that effective pathlengths range from a few to $\sim$10km, just as was indicated

AMTD
6, C3968–C3976, 2014

by the modeling described in the first few figures. The actual path of photons is more complex, and thus the concept of "pathlength" is more qualitative than the term would suggest.

3) The manuscript seems to go back and forth between considering the O4 measurement as a dSCD and as a dAMF. For example, on P8132, L 15, the O4 measurement is said to be a dSCD, which I would agree with – from the optical spectroscopy one gets a differential slant column between the 1 degree elevation path and the zenith path. However, on P8133, L2, the manuscript indicates that there is a measured O4 dAMF. One does not experimentally measure the O4 dAMF; the measurement is the dSCD. I presume that the authors are using the assumption that the VCD of O4 is effectively a constant, meaning that a dSCD can be converted to a dAMF, but it is confusing that the manuscript alternates between these two languages. Again, in P 8149, L3, the manuscript is back to having an O4 dSCD. I believe the more understandable language is to discuss only measured O4 dSCD. When the 1 degree referenced to 90 degree dSCD gets smaller, it is an indicator of less O4 differential absorption due to shorter light path near the surface, which makes logical sense.

4) The question of the temperature dependence of the 2O2 <-> O4 equilibrium does not appear to be discussed. If there is a temperature dependence, it will affect the O4 VCD in addition to the "normal" pressure and temperature dependence of the O2 concentration (number density). Also on this topic, the manuscript, on P8135, L6, the wording of the way we express the O4 concentration is confusing and should be more clearly worded. I believe that there are more recent works on the temperature dependence of this reaction, which are relevant to these calculations. Is a single value of c_O4 used, or is a pressure and temperature varying value calculated from meteorological data. Similarly, on P8137, L12, a single value of the O4 VCD appears to be used. Please clarify if a time-varying or constant values are used for both of these parameters.

5) Much of this manuscript describes "error analysis", or explores variability in the correction factors calculated for different aerosol amount and profile assumption, different

AMTD
6, C3968–C3976, 2014

gas profile assumption, etc. While it is good to see that the authors are considering ranges of correction factors, these ranges are not actually an "error analysis". These ranges depend completely on the ranges of assumed optical parameters or trace gas profiles. Therefore, the authors should choose an appropriate term for the results sensitivity to assumed (but in fact unknown) parameters. The ranges chosen by the authors does appear reasonable, so I think the authors have done a reasonable job here, but it is not actually an error analysis. One must also remember that the RTM is 1-D, so does not allow for horizontal variability in trace gas concentration (which is shown by the results to exist), nor does it allow for horizontal structures in aerosol or clouds. Therefore, actual errors could be larger than calculated by this method.

6) Section 6 – Error discussion, is confusing. As was discussed in the earlier point, some of these calculations (ones from the RTM) are not actually errors, but simply sensitivity to ranges of assumptions made by the authors. Additionally, many of the statistics in this section describe relative errors as percentages. These percentages are not at all clear, and often don't make sense. For example, on P8150, L14, the NO2 "error" is said to be 0.14 ppb (or 15%). The NO2 mixing ratio can vary from ppt levels to $\sim$50 ppb, so it doesn't make sense to say that 0.14 ppb is 15% of the NO2 mixing ratio. I think that the authors wish for a percent error so they can combine that with other errors to get a total error. However, it makes this section difficult to follow so as to be assured that the authors have made the calculation correctly. Please rework this section to be simpler and clearer.

7) The comparison of the data between the rapid method and the LP DOAS is a good point of this manuscript. However, the abstract, P8130, L24, indicates different R-squared values from the later discussion, on P8145, L27. These results appear to be different by rounding, but should be the same. The point that the slopes of the correlation plots are not unity is made in the middle of the manuscript, but not discussed in the abstract. It would be valuable to add to this discussion a zero-intercept-constrained fit to the data. I would expect that the slope of that line would be close to unity in each

AMTD
6, C3968–C3976, 2014

case, which may indicate that the slope difference in the unconstrained fit is possibly due to the positive intercept. I believe that the RA value discussed will be the slope of the zero-intercept-constrained fit, so this line's addition could help the discussion of AR and RA, which I found somewhat confusing. I am not sure if I would believe the argument about the lifetime of NO2 affecting these observations. Typically NO2 is lived for on the order of a day, which is long compared to vertical and horizontal transport on these length scales. Source variability appears more likely.

8) In a few places, the manuscript indicates good agreement between MODIS AOD and aerosol extinction as observed by the rapid inversion method. I do not believe that the manuscript ever shows anything more than vague qualitative consistency between MODIS AOD and surface-layer extinction. Therefore, language about agreement with MODIS should be eliminated or somehow more accurately stated. The point of agreement with visibility monitors is a good one, however. The authors seem to understand the reason for lack of relationship with MODIS, and described on P 8144, L 16 - 17. Additionally, if you examine Fig. 15, the AOD varies between 0.4 and 0.5 over these four days, which is shown on an expanded vertical axis (starting at 0.3). This is roughly 20% variation. The light path, however, is shown on an axis starting at 0, and varies more than the AOD. Overall, I think that comparison to MODIS over this short period of time and without any consideration of possible aerosol particles aloft is not useful.

9) One should not use references in the abstract of an article – because some services publish the abstract without the full paper. Therefore, on P 8130, L 4, references in the abstract need to be full references so that one can find them if the abstract is published without the article. Write out the reference here (e.g. say Sinreich, et al., Atmos. Meas. Tech., 6, 1521-1532, doi: 10.5194/amt-6-1521-2013, 2013). Later reference to this paper in the abstract should be clearly referring to the Sinreich et al., 2013 work.

10) The extrapolation of the effective light path to other wavelengths is probably not the largest source of error to these calculations. However, it is important to note that while the wavelength dependence of Rayleigh scattering is well known, the wavelength

AMTD
6, C3968–C3976, 2014

dependence of aerosol particle scattering is not well known and varies significantly with particle size. That complexity of aerosol particle properties gives me concern. Specifically, it seems that the calculation assumes an "urban" aerosol particle type in the boundary layer and "continental" above. It doesn't seem to consider any variability in aerosol particle optical properties, so this treatment seems incomplete. Therefore, a more complete treatment of aerosol light scattering might be warranted for use of this method under other conditions, or for coastal cities.

11) The authors make a point of selecting data with SZA < 60 degrees. They also indicate at times that certain RAA values are problematic. Can the authors make suggestions as to the appropriate cutoff values for SZA and RAA that give "good" data. Additionally, could the authors indicate what "good" is and suggest the increase in uncertainty associated with exceeding these limits?

12) The radiative transfer modeling must have assumed some surface albedo. Please list that value. Additionally, the parameters listed in the table are presumably a function of this assumed albedo, and if they vary strongly with albedo, they would not be appropriate to other situations. Please include discussion of albedo in this work.

Terminology problems:

The language of the article needs to be improved. Some examples of words that are used poorly or are the incorrect words are below. These words should be found and replaced or made consistent throughout the text.

Some sections say measurements were made in the "outskirt", while others use the correct term, outskirts. The phrase "surface near" should probably be near-surface. The phrase "circularly measuring" is confusing. The use of the phrase "divergences" is strange and should probably be differences. I think that divergence seems to indicate the vector operator, and that is not what is being discussed. The phrase "sensitive altitude range" is poorly worded, and probably not necessary. The article has a large number of very similar acronymns that make things confusing – RAA, RA, AR, and

AMTD
6, C3968–C3976, 2014

AA, are all used – The article could become more clear through decreased use of acronyms. Figures indicate measurement in the term "maxing ratio", which should be "mixing ratio". Many prepositions chosen are not correct. Please assure that all co-authors read the text for grammar.

Specific comments:

Note that terminology and grammar problems are described above, and many instances of these exist in the text but are specifically called out below. Only some of these instances are listed below, as well as other specific comments.

P 8130, L14: The abstract says "the uncertainties are especially small". It is not clear what uncertainties are being discussed here.

P 8130, L15-20: This section should be reworded for clarity. The section seems to imply that the correction factors are a function of modeled O4 absorption, when they actually appear to be a function of relative azimuth angle. Possibly this section of the abstract could be cut, retaining only the result of typical uncertainty?

P8130, L23: I think you want "outskirts"

P8132, L10: I think you mean "optimal estimation"

P8132, L14: There is a colon that should probably be a period.

P8133, L13: Measurement at "90 degrees" is indicated. How often is this measurement made? Later, in the fitting section, do you measure 1 degree, then 90 degrees, then next azimuth 1 degree....? Overall, this is not clear as to how the reference is dealt with.

P8134, whole page: This description of MAX-DOAS is not very clear. Please try to reword it more clearly. For example, the Frauenhofer reference does remove solar absorption features, but in this application of MAXDOAS, it also removes all stratospheric absorptions from the dSCDs observed because the stratospheric paths are similar be-

AMTD
6, C3968–C3976, 2014

tween light detected at 1 degree elevation and 90 degree elevation angle.

P8138, L4 and below: The units are missing from these equations.

P8138, end of page: the use of "real" in strange here. I would suggest using "assumed"

P8141, L11: The anti-reflection is a coating. A 2-D motor is probably two motors. What is the manufacturer of the compass and inclinometer? Is the accuracy of the inclinometer actually 0.01 degrees?

P8143, L7: The window contains two O4 absorption bands.

P8144, L26: Some RAA values are edited out of the data for comparison. As mentioned above, can the authors suggest values to use?

P8145, L27: R-squared values differ from abstract.

P8147, L9: U-shaped concentration profiles are commonly also affected by increased vertical convective mixing during midday causing dilution by entrainment of air above.

P8152, L25: Again, here the "maximum and minimum" values of correction factors are really just the range of assumptions made, not true maximum and minima.

P8154, L15: No quantitative relationship between AOD from MODIS and MAXDOAS has been shown in this manuscript, so this conclusion should be eliminated.

P8161, Table 1: The unit on the height of the BL is missing. I presume all combinations of these values were explored. Make that clearer.

P8162, Table 2: Units are missing from this table.

P8176, Fig 14: If this figure is not eliminated, its vertical axis should have the same label as the final term used for this parameter. The current figure's title disagrees with the text.

P8180-P8182, Figures 18-20 say "maxing ratio" but should say "mixing ratio"

AMTD

6, C3968–C3976, 2014

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AMTD

6, C3968–C3976, 2014