Atmos. Chem. Phys. Discuss., 7, S534–S542, 2007
www.atmos-chem-phys-discuss.net/7/S534/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nine years of UV aerosol optical depth measurements at Thessaloniki, Greece" *by* S. Kazadzis et al.

S. Kazadzis et al.

Received and published: 12 March 2007

General: We would like to thank the reviewer for the useful comments and suggestions. Most of them have been taken into account. A change in the deseasonalisation method and the calculation of trends lead to slight changes in the retrieved AOD trends. In addition the use of cloud flagging methodology to derive clear sky scans also for the Brewer MKII direct sun scans eliminated a (small) number of points that were included in the previous version of the manuscript mainly in figure 3. Figures 3, 4 and 8 are changed according to the reviewer's suggestions. All changes in the text according to the reviewer's suggestions are included in this response document together with the reviewer's suggestions in order to help understanding easier the manuscript changes.

Comments

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The only comment in that respect is the unusual notation of the Angstrom exponent a, instead of the greek letter. I would advise the authors to modify the notation.

Response:

Corrected as suggested.

The references seem complete and previous contributions to this subject are for the most part acknowledged. However reference Gröbner and Meleti, 2004 should be mentioned twice on page 541, since it uses both methodologies (absolute calibration using an extra-terrestrial spectrum in the UV, and langley- plots in the visible).

Response:

The reference was added as suggested

General comments: 1) The use of langley-plots to calibrate an instrument for atmospheric total column retrieval (be it total column ozone or aerosol optical depth) requires extremely stable atmospheric conditions during the calibration period. If that is not guarantied, Langley plots should not be used as calibration methodology. This is the reason why the ozone calibration of Brewer spectrophotometers is obtained by Langley-plots at high altitude, low latitude sites (i.e. Izana, Canary Island, Mauna Loa, Hawaii). The use of Langley plots for calibrating the MKII Brewer at Thessaloniki in the UV region where ozone and aerosols are important is thus highly questionable, and should be used with great caution. Any results from such an analysis should be used with great care and cannot be used to validate measurements by the MKIII Brewer.

Response:

The purpose of the paper was to analyze the MKIII AOD data series. Brewer MKII was used not for validation purposes but only for having a supporting hint of the negative changes observed with the MKIII Brewer. Great effort was spent on locating days (or parts of days) from the long-term record of Brewer MKII which, firstly, fulfilled requirements concerning ozone stability, angular and temporal coverage and number of ob-

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servations. The corresponding Langley plots were improved by visual inspection, and the derived extraterrestrial relative irradiances were applied to estimate the variations of the AODs at 320 nm on calibration days. Days with highly variable AOD (standard deviation $>$ 0.03) or with relatively high AOD ($>$0.6) were rejected, and the rest of the calibration days (162 days for the nine years period) were used to extrapolate linearly the extraterrestrial irradiances on certain time periods. A paragraph was added in the section where the MKII calibration is discussed:

'The use of Langley plots to calibrate an instrument for aerosol optical depth requires extremely stable atmospheric conditions during the calibration period. Especially for calibrating the MKII Brewer at Thessaloniki in the UV region, where ozone and aerosols are important, Langley plots were used only during days with stable ozone and AOD. These days are determined using a set of strict criteria in the Langley extrapolation analysis. In should be noted that the comparison of AOD derived from the MKIII and MKII is not used for validation purposes but only for investigating the long term changes in AOD derived from two instruments with different calibration principles."

2) The continuous measurement record of the MKII is however important since data gaps exist for the MKIII instrument due to the participation at various campaigns outside of its home site. One suggestion to use the MKII data would be to calibrate its measurements by direct comparison to the MKIII over the whole period, and thereby fill the gaps when the MKIII is not available.

Response:

The suggestion was taken into account and a new series of merged monthly mean data is presented in figure 3. A paragraph was added: "In figure 3 the results of a monthly mean AOD series merging the two instruments is shown. Measurements of MKII instrument were calibrated by direct comparison with the MKIII (figure 2), over the whole nine year period. In addition, the number of available days of each instrument (per month) have been taken into account in the calculation of the weighted average.

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The AOD change per year calculated from the merged data set was calculated to 2.9 ś 0.9 % for 320 nm."

3) Regarding the long term trend of aerosol optical depth at Thessaloniki, I believe that the conclusion reached by the authors is inconsistent and needs some further clarifications: The Cluster Analysis of the backtrajectories shows 5 Clusters, of which only Cluster 5 represents local sources (see page 548). As stated in the last paragraph of this section, the long term monthly means of AOD over Thessaloniki are however dominated by aerosols of Cluster 4, i.e. of origin in the eastern directions (biomass burning from the northern coast of the Black Sea and SO2 from power plants in Bulgaria and Romania). In contradiction to this statement, the authors claim on page 551 (last paragraph of section 6) that the PM10 aerosols are a major contributor to the total column of the AOD over Thessaloniki.

Response

We agree with the reviewer that the text was not clear in this point and that lead to contradicting conclusions like the one mentioned here. Aerosols in the city of Thessaloniki originate from local sources and can be found in the boundary layer, but also there are contributions from transport from longer distances. For all clusters there is always a contribution of the local aerosol sources. Cluster 5 contains all sources of aerosols that are transported over short distances and they originate predominantly from areas close to the country borders. The text describing cluster 5 was changed to "Finally the 5th cluster includes mostly air masses travelling over shorter distances and in some cases those from the Saharan desert."

The main point is that for the AOD column measurements we used some of the conclusions derived from the paper of Amiridis et al, 2005 that showed different seasonal characteristics in the aerosol profiles over the city. So, in this work we used the findings of the trajectory analysis (at the height of 1500m) to investigate the contribution of the transported aerosols only, in addition to the aerosol lower in the atmosphere that come

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from local city sources. To clarify this point section 5 and 6 were changed to 5.1 and 5.2 as sub-sections of a general section 5 named: “Origin of aerosols at Thessaloniki”. A paragraph was added in this section : “The total column AOD measured in Thessaloniki can be considered as the sum of two main contributors. The aerosols that are associated from local sources and can be found in the boundary layer and aerosols in higher altitudes that are transported and reach the city from sources outside the city limits. To explain part of the observed seasonality in AOD these two contributors have been investigated.”

Also, a paragraph was added to the summary and conclusions section 6: “The analysis of the origin of aerosols at Thessaloniki showed that in addition to the boundary layer aerosol originating mainly from local sources, transport of aerosols at higher altitudes contribute to the total AOD column, especially in the summer months. Back trajectories analysis showed that the contribution of air masses coming from the North and North Eastern directions result in high aerosol loads over Thessaloniki, while minimum AOD is associated with air masses originating from the Atlantic ocean.”

4) This contradiction also puts into question the origin of the downward trend in aod over the period, as illustrated in Figure 8. A possible cause of this downward trend could also be a reduction of aerosols from Eastern countries. My suggestion would be to: A) Investigate the long term trend in Angstrom exponent alpha. A trend in this component could indicate if there is reduction of PM10 particles or of particles of different sizes, and this might help in finding the cause of the observed downward trend. B) Make a statistical significance test of the trend to make sure that it is not an artifact. C) Make trend analyses for different seasons, since the Cluster 4 aerosols seem to be more predominant during the summer season (see page 549, lines 13-19).

Response:

Some of the points responsible for this contradiction referred here probably have been answered in the previous comment. Concerning the three suggestions of the reviewer:

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A) A sentence was added: “The long term trend of the Angstrom exponent alpha showed a small increase over the years but with no statistical significance and well within the standard deviation of the monthly mean values.” Therefore this result cannot be used to derive firm conclusions on which aerosols are responsible for the trend. Nevertheless, the PM10 are present in all clusters, and therefore are considered a major contributor in the observed trend in AOD.

B) A significance test analysis has been performed for figure 8 data and the results are reported in various sections in the revised manuscript.

Concerning the results from the Brewer instruments (figure 3) the text was changed to: “A linear regression on the MKIII diseasonalized data reveals a change of -2.9 ś 0.92 %, -3.8 ś 0.93 % and -3.5 ś 1.02 % per year for 320, 340 and 355 nm respectively. The statistical significance of this change was found better than 99% using student’s t-test. For Brewer MKII the change of AOD at 320 nm for the same period was calculated to -3.2 ś 0.6 % per year (with statistical significance similar to MKIII), which is similar to the one calculated from the MKIII data. “

Concerning the results shown in figure 8 the text was changed to: “A negative change is evident at all stations ranging from -2% to -5.6% per year, with an average of -3.8 ś 0.6 % per year. The statistical significance of this average change was calculated to better than 99.9 %, based on the student’s t-test.”

C) Results of trend analysis in different seasons showed a negative trend of ˜3%/year for spring and summer, ˜1% for Autumn and ˜5% for winter but the month to month variability of this trend is more than 3% so we decided that these results can not prove if there is an obvious downward trend only for the aerosols transported from North East and Eastern locations. The PM10 trend in figure 8 show that the boundary layer aerosols have a negative trend and this affects all (cluster analysis) cases.

5) One criterion of ACP is if the description of experiments and calculations are sufficiently complete and precise to allow their reproduction by fellow scientists (traceability

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of results)? I believe that the aod data presented in this study is not publicly available, and therefore does not meet the above criterion. However the WMO has created a World Data Centre for Aerosols, located in Ispra, Italy, where any information concerning atmospheric aerosols can be submitted and thus provide access to the data for other scientists. (http://wdca.jrc.it/) The authors might consider submitting their data to this database.

Response:

Our interpretation for the criterion of ACP about the traceability of results is that the paper should describe with sufficient detail the methodologies followed for the experiments and calculations so that any third party could apply these to another data set and reproduce the paper's findings, and not that the data are available to any third party to repeat the work presented in the paper. The most common situation in the peer reviewed papers published so far is that the data used therein are not publicly available, ether because of data policy restrictions or due to technical difficulties. We will consider the reviewer's suggestion for depositing our data to the WMO aerosol database, but we cannot take it as prerequisite for the eligibility of our paper to be published in ACP.

Specific comments:

page 545, line 7, the number of data points are much closer to 3000 than 2900 (2983), so I would suggest modifying this number to 3000. Figure 2 shows the comparison between the MKII and MKIII aod values. In that plot the fitted linear line does not show any offset of 5% as mentioned in the text, and actually goes nearly exactly through the origin. Where does the 5% of the text come from?

Response:

The number was modified as suggested. Maybe it is not easily visible with the scattered plot but the offset concerning the mean values of the synchronous measurements of

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both instruments is 5% as it is written. The sentence was slightly modified to: “Comparison of the AOD derived by the two Brewer spectroradiometers shows that the single monochromator Brewer MKII overestimates by about 5% the AOD.”

page 545, line 11. The uncertainty levels of the instruments are mentioned in the manuscript but have never been described and stated. I would appreciate a short paragraph stating the overall uncertainty budget of the aod retrieval of the MKII and MKIII Brewers. This will also strength the case with respect to the significance of the observed trend in aod over the time period.

Response:

For Brewer MKIII measurements using the described methodology a paragraph was added to section 2.2 describing the overall error on the AOD measurements as given in Kazadzis et al., 2005: “The experimental error on the AOD is estimated to within 0.07 in the UV-B and 0.05 in the UV-A for measurements at solar zenith angles between 15° and 75° (Kazadzis et al., 2005). This error is the result of the propagation of errors due to the direct irradiance measurement and calibration uncertainties, the determination of the extraterrestrial spectrum, and the measurement of ozone and SO2.” More details can be found in the reference given.

For the Brewer MKII the following text was added: “The uncertainty of the AOD retrieved from the MKII Brewer results from the error in determining the extraterrestrial irradiance (Marenco et al., 2002), the total ozone column measurement (Groebner and Meleti, 2004), front window polarization effects (Cede et al., 2006) and the temperature of the ozone cross sections (Cheymol and DeBacker, 2003). The total uncertainty for the AOD determination at 320 nm was found 0.04 for air mass 1.4 which is the mean air mass for Thessaloniki.”

page 547: The equation of the Angstrom power law is missing a negative sign in the exponent, it should be reading tau=beta lambdaĹ(- alpha). In addition, the unit of the wavelength should be mentioned, since this defines the value of beta (even if not used

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in the manuscript).

Response:

Equation and units were corrected and defined accordingly.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 537, 2007.