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Atmospheric Measurement Techniques Discussions

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Interactive Comment

Interactive comment on "Direct-sun total ozone data from a Bentham spectroradiometer: methodology and comparison with satellite observations" *by* M. Antón et al.

M. Antón et al.

manantón@unex.es

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Author's answer to Anonymous Referee #2

The authors greatly acknowledge the anonymous reviewer (Referee #2) for carefully reading the manuscript and providing constructive comments.

This paper presents application of the well established differential optical absorption (DOA) technique to Bentham spectroradiometer direct irradiance measurements to retrieve total ozone column (TOC). While DOA has been widely used on data obtained by other instruments, this is the ïňĄrst time it has been used to analyze Bentham direct sun observations. Improved TOC measurements are of interest to the scientiïňĄc community and therefore I recommend publishing the article after the authors address the questions and comments below.

Author's response: Thank you for the comments. We have tried to answer all your questions, see our detailed reply below

General comments:

Referee GC #1: This paper does not explain why a new method is needed to derive TOC from the Bentham spectroradiometer and how the DOA method is more superior to the currently used TOC retrieval techniques.

Author's response GC #1: As the reviewer has pointed out in his/her introduction, the technique used on our study is not new since it has been extensively employed to derive TOC data from different types of spectrophotometers (Dobson, Brewer, etc). The novelty of our paper is to explain in detail the steps to implement the method from the Bentham direct-sun raw data. To our knowledge and to date, only global (direct+diffuse) spectral irradiance (Brogniez et al., 2005) or the difference between global and diffuse irradiances (Lenoble et al., 2004) have been used to obtain ozone values from this equip. Additionally, our paper also shows the reliability of the Bentham TOC retrieval measurements by means of a detail comparison with satellite data. The results of these inter-comparisons showed that the Bentham instrument has a high potential for the retrieval of reliable direct-sun TOC data, being a viable alternative to the widely utilized Brewer and Dobson spectrophotometers. Regarding how the DOA method is more superior to the currently used TOC retrieval techniques, WMO has established that this technique is the most accurate and the bestâĂŚdefined method for determining the total ozone column. In order to clarify this issue, the following comment has been added to the text (Section 1 "Introduction"); "Although there are several methods to derive total ozone from the ground (e.g., direct moon, zenith sky irradiances, look-up-table using experimental and modeled global irradiances), WMO

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has established that the differential optical absorption technique applied to direct solar radiation at UV wavebands between 305 and 340 nm is the most accurate and the bestâĂŚdefined method for determining the total ozone column (TOC) (WMO, 1996)"

Referee GC #2: The Bentham instrument is not described sufïňĄciently to understand its capabilities and limitations. No information is provided about the detector used, entrance and intermediate slit sizes, stray light characterization, its ïňĄber optics, and its effect on irradiance measurements as a function of pointing angle, instrument function characterization, and signal to noise ratio at the selected wavelengths per measurement. It would also be beneïňĄcial to discuss which atmospheric properties are typically retrieved from the Bentham instruments and how many of the instruments are in service around the world.

Author's response GC #2: An expanded description of the instrument has been included in the new version of the manuscript. The detector is a photomultiplier tube (R1527 Hamamatsu) with a blue filter to reject visible stray light. Optical fibres are the model LI-J1010. Regarding the signal to noise, the following sentence has been included in the text (Subsection 2.1 "Ground-based measurements"): "The signal to noise ratio at 300 nm is 0.026 and 0.01 at 350 nm for SZA equal to 30°, and when SZA is 60° this ratio rises to 0.04 at 300 nm and it is still 0.01 at 350 nm (Bernhard and Seckmeyer, 1999)."

We only find in literature a work in which a Bentham instrument is used to retrieve atmospheric properties. Lenoble et al. (2008) describes a method to derive the aerosol optical depth in UV region using a Bentham double monochromator. This reference has been included in the text: Lenoble, J., Brogniez, C., de La Casiniére, A., Cabot, T., Buchard, V., and Guirado, F.: Measurements of UV aerosol optical depth in the French Southern Alps, Atmos. Chem. Phys., 8, 6597–6602, 2008. The number of operative Bentham instruments around the world would be an interesting fact, but we do not have this information.

Referee GC #3: While comparison of ground-based derived TOC with satellite measurements is important, I would recommend not using the comparison as a validation tool for such ground-based observations. Why not validate the Bentham TOC measurements side by side with a Brewer or a Dobson instrument? Many uncertainties arise from validation of direct sun ground-based TOC measurements that sample a narrow cone of the atmosphere from sunrise to sunset with equal sensitivity to tropospheric and stratospheric optical absorption (SZA <75°) using satellite observations with large footprints, reduced sensitivity near surface, and at best two overpasses a day. In addition, the DOA method itself does not require validation, while the Bentham instrument measurements do.

Author's response GC #3: We agree that ground-based instruments should be validated by a well established similar ground-based instrument to assure high quality of TOC data. However, we do not have any Brewer or Dobson instrument at Granada or in its nearness. So, it is not possible to validate our Bentham TOC data against other spectrophotometer's measurements. We think that comparison exercises against satellite instrument performed in our paper allow to readers get the picture on the reliability of the TOC data derived from the Bentham instrument at the study location. Throughout the manuscript, we have replaced the word "validate" by "compare" since we compare (not validate) our Bentham TOC measurements against satellite data.

Referee GC #4: The authors make an observation that cloud-free days have a "typical" diurnal pattern characterized by a "ïňĆat" morning and gradually increasing afternoon TOC, amounting to an unrealistically large column change (up to 40-50 DU) over a few hours. They attribute this large TOC increase in the afternoon hours to O3 photochemical production due to anthropogenic emissions in the lower troposphere. The authors need to convenience readers that the observed diurnal pattern is real and not the result of instrumental errors.

Author's response GC #4: Please read Author's responses SC #20 and SC #23

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SpeciïňĄc comments:

Referee SC #1: Page 8132: Line 13. Do the authors mean different behavior?

Author's response SC #1: The sentence has been changed by: "The analysis of the diurnal TOC variations during cloud-free days showed different TOC values between the morning and afternoon periods."

Referee SC #2: Page 8133: Line 10. The authors here state that high quality ground-based TOC measurements are necessary for satellite TOC validation. Later, they use satellite measurements to prove the high quality of the Bentham ground-based TOC observations. This is circular logic.

Author's response SC #2: Please read Author's response GC #3

Referee SC #3: Page 8133: Line 14. "is SO called differential OPTICAL absorption"

Author's response SC #3: Done.

Referee SC #4: Page 8133: Line 27: This instrument CAN record

Author's response SC #4: Done.

Referee SC #5: Page 8135: Line 4. Is this a scanning system with a 0.5 nm step?

Author's response SC #5: Yes, it is, but because it has been configured in this mode.

Referee SC #6: Page 8135: Line 5. Does 0.48 nm spectral resolution refer to FWHM with the narrowest slit available, and FWHM (Line 5) of 1.05 nm to actual spectral resolution? Please clarify.

Author's response SC #6: The spectral resolution of 0.48 nm is the resolution given by manufactured, and it is removed in the new text.

Referee SC #7: Page 8135: Line 8. Reword sentence starting with "The double-monochromator. . . ". It is unnecessarily long.

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Author's response SC #7: The sentence has been divided and rewritten, given: "The input of the double-monochromator is connected to two diffusers installed at the CEAMA rooftop by means of two optical fibers (LI-J1010). One diffuser is used to measure global and diffuse irradiances (don't used in this work). The second diffuser takes direct irradiance measurements from a collimator tube with three optic diaphragms (field of view of 1.2°) which is mounted in a sun tracker (2AP model from Kipp & Zonen)."

Referee SC #8: Page 8135: Line 11. In order to perform

Author's response SC #8: This sentence has been rewritten following the specific comment #7.

Referee SC #9: Page 8133: Line 12. Accuracy of sun tracker is BETTER than

Author's response SC #9: Done.

Referee SC #10: Page 8135: List overpass times for OMI, GOME and SCIAMACHY.

Author's response SC #10: According to the reviewer's suggestion, we have included the following information in the text ("Comparison against satellite observations"): "In addition, the OMI overpass time ($\sim$13:30 GMT) differs with respect to overpass times for SCIAMACHY ($\sim$10:30 GMT) and GOME ($\sim$11:00 GMT)."

Referee SC #11: Page 8138: Line 5. I would recommend listing the wavelength pairs as 305.5/325.5 nm (A1/A2) and 317.5/340.0 nm (D1/D2).

Author's response SC #11: Done.

Referee SC #12: Page 8139: Line 9. "temperature changes with the height..." Do you mean measurement site altitude?

Author's response SC #12: Yes. To clarify it, we have written: "The real effective temperature changes with the latitude and height of study site and season."

Referee SC #13: Page 8139: Line 11. Is the ozone effective temperature higher in winter than summer? Explain why ïňĄg. 2 is referenced while talking about TOC effective temperature.

Author's response SC #13: Sorry for the double mistake. Firstly, the right figure is number 6 (not number 2) in Antón et al. (2008). This plot showed the evolution of the daily effective temperature for Madrid between 1995 and 2002. Secondly, the lower value (218 K) corresponds to winter and the higher (232 K) to summer. Both mistakes have been corrected in the new version of the manuscript.

Referee SC #14: Page 8140: The entire discussion starting from Line 11 is somewhat irrelevant. AMF sensitivity to effective gas height is very low (<1% for an error in effective height of +/- 5 km at SZA 75°) at SZA smaller than 75°which follows directly from the equation 9.

Author's response SC #14: We have summarized this discussion.

Referee SC #15: Page 8141: Line 16. Please specify year/years of the selected 30 days. Are they all from 2005?

Author's response SC #15: Yes, the 30 cloud-free half-days correspond to 2005 (late spring and summer). This fact has been added to the text.

Referee SC #16: Page 8141: Line 16. Please explain why only morning measurements are considered for the Langley plot analysis.

Author's response SC #16: According to the reviewer's suggestion, the following sentence has been added in the text (Section 4 "Extraterrestrial constant"): "Morning measurements have been chosen due to higher stability of atmospheric conditions during these periods as is explained in the next Section."

Referee SC #17: Page 8142: Line 22. Marenco et al. 2002 mainly focuses on the effect of the diurnal changes of aerosol OD on the ETC derivation. Since intensity ratio is used in this work, the "random" aerosol effect ("disturbances") should be minimized.

Author's response SC #17: Yes, we have used intensity ratios to derive TOC data, so the aerosol effect is minimized. However, the atmospheric optical depth can also vary in a short-term period due to ozone changes. In our paper, we have shown that this factor produce a substantial variability on ETC values. Following the Marenco's procedure, we have averaged the ETC derived from a sufficiently large number of cloud-free half-days. In order to clarify this issue, we have included the following comment in the manuscript (Section 4 "Extraterrestrial constant"): "It is well-known that the correct application of the Langley method needs that the optical depth remains constant during the measured period. Nevertheless, Marenco (2002) showed that if a sufficiently large number of cloud-free half-days is available, a reliable ETC can be performed even at locations characterized by non-negible short-term variability of atmospheric conditions. This author exhibited that these atmospheric variations act as random processes, so their influence on ETC may be minimized by averaging over a sufficiently large number of cloud-free half-days. In this sense, the average ETC value (-0.071) obtained from the 30 daily values may be assumed as representative for the real constant of the Bentham instrument, and, therefore, this mean value may be considered as the extraterrestrial constant in equation 2."

Referee SC #18: Page 8143: Line 25. the error bars

Author's response SC #18: Done. Referee SC #19: Page 8144: Line 20. monotonic

Author's response SC #19: Done.

Referee SC #20: Page 8144: Line21. Consistent change in TOC of up to 40-50 DU is too large for a typical diurnal TOC variability!

Author's response SC #20: Diurnal TOC variations up to 40-50 DU only are reached at the study site on certain days in late spring and summer. Similar diurnal TOC changes have been recently reported by Tzortziou et al. (2012) at GSFC in Greenbelt (USA) by a Brewer double monochromator and a collocated Pandora CMOS spectrometer. Thus, to support our results, the following sentence has been included in the text (Sub-

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section 5.1 “Bentham total ozone data”): “Thus, it can be observed that TOC remains almost constant until midday, suffering then a monotonic increase toward sunset that can reach up to 40-50 DU on certain days. Such strong short-term changes in total ozone may occur occasionally at mid- to high-latitude locations during spring-summer (e.g., Tzortziou et al., 2012).”

The following reference has been also included in the new version of the manuscript: Tzortziou, M., Herman, J.R., Cede, A., and Abuhassan, N.: High precision, absolute total column ozone measurements from the Pandora spectrometer system: Comparisons with data from a Brewer double monochromator and Aura OMI, J. Geophys., Res., 117, D16303, doi:10.1029/2012JD017814, 2012.

Referee SC #21: Page 8144: Line 25. On most cloud free mornings

Author’s response SC #21: Done.

Referee SC #22: Page 8144: Line 29. is the result of

Author’s response SC #22: Done.

Referee SC #23: Page 8145: Line 2: Zbinden et al. 2006 reported mean summer total tropospheric columns over major middle latitude cities ranging from about 30 to 40 DU, with about 6 to 10 DU in the boundary layer. Consistent daily change in TOC of up to 40-50 DU over afternoon hours presented in the current study cannot be explained solely by the O3 photochemical production near the surface.

Author’s response SC #23: Yes, we agree with this comment. In order to clarify this issue, we have re-phased the paragraph as: “The daytime pattern observed in Figure 3 (middle) can be partially associated with the diurnal photochemical processes in the lower troposphere related to the formation of ozone near the Earth’s surface at populated urban locations. It is known that tropospheric ozone column represents about 10% of total column (between 30 and 40 DU), with up to 10 DU in the boundary layer over major middle latitude cities (Zbinden et al., 2006). Thus, surface ozone has a

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small contribution to total column but due to its marked daytime pattern (e.g., Ribas and Peñuelas, 2004; Adame et al., 2010) present a non-negligible effect on diurnal TOC variations. In this sense, Antón et al. (2010b) analyzed the simultaneous diurnal evolution of both surface and total column ozone data at Madrid. They showed diurnal TOC variations up to 20 DU of which surface ozone changes could explain between 20% and 70%, depending on the mixing layer height. The strong diurnal TOC variations (up to 40-50 DU) found on certain days at Granada cannot be exclusively explained by photochemical processes near the surface, therefore, other factors like transport processes in the upper troposphere and lower stratosphere could also present a significant contribution.."

The following references have been included in the new version of the manuscript: — Antón, M., López, M., Serrano, A., Bañón, M., García, J.A.: Diurnal variability of total ozone column over Madrid (Spain), Atmospheric Environment, 44, 2793–2798, 2010b. — Zbinden, R.M., Cammas, J.-P., Thouret, V., Nédélec, P, Karcher, F., and Simon, P.: Mid-latitude tropospheric ozone columns from the MOZAIC program: climatology and interannual variability, Atmos. Chem. Phys., 6, 1053–1073, 2006.

Referee SC #24: Page 8145: Line 28. While satellite measurements agree within a few percent with ground-based measurements, one should realize that a boundary layer ozone column of 10 DU translates into roughly 3% of TOC. The comparison is not consistent. UV ground-based instruments have high sensitivity to gases located in the lower troposphere, while UV satellite instruments have low sensitivity. Ground-based instruments measure along the line of view; satellite instruments average over a large area. Ground-based instruments should be validated by a well established similar ground-based instrument to assure high quality of TOC data.

Author's response SC #24: Please read Author's response GC #3

Referee SC #25: Page 8146: Line 4. Why were the Bentham TOC measurements for comparison with the three satellite instruments averaged from 11:00 to 13:00 instead of at the actual satellite overpass times [9:30am (GOME-2), 10:00 am (SCIAMACHY) and 13:30 (OMI)]?

Author's response SC #25: We decided to use the same the daily average value of Bentham TOC data (from 11-13h) in the three ground-based – satellite-based comparisons. However, according to the reviewer's suggestion, in the new version of the manuscript, we have used the ground-based measurements at the appropriate satellite overpass time. Thus, the inter-comparison results given in Table 2 and Figure 3 vary slightly. Additionally, the following information has been added in the text (Subsection 5.2 "Comparison against satellite observations"): "In addition, the OMI overpass time (∼13:30 GMT) differs with respect to overpass times for SCIAMACHY (∼10:30 GMT) and GOME (∼11:00 GMT). In this sense, to comparison purposes, the Bentham TOC data derived each day between 13:00 and 14:00 GMT are averaged to be compared against punctual OMI data while the average Bentham data between 10:00 and 11:00 GMT are used to be compared against both SCIAMACHY and GOME data. Only those hourly TOC averages with a SD smaller than 10 DU (∼ 3%) are assumed as valid and, therefore, utilized for intercomparison purposes. This restrictive threshold guarantees an unobstructed solar disk during the four direct solar spectrums measured within one hour."

Referee SC #26: Figure 3. Title: Error bars,

Author's response SC #26: Done.

Referee SC #27: Figure 3: Legend: Please replace "oktas" for "cloud cover", right axes title: cloud cover (okta).

Author's response SC #27: Done

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