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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of urban pollution on UV spectral irradiances" *by* R. L. McKenzie et al.

R. L. McKenzie et al.

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We would like to thank both of the anonymous reviewers for their helpful and constructive comments on our manuscript. We appreciate that although this paper represents an important contribution to the literature, it is rather heavy-going in places. Both reviewers found the paper to be interesting and informative. However, both also recommended several changes and clarifications. Fortunately, several of the concerns raised were common to both reviews, and there were no serious conflicts between their comments (although reviewer 1 suggested adding figures, while reviewer 2 suggested deleting some).

Although the reviewers required only minor changes, we found that to address their concerns adequately, the changes were quite substantial. We have substantially reordered text, and have introduced new notations to clarify terms, such as optical depth.

The attached copy was prepared with revisions marks on. However, to improve legibility, we have accepted all deletions.

We also found an error in the calculation of NO2 in Figure 8. Values were too small previously because the absorption cross sections had not been filtered correctly to the instrument band pass. We have more thoroughly investigated the uncertainties in trace gas retrievals, and found these to be larger than previously implied. Some of the "variability" reported in the previous version was actually noise. The NO2 values derived by the simplified method are approximately 12% less than with our standard method, for which the measurement uncertainty is ˜ +/-5%.

We specifically address the reviewer's concerns below. These changes include modifications to several figures (Figures 2, 4, 8, 9, 10, and 13), and the addition of several new references. We have amended the manuscript to address most of the concerns that have been raised. In the few places where we feel that no change is needed, we have explained our reasoning.

Anonymous Referee #1 Received and published: 25 May 2008

The paper is devoted to the comparisons of UV irradiance over an urban and clean site in order to understand the role of urban pollution in UV attenuation. The authors have made significant efforts to organize this experimental study. The results are based on about 1 year of simultaneous spectral precise measurements at the urban site in Tokyo and at the pristine site (Lauder New Zealand), where the effects of urban pollution can be neglected. In addition, the RT calculations were used for better understanding of the obtained results. Unfortunately, the supplementary information about the aerosol and cloud properties was unavailable in this study that made the analysis much more complicated. However, I think that this analysis is very helpful in understanding the role of different factors in solar radiation attenuation over urban area, although not all the problems were solved and even several new ones have been arisen. The paper is well organized and the summary is quite complete. However, there are several remarks,

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which should be clarified before accepting it for the publication in ACP.

Thank you for those comments.

General comments:

Two points should be discussed in more details:

1. The possible difference (or its absence) in cloud optical thickness and cloud amount over Tokyo and New Zealand in different seasons.

We now further emphasise the point in section 5 that, because of the data selection criterion that required a low variance in UVA during the scans, the derived cloud optical thicknesses in this study cannot be interpreted as mean values. We also now note specifically that spectra with cloud transmissions less than 0.3 are excluded.

2. The absorbing properties of aerosol over Tokyo and spectral features of absorbing aerosol optical thickness. Even in model calculations the authors used only one value of single scattering albedo for the whole period of observations which is constant over the UV spectral interval. I would recommend the authors to use for this analysis additional literature sources and to discuss the uncertainty due to these factors.

We had already noted in the Introduction (top of p 7151) that uncertainties in the single scattering albedo due to absorptions by organic aerosols was likely to be a factor, as predicted by Jacobson. In fact, one of the motivations for this research was to identify spectral features due to this effect. Unfortunately, we were unable to identify such features. The fact that we were unable to identify any such effects suggests that they may be rather small, or alternatively, that the overall effect of these absorbers has little overall spectral signature - albeit perhaps due to overlap of many different absorbers with uncorrelated absorption features. That point is now emphasised further at the end of section 6.

The default single scattering albedo we have used (0.95) is taken to represent the effective value for the combined cloud/aerosol effect. We now mention in section 7

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that this is somewhat higher than recent studies have indicated for urban aerosols. For example, Bais et al (2005) found the value of ssalb=0.7 found in urban environment in Europe, while Petters et al (2003) found values as low as 0.80 at 368 nm, reducing further to 0.65 at 300 nm. We also now mention, in section 8, that with these smaller values (and larger alpha values), the calculated effects on global irradiances would have been much larger for a given optical depth, as shown by Bais et al (2005).

New Refs

Bais, A., A. Kazantzidis, S. Kazadzis, D.S. Balis, C.S. Zerefos, and C. Meleti, Deriving an effective aerosol single scattering albedo from spectral surface UV irradiance measurements, Atmospheric Environment, 39, 1093-211;1102, 2005.

Petters, J.L., V.K. Saxena, J.R. Slusser, B.N. Wenny, and S. Madronich, Aerosol single scattering albedo retrieved from measurements of surface UV irradiance and a radiative transfer model, Journal of Geophysical Research, 108 (D9), 4288, 10.1029/2002JD002360, 2003.

Specific comments:

1. p.7151, line 26 .What was the time interval for the midday conditions?

Two and a half hours. Now stated in the text

2. p.7152, line 27-28. From what sources the total ozone amount was taken? It would be helpful if the description of the seasonal features of main atmospheric parameters is included in the additional paragraph.

Ozone was from the NIWA ozone climate data base, which is essentially satellite-derived ozone but latitudinally re-normalised to match available ground-based ozone measurements (Bodeker et al., 2001). The seasonal features are now described.

I propose to add the solar angle changes in Fig.1.

Reviewer 2 felt there were already too many figures, so instead we have stated the mid

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summer and mid winter minimum SZAs in the discussion of seasonal effects above.

It will be also helpful to add some climatic features of cloud and aerosol properties from literature sources.

We have added recent reference to the seasonal variations in aerosols in Japan and China, which influences that in Japan.

For example, AERONET data from Japan (Chin et al., 2004) shows an average optical depth of 0.3 to 0.5 at 550 nm, with relatively small seasonal changes. This low seasonality contrasts with the situation in China, which is a source region for Japan. Wang et al (2008) report that AODs in NE China are significantly higher, with a strong summer maximum that can exceed optical depth 1 at 500 nm.

New refs

Chin, M., A. Chu, R. Levy, L. Remer, Y. Kaufman, B. Holben, T. Eck, P. Ginoux, and Q. Gao, Aerosol distribution in the Northern Hemisphere during ACE-Asia: Results from global model, satellite observations, and Sun photometer measurements, J. Geophys. Res, 109 (D23S90), doi:10.1029/2004JD004829, 2004.

Wang, Y., J. Xin, Z. Li, S. Wang, P. Wang, W.M. Hao, B.L. Nordgren, H. Chen, L. Wang, and Y. Sun, Seasonal variations in aerosol optical properties over China, Atmos. Chem. Phys. Discuss., 8, 8431-8453, 2008.

3. p. 7153, line 4. The difference can be also observed due to the difference in cloud properties over Tokyo and Lauder. It should be discussed in the text.

Agreed. Now noted in the text.

4. p.7154, line 9. In Fig.2a it is better to show solar time on the axis.

Figure changed as requested. This was also requested by reviewer 2.

5. p.7155 line 1. Table 3 is not very informative. All helpful information has been

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already included in the text. I propose to remove the Table 3.

The table has been removed, as also suggested by reviewer 2.

6. p.7155, line 5. Too large solar angle intervals of 5 degrees can lead to an additional bias. I would suggest the authors to make the additional model correction within the bins or to discuss this point in the text.

This point was also noted by reviewer 2. In practice, this is not important for several reasons. Firstly, a large number of scans was included. Secondly, the focus was on smaller air masses where these changes are less important. Thirdly, at Lauder, where the number of scans was smaller, the majority of scans were at precise 5-degree steps in SZA. Finally, we note that the model/measurement ratios were calculated specifically for each scan in the more detailed calculations that appear later. These points are now elaborated in the revised text.

7. p.7155, line 11. The different slope in UV-A and visible region can be attributed not only to the NO2 content but to the aerosol. Aerosol optical thickness has a distinct spectral dependence and, therefore, this can lead to the noticeable spectral dependence shown in Fig.3. This should be clarified in the text.

Here we are discussing the "differential" absorption, which is a term used by the DOAS community to describe more localised spectral features. We now explain this in more detail, and add a further point that the more slowly varying differences are likely to be due to clouds, aerosols, or the geometry of local horizon.

8. p.7155, line 23. I propose to emphasize here or earlier (at line 19) that this spectral dependence takes place in situation with higher ozone over Tokyo.

We have added a comment that not only is the total column of ozone greater at Tokyo, but the effective light path is also greater by virtue of the larger fraction being in the troposphere, where multiple scattering caused by pollution further increases its absorption (as discussed in more detail later).

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9. p.7155, line 25. Does this unexpected increase lie out the error bars? It should be clarified.

Although the absolute uncertainty of the combined measurement set is similar in magnitude to the unexplained differences, any systematic errors are dominated by changes in instrument stability and by uncertainties in the reference lamps, and do not have a strong wavelength dependence. We have re-phrased this discussion.

10. p.7156, line1. It can be also the effect of higher cloud optical thickness and cloud amount. During summer conditions Japan is located in the area of heavy monsoon, which is characterized by significant increase of cloudiness.

Point noted, and text amended to state that in the summer period mid-June to mid-July, Tokyo experiences a rainy season.

11. p.7156, line 5. Is the difference statistically significant?

The differences between the summer and winter ratios at sza=70 are statistically significant. Now stated.

12. p. 7156, line 7. Fig.4 is not clear. I would recommend the authors to show the isolines of several large SZA and use black and white scheme instead of the photo to have better contrast. I would also propose to make several calculations of the obscuration effect using at least isotropic radiance distribution and to add it in the analysis.

The figure has been redrawn to show the path of the sun past the horizon obstructions. We have also improved the labelling of both axes and the curves, and removed the time information. We feel that colour is needed to retain perspective information. Calculations of the obscuration were undertaken, assuming isotropic irradiance. The result is listed as item E5 in table 5. However, , we now discuss it briefly here as well.

13. p.7157, line 13. Do I understand correctly that the calculations were made using the exact SZA of each scan measurement?

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Yes. No change necessary.

14. p.7158, line 26. I propose to show the error bars on the curves of Fig.7.

Error bars are already included for one curve from each site. The error bars are similar for the other curves. This is now stated explicitly in the caption.

15. p.7159. Section 6. It would be helpful to have the direct comparisons between the two methods using the additional figure.

Figure 9 now compares retrievals from individual spectra. The two retrieval methods give consistent results

16. p.7160, line 23. The year should be 1984 as I understand from the reference list.

Actually, both the 1984 and 1989 references are relevant here. Now modified accordingly.

17. p.7161, line 5. I would recommend including the value of the mean tropospheric NO2 content in the caption of Fig.5.

I think the reviewer is referring to Figure 10. We have added the values.

18. p.7163, line 9. From what source the value of the asymmetry factor was taken? It should be a reference here.

We used g=0.61 throughout. It is the default value in the tuv code (Madronich and Flocke, 1995). That paper was cited earlier, but was missing from the reference last. Now added.

New Refs

Madronich, S., and S. Flocke, Theoretical estimation of biologically effective UV radiation at the earth's surface, in Solar Ultraviolet Radiation. NATO, Series I: Advanced Study Institute, edited by C.S. Zerefos, and A.F. Bais, pp. 23-48, Springer, Berlin, 1995.

19. p.7163. line 14. I propose to show the value of total ozone content in the caption

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of Fig.12.

Three ozone amounts are 290, 287.7, and 300.1 DU, as given in Table 4 (now Table 2). Since these ratios are not strongly dependent on total ozone amount, we have added a reference to this table in the figure caption.

20. p.7164, line 7. It is not clear, how it is possible to calculate the combined effect of cloud and aerosol without including the cloudiness in the model? I assume that the authors thought to exclude the effect of cloudiness by having a ratio but this will happen only in the case of the same cloud properties at both sites. This point should be discussed in more details.

We agree that it is not possible to separate these components. We are limited to calculating their combined effects. This point is now clarified further.

21. p. 7164, line 16. Why the asymmetry factor here is 0.67 and earlier it was 0.61? Is it an erratum? What is the value of Angstrom parameter?

We used a value of 0.61 throughout. The value of 0.67 was a typographical error, which has now been corrected. Thank you for spotting that. The Angstrom parameter was taken as -1 throughout, except for in the new discussion - 2nd last para in new Section 8.

22. p. 7164, line 24. It is not always clear whether the aod equals to 0.5 or 0.2 in summer and at what wavelength.

Unless otherwise stated explicitly, all optical depths are specified at wavelength 1 micron. Reviewer 2 also raised this question, so we have clarified that point further earlier in the preceding paragraph.

23. p. 7165, line 21. Fig.13. I assume that the measurements will be within the model calculated values if we account for the uncertainty of measurements at the shortest wavelengths. I propose to add the marks of different aods on the model curves.

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That is correct. The error bars become large as the wavelength approaches its lowest value of 290 nm. This is now stated in the text. The figure has also been altered to include curve labels for each optical depth.

There is an erratum in the caption of Fig. 13: it should be ..around.. instead of ..about..

The caption has been re-written.

It is not clear if the NO2 content has been accounted for in the model calculations.

Corrections for NO2 and SO2 content are included in these model calculations. This was already stated in the sentences immediately before the introduction of Figure 13.

24. p. 7165, line 24. I agree concerning the effects of surface albedo. But I am not sure that this is right if speaking about Angstrom parameter exponent. If you specify aod=0.5 at 1 mkm (like in summer conditions), the transition to 308nm, for example, would lead to the difference of 0.34 in aods calculated with the Angstrom parameter exponents of 1 and of 0.8. And this difference gives about 5-10% change in the UV flux at 308 nm. At the same time, Angstrom parameter exponent can be much higher (about 1.4-1.6) over the urban area due to increasing of the fine mode aerosol particle distribution. This should be at least discussed in the text.

In all calculations the value of alpha has been specified at 1 micron.

Thank you for bringing this point to our attention. We had not expected alpha to be large in urban environments, since that is about as far from a Rayleigh atmosphere that one can get. However, papers that have recently appeared in the literature indeed give much larger values of alpha under polluted conditions. For example, Wang et al (2008), the range of alpha can be very much larger than the range that we tested. For example in NE China, their values ranged from alpha $<$ 0.5 to alpha $>$ 2.

The effects on global irradiances of changes in the Angstrom parameter depend strongly on the assumed single scattering albedo. In this case, where we are modelling the combined effects of clouds and aerosols, we have assumed a single scat-

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tering albedo of 0.95m (already stated early in this section), which admittedly would probably be too large when considering aerosols alone. With this choice, the effect of changing optical depth is rather small, since most of the scattering results simply in a redistribution of the radiation, mainly in the forward direction. The point the reviewer is raising is that many different combinations of aerosol properties could be investigated with the radiative transfer model, and it is likely that some of these choices could significantly improve the agreement between model and measurement. In particular, it could be argued that the large seasonal variability in alpha reported by Wang et al, could be a factor in the poorer agreement between measurement and model in Fig 13c (winter, sza = 70). However, this seems unlikely, since their lowest values of alpha (most different from the value assumed here) occurred in summer. Their values in winter, when there is poorer agreement are more similar to our assumed values.

We have carried out further RT calculations to investigate this further, using a wider range of alpha (0.6 to 2.0) and a wider range of single scattering albedo (0.60 to 0.95), and have found the behaviour in winter is more consistent with using a larger value of alpha and a corresponding smaller value of beta. Unfortunately, we were unable to find a realistic combination of parameters to match the observations, which we still think are more influenced by horizon obscuration during the sunnier winter period. We now discuss this point further in the text.

We cite the paper by Bais (2005) which explores this space more fully. With smaller ssalbs, the effects of differences in the Angstrom parameter can lead to much larger changes in the spectral slope, so in principle, it should be possible to find a combination of aerosol parameters to adequately model the measured ratios in winter (such as those shown in Fig 13 c).

We have shied off the task of using the model to explore all possible combinations of alpha, beta, ssalb, and g, since this would be a huge task in its own right, and the plausible range of variabilities is not known.

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We have also corrected a typo, where we had referred to the Angstrom exponent as beta instead of its more usual notation of alpha.

25. p.7167, line 22. This is true only if the sun is obscured by clouds. If not, the signal can be quite stable even in cloud conditions. And, vice versa, in conditions with high aerosol loading the signal can be very unstable.

We agree with those possibilities, and have deleted the sentence.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 7149, 2008.