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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 #2 Received and published: 6 May 2008

The paper aims at investigating the effect of tropospheric pollution on surface UV irradiance, by comparing measurements at an urban site (Tokyo) with those at a clean site (Lauder). Quantitative results are presented for the summer and winter seasons, and the effects of the various UV influencing factors are discussed in adequate detail. The data to support the findings of the paper are limited to one year only for Tokyo and to one and a half year for Lauder. The limited length of the dataset reduces the robustness of the results quantitatively. The methodology that is followed to analyze the measurements is presented clearly. I think that the paper should be accepted for publication with some revisions.

Thank you. It is true that the temporal coverage of data is rather limited, and is therefore unsuitable for trend studies. However, the study includes all four seasons, and a very large number of scans.

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Specific comments

The paper has many figures. Figure 1 may be omitted since the reader can get only a rough idea of the irradiances measured at the two sites. The comparison between the two sites is illustrated repeatedly in figures 2 and 3and it can be inferred form figures 6, 7 and 13. Figure 4 is very informative for the operators, but may not be necessary for the reader of the paper.

We think the figures are necessary to orientate the reader. Fig 1 contains additional information about the huge seasonal and cloud-induced changes in irradiances. It also includes additional information about ozone, showing that compared with sza and clouds, ozone is less important. These points have now been further emphasised.

Figure 4 is important and central to much of the discussion. We also feel that inclusion of this effect, and our manner of treatment for it is quite novel. The paper shows that horizon obscuration is an important consideration in urban environments such as Tokyo. However, the figure has been simplified and more clearly labelled to improve legibility.

The information contained in tables 2 and 3 may be incorporated in the text.

Done.

The retrieval of column amounts of NO2 and SO2 is marginally innovative, in the sense that the DOAS method is not new and there is no way to judge whether the retrieved columns are correct or not. Although the absorption signatures of the two gases are evident in one particular case (Figure 10), it is hard to say that this constitutes a new scientific result. In absolute sense, the retrieved slant columns are not validated and only qualitative statements are made for the levels of the retrieved quantities. Are the authors confident that the absolute levels of Figure 9 are correct? How large can be the uncertainty of these results? On the other hand, some meaningful information is given in Figure 8, which shows the seasonal differences of NO2 in Tokyo, but with respect to

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Lauder.

Now compared, and corrected as a result (see introductory comments) The actual values of these trace gas column amounts are not the central issue in this case. Rather, we are concerned here with identifying their radiative effect on UV irradiances. This has been emphasised by combining sections 7 and 8, and renaming the section "Absorptions by Trace Gases". Error bars on the NO2 and SO2 retrievals are given. Originally these included just the statistical error in fitting, but we have now revised them to include systematic errors. We are confident that these are realistic in the case of NO2, where we have considerable experience - our group pioneered the DOAS method (before it was known as that). We are less confident about the error bars for SO2 since there can be cross talk between O3 and SO2 absorption cross sections. We have expanded that discussion to bring out those points as noted above.

1. 7151, 2: The reference to aerosol single scattering albedo here does not fit with the discussion of the paragraph. I suggest rephrasing.

Agreed. Now rephrased in terms of reduction in irradiance. The effect on single scattering albedo is now described in the next sentence.

2. 7151, 3: Absorption effects of air pollutants can be enhanced in the presence of aerosols through increasing scattering. I think this should be mentioned here.

OK

3. 7151, 27: Explain why one scan is made at midnight.

The sentence has been removed (the midnight scan is done for QA/QC purposes).

4. 7152, 13: Actinic flux and irradiance are influenced differently by aerosols. This makes the direct comparison of the two quantities dependent on the actual conditions. Was this taken into account in the back-correction of the irradiance measurements?

It is true that aerosols affect irradiances differently from actinic fluxes. For high sun

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conditions, the presence of aerosols tends to increase the ratio of actinic flux to irradiance, whereas for low sun it tends to decrease the ratio, though the effect is smaller at larger sza. We estimated the magnitude of this effect using the tuv RT model, with sensitivity study of the effect of an increase in AOD from zero to 0.235 at 1 micron. In this case, where the back-correction was made by comparing irradiances and actinic fluxes measured at sza 60- 65 degrees, the additional uncertainty due to variability in AOD is relatively small, since the effects of aerosols are then quite similar for both quantities. The increased error bars encompass any additional uncertainties from this effect, which are estimated to be less than 4%.

5. 7154, 8: How the transmissions were calculated? Are they normalized to SZA=0?

These tropospheric transmissions are now defined more carefully. They are analogous to Cloud Modification Factors (CMFs) that have been used previously, but including aerosol effects. They are not normalised to SZA=0

6. 7154, 11: Was total ozone stable during these days? Would it be possible that the asymmetry is caused by changing ozone column?

The asymmetry is not due to changes in ozone, since it is present for both the UVA and UVB irradiances. This was not shown, so is now stated explicitly. We have also added further information about the ozone variability. Ozone variations through the Lauder day were similar to those for the Tokyo day: approximately 5%. At Tokyo, the maximum ozone occurred at about 10 am, whereas at Lauder it showed a monotonic increase throughout the day.

7. 7155, 5: The irradiance in each 5_ bin can change significantly, especially at large SZA.

That is true, especially for the SZA=70 observations. However since we have averaged a large number of scans, we would expect these effects to average out. If this were an issue, it would appear as noise in the sza-dependence. It may however be a factor in

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the lower than expected values for sza=70 at Lauder in Fig 7. Although many of the observations are at 5-degree steps in sza, that is not so for the observations during the noon time interval. Lauder data are more likely to be affected since there were a smaller number scans available. We have added a sentence to this effect.

8. 7155, 11-13: I suggest plotting the absorption cross sections of the three gases on figure 3. This would highlight the discussed absorption features.

Since there is already a large number of figures, and the cross sections of NO2 and SO2 are already implicit in Fig 10, we have added the absorption signature of ozone to that figure, and refer the reader to that figure here.

9. 7155, 24: What could cause this increase in the ratio? Different cloud patterns of cloud thicknesses? More pollutants?

We have added here that the ratio could conceivably increase from changes in clouds or aerosols (trace gas effects are unlikely), but we argue later that it is most likely due to horizon obscuration at Tokyo.

10. 7156, 27: If the comparison was repeated for other wavelengths, one could estimate better if the effects are from clouds or from aerosols.

Its doubtful that extra wavelengths would clarify this, since the wavelength dependencies of clouds and aerosols are both similar and smooth.

11. 7157, 5: Why these calculations (reduction of diffuse irradiance) are not shown in the paper?

They are included in Table 5 (item E5). This is now discussed briefly here as well.

12. 7157, 14: How good was the agreement between measurements and model?

The agreement between this model and NIWA measurement is excellent for clear skies, as has been assessed previously (e.g., see Badosa et al., 2007 now cited here). In this case we are comparing cloud affected measurements with a clear sky model, so we

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do not expect close agreements. The comparison is shown for two wavelength regions in Figure 7.

13. 7157, 23: The SZA of 68_ is not small!

True. We meant that the air mass is not large, and have now stated this (AMF $<$ 3 for sza=3).

14. 7158, 20-25: Why the average transmission in Lauder is the same in the UV-B and UV-A, whereas this does not happen in Tokyo? In addition, Tokyo has much smaller SZA dependence than Lauder. Could this be attributed to the effect of the aerosols in Tokyo which decreases the contribution of the direct component and redistribute the diffuse radiation? Are similar effects evident in the actinic spectra?

We now point out the difference noted, and speculate on possible causes. Unfortunately we do not have corresponding actinic spectra from Lauder to investigate this further. As noted above, there should be no sza-dependent or wavelength-dependent bias in irradiances arising from the way we have back–corrected our data. The back-correction was based on UVA irradiances for sza from 60 to65.

15. 7159, 9: Describe how these ratios are formed. Are these average transmission spectra, or individual measurements? Are the spectra measured at Lauder used as "reference" (free of effects from pollutants) spectra?

Now clarified. Yes, the Lauder spectra are used clean reference values. Also clarified.

16. Mention that T(Tokyo) or T(Lauder) are transmission spectra.

OK. Now done.

17. 7159, 19: The retrieved tropospheric ozone effect is not discussed. Since it is included in Table 5, a reference to this table should be given here. This discussion is partly repeated in section 8.

All discussion about ozone effects has now been moved to section 8. As suggested

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below, sections 6 and 7 are now combined under a single heading: "Absorptions by Trace Gases"

18. 7160, 21-27: Why Figure 9 was made with data derived from the "standard" DOAS method and not from the alternative method described before in this section?

Now done with both methods. The standard method was used as a check. It should be more accurate, since it has been well validated against other measurements (e.g., Hofmann et al., 1995). Its main advantage is that it uses logged spectra rather than the irradiances retrieved from those spectra, which have a higher noise level due to the relatively small signal from the calibration lamp. It is also designed for use with individual spectra rather than mean values. A further reason for including this version is to show the consistency between results from the new method and the standard DOAS method. These points are now discussed in an additional paragraph in section 6 (old section 7). In the original version of Fig 8, the error bar included only the statistical error in fitting. We have expanded the error bar to include the effects of errors in the cross sections, and uncertainties in the residual absorption in the Lauder reference spectra. Figure 9 has been redrawn because when checking these points, we found that all 26,000 scans had been included, rather than just the scans with low diode noise.

New Ref.

Hofmann, D., P. Bonasoni, M. De Maziere, F. Evangelisti, G. Giovanelli, A. Goldman, F. Goutail, J. Harder, R. Jakoubek, P. Johnston, J. Kerr, W.A. Matthews, T. McElroy, R. McKenzie, G. Mount, U. Platt, J.P. Pommereau, A. Sarkissian, P. Simon, S. Solomon, J. Stutz, A. Thomas, M. Van Roosendael, and E. Wu, Intercomparison of UV/visible spectrometers for measurements of stratospheric NO2 for the Network for the Detection of Stratospheric Change, Journal of Geophysical Research, 100 (D8), 16,765-16,791, 1995.

19. 7160 (actually 7161), 10: The lowest transmission is about 0.6 so I suggest changing the absorption of 50% to 40%.

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OK.

20. 7160 (actually 7161?) , 19-20: This statement is not supported by the discussion and the results presented in sections 6 and 7.

We presume the reviewer is referring to page 7161. The absorptions in Moscow are indeed comparable in the means found here (˜ 2% at 340 shown Figure 1 of Chubarova, 2008), though the peak absorptions are not quantified. We've now clarified to state that just the mean absorptions are comparable with Chubarova's.

21. 7161, 3: Is it really necessary to have this section 7 separated from section 6?

No. Now combined to emphasis the radiative effects of NO2 and SO2, which is the main point at issue.

22. 7162, 23-26: This sentence should be rephrased. (The effect of differences.... on spectral UV where modeled using the DISORT ....)

OK.

23. 7163, 4-7: If the irradiance calculations were made for Tokyo, why ozone columns measured at Lauder were used? I suggest deleting in line 5 the words "for Tokyo".

OK.

24. 7163, 7: Mention the wavelength for which the aerosol optical depth is considered.

The optical depth is specified at 1 micron.

25. 7163, 9-10: Delete the word "integral" in both occurrences. Figure 12 shows spectra and not integrals.

The integral actually refers to Table 5. But, it can also be used when referring to Figure 12. We have replaced "integral" with "region".

26. 7163, 18: This reversal is theoretically possible, but it may be also caused by stray light effects or the low signal at these wavelengths.

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The point is now noted that both model and measurement are less accurate at these shortest wavelengths.

27. 7164, 2: Mention here for clarity that the measured spectral ratios are averages.

OK.

28. 7165, 9-10: I think that the quoted better agreement of the afternoon ratios with the model results in winter is not justified by figure 13. The spectral dependence is still opposite from that of the model. There should be other reasons to explain this spectral effect. One possibility could be effects from clouds and the fact that these ratios are averages of spectra measured over a period of 6 months each acquired under different conditions (ozone, aerosol, clouds, tropospheric absorption, etc). I wonder whether this pattern remains the same if instead of 6 months the authors would have used 3 month averages (DJF) for winter.

We tend to agree, and the section has now been revised by removing the offending phrases. The seasonality question is addressed to some extent, at least for the summer months, by comparing results for different sza-ranges. For example, SZAs less than ˜30 occur only for the 3 summer months, whereas SZA of 70 can occur in all months. No significant anomalies arising from these changes in averaging periods were apparent in Figures 7 and 8. The results are quite consistent for all choices of SZA.

29. 7166, 11-13: The model was run for 2 different values of the ozone column, while the averaged spectra were measured under different ozone columns. The combined ozone effect on the averaged irradiance cannot be the same with the effect of the averaged ozone on irradiance, especially if one considers also the combined effect of the high aerosol load in Tokyo. This may be one reason for the higher measured ratios in the UVB. One way to investigate more deeply this effect would be to compare individual spectra (measured and modeled) and not averages.

Individual spectra from Tokyo are too noisy, even on the clearest days, due the effects

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of changing aerosols and clouds during scans. This is why we decided to focus on averaged spectra.

30. 7174, Table 4: The header or at least the caption should mention clearly what quantities are shown in Table.

Now added in the caption. Hopefully this revised version will not have a typographical error (introduced by the typesetter, we think?), where day 177 appeared as day 1770.

31. 7178, Figure 2: The upper panel should be labeled in hours instead of decimal day of the year.

We have now changed the figure according to this suggestion.

Technical comments

7151, 1: Replace "absorptions" with "absorption"

OK.

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