Interactive comments on:

"Effect of aerosols and NO_2 concentration on ultraviolet actinic flux near Mexico City during MILAGRO: measurements and model calculations"

by G. G. Palancar et al.

Anonymous Referee #1

We thank the anonymous reviewer for the insightful and helpful suggestions which resulted in an improved manuscript.

General Comment

The target quantity mainly used in this study, i.e. spectral actinic flux integrated over a wavelength range 330-420 nm, is somewhat unusual. A more natural choice would have been to calculate j(NO2) in the first place because the photochemical background for this investigation is clearly stated by the authors. j(NO2) is also not affected by the ozone column and the influence of NO2 concentrations on j(NO2) is obviously more direct. Moreover, the integrated 330-420 nm AF is not purely UV related as indicated by the title and in the abstract. As is evident from Fig. 6 roughly one third of the photon flux densities are from the visible range. If there are reasons why j(NO2) was not calculated, selecting the UV-A range, e.g. 320-400 nm, would have been more conventional. Also UV-A is hardly affected by ozone columns. The authors should at least show that their target quantity is highly correlated with j(NO2) independent of NO2 concentrations and aerosol loads which is probably the case in the solar zenith range considered here. However, my guess is that in this sense 320-400 nm would have been a better choice than 330-420 nm. Anyway this is not a major problem.

Answer: The choice of the upper limit (420 nm) was dictated by the upper limit of the SAFS instrument, while the lower limit arose (as the reviewer correctly recognized) from our desire to stay away from O3 absorption (which would carry considerable uncertainty in this polluted region). In addition to NO2, many other molecules can be photo-dissociated in this wavelength range, including HONO and organics such as the alpha dicarbonyls. By considering actinic flux rather than any specific molecular photolysis coefficient, we avoid the separate issue of uncertainties in photolysis cross section and quantum yield data, and their frequent updates. We also prefer to avoid strictly using UV-A, which is ambiguously defined in the literature as 315-400 nm or 320-400 nm, and is usually for irradiance rather than the actinic flux here.

In practice, most of the quantities being discussed are highly correlated and the exact choice of wavelength range is not critically important. As suggested by the reviewer, we calculated both J_{NO2} and the integrated actinic flux, for a few times of the day and for both polluted and clean cases, showing that their ratio is constant to within a few percent (last column of the table below)

Time	SZA	F	JNO2	JNO2/F
LT	degr.	1e16 cm-2 s-1	1e-3 s-1	1e-19 cm2
(polluted)				
12n	19	2.39	9.20	3.85
1pm	24	2.33	8.96	3.85
2pm	37	2.10	8.08	3.85
3pm	52	1.69	6.45	3.82
4pm	68	1.02	3.83	3.75
5pm	84	1.84	6.93	3.77
(clean)				
12n	19	2.45	9.45	3.86
1pm	24	2.41	9.29	3.85
2pm	37	2.26	8.70	3.85
3pm	52	1.97	7.45	3.78
4pm	68	1.43	5.37	3.76
5pm	84	0.35	1.22	3.52

[Calculated with TUV5.0 for 15 March 2006, 19N, 2.2 km asl, O3 column = 300 DU, delta-Eddington radiative transfer. Polluted case includes 1 DU NO2 column and AOD = 0.4 at 340 nm, T=273K]

For these reasons, we believe that our use of the 330-420nm integral is sufficient to elucidate the physics of how pollutants affect actinic fluxes, and using alternative definitions would give essentially identical results. We have added the following sentence to section 3 "Radiative transfer model":

The actinic flux integrated over 330-420 nm is essentially proportional to the NO_2 photolysis coefficient, but less sensitive to the temperature-dependent NO_2 quantum yield and cross sections, with a ratio of J_{NO2} to the AF of $(3.80\pm0.05)x10^{-19}$ cm^2 using the NO_2 spectral data at 273 K from Sander et al. (2011).

Specific Comments

Page 19245, line 4: "UV actinic fluxes (AF)" should be more specific, e.g. actinic fluxes (AF) in a wavelength range 330-420 nm. As mentioned above UV does not strictly apply.

Answer: The sentence was changed as follows: "*We present the first detailed comparison between actinic fluxes (AF) in the range 330-420 nm measured in highly polluted conditions and simulated with the Tropospheric Ultraviolet-Visible (TUV) model.***"**

Page 19246, line 24: "(AF)" should be deleted here because AF is later used for the integrated quantity.

Answer: AF is used throughout the paper as the linguistic abbreviation of Actinic Flux, not as a mathematical symbol of spectral or integrated Actinic Flux. Also for this reason the font used for AF in normal rather than math/italics font.

Page 19251, lines 4 and 7: Replace total, direct and diffuse "voltages" by "spectral irradiances".

Answer: done

Page 19255, line 2: Replace "total" by "spectral".

Answer: done

Page 19255, line 5: Add "spectral" to actinic flux to distinguish from the quantity considered so far.

Answer: done

Page 19252, line 11: Replace "JPL, 2006" by "Sander et al., 2011".

Answer: done

Page 19256, line 9 and Fig. 6: Why wasn't the apparent wavelength shift corrected for the SAFS instrument based on the positions of the Fraunhofer lines?

Answer: The wavelength shift observed in figure 6 is now corrected and the issue clarified in the text. As a consequence the average agreement for downwelling AF changed to (0.94 ± 0.06). The text between lines 9 and 13, page 19256 was changed as follow:

A shift in wavelength registration, by about one nm, was evident in the upward-facing aircraft SAFS instrument (downwelling radiation) compared to the other two instruments and the TUV model. Thus, this shift was corrected before processing the average spectral agreement. However, when integrated over a wavelength range (330-420 nm) the error introduced by this shift is minor, but increases the overall error estimate.

Page 19263, line 12: Remove citation "Jet propulsion laboratory: : :"

Answer: done

Fig. 4: I suggest that the authors add the term "spectral" and use the same units for spectral irradiance and spectral actinic flux, e.g. spectral photon flux densities for better comparability. For a single wavelength this conversion is explicit.

Answer: The word spectral was added. We also changed figure 4, which now shows the spectral irradiance and the spectral actinic flux in the same units. However, as the "usual" or more familiar units for spectral irradiance are W m^{-2} nm^{-1} we decided to

show both magnitudes in these units. To point out this, we added the following sentence in the text (page 19255, line 6):

Spectral AF measurements and calculations at 368 nm are also included in the figure, and were converted to W m^{-2} nm^{-1} for easier comparison to the irradiance. Note that the absolute value of the AF is larger (by about a factor of 2 for SZA = 60° in accord with simple theory, e.g. Madronich, 1987) because, contrary to the irradiance, it is not weighted by the cosine of the angle of incidence.

Also, following a comment by Reviewer #2 we realized we made a mistake in this figure. Data for actinic flux did not correspond to 13 March but to 12 March. Figure 4 is now corrected (Actinic flux agreement is now consistent with Fig. 2).

Fig. 9: Captions to a) and c) should be exchanged.

Answer: done