

## ***Interactive comment on “Use of filter radiometer measurements to derive local photolysis rates and for future monitoring network application” by Hannah L. Walker et al.***

### **Anonymous Referee #2**

Received and published: 17 July 2020

General Comments: The work ratios long-term filter radiometer jNO<sub>2</sub> measurements to a cloud-free radiative transfer model (TUV) to calculate a measurement-driven adjustment factor (MDAF). Modeled jO<sub>3</sub>, jHONO, jCINO<sub>2</sub> and jHNO<sub>3</sub> are then multiplied (presumably) by the MDAF correction factor to determine the environmentally impacted local values. Combining these rates with local measurements, the authors calculate radical products and find a significant reduction in OH compared with the cloud-free modeled products. The authors suggest that such local radiation measurements would significantly improve chemical model calculations of important species.

The local impact of clouds, aerosols and changing albedo on photolysis rates and

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the resulting impacts on radical chemistry require additional validation in models. The long-term dataset discussed provides the opportunity to examine a range of conditions to test model sensitivity to changes in local conditions and the incentive to set up additional jNO<sub>2</sub> measurement locations. The paper is relevant but lacks rigor in some areas. With revisions and additional discussion the paper would be worthy of publication.

Specific comments: Line 13: 40% lower OH production rate compared to what? Presumably compared to the cloud-free model. Best for clarity to be explicit. Some discussion of cloud-free vs cloud resolving models would be useful.

Line 35: I suggest an alternative PAN reference:

Singh, H. B., Herlth, D., O'Hara, D., Zahnle, K., Bradshaw, J. D., Sandholm, S. T., Talbot, R., Crutzen, P. J., and Kanakidou, M.: Relationship of Peroxyacetyl nitrate to active and total odd nitrogen at northern high latitudes: Influence of reservoir species on NO<sub>x</sub> and O<sub>3</sub>, *J. Geophys. Res.*, 97, 16523–16530, 1992.

Line 70: Many molecules include a pressure dependence. I suggest adding pressure (p) to equations.

Line 72: The definition attributed to Madronich is confusing. Actinic flux is more simply the spherically integrated radiation through a sphere or the radiant energy density incident on a unit spherical surface.

Line 78: Here and throughout the paper, the authors refer to applying or using the MDAF. However, I don't see the "application" is ever explicitly defined. I am left to presume (perhaps obviously) that MDAF is a simple multiplication factor. That should be stated here.

Line 86: This seems like an odd line noting that jNO<sub>2</sub> instruments have "limited potential to estimate the photolysis frequencies of other atmospheric species." Isn't that the point of this paper? I think you are saying that they cannot \*directly\* measure the other

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species without MDAF or a similar correction.

Line 103: This is an incorrect oversimplification. The Palancar data was screened for clouds and included PBL heights in addition to AOD and NO<sub>2</sub> concentrations.

Line 127: The difficulties of a 4 pi spectrometer should be discussed. In particular, the upwelling can be significantly influenced by the support tower, any nearby equipment and the local albedo (e.g. a bush or rock below the tower). This can add significant uncertainty, particularly when the upwelling is large (e.g. snow). Also, the errors in the two 2-pi optics are particularly large near the horizon. This is noted throughout the literature (e.g. Hofzumahaus, et al, (2002) doi:10.1029/2001JD900142 and references therein). See the comment at lines 258-61.

Lines 146-9: I recommend being explicit about the averaging for clarity. The filter radiometer is broadband and continuously measures the full jNO<sub>2</sub> spectrum. The scanning spectrometer measures one narrow wavelength band linearly in time over the 3 minute duty cycle. Thus, the measurements are not equivalent in rapidly changing conditions.

Line 158: Be consistent TUV 5.3 or 5.3.1 (as noted in the abstract).

Line 161: The model is not technically “clear-sky” because aerosols are included. I suggest using “cloud-free” throughout.

Lines 163-4: List the default values for albedo and aerosols (presumably 0.10 and the Elterman (1967) continental aerosol profile).

L. Elterman UV, Visible and IR Attenuation to 50 Km, AFCRL-68-0153, Environ. Res. Papers, (No. 278) (1968), Bedford, Mass.

Lines 168-9 : I believe default TUV uses a different cross section (Vandaele et al., 1998). According to JPL 2015, Vandaele and Mérienne are within 2-3% across their measurement spectral range.

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Line 190: Describe  $\text{KH}_2\text{O}$ ,  $\text{KN}_2$  and  $\text{KO}_2$ .

Line 207: Eq 2 is specific to  $\text{jNO}_2$ . One option would be to make Eq 2 generic and adjust the (“in this case”) text in line 75.

Line 215: Discuss expected seasonal variation in  $\text{ClNO}_2$  as Sommariva was summer-time only.

Line 258-61: A figure would be helpful to understand the SZA relationship to MDAF. At low sun, the angular response of the radiometer optics will contribute significantly to the MDAF. How does the filter radiometer data compare to the Bentham at low sun? In addition, the model will be particularly sensitive to the aerosols applied in TUV (Elterman). These will generally differ from the actual profile (including the PBL height). I expect the MDAF analysis is not particularly effective at very low sun (perhaps  $>85$  deg sza). This topic should be addressed and perhaps such data needs to be excluded.

Line 287: The discussion of  $\text{HNO}_3$  is a bit odd. Either it should be given the full analysis of the other molecules or it should be excluded because it is not relevant to the OH analysis. The low OH production from  $\text{HNO}_3$  is not even mentioned in the conclusion, perhaps because it is not an interesting finding of the study. If  $\text{HNO}_3$  is included, why not a list of other photolysis frequencies?

Line 289 and Table 3: I think this must state “cloud-free TUV” for clarity.

Line 295:  $\text{jNO}_2$  and  $\text{jO}_3$  cover significantly different spectral ranges resulting in differing diurnal profiles and interactions with clouds and aerosols. More importantly,  $\text{jO}_3$  is highly dependent on the  $\text{O}_3$  column. Applying the MDAF to  $\text{jO}_3$  will result in significant uncertainty and I question whether it is a valid method. For more, see Lefer et al., 2003 (doi:10.1029/2002JD003171). The  $\text{jClNO}_2$  and  $\text{jHONO}$  have spectral parameters similar enough to  $\text{jNO}_2$  that MDAF is likely ok. The paper lacks any discussion of uncertainties resulting from the MDAF results for each of the molecules.

Lines 364-70: This would be a good place to discuss using MDAF for an expanded set

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of photolysis frequencies beyond those listed in this paper. The metric would be useful for other studies.

Fig 6: What happened in June? Does this affect the annual averages shown in figure 7?

Technical corrections: Line 76: Change “and temperature” to “temperature (T) and pressure (p)”

Line 144: Shouldn't this be “>9V”

Line 166: Perhaps, note that is below, “shown in Eq. (3) (below in Section 2.4.1)”

Lines 174-5: Remove “it has been demonstrated that” and change “could” to “can”

Line 258: Change “was” to “were”

Line 316 and Fig 10: Previously used only “p(Cl)ClNO<sub>2</sub>” but now shortened to “p(Cl)”. I suggest sticking to the long version.

Line 328-9: Reword for clarity “compared with when the diurnal cycle of ClNO<sub>2</sub> is unaccounted for” to “compared with the constant ClNO<sub>2</sub> concentration”

Fig 8: Legend should say “HONO (cloud-free TUV)” and “O<sub>3</sub> (cloud-free TUV)” for the dotted values

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