

Interactive comment on “Photolysis frequencies in water droplets: Mie calculations and geometrical optics limit” by B. Mayer and S. Madronich

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The reviewer made a number of constructive comments for which we like to thank him. In particular, following his suggestion, we included a table of enhancement factors for typical droplet size distributions which can be directly applied in chemistry models to correct the photolysis frequency of any tropospheric species. Below are our detailed replies to the points the referee raised. The original referee comments are printed in italic.

Most of my comments apply to the presentation of the material. The derivation of the various equations in the paper is a bit confusing, especially for a non-specialist reader. This should be improved (Section 2; Appendix A). In fact, I like the use of Appendices: then the focus in Section 2 can be on the information that is relevant to the non-specialist. However, currently the rationale behind the separation between information in Section 2 and in the Appendix A is unclear to me. Section 2 cannot be understood without reading first the Appendix. Appendix

B relies on information (equations) that is given in Section 2. Below I give a few concrete suggestions that could improve readability.

Is answered below.

Secondly, it is a pity that the authors did not take this opportunity to include a few typical enhancement factors for some key photolysis rates, e.g., ozone, peroxides, for a few effective radii or other parameters describing cloud droplet size distributions. These results can easily be obtained by integration of the presented results as a function of size parameter over wavelength and cloud droplet radius. Examples could help the application of the results presented in this paper by atmospheric modellers, e.g. for sensitivity tests. It seems unrealistic to assume that full Mie calculations can always be included in atmospheric chemistry models.

This is an extremely valuable suggestion. We included a table providing enhancement factors for typical cloud droplet sizes, for wavelengths between 300 and 800nm. As the variation with wavelength is rather slow, the table can be used to determine the enhancement factor for any species, by selecting an appropriate wavelength from the table.

Section 1, lines 4-5: Maybe you can add, next to these modelling papers, one or more references to observations of the effects of clouds on the actinic flux?

Done. We included some more references to experimental and theoretical papers.

To start the derivations with the introduction of radiant power and definitions of the (un)perturbed fluxes is not needed and is confusing to the reader. Better start with a definition of the actinic flux, the focus of the paper. In my view the first paragraphs of Section 2 would need only three equations for the reader that is less interested in computational details: 1) A definition of the actinic flux (integral of the radiance over 4π) within a medium as the product of the energy density and the speed of light in the medium. 2) A definition of the actinic flux enhancement factor in terms of a ratio of energy densities in both media and the refractive index. 3) The relation between the photolysis rate and the actinic flux (equation 20),

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with reference to Appendix A for its derivation.

The rationale for presenting the equations this way was to provide the basics and the relevant results in the main part and the complex derivations in the appendices. After correcting a small error pointed out by the reviewer (see below) and some small re-arrangements we think that the structure is much more logical now. Furthermore, both appendices are independent; that is, the reader may read and understand Appendix B even if he hasn't read A. This requires some of the formulas in the main part, if we don't want to repeat equations.

Currently the reader is likely to apply the definition of Q_{abs} in equation (5). This leads to a dimensionless enhancement factor that is inverse proportional to the number of absorbing molecules in a droplet ($\eta=1/N$) (??).

This confusion is caused by the accidental use of the same symbol σ_{abs} for two different quantities, the absorption cross section of the individual molecule and that of the droplet. This is clarified now, by using symbols σ_{abs} and Σ_{abs} .

Further, SI units of the various quantities are not given. E.g. I guess the actinic flux is in Watts per square meter.

We think, as long as no real numbers are introduced, it is not necessary to define units for all quantities because they are straightforward to derive.

I propose to define most of the given quantities only in the Appendices when these are needed in the derivation of the photolysis rate equation (equation 20, Appendix A) and/or geo-metric optics limit (equation 6, Appendix B).

See above. After correcting a small error pointed out by the reviewer (see above) and some small re-arrangements we think that the structure is much more logical now.

In Section 2.1 I recommend to rename the real part of the refractive index n into n_r , to distinguish from $n = n_r - i n_{im}$.

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This was certainly confusing. We adopted the definition of Born and Wolf now who use n for the real part and $n\kappa$ for the imaginary part.

In Section 2.2 it seems more logical to start the derivation of the Mie-derived enhancement factor with the energy density ratio as derived by Bott and Zdunkovski, followed by equation (9). Equation (8) is not needed here. The authors should consider to add a reference to the formula for Q_{abs} as a function of the complex part of the refractive index and the size parameter as given in: Van de Hulst, H.C., 1981, Light Scattering by Small Particles, p.181., Dover, New York.

The presented order of equations is a logical proof of the main point, that an n is missing in the Ruggaber et al. paper. After some changes it should be straightforward to understand now.

Appendix A: I would suggest to move the relation between actinic flux and energy density to Section 2. Appendix A should focus on the absorbed radiant power, the net flux divergence and the actinic flux to arrive at equation (20)

Again, the rationale for presenting the equations this way was to provide the basics and the relevant results in the main part and the complex derivations in the appendices. To move the relation between actinic flux and energy density to the main part would imply to move also several of the definitions, including that of the radiance. We think that would make the paper much more complicated to read. In the main part, the relation is stated (Eq. 2), together with a reference; the interested reader may read the appendix to understand why this is so, but those only interested in the implications for chemistry don't need to do so.

Technical corrections

All considered. Thanks!

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