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Atmospheric Chemistry and Physics Discussions

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

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General comments

The paper reports on the enhancement of the actinic flux inside cloud water droplets in comparison to the actinic flux in the surrounding interstitial air. The authors argue that photolysis of water-soluble components inside droplets may play an important role in atmospheric chemistry.

The authors have produced a concise and interesting paper. The science is sound. Important result is that by re-interpretation of, and corrections to the existing scientific literature a consistent picture has emerged from both Mie theory and in the geometrical optics limit. This is a very valuable result and I strongly recommend the paper for publication.

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:

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.

Specific comments

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? Section 2, lines 1-25: 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 4 pi') 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

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photolysis rate and the actinic flux (equation 20), with reference to Appendix A for its derivation.

Currently the reader is likely to apply the definition of Qabs 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) (??). Further, SI units of the various quantities are not given. E.g. I guess the actinic flux is in Watts per square meter. 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 geometric optics limit (equation 6, Appendix B).

In Section 2.1 I recommend to rename the real part of the refractive index n into nr, to distinguish from n = nr - i nim.

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 Qabs 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.

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)

Technical corrections Abstract, line 4: remove 'the' P4107, L19: 'droplet size distributions' (plural) P4107, L20: 'types of clouds' (plural) P4108, L23: remove 'either' P4111, L17: 'given by Eq.(6). P4112, L8: add to equation (10) the condition '(for nim $<<$ nr)' Appendix A, L16: next => net Figure 3: The x-axes should be the same (either remove or add an 'x') Figure 4: The figure caption is not very precise. Maybe use: .... REFWAT (Wiscombe, 1994) in comparison with data taken from Hale and Query (1973). Figure 6: Add to the figure caption a warning on the different scales used for the y-axes

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