

Title: The impact of parameterising light penetration into snow on the photochemical production of NO_x and OH radicals in snow

Authors: H.G. Chan, M.D. King, M.M. Frey

Review:

General comments

This paper presents a snowpack actinic flux parameterization that has the potential to be incredibly useful for simulating snow photochemistry in global climate models and global chemical transport models. Although the analysis is robust, the manuscript will be more structured once it is reorganized and some of the figures are elaborated on more in the text.

It is great to read that e-folding depth method agrees well with the RT method under most circumstances and when the e-folding depth and surface actinic flux values from TUV snow are used. However, for the ze method, where will e-folding depths and surface actinic flux values come from going forward? Surface direct and diffuse downwelling irradiance from global chemical transport models and global chemical transport models can be used to calculate the surface actinic flux, but these models will not be able to calculate e-folding depths in snow. Please address this topic in the manuscript.

Towards the end of the paper, the importance of nonBC absorbers in the UV wavelength region is acknowledged. If this parameterization is ultimately going to be incorporated into large scale models, it is worth making sure that this parameterization can successfully calculate photolysis rates in snowpacks with nonBC absorbers. If it is not possible to perform sensitivity studies with nonBC absorption in this study, please outline a plan to perform these sensitivity studies before the parameterization is included in global chemical/climate models. Also, will this parameterization only be valid for deep snowpacks found in polar regions? In shallower snowpacks, actinic flux drops off more than exponentially right near the underlying surface (e.g. soil, rock). It would be very useful to develop this parameterization for shallow snowpacks as well for incorporation in large-scale models.

This paper should be accepted with major revisions.

Specific comments:

P8610, L12: I think that the transfer velocity should be referred to as the “depth-integrated photolysis rate constant” throughout the text. It helps the reader understand what this term physically represents.

P8610, L16: Mention that RT is short for radiative transfer

P8610, L24: reduces instead of reduce

P8611, L8: Mention that OH is the hydroxyl radical.

P8611, L11-12: add “the” before atmosphere and “a” before source and also before sink.

P8611, L15: the “to be” is not needed before preserved

P8611, L15: Mention that NO_x is nitrogen oxides

P8611, L17: Add that “photochemically-active” species (e.g. NO₃) are the proxies to be more specific

P8612, L15-16: The enhancement in actinic flux in the top few cm is only for certain solar zenith angles (SZA), less than ~50 degrees.

P8612, L25: check tense of ‘providing’. I’m not sure if ‘provided’ would be better

P8614, L6: Provide the range of wavelengths that these reactions each occur over

P8614, L13: Provide range of quantum yield and also absorption spectrum

P8614, L16: How deep in the snow is it expected that NO₂ would produce O₃ by R4 and R5?

P8615, L5-8: This sentence needs to be ended with something like “are calculated in this study, with and without an algorithm designed to improve X”.

General comment: Are you taking into account the fact that R6 and R7 occur in the condensed phase while R4 and R5 occur in the gas phase?

General comment: Is it possible to include non-black carbon species in this analysis (e.g. insoluble organics (HULIS), dust, brown carbon).

P8615, L15: Are these all polar studies, or are some mid-latitude studies as well?

The semi-infinite snowpacks are most commonly found in polar regions, so will this parameterization be limited to deep snowpack (e.g. > 3 meter depth) regions? It would be really useful to make the parameterization valid for shallower snowpacks as well.

P8616, L10: Although black carbon is the most effective absorber by mass, non-black carbon species (e.g. HULIS) dominate the ultraviolet absorption (e.g. nonBC material absorbs 89% of radiation at 307 nm) – see Zatko et al. [2013], likely because there is much more nonBC material in the snow. The parameterization would be most realistic if nonBC absorbers were also included, but perhaps this can be included in the future.

P8616, L18: Zatko et al. [2013]

P8616, L20: remove “a” after has. Also, add reference for this sentence.

P8616, L24: stratosphere does not need a capital

P8617, L24: add “an” before ice

P8618, L4: what is the vertical resolution from the snow surface to 20cm? Is it also 1 cm?

P8618, L6: Going forward, will the e-folding depth always be obtained from TUV? Or is the goal to always use field measurements of e-folding depth? If the goal is to use field e-folding depths, a robust study of field e-folding depths compared to TUV model output should be performed, similar to the Appendix in Zatko et al. [2013].

P8618, L19: species instead of specie

P8618, L16: Similar to my e-folding depth comment, will the surface J value ultimately be obtained using actinic flux values from the field?

P8619, L8: coefficient should be plural

P8619, L13: parentheses around ‘i.e. total column ozone’

P8619, L19-21: By how much do the ze differ across that wavelength range and solar zenith angle range for all the different snowpack types? If nonBC absorbers were included, there would be a distinct wavelength dependence on e-folding depth

even from 321 to 375 nm. The snowpack types included likely have different density profiles, which should influence z_e (Figure 2a).

General comment: Will this parameterization be able to account for varying amounts of direct and diffuse radiation? Please specify the fraction of direct to diffuse incoming solar radiation in TUV.

P8620, L4-5: Increased scattering leads to increased chance of absorption by black carbon and other absorbers

P 8620, L22: Please include which SZA that the purple line represents in the text. Also, which figures in Figure 4 show that the purple line ($sza=76$) overestimates relative to the RT method? In the figures on the left (top and bottom), the purple dashed and solid lines are similar; it's hard to see that the z_e method overestimates relative to the RT method in this case. In the figures on the right (top and bottom) the purple line is in the middle of the other lines, so it seems like the purple line shouldn't stand out.

General comment: The discussion switches back from Figure 3 and Figure 4 throughout this section. Please try to discuss all of Figure 3 first and then discuss Figure 4 so that the reader doesn't need to keep switching back and forth between figures.

P8621, L5: The sentence about anthropogenic pollution is interesting, but out of place. Please move it to a section that is more relevant (3.2.3?).

P8621, L19-20: add "and" and remove comma between fluxes and photochemical
P8621 (bottom) and P8622 (top): Refer to the figures that are relevant to each statement. Also, NO_3^- , H_2O_2 , and NO_2 disagreements are given in percent but for NO_2^- , the disagreement is expressed in factor form. Change NO_2^- to percent to be consistent.

General comment: Please add letters to all multipanel plots (e.g. a, b, c, d)

P8622, L5: change 'negligible' to 'negligibly'

P8622, L6-7: There are some missing words in this sentence.

P8622, L9-10: Remind readers what the action spectrum for each species is.

P8622, L15: coefficients

P8622, L16: add "that" before have

P8622, L13-22: Please include figures that show changes in J when the ozone column is changed.

General comment: Section 3.2.2. should be split up into two separate sections, or at least include the discussion of how different chemical species are influenced by SZA in the previous section about SZA. The ozone sensitivity studies should be its own section.

P8623, L1-6: This paragraph needs to be moved to an earlier section where Figure 3 is first described. Figure 3 should somehow be color-coded so that these key points stand out clearly in the figure (see Figure 3 comment below).

P8623, L10-11: State the depth where the maximum in J occurs for clean snowpacks and describe why the J maximum depth varies for each different snowpack type.

P8623, L15-16: Please describe what would happen to the agreement with RT and z_e when the density is lowered. The agreement between the RT and z_e approaches is important and it would help the reader understand these concepts better.

P8623, L24: compounds

General comment: Section 3.2.3. should be renamed “variations in snow physical properties” and then you should add a section after that titled “variations in optical properties” and include the “variation of asymmetry factor section” into it. In the “physical property” section, describe Figure 6 (it isn’t described anywhere else in the text). In the optical section, include the direct vs. diffuse discussion from section 3.2.2.

P8624, L11: I think the transfer velocity should be referred to as the “depth-integrated photolysis rate” throughout the document. This will make more sense to the reader. Also, change the axis labels in Figure 7 and Figure 8 accordingly.

P8625, L2: velocities to velocity

P8625, L8 and L10: factor should be factors, or add ‘a’ in front of factor

P8625, L11: snowpack to snowpacks

General comment: In section 3.3., Figures 7 and 8 need to be introduced and described in more detail.

Tables and Figures:

Table 2:

Meusinger et al. [2014] and Zhu et al. [2010] also report quantum yields for NO₃-photolysis. The use of the Chu and Anastasio [2003] quantum yield should be justified or additionally sensitivity studies should be performed using the reported quantum yields from these two studies.

Table 3 and 4: Why doesn’t H₂O₂ have a, b, and c coefficients?

Figure 1:

- Why is the wavelength 451 nm used for comparison instead of a more photochemically-relevant wavelength (such as from 298-345nm)?
- Is the snowpack typically for polar regions or for other regions too?
- angle should be plural

Figure2:

- The red line description is missing in the figure caption

Figure 3:

It might be helpful to give each of these scenarios a name (e.g. like you’ve done for BC₁₂₈ on Figure 4). Right now it’s hard to extract information from the figure. It would be effective to use the same color but different line style for similar runs that are slightly different. For example, instead of having dark blue, dark green, and black represent different densities but otherwise same BC and scattering, you could have these lines all be dark blue but vary the line style – e.g. one solid, one dashed, one starred.

Figure 4:

- Purple doesn’t need to be capitalized

-It would be helpful to add a line on the two left plots that distinguishes which SZA leads to the ze method underestimating with respect to the RT method compared to leading to overestimates.

Figure 6:

-The caption suggests that the transfer velocity is being plotted, but it looks like it is the photolysis rate constant that is being plotted instead. The second sentence should describe what the different colored lines are to orient readers.

-I don't understand or see from the plot what the second sentence of the caption is getting at or referring to.

-For the Scatt2 case, shouldn't the density of melting snow be higher than the standard case? It looks as though you account for changes in grain size though by lowering the scattering cross-section.

-There are several sentences in this caption that describe results of the figure. These sentences belong in the part of the main text where Figure 6 is introduced and described.

Figure 7 and 8: make angle plural. In Figure 7, the last two sentences can go into section 3.3. The R squared sentence can go into section 3.3 instead of the caption in Figure 8.