

Reply to referee 2, RC C1721 for: The impact of parameterizing light penetration into snow on the photochemical production of NO_x and OH radicals in snow

We thank the reviewers for their reviews and recommendation to publish. We have considered every point and corrected the paper to include their points.

Abstr, line 16: please define the abbreviation 'RT'.

Fixed, Abstr, line 11-12, now reads "... calculated (a) explicitly with TUV, a physical radiative transfer (RT) model, ..."

Abstr, line 19: 4 chemical species are mentioned, but only 3 ranges are given for the ratio result. This is confusing. Please clarify (also in numerous other locations in the manuscript). If NO₃⁻ and H₂O₂ have indistinguishable results, might it help to define an aggregate term for the two species?

The parameterization of nitrate and hydrogen peroxide were done together as the action spectrum for both species peaks at 321 nm. For clarification, new coefficients for nitrate and hydrogen peroxide were calculated separately and listed in Table 3 & 4. However, the R² remain the same with the new found coefficients compare to previous calculated coefficients. The ratio results are updated according to the new calculated coefficients.

Pg8611, line 10: Does the emission flux from the snowpack depend also on other factors such as dry deposition or wind-pumping? Perhaps such factors could be briefly mentioned and their potential impact discussed? Please also briefly discuss the inherent assumptions about the nature of the ion-containing medium (snow/ice/brine), with references. (Perhaps the recent paper by Domine et al is relevant here). These assumptions could affect both the absorption characteristics of the molecules and the transfer of photolysis products out of the medium. Of course these factors are the same whichever photolysis model is used, however they do have a bearing on the subsequent chemistry, so should be mentioned.

In the Introduction, now reads

"... The porous structure of snowpacks allows the exchange of gases with the atmosphere, that is the release and uptake of atmospheric gases and particles by the surface snow. The exchange between snowpack and overlying atmosphere depends on dry and wet deposition, transport (including wind-pumping and diffusion) and snow microphysics (Bartels-Rausch et al., 2014). Thus snow can act as both a source and a sink of atmospheric chemical species as summarised in Bartels-Rausch et al., 2014 and Grannas et al., 2007. ..."

Pg 8611, line 17: The last sentence in the paragraph needs a subject.

Fixed, now reads "Photochemistry in the snowpack needs to be fully understood because: (1) emitted photolysis products play an important role in determining the oxidizing capacity of the lower atmosphere, e.g. concentration of O₃, HO_x, H₂O₂, and (2) chemical preserved in ice cores, and potential paleo-climate proxies, may be altered by reactions with OH radicals, photolysis or physical uptake and release (Wolff and Bales, 1996)."

Pg 8612: The discussion of the two snowpack layers could be better organized. Please introduce the names of the two layers more explicitly early on (e.g. in line 6) and use them consistently.

Fixed, now reads “Under clear sky conditions, a homogeneous snowpack can be separated into two optical layers based on the propagation of actinic flux from the surface into the snow: the near-surface layer, i.e. the top few centimetres of the snowpack, where direct solar radiation is converted into diffuse radiation. Below the near-surface layer is the asymptotic zone where all solar radiation is diffuse and will decrease exponentially with depth (Warren, 1982).”

P8612, L23 – 25, now reads “In the asymptotic zone radiation is diffused, and provided that the snowpack is semi-infinite, i.e. the albedo of the surface underlying the snow does not affect the calculation of the actinic flux within the snowpack as the radiation decreases exponentially according to Beer–Lambert law. (France et al., 2011 define semi-infinite as 3-4 e-folding depths)

Pg 8613, line 11: add (θ, z) to $J_{\text{sub}} z_{\text{sub-sub}} e$.

Fixed

Pg 8613, line 16: the term “transfer velocity” suggests (at least to this reviewer) some contributions from physical processes (inter-medium transfer, wind-pumping and so on). I don’t believe you mean to imply this. “Depth-integrated photolysis (or production) rate” is more descriptive.

Fixed, the term “transfer velocity” is changed to “depth-integrated photolysis rate” within this manuscript.

Pg 8613, line 17: do you mean “which may be CONSIDERED approximately proportional”?

Fixed as suggested.

Pg 8613, line 23: Does this also suggest the requirement that photolysis makes negligible difference to $[A]$?

Yes and it is now fixed

Pg 8614, R1-R4: It would be helpful to include notes on the wavelengths of maximum effectiveness for each reaction, on the same line as the reaction itself.

Edited the wavelengths of maximum effectiveness for each reaction together with the reaction itself.

Pg 8616 line 24: “stratospheric” should not be capitalized.

Fixed

Pg 8618: Please introduce Figure 2, and discuss the variation of e-folding depth with density.

Fixed. The following sentence has been added to Section 3.1

“At all wavelengths, the e-folding depth decreases with increasing snow density and increasing the mass ratio of the black carbon increased the absorption of incident radiation.”

Pg 8619, lines 2-4: This sentence is a little confusing.

Fixed. Now reads

“Depth-integrated photolysis rate coefficients of the four chemical species considered (NO_3^- , NO_2^- , H_2O_2 and NO_2) were calculated by the RT method and the z_e method. To evaluate the accuracy of the approximation by the z_e method, the ratio Q , (V_{TUV}/V_{z_e} , using Eq. 9), is calculated and considered independently.”

Pg 8619, line 13: do you mean ‘e.g.’ instead of ‘i.e.’?

Fixed

Pg 8620, line 13: Please introduce Figure 3. It doesn’t appear to be mentioned in the text until later, after the discussion of Figure 4.

Figure 3 has been first introduced and discussed in Section 3.2.1

Figure 4 has now taken out from the manuscript

Pg 8621, line 22: Please explicitly introduce Figure 6.

Yes, Figure 6 has now been introduced and discussed in Section 3.2.3

“Scattering cross-section of the snowpack: Lower values of the scattering cross-section implies longer path length of the photon between individual scattering events. Hence, the maximum photolysis rate coefficient tend to occur deeper into the snowpacks, as shown in blue in Fig. 6 (Scatt2, i.e. melting snow), compared with snowpacks that have a larger scattering cross-section (magenta in Fig. 6, BaseC, i.e. cold polar snow). Thus for snow- packs with a small scattering cross-section the agreement between the RT and z_e methods is likely to be poor as the z_e method will not capture the behaviour in the near-surface layer accurately.”

“In Fig. 6, black lines representing the extreme polluted case - BC128, the photolysis rate coefficient calculated by the two methods matches at around 2 cm depth for the NO_3^- anion, but ~4 and ~5 cm for the NO_2^- and NO_2 respectively.”

Pg 8624, lines 15 & 18, and Table 3: The first snowpack type is inconsistently named: is it “windpack and cold polar”, “windpack” alone, or “general”? The name “general” should be defined in the text and/or in an earlier Table, if it is to be used.

Fixed. “general” and “standard” snowpack has now renamed as the “BaseC” (base case snowpack) and is defined in Table 1.

Pg 8625, lines 18-20: This is a very good point, which makes your results more useful in the general case.

Thank you

Pg 8626, Conclusions: You focus on under/overestimation, which is important. Can you also say whether you actually recommend your method, and under which conditions it works best? Please add a discussion of how the input parameter e-folding depth might be assessed and distributed for the types of large-scale modeling studies where your parameterization would be applied, and

discuss, based on your results, how this would contribute to uncertainty in the chemical production rates.

[Suggestion on how to apply the correction factors are now included to Section 3.3](#)

[The following texts had been added](#)

[“Snowpacks with a large e-folding depth, i.e. > 30 cm, e.g. either have small scattering cross-section or contain small amount of light absorbing impurities, are suggested to apply correction factors for “melting and clean snow” when solar zenith angles are smaller than 50° and larger than 80° to reduce the error by 10–30%. For snowpacks that have an e-folding depth smaller than ~ 30 cm should apply correction factors for “windpack and cold polar snow” when solar zenith angles are smaller than 30° or between 60–70°, which could reduce the error by up to 15%.](#)

Tables in general: It would be helpful to include an initial table (I'll refer to it as Table 0) listing the seven hypothetical homogeneous snowpacks (or at least the 3 main types used) by name, along with their characteristics. This would enable a lot of simplification in the text, in the Figure captions, and in

Table 1, which could then focus more clearly on how variables change in the sensitivity studies. Table 1: Since e-folding depth appears to be a major input to your parameterization, it would be helpful to list the calculated reference e-folding depths at 321 nm either in this table or in a summary “Table 0”.

[Table has now been added](#)

Table 1: the “Case” names defined the Table are not used in the text. Please consider doing so, to help orient the reader.

[Improved regarding to the labelling different hypothetical snowpacks.](#)

Table 3: Why do you not list a,b,c for H₂O₂?

[The parameterization of nitrate and hydrogen peroxide were considered together as the peak in the action spectrum for both species is the same, 321 nm. For clarification, new coefficients for nitrate and hydrogen peroxide were calculated separately and listed in Table 3 & 4. However, the R² remain the same with the newfound coefficients compared to previous calculated coefficients.](#)

What is a “general” snowpack? Is it the same as a “standard” one?

[Please refer to reply for Pg 8624, lines 15 & 18](#)

Table 4: See comments for Table 3.

[Please refer to reply for Table 3.](#)

Table 5: this table should be either referred to in the Introduction (and renumbered accordingly) or moved to Supplementary Information.

Figures in general: Please add figure keys if possible, in keeping with ACP style. Yellow lines do not show up well. Can you find a stronger alternative color? The captions contain lots of detail. Perhaps this could be simplified by strengthening

the Tables, enabling the Figure captions to use snowpack /snow type names as shorthand for the specific parameter lists.

Fixed, figures keys were added within the figures

Figure 3: Please include the term 'Q' explicitly in the first line of the caption. The term 'Q' is added to the caption, now reads "The ratio of depth-integrated photolysis rate, $Q = \nu TUV / vze$, for the two different methods ..."

Figure 4 caption should read "The effect. . . on photolysis rate coefficient"
Fixed.

Reference:

- Bartels-Rausch, T. and Jacobi, H.-W. and Kahan, T. F. and Thomas, J. L. and Thomson, E. S. and 20 Abbatt, J. P. D. and Ammann, M. and Blackford, J. R. and Bluhm, H. and Boxe, C. and Domine, F. and Frey, M. M. and Gladich, I. and Guzmán, M. I. and Heger, D. and Huthwelker, Th. and Klán, P. and Kuhs, W. F. and Kuo, M. H. and Maus, S. and Moussa, S. G. and McNeill, V. F. and Newberg, J. T. and Pettersson, J. B. C. and Roeselová, M. and Sodeau, J. R.: A review of air-ice chemical and physical interactions (AICI): liquids, quasi-liquids, and solids in snow, *Atmospheric Chemistry and Physics*, 3, 1587–1633, doi:10.5194/acp-14-1587-2014, 2014.
- Grannas, A. M. and Jones, A. E. and Dibb, J. and Ammann, M. and Anastasio, C. and Beine, H. J. and Bergin, M. and Bottenheim, J. and Boxe, C. S. and Carver, G. and Chen, G. and Crawford, J. H. and Dominé, F. and Frey, M. M. and Guzmán, M. I. and Heard, D. E. and Helmig, D. and Hoffmann, M. R. and Honrath, R. E. and Huey, L. G. and Hutterli, M. and Jacobi, H. W. and Klán, P. and Lefer, B. and McConnell, J. and Plane, J. and Sander, R. and Savarino, J. and Shepson, P. B. and Simpson, W. R. and Sodeau, J. R. and von Glasow, R. and Weller, R. and Wolff, E. W. and Zhu, T.: An overview of snow photochemistry: evidence, mechanisms and impacts, 7, 4329–4373, doi:, 2007.
- France, J. L., King, M. D., Frey, M. M., Erbland, J., Picard, G., Preunkert, S., MacArthur, A., and Savarino, J.: Snow optical properties at Dome C (Concordia), Antarctica; implications for snow emissions and snow chemistry of reactive nitrogen, *Atmos. Chem. Phys.*, 11, 9787–9801, doi:10.5194/acp-11-9787-2011, 2011.
- Wolff Eric W. and Bales Roger C. : *Chemical Exchange Between the Atmosphere and Polar Snow*, NATO ASI Series, 43, Springer Berlin Heidelberg, 1996
- Warren, S. G.: Optical properties of snow, *Rev. Geophys.*, 20, 67–89, doi:10.1029/RG020i001p00067, 1982.