Reply to the review of Anonymous Referee #1

Replies to his/her remarks and suggestions are given below. For the sake of clarity, the reviewer's comments are in blue italics and our response is in black font.

Bock et al. use a model to examine the impacts of the physical exchange processes, adsorption, bulk diffusion and co-condensation, on the depth profiles of nitrate concentration in snow at Dome C, Antarctica. They find that bulk diffusion and co-condensation alone can explain the observed profiles.
This paper was not clearly written and thus very frustrating to read. I don't understand how one can model snow nitrate without including important processes such as atmospheric deposition and the photolysis of snow nitrate. The former is how nitrate gets to the snow in the first place, and the latter has been shown to be the dominant loss process of snow nitrate at Dome C.

Anonymous Referee #1 states that the photolysis of snow nitrate is the dominant loss process of snow nitrate at Dome C, and should be included in a modelling framework dedicated to the study of snow nitrate. We are aware that some studies concluded that the photolysis is the dominant loss process. We already highlighted these results in the submitted manuscript (L92-93). However, as explained in our paper (L50-54), the dramatic increase of nitrate concentration in surface snow during the summertime implies that uptake processes have a much stronger magnitude than sinks (which is mainly the photolysis).
In order to strengthen this point, we added a calculation of photolysis and co-condensation fluxes in the Supplementary Information (Sect. 3):
In this section, an estimation of loss and uptake fluxes is presented. Both calculations are based on the following assumptions: a skin layer thickness of 3 mm, with a snow density of 0.3 kg m^{-3}. The fluxes are calculated for an area of 1 cm^2.
The photolysis flux is calculated for a single nitrate concentration of 1200 ng g^{-1}, which results in 9.7×10^{14} molecules in the 1 cm^2 × 3 mm volume. France et al. (2011) reported a photolysis rate for nitrate of about 1×10^{-7} s^{-1} in Dome C surface snow, for a solar zenith angle (SZA) of 52° which is the maximum solar elevation at Dome C. The resulting photolytic loss flux is 9.7×10^{7} molecules cm^{-2} s^{-1}.
The uptake flux resulting from the co-condensation process is calculated by assuming that the 1 cm^2 × 3 mm volume is filled with ice spheres of radius 85 µm (the value used in this study) up to the prescribed density. This results in ~ 37200 spheres. In the theoretical study by Dominé and Thibert (1996), the average concentration in the condensed layer immediately before another layer condensates and isolates the previous one, is given by the integral of Eq. (15):

$$X_{average} = X_{kin} + \frac{X_{eq} - X_{kin}}{\Delta r} \int_{0}^{\Delta r} erfc\left(\frac{x}{2\sqrt{(D \cdot t)}}\right) dx$$

Using the same input data as in the model, and assuming that this average concentration multiplied by the condensed volume corresponds to the quantity of nitrate actually taken up by the snow, we calculate an average uptake flux of 5.4×10^{9} molecules cm^{-2} s^{-1} over the December 2009 to January 2010 period. The minimum and maximum values are 1.6×10^{8} molecules cm^{2} s^{-1} and 2.7×10^{10} molecules cm^{-2} s^{-1}, respectively.
As a conclusion, the uptake flux due to the co-condensation appears to be ? 56 times larger, on average, than the photolysis flux calculated for the highest solar elevation condition. This confirms that photolysis loss can be neglected when studying the nitrate concentration in the skin layer.
We added the following paragraph in the main text to refer to this comparison:
(L665-671)
The photolysis has not been included in this study, because the dramatic increase of summer nitrate concentration in the skin layer demonstrate

`that uptake processes overtake loss processes in this specific layer. In order to refine this comparison regarding the budget of nitrate in the skin layer, an estimation of the uptake and destruction fluxes is presented in the supplementary information (Sect. 3). It appears that the uptake flux calculated with the BC3 parameterisation is 1.5 orders of magnitude larger than the maximum loss flux due to photolysis. This confirms that photolysis loss is negligible as compared to the co-condensation uptake when studying the skin layer concentration.`

Anonymous Referee #1 states that atmospheric deposition is an important process which should be taken into account. We are not sure of the exact processes he/she is referring to. However, we emphasise that our study focuses on air – snow exchanges processes on the scale of a snow grain. The study of atmospheric processes is much beyond the scope of our work. However, we also stress that the air-snow uptake processes studied here are able to explain "how nitrate gets to the snow in the first place".

Although I agree that their study is important, as such physical processes will influence the distribution of nitrate in the snow column and the snow grain, the latter of which may influence e.g., how photolabile snow nitrate is, I don't see how they can ignore these other important processes. It seems that it would be better to use their model to examine the results in a laboratory, where the processes they ignore can be controlled. Since the manuscript is not clearly written, it is possible that I am misunderstanding something important about their modeling framework

We agree with Anonymous Referee #1 that the developed modelling framework allows us to infer the distribution of nitrate inside the snow grain, which is likely to influence how photolabile snow nitrate is. We emphasised this aspect in the conclusion by adding the following sentence:
(L743-749)
`In this study focused on skin layer snow, nitrate photolysis inside the snow grain has not been implemented since nitrate loss is much weaker than uptake for this specific layer, as demonstrated by the dramatic increase of nitrate concentration during summer. This is not true for the whole snowpack, and photolysis should be included in a 1-D snow chemistry model. For that purpose, the description of a snow grain as a layered medium will enable using different quantum yields, after some studies suggested that it span more than 2 orders of magnitude depending on the availability of nitrate inside the ice matrix (Zhu et al. 2010, Meusinger et al, 2014).`

Cited Papers:

Dominé, F. and Thibert, E.: Mechanism of incorporation of trace gases in ice grown from the gas phase, Geophysical Research Letters, 23, 3627–3630, doi:10.1029/96GL03290, 1996.

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, Atmospheric Chemistry and Physics, 11, 9787–9801, doi:10.5194/acp-11-9787-2011, 2011.

Meusinger, C., Berhanu, T. A., Erbland, J., Savarino, J., and Johnson, M. S.: Laboratory study of nitrate photolysis in Antarctic snow. I. Observed quantum yield, domain of photolysis, and secondary chemistry, The Journal of Chemical Physics, 140, 244 305, doi:10.1063/1.4882898, 2014.

Zhu, C., Xiang, B., Chu, L. T., and Zhu, L.: 308 nm photolysis of nitric acid in the gas phase, on aluminum surfaces, and on ice films, The Journal of Physical Chemistry A, 114, 2561–2568, doi:10.1021/jp909867a, 2010.