Reply to Quentin Libois (Reviewer #1):

We gratefully thank the reviewer for the detailed review and the numerous larger and smaller suggestions. The comment guided us easily to improve the manuscript. We would like to highlight the efforts of the reviewer, for reading the manuscript very carefully and identifying many typos.

Detailed replies on the reviewer's comments are given below. Our replies are given written with indention. Citations from the revised manuscript are given in italic and quotation marks.

General comments

This study aims at estimating the radiative impact of black carbon (BC) particles suspended in the atmosphere and contained in the snowpack in the Arctic. It simultaneously and consistently computes the radiative forcing of BC in both the snowpack and the atmosphere. To this end it couples an atmospheric and a snow radiative transfer model. The BC atmospheric concentrations are taken from three aircraft campaigns that explored various atmospheric conditions, from early spring to summer. A variety of radiative transfer simulations are performed, where snow properties and BC mass concentrations are varied to cover the range of Arctic conditions reported in the literature. The main conclusion is that the radiative impact of BC is marginal in typical Arctic conditions, amounting to about a few percent of the total heating rates and to less than 1 W m-2 in terms of surface forcing. The authors also point a competition between shading of the surface by atmospheric BC that counteracts the warming effect of BC in snow. The impact of clouds is investigated, also showing complex interactions, where depending on their altitude and optical thickness, clouds can either enhance the effect of BC through multiple scattering, or reduce it by shading. In any case, the authors highlight that other drivers of the Arctic energy budget are more significant than BC, such as absorption by water vapour, snow metamorphism and clouds.

The topic of the study is relevant to ACP because it combines numerical simulations and field observations to provide a geophysical analysis. The paper is well written and easy to follow. There is much relevant physical insight and the conclusions are drawn rigorously from the computations. The findings are not a breakthrough but they have the merit to provide a self-standing investigation of the total BC impact in Arctic conditions, where previous studies have either focused on the atmosphere or in the snow. This is probably the greatest added value compared to previous work. We may regret the lack of field data for the snow. Likewise, the fact that only offline radiative computations are performed precludes a rigorous quantification of the impacts on atmospheric dynamics and snow evolution. As a consequence, the numerous conclusions on the impact of BC with a dynamic perspective appear quite weak and should be better motivated with appropriate references. Practically, data from aircraft campaigns are only used to derive average profiles of temperature, humidity (in a manner that should be more detailed) and BC, but snow properties are chosen based on other studies and more as varying parameters. This is not an inappropriate approach but this makes the importance of novel data quite limited in this work. Based on the comments above, I recommend this paper be published after the corrections suggested below are tackled.

> Again, we thank the reviewer for summarizing the open issues of the original manuscript. The replies on the following specific comments hopefully consider also the general concerns raised by the reviewer.

Specific comments

1) It is clear that the study focuses on BC and the conclusion is that BC is not so critical with the amounts currently observed in the remote Arctic. However, recently there have been plenty of studies clearly showing altered surface albedos because of light absorbing impurities. The latter could then be dust, micro-organisms or anything else. It might be worth insisting that you only deal with BC, which is one amongst many others light absorbing species, so that the conclusion should not be over-interpreted as ≪ there is no impact of impurities in the Arctic ≫. Likewise, the geopgraphical area to which the work is relevant should be better identified.

> We agree with the reviewer, that the estimates of the BC radiative effect calculated in our study cannot be generalized. Other impurities might give a more significant signal. Also we restricted our analyses to snow on sea ice. The Effects of BC might accumulate as BC particles accumulate when snow melts and bare sea ice is left. Ever more important is BC on glaciers where the accumulation does last more than the 1-3 years before sea ice typically melts. In the revised manuscript we emphasized the limitations of our calculations at several instances:
>
> *"For the conditions over the Arctic Ocean analyzed in the simulations, it is found, that…"*
>
> *"This study analyzed the instantaneous solar radiative effect at the surface of Arctic BC particles (suspended in the atmosphere and embedded in the snow pack) over the sea ice covered Arctic Ocean."*
>
> *"It needs to be considered, that this picture might change if the accumulation of BC particles is more efficient than it is over the snow covered Arctic sea ice, where the sea ice and snow pack does not last more than one to three years. Accumulation of BC on e.g. the Greenlandic glaciers will amplify the radiative forcing on a local scale. Furthermore, BC particles are not the only light absorbing impurities, which are transported into the Arctic. The relevance of dust particles and micro-organisms is currently subject of the scientific discussion and may exceed the effect of BC particles (Kylling et al., 2018, Skiles et al., 2018)."*

2) The paper focuses on energy budgets (of the atmosphere and snow). Although the impact of BC on these budgets is very limited, BC strongly impacts the light penetration depth in snow, or equivalently snow transmittance. For instance, if a 20 cm snow layer in the Arctic has a transmittance of 1 %, adding BC may decrease this value down to 0.5 %. This is nothing for the snow budget, buth this makes a huge difference for the amount of energy transmitted. This will for instance be critical for photosynthesis within or under the snowpack. Maybe this should be mentioned somewhere so that again readers don't think ≪ BC does not matter ≫. The paper by Tuzet et al., (2019) may be a useful reference for that.

> Thanks for pointing at this relevant aspect which we did not consider so far. Indeed, our simulations show a significant decrease of transmissivity below the snow layer. For the homogeneous snow layer, the transmissivity in 20 cm depth for the unpolluted case is about 0.3, while adding a BC concentration of 5 ng g^{-1} reduces the transmissivity to almost 0.2. This obviously may have an impact on the radiative processes in and below the sea ice. In the revised manuscript, we added an additional panel to Figure 8 showing the transmissivity profile and added a short discussion.

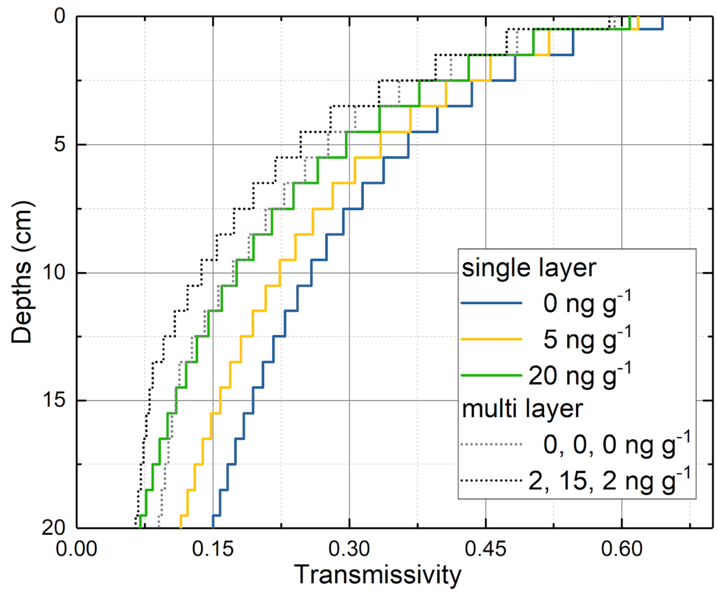

Fig. 8a: Transmissivity profiles of solar radiation within the snow pack for three single layers and one multi-layer case assuming ACLOUD conditions.

“Figure 8a shows the transmissivity profiles of solar radiation within the snow pack. The homogeneous single layer reference case without BC particles (SSA = 20 m^2 kg^{-1}) illustrates the general decrease of transmissivity, which is reduced to 0.3 in 20 cm snow depth. Adding a typical Arctic BC concentration of 5 ng g^{-1} reduces the transmissivity to almost 0.2. This obviously may have an impact on the radiative processes below the snow pack, in and below the sea ice as discussed by, e.g., Tuzet et al (2019) and Marks and King (2014). The inhomogeneous multi-layer case shows in general lower transmissivities due to the enhanced reflection of the smaller snow grains at top of the layer (SSA = 60 m^2 kg^{-1} down to 5 cm depth) but also indicates a significant dimming effect of the BC particles.”

3) The iterative coupling between libRadtran and TARTES is a first valuable step towards consistent radiative transfer simulations. I can only encourage the authors to fully incorporate the scattering snowpack in libRadtran for their future work. This can be done simply by providing the single scattering properties computed by TARTES to create new ≪ atmospheric ≫ layers in libRadtran which would be extremely thin. Such strategy would avoid the iterative coupling and be overall more elegant. See for instance Blanchet and List (1987) for a very similar study.

We agree, a full coupling of both models is the final goal for further studies combining atmosphere and snow radiative transfer. For this study we first aimed to test, if the coupling is possible in general and how large the interaction is. As the iterative coupling shows a very quick convergence, we concluded that this iterative coupling is sufficient for this study. However, for future studies, we will consider the advice of the reviewer.

4) The evaluation of BC contribution to heating rates or total absorption is sometimes misleading. The authors often conclude that BC contribution being a few percents its impact is negligible. However think in terms of CO2 forcing, where a few W m-2 (in addition to hundreds of W m-2) can fully change the face of the Earth. I simply mean that it is hard to conclude that

a few percents perturbation of the energy budget due to BC is insignificant. Be more cautious in the conclusions, unless you have strong and better argumented reasons to think that it is indeed negligible.

> We agree that also only a few W m^{-2} radiative effect can be significant in terms of the total Arctic wide energy budget. As our study is based on three campaigns, the Arctic wide absolute BC radiative effect (or even forcing) cannot be assed. That's why comparing the W m^{-2} of our study to the $C0_2$ forcing would be misleading. Also because the BC radiative effect is a local instantaneous radiative effect, while the climate effect would include all relevant feedbacks. Therefore, we always compared the BC radiative effect to other radiative effects of other properties, e.g., atmospheric water vapor, clouds, snow grain size. At no point in the manuscript we claim, that the BC effect is negligible for the total energy budget. Our conclusion is, that compared to BC radiative effects, other drivers are more important and these other parameters first need to be constrained more precisely to improve e.g., Arctic climate models.
>
> In the revised manuscript, we tried to check all our conclusions and adjusted the wording if needed:
>
> *"The magnitude of solar radiative effects (cooling or warming) of black carbon (BC) particles embedded in the Arctic atmosphere and surface snow layer were quantified on the basis of case studies."*
>
> *"However, in other Arctic regions characterized by higher atmospheric BC particle concentrations due to local fires, e.g., northern Siberia, a stronger impact can be expected. "*
>
> *"These results indicate, that the microphysical properties of the snow pack (mainly snow grain sizes) are more important drivers for the degree/strength of the snow metamorphism. It needs to be considered, that this picture might change if the accumulation of BC particles is more efficient..."*

5) The paper somehow lacks a bit of discussion, where the limits of the study and recommendations for future work would be provided. In particular, the importance of BC in locations where it is much more concentrated could be discussed. The representativity of the BC atmospheric profiles used as well. The use of daily averages to asses a radiative impact may not be relevant (maximum values matter as well). The link with snow metamorphism is only qualitative why models allow to explicitly simulate the impact of these heating profiles of metamorphism, etc. All these points should be brought to the reader and further investigated in future work, if not already further discussed in the present paper.

> We agree, that the discussion of our results was lacking in detail. In the revised manuscript we tried to address all issues raised here by the reviewer. As all of the single issues are listed in the technical correction, we did not explicitly reply here and refer to the replies given below.

Technical corrections

title : would **forcing** be more appropriate than ≪ effects ≫ ? Consider also removing ≪layer≫

> For the title, we do not think that forcing is appropriate. We calculate the radiative effects (forcing) on the surface radiation budget but also the effect on heating rate profiles. To our understanding, the term "forcing" is linked to the energy budget only. So we would keep "radiative effect" in the title.
>
> "layer" is removed.

p.1

l.1 : the abstract could be written using the present. More generally there is no consistent use of present or past in the manuscript. Some homogenization would be recommended.

> We are sorry that we often struggle with the use of the correct tense. We tried to follow our experience publishing in Copernicus journals, where Copy-Editing mostly changes tense into past, when things are done in past. We tried again to homogenize the text and will hope for advice from the final copy-editing process.

l.2 : BC particles are not really ≪ suspended ≫ in the snowpack. They're rather embedded or contained. Consider changing this throughout the paper.

> Changed to embedded throughout the paper

l.2 : ≪ by ≫ → using

> Changed as suggested.

l.4 : ≪ interactions ≫ is unclear. Maybe use multiple scattering or coupling

> We kept "interactions" as multiple scattering is only one process which is considered when coupling the two models. E.g. also the change of direct to diffuse incoming radiation, which is not driven by multiple scattering alone, changes the radiative properties of the surface.

l.4 : ≪ a snow layer ≫ should be replaced by ≪ a snow ≫ because multi-layer snowpacks are explored. Maybe write ≪ An atmospheric and a snow radiative... ≫

> Changed as suggested.

l.6 : this radiative effect is very dependent on the SZA chosen. Please clarify

> We calculated daily mean values considering the diurnal change of the solar zenith angle. Sure, still the results depend on the location, time of year. We therefore added the minimum solar zenith angle and pointed out that the numbers give daily mean estimates of the BC radiative effects.
>
> *"For pristine early summer conditions (no atmospheric BC, minimum solar zenith angles of 55°) and a representative BC particle mass concentration of 5* ng g^{-1} *in the surface snow layer, a positive daily mean solar radiative effect of +0.2* W m^{-2} *was calculated for the surface radiative budget."*

l.9 : counteracting ≪effect ≫

> Changed to:
>
> *"The total net surface radiative forcing combining the effects of BC embedded in the atmosphere and in the snow layer strongly depends on the snow optical properties (snow specific surface area and snow density)."*

l.10 : technically snow density also impacts snow optical properties

> Density was added as suggested.

l.10 : ≪ however ≫ does not really oppose to anything

Changed as suggested.

l.12 : I think ≪ ice ≫ could be used instead of ≪ ice water ≫

Changed as suggested.

l.19 : absorbs, scatters

Thanks! changed as suggested.

L.24 : predominantly

Thanks! changed as suggested.

p.2

l.1 in higher → to higher

Changed as suggested.

l.4-5 : using ≪ nevertheless ≫ and ≪ still ≫ in two consecutive sentences makes it difficult to follow

We changed the sentences to:

"In future, a strong intensification of the ship traffic in the Arctic Ocean and further polluting human activities are expected (Corbett et al.,2010). Still, the direct …"

l.7 : ≪ of suspended ≫ is awkward

We corrected this typo.

l.9 : can be expected → are observed

Changed as suggested.

l.14 : double ≪ the ≫

We corrected this typo.

l.20 : associated with → , thus increasing the amount…

We changed the sentence to:

"The absorption effect can add to the warming of the atmosphere or the snow pack, when the BC particles are suspended either in the air or embedded in the snow. Furthermore, the BC particles may lead to a reduction of the snow surface albedo if the BC sediments on or into the snow pack (Sand et al. 2013)."

l.29 : there is no ≪ novel ≫ feedback described here. BC is just shown to trigger the snow metamorphism feedback. There is actually a feedback because BC impact will be stronger for lower SSA, but this should be described here if this is what you actually mean.

Yes, we did not precisely distinguish booth feedback mechanisms. In the revised manuscript, we changed this into:

“As a further consequence, the absorption by BC particles supports the melting of snow and increases the snow grain size due to an enhanced snow metamorphism, leading to further reduction of the surface albedo. The increase of the snow grain size also feeds back to the absorption by BC particles, which is more efficient for larger snow grain sizes (Warren and Wiscombe, 1980).”

l.34 : warming

Changed as suggested.

l.34 : ≪ the atmospheric layer containing BC ≫

Changed as suggested.

p.3

l.4-10 : this paragraph is not very clear and could probably be removed

As we would like to keep this model aspect in the introduction, we did rewrite the paragraphs as follows to make the statements more clear.

Several regional and global climate models account for the opposite radiative effects of atmospheric BC particles and snow-embedded BC particles (Samset et al., 2014). However, estimates of the total net forcing rely on the accuracy of the distribution of the BC particles assumed in the particular model. Samset et al. (2014) compared 13 aerosol models from the AeroCom Phase II; all of them included BC. They found that modeled atmospheric BC concentrations often show a spread over more than one order of magnitude. In remote regions, dominated by long range transport, these models tend to overestimate the atmospheric BC particle mass concentrations compared to airborne observations. On the other hand, an underestimation of deposition rates induces a lower BC mass fraction in snow (Namazi et al., 2015). While this may introduce significant local and temporal uncertainties of the BC concentration and related radiative effects, long-term trends and mean multi-model results are representative for Arctic-wide observations (Sand et al., 2017).”

l.15 : why only ≪ local ≫ ? Not clear whether this refers to local pollution or not

Our aim was to clarify, that our estimates are not general for the entire Arctic. As local obviously can be misleading, we changes the sentence into:

“On the basis of measured Arctic BC particle mass concentrations for spring and summer months, the instantaneous radiative forcing of BC particles embedded in the snow surface layer and in the atmosphere were quantified for specific cases.”

l.16 : With

Changed as suggested.

l.15 : ≪ interactions ≫ is not very appropriate

We changed this sentence into:

“With help of the coupled model, the interaction of radiative effects in the atmosphere and the snow pack was considered.”

p.4

l.5 : ≪ relevance ≫ is not well chosen → contribution

We changed the last two sentences into:

“Vertical profiles of heating rates in the atmospheric and in the snow pack are presented for clean and polluted conditions. To estimate the impact of BC particles, effective heating rates are calculated by separating the BC radiative effect from the total heating rates.”

l.7 : ≪ setup ≫ suggests there is some evolution from an initiation which is not the case. ≪ Configuration ≫ would be better.

Changed as suggested.

l.8 : change title to ≪BC profiles from aircraft campaigns ≫

Changed as suggested.

l.9 : not clear what this ≪ atmospheric ≫ model is

We changed this sentence into:

“The input for the radiative transfer simulations was adapted to campaign-specific conditions.”

l.23 : are these ≪ snow properties ≫ used later on ?

Yes, these measurements were partly used in the simulations. This is mentioned in Section 2.3, where the snow pack radiative transfer model is introduced.

p.5

l.5 : **is** available

Changed as suggested.

l.10 : consider adding some information about the thermodynamical profiles measured during the flights, if actually used further

Yes, the humidity profiles are used to explain the heating rate profiles and should be added. We included the figure as a second panel to Figure 2, which shows the BC profiles. The discussion of the atmospheric profiles was extended to:

“Fig. 1b shows the profiles of relative humidity, used for the simulations. PAMARCMiP was characterized by rather dry air. Only in the boundary layer, an average humidity up to 60 % was observed often linked to boundary layer clouds. ACLOUD and ARCTAS showed a higher relative humidity in higher altitude of up to 6 km, which indicates the influence of higher level clouds.”

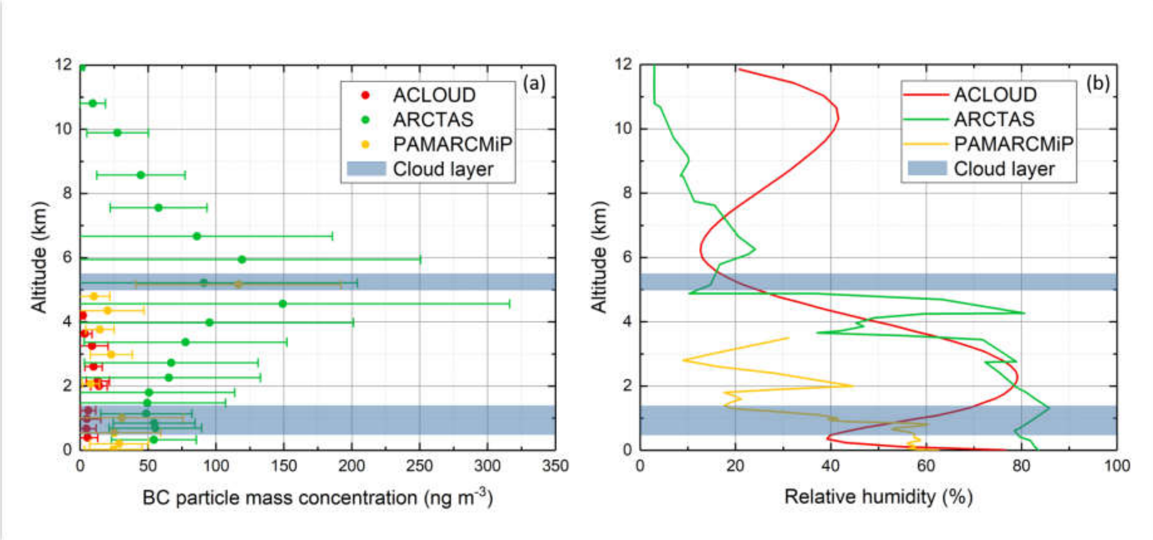

Figure 1. Mean profiles of atmospheric BC particle mass concentration (a) and relative humidity (b) averaged for each the three campaigns (ACLOUD, ARCTAS and PAMARCMiP) as used for the radiative transfer simulations. Horizontal bars indicate the standard deviation. The positions of the two implemented cloud layers (blue shaded area) are marked.

l.13 : url for libRadtran download should be provided here or in the Data availability section

The URL was added as suggested.

l.15 : is reference to ≪ Evans 1998 ≫ relevant here ?

Thanks for identifying this mistake. The reference was removed.

l.15 : precise that this assumes a plane-parallel atmosphere

In the revised manuscript we added a short justification of the assumption of a plan-parallel atmosphere:

"For the calculations, a plane-parallel atmosphere was assumed, which is justified for the Arctic conditions during the three campaigns. Using a pseudo-spherical geometry in libRadtran would change the broadband downward irradiance by less than 0.1 % (0.7 %) for a calculation with a SZA of 60° (75°)."

l.19 : mention explicitly humidity (or water vapor)

Added as suggested

p.6

l.1 : ≪ adjusted ≫ is unclear. Do you mean that profiles from the mid-campaign were used ?

Yes, we used values representative for the campaign, which was the mid-campaign period. The sentence was changed into:

"Corresponding to the campaign average BC profiles, the range of the SZA values was set to values representing the campaign conditions (see Table 2)."

“The standard profiles were adapted to observations from radio soundings near the airborne observations or dropsondes released during the flights and represent the middle of the individual campaign periods.”

l.7 : where do the cloud optical properties come from ?

Yes, this important information was completely missing. In the revised manuscript ee added:

“Optical properties of the liquid cloud were calculated from Mie-Theory, while the ice crystal optical properties are based on (Fu, 2007).”

l.9 : can you provide optical thickness values ?

Sure, we should add the optical thickness and did so in the revision:

“The assumed cloud properties correspond to a cloud optical thickness of 15 for the water cloud and 0.2 for the thin ice cloud.”

p.7

l.2 : provide url for TARTES

We added a web link.

l.6 : The reference provided is not about delta-Eddington approximation. Prefer Joseph et al. (1976)

Thanks for identifying this mistake. We changed the reference as suggested:

“To solve the radiative transfer equation, the delta-Eddington approximation (Joseph et al.,1977) is used.”

l.8 : SSA should not be italic (throughout the text)

Changed as suggested.

l.9 : there are two shape parameters (B and g). Please provide the values used.

In the revised manuscript, these parameters were added.

“Furthermore, the specific values of the so-called absorption enhancement parameter B= 1.6 and the geometric asymmetry factor g^G= 0.85 were applied.”

l.12 : ot → to

Changed as suggested.

l.13 : another important point is that impurities are assumed to be Rayleigh scatterers

We added this important fact to the revised manuscript.

“The impurities are externally mixed and assumed to interact by Rayleigh scattering.”

l.23 : please provide some references for the SSA values assumed

The values are based on our measurements during PASCAL and PAMARCMiP. We added this in the revised manuscript.

"The default values of snow density and SSA were based on measurements during PASCAL and PAMARCMiP and were set to 300 kg m^{-3} and 20 m^2kg^{-1}, respectively."

l.33 : SSA for fresh snow could be larger

Yes, we agree, that fresh snow can have larger values of SSA. However, we chose this value based on the measurements during PASCAL (ACLOUD), where in late spring those values were reported. In the revised manuscript we clarified that the assumption is based on measurements.

"The top layer was assumed to be of fresh and clean snow with …. representing measurements from the PASCAL campaign."

p.8

l.5 : no, spectral albedo does not depend on the spectral distribution of irradiance. Broadband albedo does

Sure, this only refers to broadband albedo. We removed "spectral" in the revised manuscript.

l.7 : ≪ shifts ≫ suggests a conversion of some wavelengths to some others. Maybe say ≪ filters/absorbs longer wavelengths so that the downward irradiance spectrum is shifted towards shorter wavelengths ≫

We reformulated this sentence to:

"The transition from cloudy to cloudless atmospheric conditions increases the direct-to-global ratio ($f_{dir/glo}$) and the contribution of short wavelengths to the broadband downward irradiance (Warren, 1982)."

l.9-10 : for which snowpack ?

We added this information in the revised manuscript:

"For example, simulations with TARTES assuming cloudless and cloudy conditions changed the broadband snow surface albedo from about 0.8 to 0.9 for a SZA of 60° and a snow pack (no impurities) characterized by SSA= 20 m^2 kg^{-1}."

p.9

Figure 2 : ≪ adjusted ≫ parameters is not clear. Do you mean that they can vary ? Maybe specify that the procedure is done at high spectral resolution so that the figure holds for a single wavelength. Consider adding a title with TARTES/libRadtran (or SNOW/ATMOSPHERE) on top of the colored boxes.

Thanks for the hint. We adjusted the scheme and figure caption:

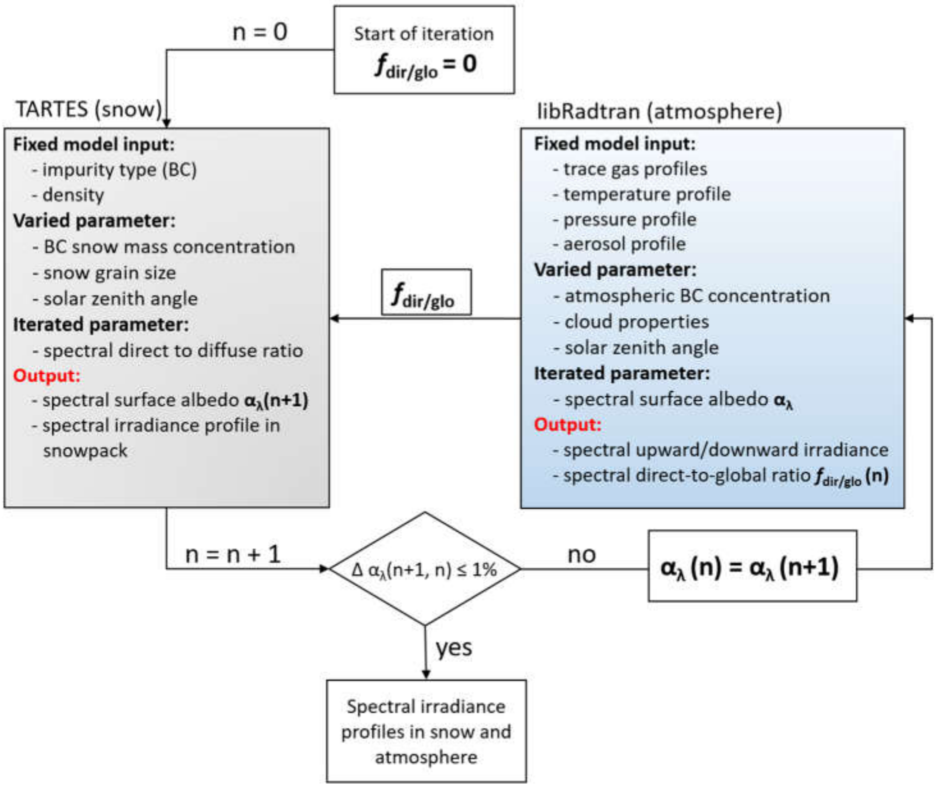

Figure 2. Schematics of the coupling of TARTES (gray box) and libRadtran (blue box) by exchanging the spectral surface albedo and the direct-to-global ratio. The list of varied parameters addresses the variables which were changed between the different realizations. Only the iterated parameters $f_{dir/glo}$ and α_λ are adjusted within an individual iteration cycle.

l.4 : surface radiative effect is not clear (radiative forcing ?)

In the revised manuscript, we distinguish between the surface radiative forcing, which has a well-established definition and the BC radiative effect on the vertical heating rate profiles. We did go through the entire manuscript and exchanged “effect” by “forcing” wherever it refers to the surface radiative forcing. We hope that this makes it more clear, which quantity BC affects in the different discussions.

l.7 : specify what the default cases are when snowpack is considered (what BC in atmosphere ?) or atmosphere is considered (what BC in snow ?)

Yes, this was not fully described. Now we added the definition of the clean reference cases:

“For the separated forcings, $F_{net,clean}$ refers to either a clean atmosphere or a clean snow layer, while the other part does consider BC particles. The default case of a clean atmosphere uses a BC mass concentration in the snow layer of 5 ng g^{-1}. Vice versa, the default case of a clean snow layer assumed the atmospheric BC profile of the ACLOUD campaign. For ΔF_{tot}, the clean reference assumed both a pristine atmosphere and pristine snow layer.”

l.11 : can ? Should be ≪ does ≫ ?

Changed as suggested.

p.10

l.5-7 : the details about the vertical resolution of both radiative transfer codes should be given earlier in the presentation of the models configurations.

> As suggested, we moved this description into the model configuration section.

l.13 : daily means may hide much larger instantaneous values which are very relevant both for snow metamorphism and atmospheric dynamics. Adding the max values on the subsequent plots would be very useful

> We decided to analyze daily mean values to have a better quantification of the total daily effect with respect to the surface energy budget. Heating rates are given in K per "day". Showing the maximum values can be misleading, as the reader may conclude from the unit, that these values are relevant for the complete day. This, we aim to avoid, although we are aware that the maximum heating rates can be higher. Adding the maximum values is also no option as the range of the scale would need to be enlarged and reduce the visibility of the daily mean values.

p.11

l.2 : should be HR_BC ?

> Here we mean heating rates in general including the total heating rates and the efficient heating rate of BC. In the revised version we listed all calculated quantities.

l.6 : what kind of dependence ?

> We changed this sentence into:
>
> *"The reduction of the snow surface albedo by BC impurities depends on the snow grain size."*

l.6 : ≪ respectively ≫ is awkward

> We deleted the bracket.

l.15 : I don't see any zoom of the Figure 4, but definitely this would be useful

We are sorry for the confusion. We included the wrong image file in our first version. It is updated as follows:

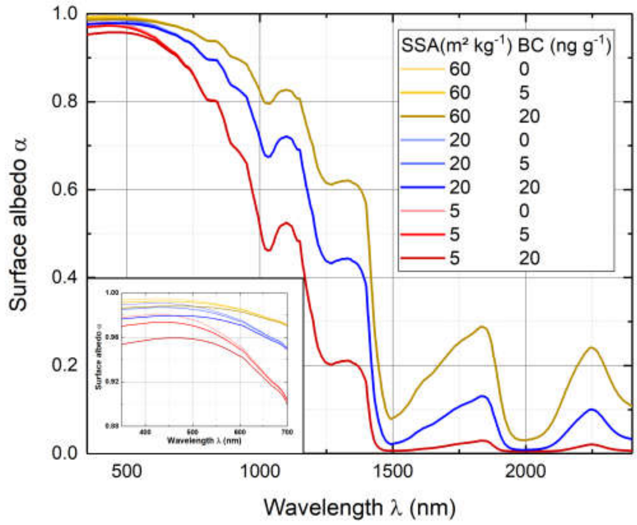

Figure 4. Spectral surface albedo of snow for cloudless conditions and a SZA of 60° for different SSA and BC particle mass concentrations. The inlay shows an enlargement of the spectral albedo between 350 and 700 nm.

l.31 : please specify that this is the impact of BC on the broadband albedo. Other optical quantities might be much more altered

We reworded this sentence as suggested:

"Therefore, for Arctic conditions, the impact of BC impurities on the broadband snow albedo is of minor importance, compared to the impact of modifying the snow grain size."

p.12

Table 4 : particle

Changed as suggested.

l.3 : the distinction between titles 3.1.1 and 3.1.2 is not obvious. Say radiative forcing ?

In the revised manuscript we changed the terminology and used "forcing" for the instantaneous effect of BC on the surface radiative energy budget.

l.5 : twice ≪ effect ≫

Thanks for identifying this typo. We corrected this sentence.

l.5 : **first** calculated… (because standard is with daily cycle). Note also that sometimes the past is used, sometimes the present. It'd be worth homogenizing this.

We changed this sentence into:

“To quantify these radiative effects, ΔF_{snow} was first calculated for a fixed solar zenith angle of 60° only. A typical Arctic range of BC particle mass concentrations in snow and SSA values assuming the ACLOUD atmospheric conditions were applied.”

p.13

l.8 : are **then** analyzed

Changed as suggested.

p.14

l.1 : how much in %? Using relative contributions rather than absolute forcing may be instructive to compare campaigns. This holds also when evaluating the contribution of clouds. Of course they shade the surface, but how does the relative forcing of BC vary ?

We tried to avoid using relative numbers as these might be misleading. Even small absolute effects may be large in relative numbers but still not relevant for the radiative energy budget. There is also no reference to what the radiative forcing can be compared to. In clean cases the forcing is zero, which makes it difficult to calculate relative numbers. The relative effect of clouds can be easily read from Figure 6 and does not need to be given in % in the text to our opinion.

p.16

l.5 : of by

Changed as suggested

l.15-16 : already said in the introduction

We removed this sentence in the revised manuscript.

l.19 : remove ≪ were applied ≫

Changed as suggested

l.32 : ≪ is less pronounced … significantly ≫

Changed as suggested

p.17

l.2 : one order of

Changed as suggested

l.15 : slight

Changed as suggested.

l.27 : ≪ to access ≫ is awkward

This sentence was changed due to another comment.

l.32 : ≪ transmittance ≫ is unclear. Do you mean irradiance with respect to surface irradiance ? Then relative illumination or relative irradiance is better.

Following an earlier comment, we included profiles of the transmissivity in the revised manuscript and extended this discussion.

l.33 : what is ≪ quickly ≫ for a heating rate decrease?

This sentence was changed into:

"For all cases, the total heating rate rapidly decreases by one magnitude within the first 10 cm of depth."

p.19

l.2 : I'm surprised not to see the shading of the lower layers by BC in the topmost layers. Did you observe that below?

We think that the shading is only hard to identify in the profiles of heating rates. More suited are the transmissivities which are now included in the revised Figure 8 (see comments above). Only for the multi-layer scenario, the shading is obvious in the lowest snow layer. Here, the heating rates decrease toward zero, while the top layer (same amount of BC) shows a non-zero heating rate. With the revised Figure 8, this is also visible in the transmissivities.

l.12 : = 0 or ≈ 0 ?≫

Changed as suggested.

l.17 : I think the conversion from a contribution to a snow heating rate into a metamorphism rate is not that straightforward, especailly with daily means. Providing snow physics references would be helpful to strengthen your conclusions

Yes, our conclusion was not well justified. In the revised manuscript, we extended the discussion and compared to results for alpine snow.

"Therefore, in Arctic conditions the snow grain size typically plays a larger role than the concentration of BC particles embedded in snow. To estimate if BC particles can accelerate the snow metamorphism, coupled snow physical models need to be applied (Tuzet et al., 2017). However, compared to the results reported by Tuzet et al. (2017) who studied alpine snow with at least a magnitude higher BC mass concentrations, for Arctic conditions it is likely, that the self-amplification of the snow metamorphism is dominated the reduction of the surface albedo."

l.33 : were used **in** the

Changed as suggested.

p.20

l.3 : ≪ other parameters ≫ is awkward. Please clarify. Maybe mention reference/unpolluted conditions

The sentence was changed to:

"For the heating rate profiles, the effective contribution of BC particles to the total heating rates was derived and compared to further atmospheric and snow parameters also leading to a warming or cooling (e.g., water vapor, clouds, snow grain size)."

l.5 : again, ≪ local ≫ is unclear

As explained above, “local” refers to a local instantaneous effect which cannot be used for the entire Arctic. The changes the sentence to:

“The simulations suggest, that for the specific Arctic cases investigated in our study, the radiative forcing of BC is small compared to the radiative impact of other parameters (water vapor, clouds, snow grain size).”

l.6 : why ≪ therefore ≫ ?

We deleted “therefore”.

l.6 : shows

Changed as suggested.

l.7 : ≪ driver ≫ means that its variability controls the variability of the heating rates. Is that the case (then it should be detailed) ? You could have varying BC for constant water vapour, then the variations of the heating rates would be driven by BC.

Yes, our conclusion was not expressed precisely and might be misleading. We changed the sentence to:

“In cloudless conditions, the absorption by atmospheric water vapor shows a much stronger contribution to the atmospheric heating rates than the radiative effect of BC particles.”

l.15 : lapse-rate feedback refers to the response of the atmosphere to a surface temperature change. In terms of vertical gradient of temperature. I’m not sure you really mean this here (as a feedback).

Thanks again! We remove “feedback” as we can only assume what happens to the lapse rate without all feedback mechanisms.

l.16-17 : again, what is ≪ small ≫ ? can you provide elements to support the fact that 0.1 K day-1 cannot change atmospheric stability ?

Of course, also 0.1 K numerically changed the atmospheric stability. However, the effect is about two magnitudes smaller than calculated for polluted regions (8 K day^{-1} reported by Wendisch et al. 2007). In these polluted cases, changes of the temperature profile by advection, radiative cooling might be slower than the heating by BC particles. But the rate of 0.1 K per day is too slow compared to other processes. We changed the sentence to make this more clear.

“For example, studies investigating strong pollution conditions in northern India or China reported on BC heating rates in the atmosphere larger than 2 K day^{-1}, which may significantly influence the lapse rate and the atmospheric stability (Tripathi et al., 2007; Wendisch et al., 2008). For the rather pristine Arctic, this study showed significantly lower daily mean BC heating rates of maximum 0.1 K day^{-1}, which have not the potential to significantly modify the atmospheric stability.”

l.17-18 : this could be moved to the introduction, that the study focuses on remote Arctic locations, not on locally polluted areas.

To make this more clear, we adjusted the introduction. But we like to keep this sentence also for the discussion in the conclusion section.

"The area of interest is the remote sea ice covered Arctic Ocean in the vicinity of Spitsbergen, northern Greenland and northern Alaska typically not affected by local pollution."

l.33 : these **two** cloud

Changed as suggested.

p.21

l.4 : ≪ Atlantic Arctic ≫ should be emphasized in the introduction

Atlantic Arctic was not correct, as we also use data from ARCTAS (Alaska). Therefore, we changed the abstract to:

"The area of interest is the remote sea ice covered Arctic Ocean at latitudes of Spitsbergen, northern Greenland and northern Alaska typically not affected by local pollution."

l.5 : cooling is at the surface, please clarify

Yes, this effect refers to the surface warming/cooling. We added "surface" in the revised manuscript.

l.10 : some elements should be provided about other types of impurities which may eventually be more critical than BC in the Arctic

Based on another comment, we extended the discussion with:

"Furthermore, BC particles are not the only light absorbing impurities, which are transported into the Arctic. The relevance of dust particles and micro-organisms is currently subject of the scientific discussion and may exceed the effect of BC particles (Kylling et al., 2018, Skiles et al., 2018)."

Reply to Reviewer #2:

We gratefully thank the reviewer for the detailed review and her/his valuable suggestions to improve the manuscript. Detailed replies on the reviewer's comments are given below. Our replies are given written with indention. Citations from the revised manuscript are given in italic and quotation marks.

Page 3, caption table 1: There are various ways of defining BC. Please include a reference to for example Petzold et al. (2013) to clearly define your use of BC. Also please mention how EC values compare with BC values.

> Thanks to the reviewer to bring this up. We are aware that the definition of BC in literature is not consistent. However, Petzold et al., 2013 provided an excellent overview on that topic. Since the terminology depends on the different measurement techniques, we added the applied measurement method in Table 1.

Table 1. Typical values of the black carbon mass concentration in snow pack observed in different regions and seasons in the Arctic. Note, that Pedersen et al. (2015) and Forsström et al. (2013) derived the mass concentration of elemental carbon applying a thermal-optical measurement method.

Location	Season	BC mass concentration (ng g^{-1})	Method	Source
Svalbard region	March/April	13	filter transmission	Doherty et al. (2010)
Arctic Ocean snow	Spring	7	filter transmission	Doherty et al. (2010)
Arctic Ocean snow	Summer	8	filter transmission	Doherty et al. (2010)
Northern Norway	May	21	filter transmission	Doherty et al. (2010)
Central Greenland	Summer	3	filter transmission	Doherty et al. (2010)
Svalbard region	March/April	11 - 14	thermal-optical	Forsström et al. (2013)
Corbel, Ny-Ålesund	March	21	thermal-optical	Pedersen et al. (2015)
Barrow	April	5	thermal-optical	Pedersen et al. (2015)
Ramfjorden, Tromsø	April	13	thermal-optical	Pedersen et al. (2015)
Valhall, Tromsø	April	137	thermal-optical	Pedersen et al. (2015)
Fram Strait	April	22	thermal-optical	Pedersen et al. (2015)

> Further, we cited Petzold et al. (2013) in the introduction:
>
> *"Black carbon (BC) aerosol particles, which mostly originate from incomplete combustion of organic material (Bond et al., 2013; Petzold et al., 2013), absorb and scatter solar radiation in the visible wavelength range and, therefore, influence the atmospheric solar radiative energy budget."*
>
> and revised the manuscript accordingly:
>
> *"The numbers given in Table 1 were derived from different measurement methods. More precisely, thermal-optical techniques were applied in Forsström et al. (2013) and Pedersen et al. (2015) provide the elemental carbon (EC) mass concentration, while filter transmission methods result in BC concentrations (Doherty et al., 2010). As a consequence of the different measurement methods, the ratio of the BC to EC concentration in snow can reach values of 1.3 as reported by Douet al. (2017). A full discussion of the EC/BC terminology can be found in Petzold et al. (2013)."*

Page 4, lines 17-19: Sentence is unclear. Please reformulate.

> We rephrased the sentence:

"In this paper, measurements from the PAMARCMiP 2018 observations conducted from 10 March to 8 April 2018 were analyzed. The research flights, starting from Station Nord/Greenland, were performed above the sea ice in the Arctic ocean north of Station Nord and the Fram Strait."

Page 5, line 15: The reference to Evans (1998) and the SHDOM code appears to be out of place. Should it be Stamnes et al. (1988) instead?

We cite Stamnes et al., (2000) here which explicitly refers to DISORT2.0 as also indicated in Mayer and Kylling (2005):

"As a solver for the radiative transfer equation, the Discrete Ordinate Radiative Transfer solver (DISORT) 2 (Stamnes et al., 2000) routine running with 16 streams was chosen."

Page 5, lines 12-18: 1. How many streams was used for DISORT? 2. The solar zenith angle is large for all regions considered. Did you make any spherical corrections? If not, why not, and how do you expect this to affect your results? 3. What was the vertical resolution of your model atmosphere?

The number of streams (16) is given in the sentence before. Further, the reviewer raises a good question concerning the plane-parallel assumption we applied here. For testing the effect, we compared pseudo-spherical and plane-parallel for SZA ranging between 60 - 75° giving an uncertainty of less than 0.7% in downward irradiance. We added:

"*For the calculations, a plane-parallel atmosphere was assumed, which is justified for the Arctic conditions during the three campaigns. Using a pseudo-spherical geometry in libRadtran would change the broadband downward irradiance by less than 0.1 % (0.7 %) for calculations with SZA = 60° (75°). The vertical resolution of the simulated irradiances was adjusted to the measured BC profiles, ranging between 100 m and 1 km."*

Page 6, Fig 1: The profiles shown are averages. Please also include the standard deviation (or other measure of variability) of the profiles to give an idea of how the profiles varied for the different campaigns.

Thanks for the suggestion. We added the standard deviation in Figure 1 and included profiles of the relative humidity in a second panel as suggested by the other reviewer.

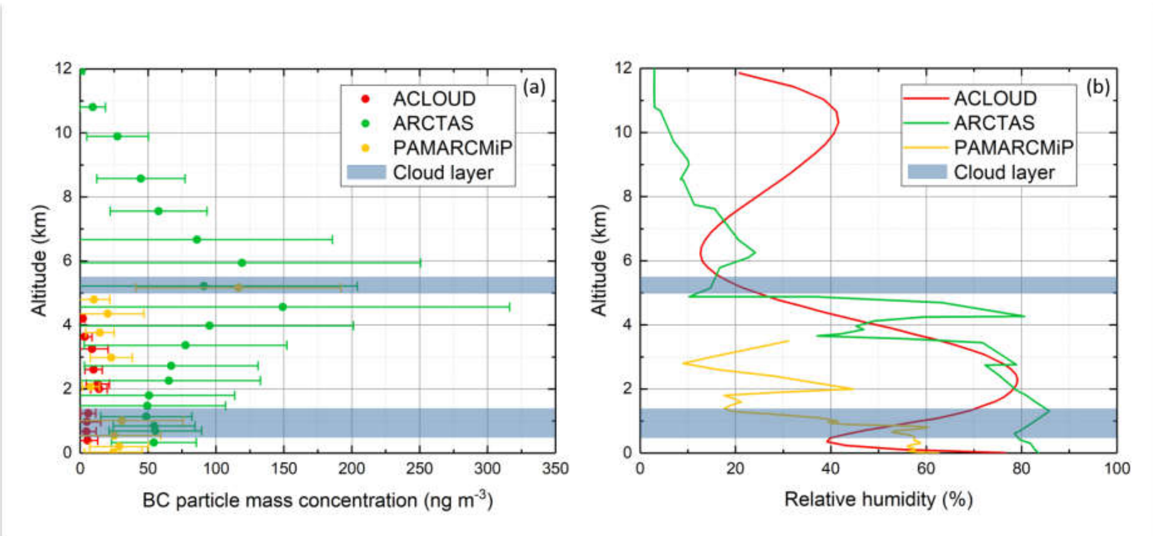

Figure 1. Mean profiles of atmospheric BC particle mass concentration (a) and relative humidity (b) averaged for each the three campaigns (ACLOUD, ARCTAS and PAMARCMiP) as used for the radiative transfer simulations. Horizontal bars indicate the standard deviation. The positions of the two implemented cloud layers (blue shaded area) are marked.

Page 7, line 2: In the snow a two-stream model is used. Presumably more streams were used for the atmospheric radiative transfer. Why is it sufficient to use only two streams in the snow pack?

The number of streams is related to the number of angles where the radiance is calculated. For up- and downward irradiance calculations often two-stream models are applied. In particular, for the radiative transfer simulations snow models apply the two-stream approximation. Dang et al. (2019) compared the DISORT calculation using 16 streams as benchmark with three two-stream models to identify the uncertainty of albedo simulations. Figure 2 from Dang et al. (2019) shows the simulated snow albedo for the tested models, illustrating the sufficient accuracy of the two-stream approximation.

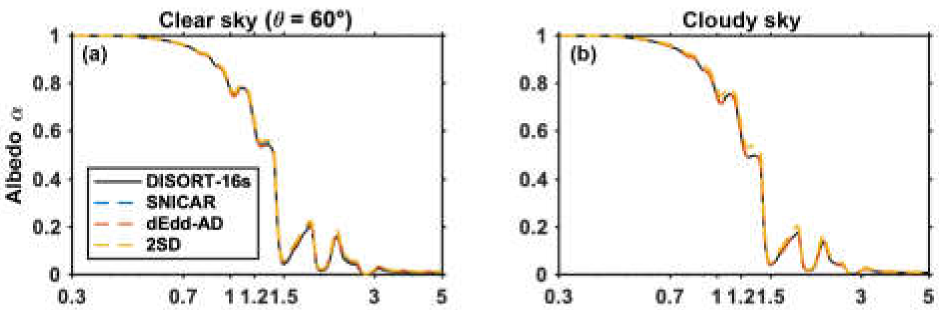

Figure 2 from Dang et al. (2019)

They conclude: “Compared with a 16-stream benchmark model, the errors in snow visible albedo for a direct-incident beam from all three two-stream models are small (<±0.005) and in-crease as snow shallows, especially for aged snow. The errors in direct near-infrared (near-IR) albedo are small (<±0.005) for solar zenith angles θ <75°, and increase as θ increases.”

We are aware that for SZA > 75° the uncertainty by using the two-streams approximation might be higher than 0.005.

Dang, C., Zender, C. S., and Flanner, M. G.: Intercomparison and improvement of two-stream shortwave radiative transfer schemes in Earth system models for a unified treatment of cryospheric surfaces, The Cryosphere, 13, 2325–2343, https://doi.org/10.5194/tc-13-2325-2019, 2019.

line 6: Stamnes et al. (1988) is not a reference for the delta-Eddington approximation. Maybe rather cite Joseph et al. (1976)?

Thanks for identifying this mistake. We changed the reference as suggested:

“To solve the radiative transfer equation, the delta-Eddington approximation (Joseph et al.,1977) is used.”

Page 7, line 14: In the snow the BC optical properties are from Bond et al. (2013) while in the atmosphere they are from Hess et al. (1998). Hence, the BC particles are different in the atmosphere and the snow. What is the rationale behind this choice other than what is available in the models used?

The refractive index of BC can vary a lot and is reported differently in various publications. Bond et al., 2013 writes exemplarily: “A variety of values for the refractive index of BC has been used in global climate models including the OPAC value of 1.74 +- 0.44i [Hess et al., 1998].” In this study we decided to use the data of BC optical properties as proposed by the two separate models for radiative transfer simulations in snow and in atmosphere, respectively.

Page 8, Table 3: Should the first row in the table be named „Thickness" instead of „Depth"?

In literature we found both terms. However, we stick to the term “depth”. It makes sense following the explanation on https://wikidiff.com/depth/thickness: “As nouns the difference between depth and thickness is that depth is the vertical distance below a surface; the degree to which something is deep while thickness is (uncountable) the property of being thick (in dimension).”

Page 8, lines 20-21: The sentence „This procedure is repeated until the deviation between previous (step n) and revised surface albedo decrease below 1 %" is unclear. Please reformulate.

The sentence is revised as follows:

“This procedure was repeated until the deviation of the surface albedo calculated in the previous step (n) and calculated in the revised step (n+1) decreases below 1 %.”

Page 10, Fig. 3: May it be concluded from the plot that the iteration procedure has no impact on the surface albedo in the wavelength region where BC absorbs?

The reviewer is right. The coupling is of minor importance for the surface snow albedo in the wavelength range where BC in snow absorbs. In TARTES, the calculation of the snow albedo requires the direct-to-global ratio as boundary condition. The difference of the calculated snow albedo from one iteration step to the next depends strongly on the change of the direct-to-global ratio. As indicated in Fig. 3, the initial step assumes a ratio of 0, which is more appropriate for the visible spectral range than for the near-infrared. For cloudless conditions the direct-to-global ratio is almost one in the near-infrared. Therefore, the largest effect of coupling is observed in the near-infrared spectral range.

We added:

“The assumption of a pure diffuse illumination in the initial run caused no significant difference of the calculated visible snow albedo to the first and second iteration step. In contrast, the iterated direct-to-global ratio adjusts the snow albedo in the near-infrared, because the direct fraction is quickly approaching unity in this spectral range.”

Page 11, lines 1-2: The upward and downward irradiances were averaged and from these the averaged heating rates were calculated. This appears as a rather unusual and unphysical approach. Would it not be more appropriate to calculate the instantaneous heating rates and then average these?

In this study we were focused on the daily averaged heating rates. The averaging gives exactly what the unit of heating rates, K/day, is expressing. From the mathematical point of view, there is no difference between the temporal averaging of the irradiances and calculating the mean heating rate out of it, or averaging the temporal resolved heating rates over the day.

Since the numerator is a linear term and the arithmetic mean has linear correlations, the results will not change when swapping the order of operation. We try to illustrate that by the following equation:

$$\overline{HR(z)} = \frac{\frac{1}{n}\sum_{i=1}^{n}(F_{net}z_t,i - F_{net}z_b,i)}{\rho(z)c_p(z_t - z_b)} = \frac{\frac{1}{n}\sum_{i=1}^{n}(F_{net}z_t,i) - \frac{1}{n}\sum_{i=1}^{n}(F_{net}z_b,i)}{\rho(z)c_p(z_t - z_b)} = \frac{1}{n}\sum_{i=1}^{n} HR(z),i$$

Page 11, line 15: The enlargement of Fig 4. seems to be missing. As Fig. 4 is, it does not make sense to have many overlapping lines. Please provide a zoom in of the visible wavelength region (lambda < 700 nm).

We are sorry for the confusion. We included the wrong image file in our first version. It is updated as follows:

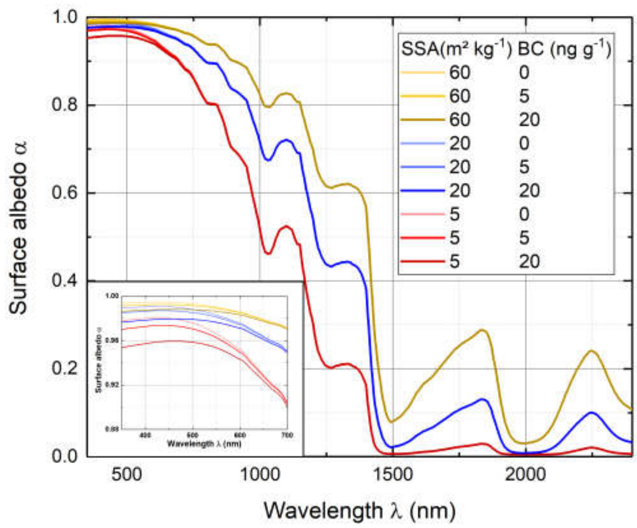

Figure 4. Spectral surface albedo of snow for cloudless conditions and a SZA of 60° for different SSA and BC particle mass concentrations. The inlay shows an enlargement of the spectral albedo between 350 and 700 nm.

Page 11-12, line 31-1: In the introduction it is stated that „the radiative effects of atmospheric BC particles and BC suspended in snow shows an opposite behavior" and „these two effects balance each other". Here it says „the impact of BC particles suspended in the snow pack is assumed to be of minor importance for Arctic conditions". These statements appears to be contradicting each other. Please clarify.

Indeed, our statement was misleading. We adjusted the introduction part and removed the "balance statement" which has not been properly expressed.

"Many regional and global climate models do account for the opposite radiative forcing of atmospheric BC particles and BC particles embedded in snow (Samset et al., 2014). However, estimates of the total net forcing rely on the accuracy of the distribution of the BC particles assumed in the particular model."

Page 12, lines 1-2: The paper by Warren (2013) discussed remote sensing of BC in the snowpack. I can not see how it justifies the claims made here?

We weakened our statement here that BC in snow is of low importance by relating directly to the snow grain size effect. The numbers of albedo reduction due to BC in snow given in Warren (2013) are in good agreement with our calculation. Therefore, we cited the paper here. For clarification we added some more details as follows:

"Therefore, for Arctic conditions, the impact of BC impurities on the broadband snow albedo is of minor importance, compared to the impact of modifying the snow grain size. Also Warren and Wiscombe (1980) and Warren (2013) found only a small reduction of the broadband albedo between 0 - 1 % for fresh snow and 0 – 3 % for aged snow when adding BC with a mass concentration of 34 ng g^{-1} to the clean snow."

Page 14, line 2: Is the factor of about 3 mostly due to differences in solar zenith angle?

Yes, the difference between the three cases is the diurnal pattern of available radiation.

We stated in the original manuscript: "This difference is caused by the lower maximum Sun elevation during PAMARCMiP (location in higher latitude) resulting in a lower amount of available incoming solar irradiance compared to ACLOUD and ARCTAS (see range of SZA in Tab. 2)."

Page 16, line 32: Sentence starting with „Absorption in the ..." is unclear. Please reformulate.

We rephrased the sentence:

"The absorption in the ice cloud is less pronounced, and the increase of $HR_{tot}(z)$ is significantly lower."

Pages 19-21: In the conclusions please discuss how the results from this study compare with previous studies mentioned in the introduction.

We compared the derived atmospheric heating rates due to BC already with findings from other regions to relate the numbers to more polluted conditions:

"For example, studies investigating strong pollution conditions in northern India or China reported on BC heating rates in the atmosphere larger than 2 K day^{-1}, which may significantly influence the lapse rate and the atmospheric stability (Tripathi et al., 2007; Wendisch et al., 2008). For the rather pristine Arctic, this study showed significantly lower daily mean BC heating rates of maximum 0.1 K day^{-1}, which have not the potential to significantly modify the atmospheric stability."

Further we added a comparison of BC radiative effects with Wendling et al. (1985) for atmospheric BC and Dou and Xiao, 2016 for BC embedded in snow:

"The magnitude of the atmospheric BC radiative forcing at the surface derived in this study (up to -0.2 W m^{-2}) agrees quite well with findings from Wendling et al. (1985). They reported a BC induced solar cooling in the range of 0.0 to -0.5 W m^{-2} for spring measurements in the Svalbard area. Further, the solar surface radiative effect due to BC embedded in snow has shown solar warming between 0.05 and 0.7 W m^{-2} depending on the BC mass concentration and incident solar irradiance. For comparison, Dou and Cun-De (2016) deduced an averaged solar warming over Svalbard in spring of 0.54 W m^{-2} based on a BC mass concentration of 5 ng g^{-1} in snow."

Language corrections

Page 2, line 7: change 'of suspended' to 'suspended'.

Changed.

Page 2, line 34: change 'will warming' to 'will warm'.

Changed.

Page 3, line 16: remove '.' after 'quantified.'.

Changed as suggested.

Page 9, line 1: Should it be „converge" instead of „conversion"?

We changed it to: *“This quick convergence of …”*

Page 11, line 6: Remove “(SSA respectively)".

Changed.

Combining atmospheric and snow ~~layer~~ radiative transfer models to assess the solar radiative effects of black carbon in the Arctic

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Abstract. ~~Solar~~ The magnitude of solar radiative effects (cooling or warming) of black carbon (BC) particles ~~suspended~~ embedded in the Arctic atmosphere and surface snow layer were explored ~~by~~ on the basis of case studies. For this purpose, combined atmospheric and snow radiative transfer simulations were performed for cloudless and cloudy conditions on the basis of BC mass concentrations measured in pristine early summer and more polluted early spring conditions~~under cloudless and cloudy conditions~~. The area of interest is the remote sea ice covered Arctic Ocean in the vicinity of Spitsbergen, northern Greenland and northern Alaska typically not affected by local pollution. To account for the radiative interactions between the black carbon containing snow surface layer and the atmosphere, ~~a snow layer and an atmospheric~~ an atmospheric and snow radiative transfer model were coupled iteratively. For pristine summer conditions (no atmospheric BC, minimum solar zenith angles of 55°) and a representative BC particle mass concentration of 5 ng g^{-1} in the surface snow layer, a positive ~~solar radiative effect~~ daily mean solar radiative forcing of +0.2 W m^{-2} was calculated for the surface radiative budget. ~~Contrarily, a~~ A higher load of atmospheric BC representing early springtime conditions, results in a slightly negative ~~radiative effect~~ mean radiative forcing at the surface of about -0.05 W m^{-2}, even when the ~~same~~ low BC mass concentration ~~is suspended~~ measured in the pristine early summer conditions was embedded in the surface snow layer. ~~This counteracting of atmospheric BC and BC suspended~~ The total net surface radiative forcing combining the effects of BC embedded in the atmosphere and in the snow layer strongly depends on the snow optical properties ~~determined by the~~ (snow specific surface area ~~. However~~and snow density). For the conditions over the Arctic Ocean analyzed in the simulations, it was found, that the atmospheric heating rate by water vapor or clouds is one to two orders of magnitude larger than that by atmospheric BC. Similarly, the daily mean total heating rate (6 K day^{-1}) within a snow pack due to absorption by the ice~~water, was found to be~~, was more than one order of magnitude larger than ~~the heating rate of suspended~~ that of atmospheric BC (0.2 K day^{-1}). ~~The role of clouds in the estimation of the combined direct radiative BC effect (BC in snow and in atmosphere) was analyzed for the pristine early summer and the polluted early spring BC conditions. Both, the cooling effect~~ Also it was shown that the cooling by atmospheric BC of the near-surface air, as well as the warming effect by BC ~~suspended~~ embedded in snow are reduced in the presence of clouds.

1 Introduction

Black carbon (BC) aerosol particles, which mostly originate from incomplete combustion of organic material ~~. They strongly absorbs and scatters~~ (Bond et al., 2013; Petzold et al., 2013), absorb and scatter solar radiation in the visible wavelength range and, therefore, influence the ~~Arctic~~ atmospheric solar radiative energy budget. The manifold sources of BC particles and their atmospheric transport paths ~~are well known~~ have been studied extensively (Law et al., 2014). However, the source strengths of the emissions are hard to quantify, which makes it challenging to ~~reproduce~~ quantify the transport of BC particles into the Arctic by simulations (Stohl et al., 2013; Arnold et al., 2016; Schacht et al., 2019). Major sources of BC particles are forest fires, ~~industry, and traffic predominately located~~ industrial activities, and traffic-related emissions, which are main factors in lower latitudes; northern parts of Europe~~and Americaas well as Siberia. Long-range transport in higher altitudes brings these emitted BC particles~~ , America, and Siberia. The BC particles emitted at the surface of the mid-latitudes are lifted and transported into the Arctic, where they can stay for several days and longer (Liu et al., 2011). Contrarily, particles ~~locally produced~~ produced locally in the Arctiv through ship traffic emissions~~and~~ , flaring from the oil industry or other ground-based activities, settle down quickly on the surface and may alter the radiation budget within the snow pack (Bond et al., 2013). Nowadays, local sources are only a minor component. ~~Nevertheless~~In future, a strong intensification of the ship traffic ~~is expected in the future~~ in the Arctic Ocean and further polluting human activities are expected (Corbett et al., 2010). Still, the direct radiative impact by these future additional BC particle emmissions is assumed to be of minor importance (Gilgen et al., 2018).

The BC ~~particle mass concentration~~ magnitude of the atmospheric particle mass concentrations (in units of $\mathrm{ng\,m^{-3}}$) ~~of suspended in the atmosphere is highly variable depending~~ depends on the season and general meteorological conditions. In ~~the~~ case of BC particle plumes reaching the Arctic by long-range transport, atmospheric concentrations of up to $150\,\mathrm{ng\,m^{-3}}$ ~~can be expected~~ were observed (Schulz et al., 2019). Sharma et al. (2013) compared atmospheric BC particle mass concentrations measured during different Arctic campaigns. They identified large differences depending on region and season. Measurements in spring 2008 covering Alaska and northern Canada, showed values above $200\,\mathrm{ng\,m^{-3}}$ in higher altitudes, while in spring 2009 more pristine air masses were encountered ~~. In this period, the Arctic-wide airborne measurements indicated~~ showing BC particle mass concentrations of less than $100\,\mathrm{ng\,m^{-3}}$ ~~in~~ integrated over the entire vertical column.

To quantify the ~~the~~ amount of BC particles in a snow pack volume, the BC mass ~~fraction (in units of ng g $^{-1}$~~concentration (ng of BC in 1 g of snow) is used commonly. Typical values observed in Greenland range between 1 and $10\,\mathrm{ng\,g^{-1}}$, in the Canadian Arctic between 5 and $20\,\mathrm{ng\,g^{-1}}$, and in the northern parts of Russia values may reach $100\,\mathrm{ng\,g^{-1}}$. Table 1 summarizes observational data of ~~typical BC mass fractions~~ measured BC mass concentrations in snow for different Arctic regions, as reported by Doherty et al. (2010), Forsström et al. (2013), and Pedersen et al. (2015). The numbers given in Table 1 were derived from different measurement methods. More precisely, thermal-optical techniques were applied in Forsström et al. (2013) and Pedersen et al. (2015) provide the elemental carbon (EC) mass concentration, while filter transmission methods result in BC concentrations (Doherty et al., 2010). As a consequence of the different measurement methods, the ratio of the BC to EC concentration in snow can reach values of 1.3 as reported by Dou et al. (2017). A full discussion of the EC/BC terminology can be found in Petzold et al. (2013).

Due to ~~their absorbing effect~~the absorption of solar radiation, BC particles may contribute to the currently ongoing drastic Arctic climate changes ~~, namely the Arctic Amplification . They can directly~~ (called arctic amplification, e.g., Wendisch et al., 2017). The absorption effect can add to the warming of the atmosphere ~~when suspended~~ or the snow pack, when the BC particles are suspended either in the air or ~~to the~~ embedded in the snow. Furthermore, the BC particles may lead to a reduction of the snow surface albedo if the BC ~~is sedimented on~~ sediments on or into the snow pack ~~associated with a higher amount of absorbed radiation within the snow layer~~ (Sand et al., 2013). ~~Exemplarily,~~ Warren (2013) estimated a decrease of 2 % in snow albedo in the visible spectral range for a snow pack with a BC mass ~~fraction~~ concentration of 34 ng g^{-1}, which corresponds to the maximum value observed on the Greenland ice sheet (Doherty et al., 2010). More typical BC mass ~~fractions~~ concentrations in Arctic snow range between 5 and 20 ng g^{-1} (Tab. 1), which would lead to a reduction of the snow surface albedo of around 1 %. For typical Arctic summer conditions with a downward irradiance of 400 W m^{-2} at the surface, a snow surface albedo reduction by one percent would ~~lead to~~ cause an additional absorption of solar radiative energy of 4 W m^{-2} (Flanner et al., 2007). ~~The additional~~ As a further consequence, the absorption by BC particles supports the melting of snow and increases the snow grain size due to an enhanced snow metamorphism, ~~which may lead to a~~ leading to further reduction of the surface albedo~~and, as a consequence, even more incoming solar radiation being absorbed. This positive feedback represents a self-amplifying process due to .~~ . The increase of the snow grain size also feeds back to the absorption by BC particles~~in snow . So far the relevance of this feedback was not quantified~~, which is more efficient for larger snow grain sizes (Warren and Wiscombe, 1980).

BC particles suspended in the atmosphere, ~~are known to~~ influence the absorption and scattering of the incoming solar radiation. If atmospheric BC particles are located in high altitudes, enhanced backscattering and absorption of incoming solar radiation by the BC layer leads to a reduction of the solar radiation reaching the surface. At the same time, the absorbed radiation ~~will warming~~ warms the atmospheric BC layer. In extreme cases, the ~~presence of atmospheric BC can effect the atmospheric stability~~ absorption due to atmospheric BC particles can affect the thermodynamic stability of the BC containing atmospheric layer (Wendisch et al., 2008). The radiative heating of the lofted BC layers and the local cooling of the surface may enhance the ~~already strongly stratified~~ thermodynamic stability of the Arctic boundary layer over the snow and ice-covered areas ~~, such that the atmospheric stability increases~~ (Flanner, 2013).

~~In general, the~~ Several regional and global climate models account for the opposite radiative effects of atmospheric BC particles and ~~BC suspended in snow shows an opposite behavior . Model estimates of how these two effects balance each other,~~ snow-embedded BC particles (Samset et al., 2014). However, estimates of the total net forcing rely on the accuracy of the ~~assumed~~ distribution of the BC particles ~~,~~ assumed in the particular model. Samset et al. (2014) compared 13 aerosol models from the AeroCom Phase II; all of them included BC~~as an aerosol species~~. They found that modeled atmospheric BC particle mass concentrations often show a spread over more than one order of magnitude. In remote regions, dominated by long range transport, these models tend to overestimate the atmospheric BC concentrations compared to airborne observations. On the other hand, an underestimation of deposition rates induces a lower BC mass ~~fraction~~ concentration in snow (Namazi et al., 2015). ~~However~~While this may introduce significant local and temporal uncertainties of the BC concentration and related radiative effects, long-term trends and mean multi-model results ~~were~~ are representative for Arctic-wide observations (Sand

Table 1. ~~Typical values~~ Values of the ~~BC particle~~ black carbon mass ~~fraction~~ concentration in snow pack observed in different regions and seasons in the Arctic. Note, that Pedersen et al. (2015) and Forsström et al. (2013) derived the mass ~~fraction~~ concentration of elemental carbon applying a thermal-optical measurement method.

Location	Season	BC mass ~~fraction~~ concentration (ng g^{-1})	Method	Source
Svalbard region	March/April	13	filter transmission	Doherty et al. (2010)
Arctic Ocean snow	Spring	7	filter transmission	Doherty et al. (2010)
Arctic Ocean snow	Summer	8	filter transmission	Doherty et al. (2010)
Northern Norway	May	21	filter transmission	Doherty et al. (2010)
Central Greenland	Summer	3	filter transmission	Doherty et al. (2010)
Svalbard region	March/April	11 - 14	thermal-optical	Forsström et al. (2013)
Corbel, Ny-Ålesund	March	21	thermal-optical	Pedersen et al. (2015)
Barrow	April	5	thermal-optical	Pedersen et al. (2015)
Ramfjorden, Tromsø	April	13	thermal-optical	Pedersen et al. (2015)
Valhall, Tromsø	April	137	thermal-optical	Pedersen et al. (2015)
Fram Strait	April	22	thermal-optical	Pedersen et al. (2015)

et al., 2017).

Most previous studies quantifying the radiative impact of BC particles ~~either focused~~ focused either on estimates of cooling/heating effects in the atmosphere (e.g., Wendling et al., 1985; Samset et al., 2013), or on radiative effects of BC in the snow surface layer (Dou and Cun-De, 2016). In contrast, this paper ~~will combine~~ combines both effects by iteratively coupling radiative transfer simulations in both compartments, the atmosphere and the snow pack. ~~For typical Arctic BC distributions and~~ On the basis of measured Arctic BC particle mass concentrations for spring and summer months, the ~~local direct radiative effects~~ instantaneous radiative forcing of BC particles ~~suspended~~ embedded in the snow surface layer and in the atmosphere ~~are quantified . with this approach, the interactions between the BC~~ were quantified for specific cases. Here, the instantaneous radiative forcing refers to the change of the surface radiation budget caused by the presence of BC particles. With help of the coupled model, the interaction of radiative effects in the atmosphere and the snow pack ~~will be~~ was considered. In particular, the role of clouds on the cooling/heating effect caused by BC particles ~~will be~~ was examined. Due to the fact that clouds enhance the atmospheric multi-scattering between surface and cloud layer, but also enhance the surface albedo (Choudhury and Chang, 1981), it is expected that clouds alter also the radiative impact by BC particles. To our knowledge, this interaction was not explicitly discussed in previous publications.

The radiative transfer simulations used in this study ~~are~~ were based on airborne observations of atmospheric BC concentration in the Arctic, which were taken during three field campaigns in the European and Canadian Arctic. The applied models and observations are introduced in Section 2. Section 3 discusses the radiative ~~effects~~ forcing of BC particles on the surface solar radiative budget. Vertical profiles of heating rates ~~induced by the atmospheric BC particles and BC particles~~ in the atmospheric and in the snow pack are presented for clean and polluted conditions. To estimate the ~~relevance~~ impact of BC particles, effective heating rates are calculated by separating the ~~pure BC~~ BC radiative effect from the total heating rates.

2 ~~Setup~~ Configuration of radiative transfer simulations and iterative model coupling

2.1 ~~Aircraft campaigns and~~ BC ~~data~~profiles from aircraft campaigns

The ~~atmospheric model setup~~ input for the radiative transfer simulations was adapted to ~~campaign specific conditions.~~ campaign-specific conditions. The atmospheric BC particle mass concentrations were derived from airborne measurements with a Single Particle Soot Photometer (SP2, Moteki and Kondo, 2007). Measured profiles of the atmospheric BC ~~taken~~ were used from three aircraft campaigns ~~were taken into account, which represent~~ representing typical cases with higher BC concentrations (polluted case) in early spring with low sun, and lower BC concentration (pristine conditions) in early summer during the polar day. The ~~atmospheric BC particle mass concentrations were derived from airborne measurements with a Single Particle Soot Photometer. The~~ Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) spring campaign was performed in April 2008 (Jacob et al., 2010; Matsui et al., 2011). The aircraft operation of ARCTAS mainly took place in northern Alaska and the Arctic Ocean. Similar SP2 measurements were performed during the Polar Airborne Measurements and Arctic Regional Climate Model Simulation Project (PAMARCMiP) campaigns ~~which is a series of aircraft observations performed within the Arctic region~~ (Herber et al., 2012; Stone et al., 2010). ~~Here data from~~ In this paper, measurements from the PAMARCMiP 2018 ~~are analysed which was based at the Villum Research Station (Station Nord/Greenland) and conducted flights~~ observations conducted from 10 March to 8 April 2018 ~~in the European Arctic~~ were analyzed. The research flights, starting from Station Nord/Greenland, were performed above the sea ice in the Arctic ocean north of Station Nord and the Fram Strait. In contrast to both spring campaigns, the Arctic CLoud Observations Using airborne measurements during polar Day (ACLOUD) campaign was conducted in early summer 2017 characterizing the atmosphere over the Arctic Ocean north and west of Svalbard (Wendisch et al., 2019; Ehrlich et al., 2019). ACLOUD was coordinated with the Physical Feedbacks of Arctic Boundary Layer, Sea Ice, Cloud and Aerosol (PASCAL) cruise of the research vessel Polarstern which ~~provides~~ provided a ground-based characterization of snow properties (Wendisch et al., 2019).

Mean vertical profiles of the measured atmospheric BC particle mass concentrations averaged for each of the three campaign (ACLOUD, ARCTAS and PAMARCMiP), are shown in Figure 1a. The conditions between the individual flights were highly variable (see the standard deviation of each layer in Fig. 1). ACLOUD shows rather low mean BC concentrations, which do not exceed $30\,\mathrm{ng\,m^{-3}}$. During PAMARCMiP, the background concentrations were similarly low, with the exception of measurements in about 5 km altitude, where more than $100\,\mathrm{ng\,m^{-3}}$ were recorded. For ARCTAS observations, conducted at lower latitudes, significantly higher BC concentrations of up to $150\,\mathrm{ng\,m^{-3}}$ were observed. Similar to PAMARCMiP, the maximum concentrations were observed at about 5 km altitude indicating that the BC particles were linked to long-range transport. Besides the differences in atmospheric BC concentrations, ~~also~~ the range of the daily solar zenith angle (SZA) and, ~~therefore~~thus, the available incoming solar radiation, varied significantly for the three campaign periods. When analysing the radiative impact of BC on basis of daily averages, the magnitude of the solar incident solar ~~incident~~ radiation and the length of the day play a major role. While the early summer conditions of ACLOUD ~~are~~ were characterized by the polar day and SZA between 55° and 78°, during ARCTAS the available incoming solar radiation was lower due to ~~a nighttime of about 8.5 hours and a minimum SZA~~ lower values of the SZA (minimum at noon of 62.5°). PAMARCMiP was conducted in the most northern region and earlier in the year, such that the Sun was about 9.5 hours below the horizon and the minimum SZA was 79° at noon. Table 2 summarizes the key characteristics of the three ~~analysed~~ analyzed data sets.

Table 2. Region, period, solar zenith angle range, and maximum BC particle mass concentration and mean optical depth of BC at 500 nm wavelength characterizing the three data sets obtained within ARCTAS, ACLOUD, and PAMARCMiP.

	ARCTAS	ACLOUD	PAMARCMiP
Region	Alaska/ Northern Canada	Svalbard/Arctic Ocean	Northern Greenland/ Arctic Ocean
Latitude (°)	71	78	82
Period	April 2008	May/June 2017	March/April 2018
SZA (°)	63–90	55–78	79–90
Night length (h)	8.6	0.0	9.4
Max. BC concentration (ng m^{-3})	149	13	117
BC optical depth at 500 nm	0.008	0.0003	0.006
~~Reference~~ Data reference	Jacob et al. (2010)	Ehrlich et al. (2019)	Herber (2019)

~~Campaign specific BC particle mass concentration profiles were calculated by averaging over all available aircraft measurements. Figure 1 shows the different BC profiles composed in discrete layers as implemented into the radiative transfer simulations. The ACLOUD case represents nearly pristine BC conditions with maximum values of up to 12 ng m^{-3}, while the ARCTAS and PAMARCMiP profiles show 10-20 times larger BC concentration. Due to the different flight performances of the aircrafts (ARCTAS used a DC-8 while ACLOUD/PAMARCMiP operated the Polar 5 and 6, two modified DC-3 aircraft of the type Basler BT-67), the profiles measured in the European Arctic are restricted to 5.5 km altitude . However, on the Polar 5 aircraft Sun photometer are available. The observations during ACLOUD indicate, that the extinction above the maximum flight altitude of 5.5 km does was negligible. During PAMARCMiP, additionally the upward-looking Airborne Mobile Aerosol Lidar for Arctic research (AMALi) was installed on the aircraft . Based on the backscattering signal potential aerosol layer above the maximum flight level were observed only in one of the 14 flights.~~Therefore, the constructed profiles of PAMARCMiP and ACLOUD are assumed to be representative for Arctic ~~spring and~~ early spring and early summer conditions, respectively.

2.2 Atmospheric radiative transfer model

To simulate vertical profiles of the spectral upward and downward irradiance, the library for radiative transfer routines and programs (libRadtran, Emde et al., 2016; Mayer and Kylling, 2005) was used (*http://www.libradtran.org/doku.php*). The model also provides the ratio of the direct-to-global irradiance $f_{\mathrm{dir/glo}}$, which is required as a boundary condition of the snow pack radiative transfer model. As a solver for the radiative transfer equation, the Discrete Ordinate Radiative Transfer solver (DIS-

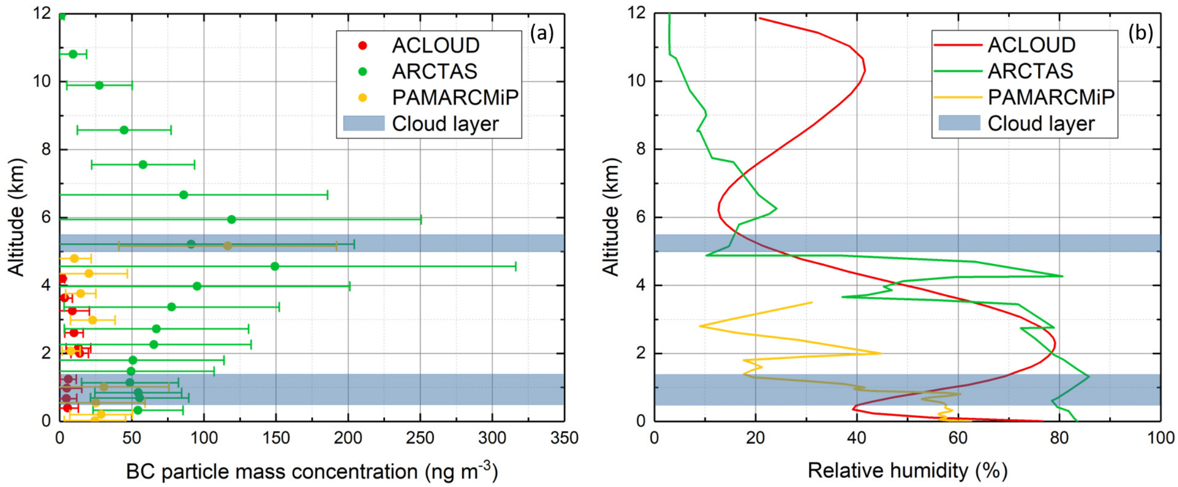

Figure 1. ~~Campaign specific mean BC~~ Mean profiles of atmospheric BC particle mass concentration (a) and relative humidity (b) averaged for each of the three campaigns (ACLOUD, ARCTAS and PAMARCMiP) as used for the radiative transfer simulations. ~~Additional~~ Horizontal bars indicate the standard deviation. The positions of the two ~~assumed~~ implemented cloud layers (blue shaded area) are marked.

ORT) 2 ~~routine was chosen from the libRadtran package~~(Stamnes et al., 2000) routine running with 16 streams was chosen.

For the calculations, a plane-parallel atmosphere was assumed, which is justified for the Arctic conditions during the three campaigns. Using a pseudo-spherical geometry in libRadtran would change the broadband downward irradiance by less than 0.1 % (0.7 %) for a calculation with a SZA of 60° (75°). The vertical resolution of the simulated irradiances was adjusted to the measured BC profiles, ranging between 100 m and 1 km. The spectral resolution of the simulations was set to 1 nm covering a wavelength range between 350 nm and 2400 nm. The extraterrestrial spectrum was taken from ~~the solar spectrum from~~ Gueymard (2004). The BC optical properties including the refractive index, density, extinction coefficient, single scattering albedo, and scattering phase function from the OPAC aerosol database were applied (Hess et al., 1998). Corresponding to the campaign average BC profiles, the range of the SZA values was set to values representing the campaign conditions (see Table 2).

The meteorological input for the model ~~is~~ was based on standard profiles of trace gases, temperature, humidity, and pressure from Anderson et al. (1986). Sub-Arctic summer conditions were chosen for the ~~summer case~~ early summer case (ACLOUD) and subarctic winter conditions for the winter and ~~early spring cases~~ spring cases (ARCTAS, PAMARCMiP). The standard profiles were ~~modified by~~ adapted to observations from radio soundings near the ~~research area~~ airborne observations or dropsondes released during the flights ~~. The range of the SZA and the atmospheric profiles were adjusted to~~ and represent the middle of the individual campaign periods.

~~To account for the impact of the campaign specific atmospheric BC profiles (~~Fig. 1~~), the BC optical properties including the refractive index, density, extinction coefficient, single scattering albedo, and scattering phase function from the OPAC aerosol database were applied .~~b shows the profiles of relative humidity, used for the simulations. PAMARCMiP was characterized by rather dry air. Only in the boundary layer, an average humidity up to 60 % was observed often linked to boundary layer clouds. ACLOUD and ARCTAS showed a higher relative humidity in higher altitude of up to 6 km, which indicates the influence of higher level clouds.

To test the sensitivity of the BC radiative effects ~~on~~ with respect to cloud occurrence, two cloud layers were ~~implemented~~ synthetically included in the atmospheric profiles as illustrated in Fig. 1. The cloud ~~layers were constructed~~ layer properties were based on observations by Bierwirth et al. (2013), Leaitch et al. (2016), and Blanchard et al. (2017) to represent typical Arctic cloud conditions. A low-level liquid water cloud was placed between 500 m and 1.4 km ~~according to observations of Arctic clouds presented by and~~ representing the humid boundary layer observed during PAMARCMiP. The liquid water content increases from 0.1 $\mathrm{g\,m^{-3}}$ at cloud base to 0.3 $\mathrm{g\,m^{-3}}$ at cloud top, the cloud particle effective radius increased from 6 μm to 12 μm. The second cloud layer represents a thin ice water cloud and was positioned between 5 and 5.5 km representing the higher level clouds observed during ACLOUD and ARCTAS. This thin cloud was assumed to be homogeneous with an ice water content of 0.006 $\mathrm{g\,m^{-3}}$ and an effective cloud particle radius of 40 μm, according to airborne measurements reported by Wyser (1998) and Luebke et al. (2013). Optical properties of the liquid cloud droplets were calculated from Mie-Theory, while the ice crystal optical properties are based on (Fu, 2007). The assumed cloud properties correspond to a cloud optical thickness of 15 for the water cloud and 0.2 for the thin ice cloud.

2.3 Snow pack radiative transfer model

The Two-streAm Radiative TransfEr in Snow model (TARTES, https://github.com/ghislainp/tartes) was used to simulate the radiative transfer ~~in~~ through the snow pack (Libois et al., 2013, 2014). In TARTES, the snow profile ~~can be~~ is constructed of a predefined number of horizontally homogeneous snow layers, which allows to account for the stratifiction of the snow pack. To ~~account for the single scattering~~ consider the single-scattering properties of each layer, ~~TARTES applies~~ the method described by Kokhanovsky and Zege (2004) is applied in TARTES. To solve the radiative transfer equation, the delta-Eddington approximation ~~is applied. The model~~ (Joseph et al., 1977) is used. As a result, TARTES computes the spectral surface albedo and the profile of the irradiance within the snow pack. As boundary condition, the SZA and $f_{\mathrm{dir/glo}}$ have to be ~~defined~~predefined. For each of the snow layers, the optical and microphysical properties have to be given, such as the snow density (ρ_{ice}), the specific surface area ($\cancel{SSA}$SSA), and the snow grain shape ~~parameter~~parameters, which represents a mixture of different grains as suggested by Libois et al. (2013)~~, were set~~. Furthermore, the specific values of the so-called absorption enhancement parameter $B = 1.6$ and the geometric asymmetry factor $g^G = 0.85$ were applied. The specific surface area can be translated into the optical snow grain size r_{opt} by:

$$r_{\mathrm{opt}} = \cancel{\frac{3}{\rho_{\mathrm{ice}} \cdot SSA}} \frac{3}{\rho_{\mathrm{ice}} \cdot \mathrm{SSA}}, \tag{1}$$

TARTES allows to ~~add impurities ot~~ consider impurities to each snow layer, which are characterized by the impurity type and mass ~~fraction. Note, that the impurities considered in TARTES are assumed to be externally mixed~~ concentration. The impurities are externally mixed and assumed to interact by Rayleigh scattering. To simulate a BC-containing snow layer, the complex refractive index and the density of BC particles given by Bond et al. (2013) ~~were applied.~~ are applied.

The input parameters of the snow pack model are summarized in Table 3. For the bottom layer, a soil albedo of 0.3 was assumed representing the ~~conditions~~ reflection properties below the snow pack. The impact of the soil albedo on the albedo of the snow surface depends on the depth of the overlying snow pack. Sensitivity studies have shown, that for snow ~~depth~~ depths of more than 20 cm the albedo of a snow surface is independent of the choice of the soil albedo below. In this study the snow pack depth was set to 1 m thickness. Reference simulations ~~of a pristine~~ assuming a pristine homogeneous snow layer were performed~~by assuming only one single homogeneous snow layer~~. Simulations including BC impurities were based on BC particle mass ~~fractions measured by several studies reported in literature as~~ concentrations summarized in Table 1 (Doherty et al., 2010; Forsström et al., 2013; Pedersen et al., 2015) and observations during PASCAL and PAMARCMiP. For the simulations of a single homogeneous snow layer, typical BC particle mass ~~fractions~~ concentrations of 5 ng g^{-1} and 20 ng g^{-1} were chosen. The default values of snow density and ~~SSA~~ SSA were based on measurements during PASCAL and PAMARCMiP and were set to 300 kg m^{-3} and 20 m^2 kg^{-1}, respectively. To analyze the sensitivity of the snow surface albedo with respect to the snow grain size, ~~SSA~~ SSA values of 5 m^2 kg^{-1} and 60 m^2 kg^{-1} were used. The vertical model resolution was set to 1 cm.

In addition to the simulations of a homogeneously mixed snow layer, a second model setup used ~~in Section 3.2.2 considers~~ to consider a multi-layer snow pack. ~~Based on snow pit~~ Pit measurements in Greenland ~~,~~ (Doherty et al., 2010) identified typical multi-layer structures, where BC accumulated in a melting layer approximately 10 cm below the surface. Referring to these measurements, the snow pack of the second model setup consists of three snow layers. The top layer is 5 cm and the BC-containing middle layer is 10 cm thick. The bottom layer below continues to 1 m depth. For this multi-layer approach, BC was ~~mainly~~ included in the middle layer, representing an aged melting layer in which, impurities ~~have accumulated ($SSA = 20$~~had accumulated (SSA = 20 m^2 kg^{-1}, snow density of 350 kg m^{-3}, and a BC mass ~~fraction~~ concentration of 15 ng g^{-1}). The top layer ~~is~~ was assumed to be of fresh and clean snow with ~~$SSA = 40$~~SSA = 40 m^2 kg^{-1}, a snow density of 250 kg m^{-3}, and a BC mass ~~fraction~~ concentration of 2 ng g^{-1}) representing measurements from the PASCAL campaign. The aged snow layer at bottom was characterized by an enhanced snow grain size and density of ~~$SSA = 10$~~SSA = 10 m^2 kg^{-1} and $\rho_{\mathrm{ice}} = 450\,\mathrm{kg\,m^{-3}}$, respectively, and a BC mass ~~fraction~~ concentration of 2 ng g^{-1}.

2.4 Iterative coupling

The surface albedo is an important boundary condition to simulate the radiative transfer in the atmosphere. ~~At the same time, the spectral surface albedo~~ It depends on the illumination conditions defined by the solar zenith angle, the spectral distribution of downward irradiance, and the ratio of direct-to-global irradiance ~~. Especially a~~ (e.g., Wiscombe and Warren, 1980; Gardner and Sharp, 2010; Stapf et al., 2019). The transition from cloudy to cloudless ~~conditions significantly~~ atmospheric conditions increases the direct-to-global ratio ($f_{\mathrm{dir/glo}}$) and ~~shifts the downward irradiance to shorter wavelengths~~ the contribution of

Table 3. Snow pack model setups for the single layer and multi-layer cases. The default ~~*SSA*~~ SSA for the single layer case is 20 $\mathrm{m^2\,kg^{-1}}$.

	Single layer	Multi-layer		
		top layer	middle layer	bottom layer
Depth (cm)	100	5	10	85
BC mass ~~fraction~~ concentration ($\mathrm{ng\,g^{-1}}$)	5 / 20	2	15	2
~~*SSA*~~ SSA ($\mathrm{m^2\,kg^{-1}}$)	5 / 20 / 60	40	20	10
Density ($\mathrm{kg\,m^{-3}}$)	300	250	350	450

short wavelengths to the broadband downward irradiance (Warren, 1982). Therefore, a cloud cover typically increases the broadband surface albedo. ~~Exemplarily, comparing~~ For example, simulations with TARTES assuming cloudless and cloudy conditions ~~with TARTES lead to a change of~~ changed the broadband snow surface albedo from about 0.8 to 0.9 for a SZA of ~~60°~~ 60° and a snow pack (no impurities) characterized by SSA= 20 $\mathrm{m^2\,kg^{-1}}$. As clouds ~~mostly~~ absorb solar radiation mostly at wavelengths larger than 1000 nm, the shorter wavelengths, where BC particles strongly absorb solar radiation, become more relevant. ~~However, it is not sufficient to consider only the two cases of pure cloudless and cloudy conditions. For optically thin clouds or the presence of atmospheric pollution layers, $f_{\mathrm{dir/glo}}$ can quickly change with slight changes of the cloud and atmosphere properties (e.g., liquid water path or aerosol optical depth). To account for the variability of these effects~~Because of the significant surface-cloud interactions, the atmospheric and ~~the~~ snow pack radiative transfer models need to be coupled, interactively. Therefore, an iterative method coupling libRadtran and TARTES via their boundary conditions, surface albedo and direct-to-global ratio ($f_{\mathrm{dir/glo}}$) of the incident radiation, was applied. Both parameters were transferred between the models as schematically illustrated in Fig. 2.

In the first iteration step, only diffuse radiation was assumed ($f_{\mathrm{dir/glo}}$ = 0) to calculate the snow surface albedo by TARTES, which subsequently serves as input for the libRadtran simulations. Then a new spectral direct-to-global ratio representing the atmospheric conditions was calculated by libRadtran, which is in turn used to re-adjust TARTES, starting a revised iteration (n+1) to calculate a new spectral surface albedo α_λ(n+1). This procedure ~~is~~ was repeated until the deviation ~~between previous (step~~ of the surface albedo calculated in the previous step (n) and ~~revised surface albedo decrease~~ calculated in the revised step (n+1) decreases below 1 %. Exemplarily, Figure 3 illustrates the change of the spectral surface albedo for a cloudless case without atmospheric BC and a SZA of 60°. The BC mass ~~fraction~~ concentration in snow was set to 5 $\mathrm{ng\,g^{-1}}$. Two iteration steps were necessary in this particular example to match the ~~termination criteria, which was~~ 1% termination criterion, which is a typical number for all studied cases. Starting with ~~pure~~ purely diffuse conditions allows faster calculations in ~~particular for~~ cloudy cases. This quick ~~conversion~~ convergence of the iteration ~~allows~~ enables considering different cloud properties and atmospheric conditions and facilitates to calculate the radiative effects of BC particles in the atmosphere and within the snow pack simultaneously. The assumption of a pure diffuse illumination in the initial run caused no significant difference of the calculated visible snow albedo to the first and second iteration step. In contrast, the iterated direct-to-global ratio adjusts the snow albedo in the near-infrared, because the direct fraction is quickly approaching unity in this spectral range.

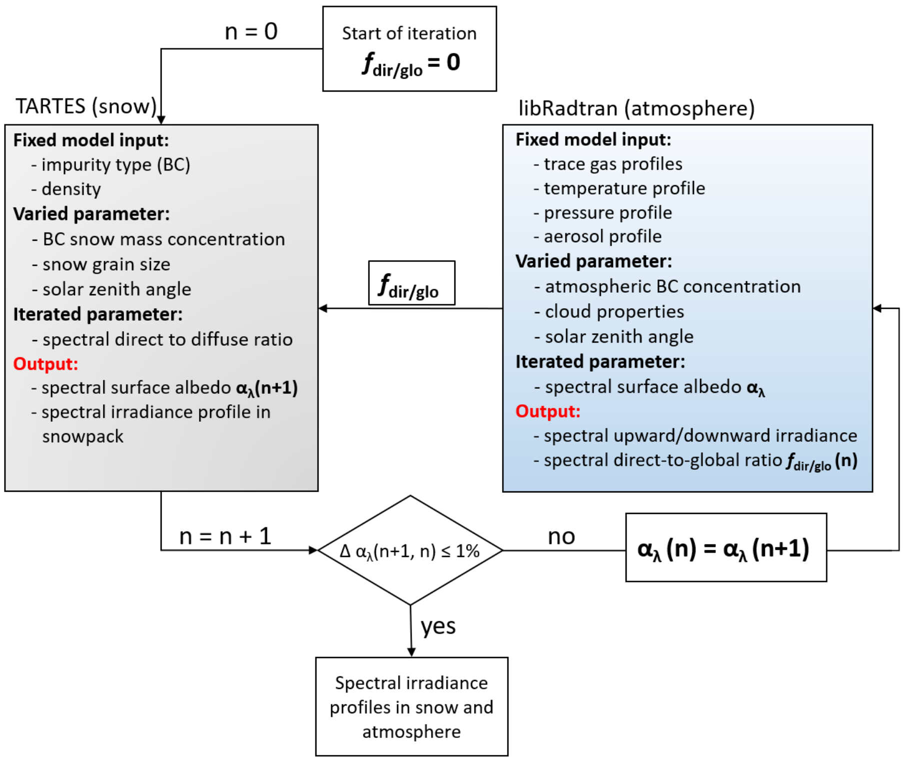

Figure 2. Schematics of the coupling of TARTES (gray box) and libRadtran (blue box) by exchanging the spectral surface albedo and the direct-to-global ratio. The ~~major model input~~ list of varied parameters ~~are listed in~~ addresses the ~~boxes~~variables which were changed between the different realizations. Only the iterated parameters $f_{\mathrm{dir/glo}}$ and α_λ were adjusted within an individual iteration cycle.

2.5 Quantities used to characterize the impact of BC particles

In ~~this study~~the following, the surface radiative ~~effect~~ forcing of BC particles and profiles of heating rates ~~were~~ are analyzed. The total radiative ~~effect~~ forcing at the surface ΔF_{tot} is separated into the ~~effect~~ forcing of BC particles suspended in the atmosphere ΔF_{atm} ~~and the effect~~ , and the forcing of BC particles deposited in the snow pack ΔF_{snow}. ~~The separated effect of BC suspended in the snow~~ ΔF_{snow} is defined by the difference of the net irradiance (~~difference of downward and~~ downward minus upward solar irradiance) ~~of the case including BC~~ if BC is considered in the snow layer ($F_{\mathrm{net,BC}}$~~and the~~) and a clean reference case without BC in the snow layer ($F_{\mathrm{net,clean}}$). Similarly, ΔF_{atm} is defined as the difference between the net irradiances derived for BC in snow and atmosphere and the atmospheric BC-free reference case:

$$\Delta F_{\underline{\mathrm{tot/atm/snow}}\,\mathrm{i}} = F_{\mathrm{net,BC}} - F_{\mathrm{net,clean}}\cancel{.}, \tag{2}$$

with index "i" standing for "tot", "atm", or "snow". For the separated ~~effects~~forcings, $F_{\mathrm{net,clean}}$ refers to either a clean atmosphere or a clean snow layer, while the other part ~~can contain~~ does consider BC particles. The default case of a clean atmosphere

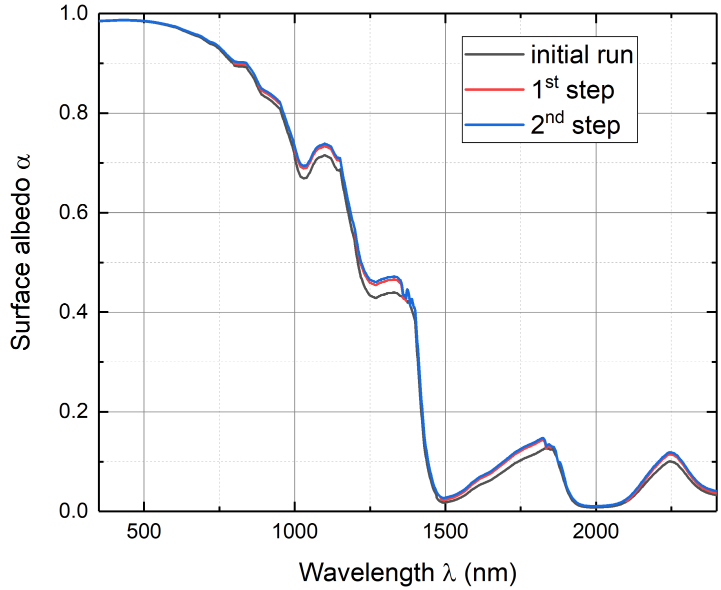

Figure 3. Change of the spectral snow albedo for cloudless conditions with a SZA of 60° due to the iterative adjustment by the coupled atmosphere and snow radiative transfer models. The initial run assumes a ~~direct-to-diffuse~~ direct-to-global ratio of zero.

used a BC mass concentration in the snow layer of 5 ng g^{-1}. Vice versa, the default case of a clean snow layer assumed the atmospheric BC profile of the ACLOUD campaign. For ΔF_{tot}, the clean reference ~~assumes~~ assumed both a pristine atmosphere and pristine snow layer.

The calculation of atmospheric and snow heating rate profiles $HR(z)$ (in K day^{-1}) ~~is~~ was based on the net irradiances at the top (t) and bottom (b) of selected atmospheric or snow layer z, the layer density $\rho(z)$, the specific heat capacity under constant pressure ~~c_{p}~~ $\mathrm{c_p}$, and the layer thickness $(z_{\mathrm{t}} - z_{\mathrm{b}})$:

$$HR(z) = \frac{\Delta T}{\Delta t}(z) = \frac{F_{\mathrm{net}}(z_{\mathrm{t}}) - F_{\mathrm{net}}(z_{\mathrm{b}})}{\rho(z) \cdot \mathrm{c_p} \cdot (z_{\mathrm{t}} - z_{\mathrm{b}})} \quad . \tag{3}$$

For atmospheric profiles, ~~$c_{\mathrm{p,air}} = 1004\,\mathrm{J\,kg^{-1}\,K^{-1}}$ the vertical resolution of the simulated irradiances was adjusted to~~ the vertical resolution ~~of the measured BC profiles , ranging between 500 m and 1 km. Increasing the vertical resolution has shown only negligible differences of less than 1 %~~ from the BC profiles was used. Similarly, the heating rate profiles within the snow pack were ~~calculate with~~ calculated applying Eq. 3 by accounting for the snow density (set to 300 kg m^{-3}) and the specific heat capacity of ice $c_{\mathrm{p,snow}}$ = 2060 J kg^{-1} K^{-1} at a temperature of 0 °C. The layer thickness within TARTES and therefore, the resolution of the heating rate profiles ~~was~~ is of 1 cm.

To separate the contribution of BC particles to the total heating rate, the effective BC heating rate $HR_{\mathrm{BC}}(z)$ ~~was~~ were calculated as the difference between the total heating rate $HR_{\mathrm{tot}}(z)$ and the heating rate of the clean reference case $HR_{\mathrm{clean}}(z)$:

$$HR_{\mathrm{BC}}(z) = HR_{\mathrm{tot}}(z) - HR_{\mathrm{clean}}(z). \tag{4}$$

If not indicated differently, radiative effects reported in this study refer to daily ~~mean estimates~~ means accounting for the change of the SZA and the night time. Therefore, simulations were performed for a full diurnal cycle with a temporal resolution of five minutes. The simulated upward and downward irradiance were averaged. Then these daily mean irradiances ~~are~~ were applied to calculate mean values of ~~ΔF and $HR(z)$~~ ΔF_{tot}, ΔF_{atm}, ΔF_{snow}, $HR_{\mathrm{tot}}(z)$, and $HR_{\mathrm{BC}}(z)$.

3 Results

3.1 Radiative impact of BC at surface level

3.1.1 Effect on surface albedo

The ~~effect of BC impurities on~~ reduction of the snow surface albedo ~~are known to depend~~ by BC impurities depends on the snow grain size~~(SSA respectively)~~. Here, changes of the snow surface albedo due to the combination of BC impurities and snow grain size variations ~~are~~ were evaluated for Arctic conditions. The single-layer snow pack setup, as defined in Section 2, was used together with atmospheric properties representing the ACLOUD conditions. The SZA was set to a constant value of 60°. Figure 4 shows the spectral snow albedo for variable BC particle mass ~~fractions~~ concentrations (0, 5, and 20 ng g^{-1}) as calculated with TARTES. The selected ~~SSA~~ SSA values represent different snow types, as freshly fallen snow with small snow grains (~~$SSA = 60$~~SSA = 60 m^2 kg^{-1}), aged snow which has undergone snow metamorphism (~~$SSA = 5$~~SSA = 5 m^2 kg^{-1}) when surface temperature approaches 0°C, and moderate aged snow without melting (~~$SSA = 20$~~SSA = 20 m^2 kg^{-1}), which ~~in this study is~~ was considered as default case. As expected, the highest values of surface albedo ~~are~~ were obtained for the case with clean and fresh snow. Adding BC particles ~~causes~~ caused a decrease in the spectral surface albedo, in particular in the visible spectral range up to 700 nm, shown in the enlargement of Fig. 4. In contrast, the near-infrared spectral range ~~is~~ was dominated by ice absorption, which is affected by the ~~SSA~~ SSA (grain size). From the simulations shown in Fig. 4 it becomes apparent that the decrease of surface albedo with increasing BC mass ~~fraction~~ concentration is stronger for aged snow than for fresh snow. Fresh snow with smaller grains leads to an enhanced backscattering of the ~~incoming~~ incident radiation, while larger grains allow for a deeper penetration of the incident radiation into the snow pack. Since the penetration depth for aged snow is deeper, the probability is higher, that the radiation gets absorbed by the BC particles leading to a decrease of the spectral surface albedo.

In the same way, the radiative ~~effect~~ forcing of BC particles ~~suspended~~ embedded in the snow layer was calculated for overcast cloudy conditions (predefined low-level liquid water cloud case) ~~in order~~ to assess the relevance of changes of the BC mass ~~fraction~~ concentration compared to variations in ~~SSA~~ SSA and the illumination conditions. To estimate the relevance

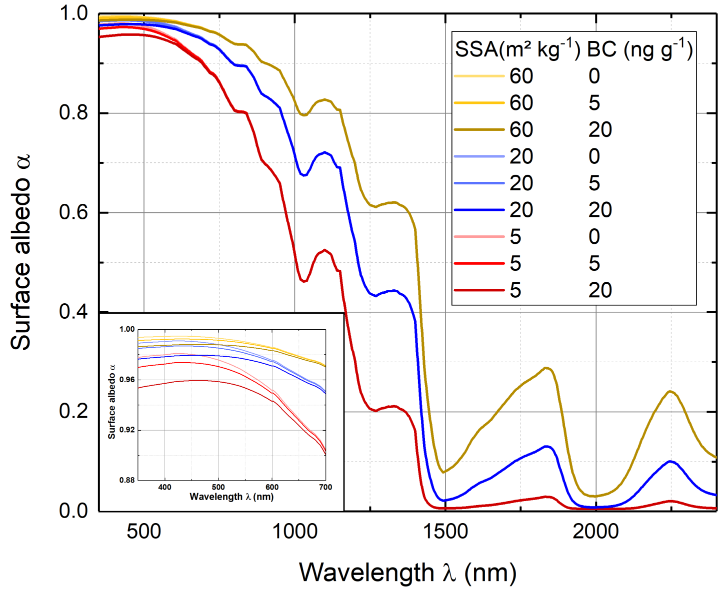

Figure 4. Spectral surface albedo of snow for cloudless conditions and a SZA of 60° for different ~~*SSA*~~ SSA and BC particle mass ~~fractions~~concentrations. The inlay shows an enlargement of the spectral albedo between 350 and 700 nm.

for the surface energy budget, the solar broadband ~~effect is~~ forcing was analyzed by calculating the broadband albedo α_{bb}. Therefore, the spectral albedo simulated by TARTES and the spectral downward irradiance $F_\lambda^{\downarrow}(\lambda)$ simulated by libRadtran ~~are~~ were used:

$$\alpha_{\mathrm{bb}} = \frac{\int \alpha(\lambda) \cdot F_\lambda^{\downarrow}(\lambda)\, d\lambda}{\int F_\lambda^{\downarrow}(\lambda)\, d\lambda}. \tag{5}$$

The calculated broadband ~~albedo~~ surface albedo values are summarized in Table 4 for the cloudy and cloudless ~~case~~cases, respectively. For both cases, ~~the highest~~ even the most extreme BC mass concentration ~~does reduce~~ reduced the surface albedo by less than 1 %. Contrarily, the snow grain size and the presence of clouds ~~show~~ cause significant changes of the snow albedo. The difference of the broadband surface albedo between fresh and aged snow ranges up to 0.12 and 0.08 for cloudless and cloudy conditions, respectively, which is in the same order ~~than the effect of the presence~~ of magnitude as the effect of clouds (0.12 for fresh snow and 0.07 for aged snow). Therefore, for Arctic conditions, the impact of BC ~~particles suspended in the snow pack is assumed to be~~ impurities on the broadband snow albedo is of minor importance~~for Arctic conditions, which is in agreement with findings by e.g.~~, compared to the impact of modifying the snow grain size. Also Warren and Wiscombe (1980) and Warren (2013) found only a small reduction of the broadband albedo between 0 - 1 % for fresh snow and 0 - 3 % for aged snow when adding BC with a mass concentration of 34 ng g^{-1} to the clean snow.

Table 4. Broadband surface albedo (α_{bb}) of fresh (~~$SSA=60$~~SSA = 60 $\mathrm{m^2\,kg^{-1}}$) and aged snow (~~$SSA=5$~~ SSA = 5 and 20 $\mathrm{m^2\,kg^{-1}}$) depending on the BC ~~parteile~~ particle mass concentration and illumination condition.

	Cloudless case α_{bb}			Cloudy case α_{bb}		
SSA ($\mathrm{m^2\,kg^{-1}}$)	BC mass concentration ($\mathrm{ng\,g^{-1}}$)			BC mass concentration ($\mathrm{ng\,g^{-1}}$)		
	0	5	20	0	5	20
5	0.76	0.76	0.75	0.88	0.87	0.87
20	0.83	0.83	0.82	0.92	0.92	0.92
60	0.87	0.87	0.87	0.95	0.95	0.94

3.1.2 Surface radiative ~~effects~~forcing

The decrease of the snow surface albedo due to ~~the~~ an increase of BC particle mass concentration ~~and~~ or snow grain size directly ~~effects~~ alters the surface radiative ~~effect of the snow pack~~ forcing ΔF_{snow}. ~~Applying Eq. 2~~To quantify these radiative effects, ΔF_{snow} was first calculated for a fixed solar zenith angle of 60°. A typical Arctic range of BC particle mass ~~fractions~~ concentrations in snow and ~~SSA~~ SSA values assuming the ACLOUD atmospheric conditions ~~and a fixed solar zenith angle of 60°.~~ were applied . Figure 5 shows a contour plot of ΔF_{snow} for ~~all combinations of SSA~~ combinations of SSA and BC particle mass ~~fractions~~concentrations. For a BC particle mass ~~fraction~~ concentration of 5 $\mathrm{ng\,g^{-1}}$ ~~representing rather clean conditions~~ in snow representing clean conditions and a SSA larger than 20 $\mathrm{m^2kg^{-1}}$, ΔF_{snow} ranges between ~~0.1 –~~0.4 – 0.7 $\mathrm{W\,m^{-2}}$. Higher BC particle mass ~~fractions~~ concentrations increase ΔF_{snow} depending on the snow grain size (~~SSA~~ SSA respectively). The strongest increase of the solar radiative warming was calculated for small ~~SSA~~ SSA values, corresponding to larger snow grain sizes. With the larger penetration depth for a smaller ~~SSA~~SSA, more radiation can be absorbed by the BC particles. ~~For typical BC concentrations in the central Arctic, which are below 10 $\mathrm{ng\,g^{-1}}$, the maximum BC radiative effect is about 1 $\mathrm{W\,m^{-2}}$.~~

To compare the radiative ~~effect~~ forcing at the surface of atmospheric BC particle profiles observed during the three aircraft campaigns ACLOUD, PAMARCMiP, and ARCTAS, the daily averaged surface radiative ~~effects are~~ forcing was then analyzed. To limit the degree of freedom, the ~~SSA~~ SSA was set to a default value of ~~$SSA=20$~~SSA = 20 $\mathrm{m^2\,kg^{-1}}$ representative for snow covered Arctic sea ice. To estimate the relevance of the atmospheric BC particles, their separated radiative ~~effect~~ forcing ΔF_{atm} was calculated. Additionally, the total radiative ~~effect~~ forcing ΔF_{tot} combining the atmospheric and snow BC was analyzed. Figure 6 summarizes the daily averaged ΔF_{snow} (panel a), ΔF_{atm} (panel b), and ΔF_{tot} (panel c) for different BC particle mass ~~fractions~~ concentrations in snow (0, 5, 20 $\mathrm{ng\,g^{-1}}$) in cloudless and cloudy conditions.

The BC particles ~~suspended~~ embedded in snow lead to warming effects of up to ~~0.9~~0.7 $\mathrm{W\,m^{-2}}$ for high BC mass ~~fractions~~ concentrations of 20 $\mathrm{ng\,g^{-1}}$ and ACLOUD conditions. For ARCTAS ΔF_{snow} ~~is~~ was slightly lower and for PAMARCMIP reduced by a factor of about 3. This difference is caused by the lower maximum Sun elevation during PAMARCMiP (location in higher latitude) resulting in a lower amount of available incoming solar irradiance compared to ACLOUD and ARCTAS (see range of SZA in Tab. 2).

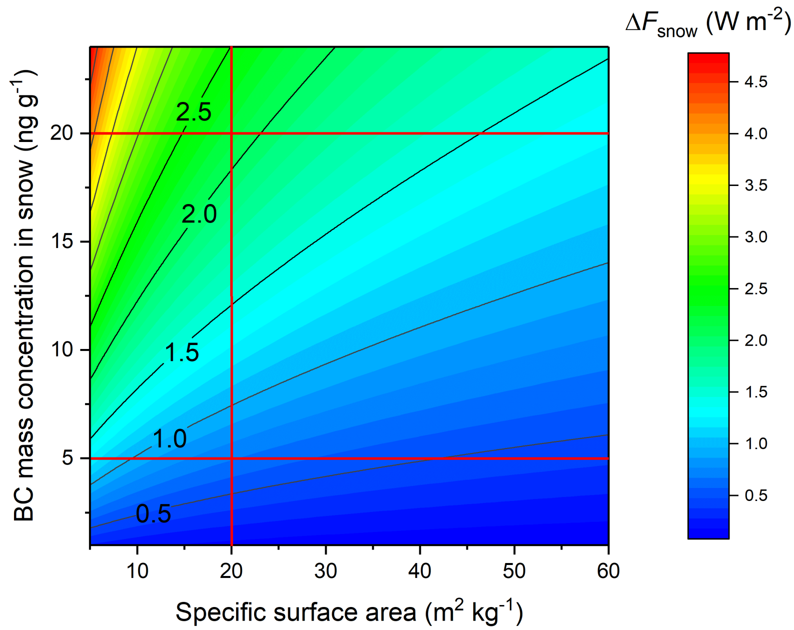

Figure 5. Solar surface radiative ~~effects~~ forcing of BC impurities in snow ΔF_{snow} calculated for different ~~*SSA*~~ SSA values and BC particle mass ~~fractions~~concentrations. The atmospheric conditions correspond to the ACLOUD case with a fixed SZA of 60°. Horizontal red lines indicate typical Arctic conditions with rather clean and more polluted snow; the vertical line represents the most typical ~~*SSA*~~SSA.

Atmospheric BC particles reduce the incident solar radiation at surface due to extinction, such that the atmospheric radiative ~~effect~~ forcing ΔF_{atm} is negative in all scenarios (Figure 6b). This cooling at the surface is strongest with values up to -0.2 $\mathrm{W\,m^{-2}}$ in cloudless conditions for the ARCTAS case, where the largest atmospheric BC particle concentrations were observed. Despite having a BC optical depth of similar magnitude, the PAMARCMiP case ($\mathrm{AOD_{BC}}$ = 0.006) shows a weaker radiative cooling compared to the ARCTAS case ($\mathrm{AOD_{BC}}$ = 0.008) caused by the higher solar zenith angles in PAMARCMiP. Minor cooling of less than $-0.02\,\mathrm{W\,m^{-2}}$ ~~is~~ was observed for the ACLOUD case, where the atmosphere ~~is~~ was rather clear with significant reduced atmospheric BC particle concentrations (factor ten lower than during ARCTAS). Comparing the simulations with different BC mass ~~fractions in snow shows~~ concentrations in snow showed only little effects of the surface properties on the radiative ~~effect~~ forcing of atmospheric BC. A slight decrease of ΔF_{atm} with increasing BC mass ~~fractions is~~ concentrations was observed for the ARCTAS case indicating, that a lower surface albedo enhances the radiative ~~effects~~ forcing of atmospheric BC particles.

The cooling effect of atmospheric BC counteracts the warming effect of BC particles in snow and can lead to a positive and negative total radiative ~~effects~~forcing. Figure 6c shows the total radiative ~~effect~~ forcing ΔF_{tot} for all cases. For BC mass ~~fraction~~ concentration of 20 $\mathrm{ng\,g^{-1}}$, all cases ~~show~~ showed a total warming effect when the warming of BC in the snow pack exceeds the cooling by atmospheric BC. The strongest warming effect of up to 0.7 $\mathrm{W\,m^{-2}}$ ~~is~~ was found for the ACLOUD case which is characterized by the pristine atmospheric conditions in the Arctic summertime. For less polluted snow (5 $\mathrm{ng\,g^{-1}}$), warming and cooling scenarios can occur depending on the concentration of atmospheric BC (ARCTAS shows a slight cooling) and the solar zenith angle (ACLOUD shows a significant warming effect). ΔF_{tot} calculated for ACLOUD even exceeds the warming effect of PAMARCMiP for the higher BC mass ~~fraction~~ concentration in the snow layer. This clearly demonstrates that the competition between the individual BC radiative ~~effects~~ forcings ΔF_{atm} and ΔF_{snow} is strongly driven by solar zenith angle and the available solar radiation and is less affected by the BC concentrations itself.

The available solar irradiance is strongly affected by the presences of clouds. Therefore, the impact of clouds on the BC radiative ~~effects~~ forcing was analyzed. Two cloud layers as defined in Section 2.2 were implemented in the simulations and considered in the calculation of $\Delta F_{\mathrm{tot/atm/snow}}$ (clean cloudy and polluted cloudy case in Eq. 2) to extract the pure BC radiative ~~effect~~forcing. In Fig. 6 the BC radiative ~~effects~~ forcing of the cloudy scenarios are shown by the shaded bars. The magnitudes of ΔF_{snow} (panel a) and ΔF_{atm} (panel b) are always reduced by the presence of clouds. ΔF_{snow} drops by about 15 % in all cases (0.1 $\mathrm{W\,m^{-2}}$ for ALCOUD and ARCTAS and high BC mass concentration in snow), while ΔF_{atm} increases by more than 50 %. $\mathrm{W\,m^{-2}}$, which amounts for ARCTAS to an absolute increase of 0.14 $\mathrm{W\,m^{-2}}$. ~~Cloud~~ Clouds reduce ΔF_{snow} because less radiation reaches the surface and can be absorbed by BC particles in the snow pack. The shift from a mostly direct illumination of the snow surface by the Sun to a diffuse illumination below the clouds is less significant as demonstrated in Table 4.

These different cloud effects counterbalance in the total radiative ~~effects~~ forcing ΔF_{atm} (Fig. 6c). To illustrate the total effect by clouds, Fig. 6d shows the difference between cloudy and cloudless simulations. In all scenarios, still slight differences between cloudy and cloudless conditions ~~are~~ were observed, but with different direction. For the ACLOUD case, the clouds reduce the warming effect of BC particles mainly due to a reduction of radiation that reaches the surface. As almost no atmospheric BC ~~is~~ was present, only ΔF_{snow} is affected.

For the ARCTAS cases, the clouds always ~~increase~~ increased ΔF_{tot}. For a BC mass concentration of 5 $\mathrm{ng\,g^{-1}}$ even the sign shifts from a total cooling to a total warming effect of BC. For ARCTAS, with high atmospheric BC concentrations, the presence of clouds mainly reduce the cooling effect of the atmospheric BC, ΔF_{atm}. As the atmospheric BC layer ~~is~~ was located mostly above the cloud, the radiative effect of the clouds, which is typically much stronger than the absorption by the atmospheric BC, reduces the significance of the atmospheric BC ~~effect~~forcing. For higher BC mass ~~fractions~~ concentrations in the snow, the increase of ΔF_{tot} by adding a cloud becomes weaker because ΔF_{snow} simultaneously slightly decreases in cloudy conditions.

The PAMARCMiP case, characterized by the low sun elevation, in general, ~~shows~~ showed a reduced effect by clouds. Here, the reduction of the cooling effect of ~~by~~ atmospheric BC, ΔF_{atm}, and the increase of the BC snow ~~effect~~forcing, ΔF_{snow}, compete each other and result in different total cloud radiative effects. Model runs with and without the upper ice cloud layer did not show any significant difference in $\Delta F_{\mathrm{tot/atm/snow}}$, which allows concluding that mainly the presence of the low liquid water clouds affects the radiative ~~effects~~ forcing of BC particles.

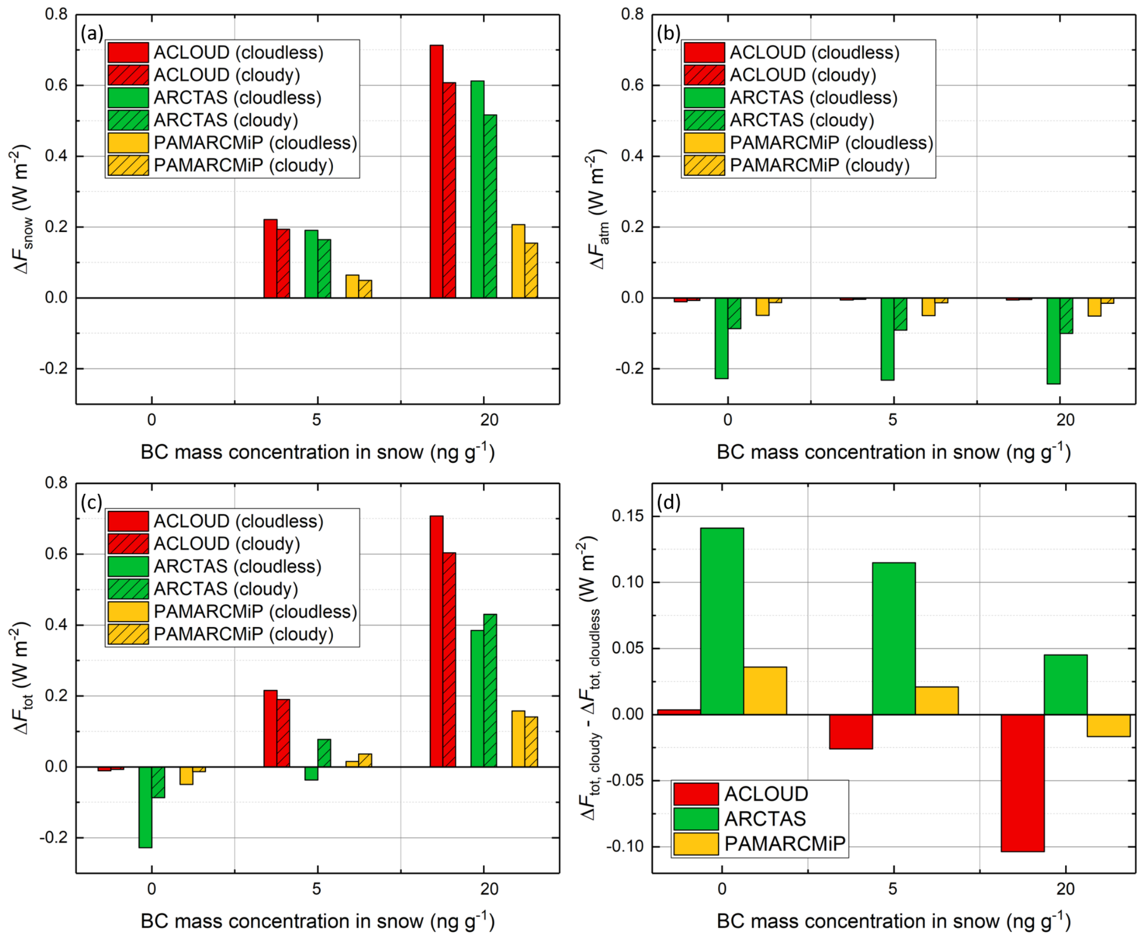

Figure 6. Daily mean of the solar surface radiative ~~effects~~ forcing simulated for the conditions of the three campaigns ACLOUD, PAMARCMiP, and ARCTAS assuming a fixed ~~SSA~~ SSA of $20\,\mathrm{m^2\,kg^{-1}}$. The separated ~~effects~~ forcings by BC ~~suspended~~ embedded in snow (ΔF_{snow}, panel a), atmospheric BC (ΔF_{atm}, panel b), and the total ~~effect~~ forcing (ΔF_{tot}, panel c) are shown. The daily mean solar radiative ~~effects~~ forcing of BC in cloudy conditions is displayed by shades bars. The difference of ΔF_{tot} between simulations with and without clouds is given in panel d.

In summary, the comparison of the radiative ~~effects~~ forcing by BC particles in snow and atmosphere with typical concentrations and mass ~~fractions~~ concentrations observed in Arctic spring and summer are rather small compared to other parameters (SZA, grain size) which are contributing to solar cooling or heating on the surface level. The highest radiative cooling of BC particles ~~is~~ was in the range of $1\,\mathrm{W\,m^{-2}}$ and ~~was~~ is estimated for low SZA, high BC particle mass ~~fractions~~concentrations, and large grains.

3.2 Vertical radiative impact of BC particles in the atmosphere and snow

3.2.1 Heating rate profiles in the atmosphere

~~In the atmosphere, BC particles can absorb solar radiation and lead to a local warming effect which might influence the stability of the atmosphere . To quantify these effects for~~ To quantify the absorption of solar radiation by Arctic atmospheric BC particles and consequent local warming effects, profiles of the heating rates were simulated for the three cases ACLOUD, ARCTAS, and PAMARCMiP. Based on simulations with and without atmospheric BC, the total heating rate $HR_{\mathrm{tot}}(z)$ and the effective heating rate of BC particles $HR_{\mathrm{BC}}(z)$ was calculated (see Eqs. 3 and 4)~~were applied~~. Figure 7 shows daily averaged profiles of $HR_{\mathrm{tot}}(z)$ and $HR_{\mathrm{BC}}(z)$ calculated for the three BC profiles. Solid lines represent the cloudless scenarios while dotted lines show simulations where the two predefined cloud layers were added. The location of the clouds is indicated by the gray shaded area. Highest total heating rates in cloudless conditions were found for the ACLOUD case, with maximum values of more than 1.2 K day^{-1} in about 2-4 km altitude. This altitude range was characterized by enhanced humidity leading to a stronger absorption of solar radiation by the water vapour. The spring campaigns ARCTAS and PAMARCMiP were characterized by lower water vapour concentrations (factor of four and ten lower than for ACLOUD, respectively) and reduced incident solar radiation due to the time of year and latitude of the observations. This ~~leads~~ lead to significant lower values of $HR_{\mathrm{tot}}(z)$ compared to the ACLOUD case. While ARCTAS ~~shows~~ showed a similar vertical pattern with maximum $HR_{\mathrm{tot}}(z)$ of 0.5 K day^{-1} in the lower troposphere below 5 km altitude, the conditions during PAMARCMiP lead to a maximum of $HR_{\mathrm{tot}}(z)$ of about 0.25 K day^{-1} located in 5-6 km altitude. This corresponds to the rather dry lower troposphere observed in spring time in the central Arctic. By adding clouds in the simulations, the highest $HR_{\mathrm{tot}}(z)$ ~~are~~ were observed within the liquid water cloud layer, where solar radiation is absorbed by the cloud particles. Similar to the cloudless scenarios, the ACLOUD case ~~shows~~ showed the highest values of $HR_{\mathrm{tot}}(z)$ with up to 4.1 K day^{-1} ~~)~~ at cloud top of the lower liquid cloud layer. ~~Absorption in the higher ice cloud less pronounces~~ The absorption in the ice cloud is less pronounced, and the increase of $HR_{\mathrm{tot}}(z)$ is ~~significant~~ significantly lower.

The profiles of the effective BC heating rate $HR_{\mathrm{BC}}(z)$ (Fig. 7b) shows a completely different pattern compared to $HR_{\mathrm{tot}}(z)$. In general, $HR_{\mathrm{BC}}(z)$ ~~is about one~~ was about one order of magnitude lower than $HR_{\mathrm{tot}}(z)$ for all three cases. Significant BC heating rates ~~are~~ were observed only for the ARCTAS and PAMARCMiP cases with values up to 0.1 K day^{-1}. The profiles of $HR_{\mathrm{BC}}(z)$ are strongly correlated with the vertical distribution of BC particles in the measured profiles. Maximum $HR_{\mathrm{BC}}(z)$ ~~are~~ were located in the pollution layers. The pollution layer observed during PAMARCMiP at 5 km and the BC layers of ARCTAS above 5 km altitude ~~show~~ showed the largest relative impact of BC particles where nearly one-fifth and one-third, respectively, of the total solar heating is attributed to BC absorption. In lower altitudes of the ARCTAS case, the enhanced absorption by water vapor ~~reduces~~ reduced the relative importance of BC particles. For the summer case of ACLOUD, $HR_{\mathrm{BC}}(z)$ ~~is~~ was rather small in all altitudes and ~~does~~ did contribute to the total radiative heating by only 10 %. However, in low altitudes, the absolute values of $HR_{\mathrm{BC}}(z)$ ~~are~~ were in the same order for both, ACLOUD and the PAMARCMiP. This illustrates that the effect of a higher BC particle concentration during PAMARCMiP ~~is~~ was compensated by the dependence of $HR_{\mathrm{BC}}(z)$ on the amount of the available incoming solar radiation and the atmospheric water vapour concentration.

Adding clouds in the simulations, ~~affects~~ affected $HR_{\mathrm{BC}}(z)$ of the three cases differently. While the clean atmosphere layer of ACLOUD and the PAMARCMiP cases show almost no differences to cloudless conditions, a minor cloud effect ~~is~~ was observed for the ARCTAS case and the polluted layer of PAMARCMiP. In both cases, the ice cloud ~~leads to a slightly~~ lead to a slight increase of $HR_{\mathrm{BC}}(z)$ by about 5 % within and above the cloud layer. This ~~is~~ was caused by the enhanced reflection of the incoming radiation which ~~leads~~ lead to additional absorption of the reflected radiation by the atmospheric BC particles. In altitudes between the ice and liquid water clouds no significant effect by the clouds ~~are~~ were observed. Within and below the liquid water cloud $HR_{\mathrm{BC}}(z)$ ~~is~~ was significantly reduced by almost 0.01 K day^{-1} for the ARCTAS case. This cloud effect ~~is~~ was caused by the strong reflection of radiation at the cloud top leading to a reduction of radiation reaching into and below the cloud layer.

Comparing all simulations, it can be concluded that the absolute radiative effects of atmospheric BC particles are potentially strongest in early spring when incoming solar radiation starts to increase and BC particle concentration is still high enough. Furthermore, the surface conditions in spring ~~are~~ were dominated by snow and ice coverage which causes an increase in the amount of upward radiation contributing to the atmospheric heating rate. In late spring and summer, the BC particle concentration ~~decreases~~ decreased rapidly, while the absorption by water vapour ~~becomes~~ became more and more dominant with increasing temperatures.

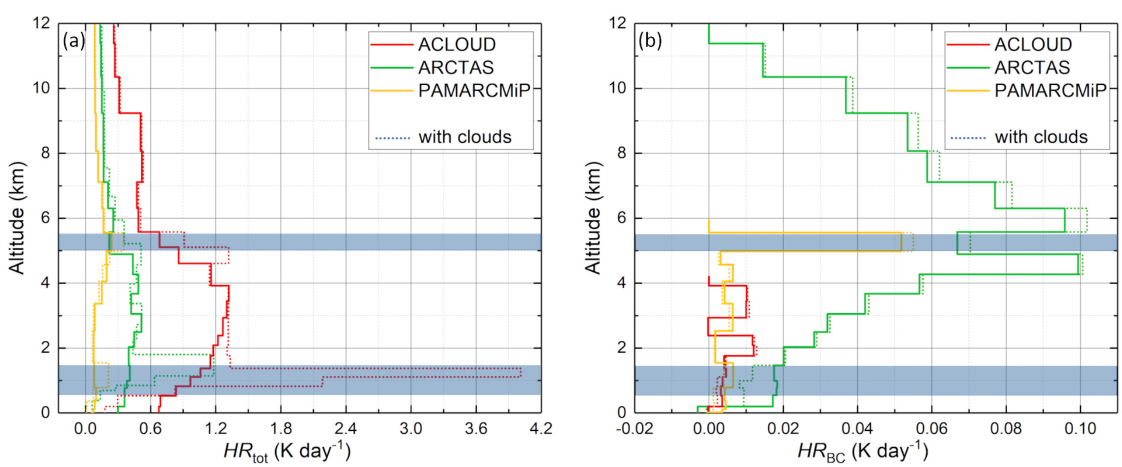

Figure 7. Daily averaged profiles of the total radiative heating rate $HR_{\mathrm{tot}}(z)$ (panel a) and the effective BC heating rate $HR_{\mathrm{BC}}(z)$ (panel b) calculated for the three cases ACLOUD, ARCTAS, and PAMARCMiP. Both, cloudless simulations (solid lines) and cloudy scenarios (dotted lines) are shown. The gray shaded areas indicate the location of the cloud layers.

3.2.2 Heating rate profiles in the snow pack

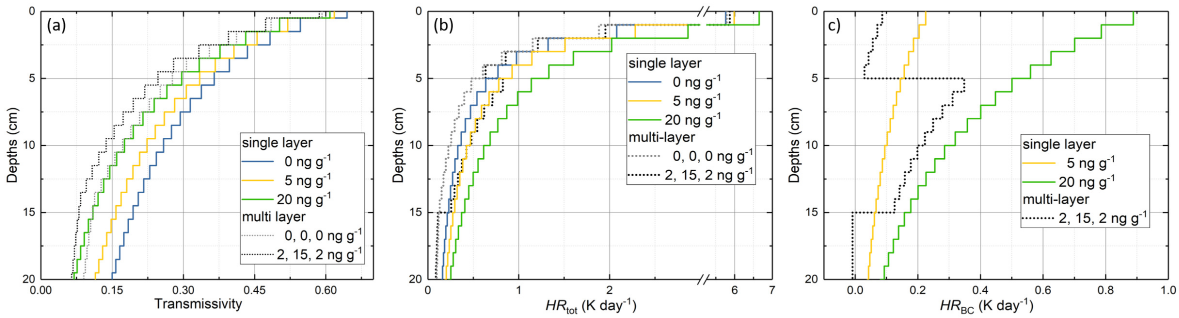

Figure 8. (a) ~~Daily mean total heating rate~~ Transmissivity profiles of solar radiation within ~~a~~ the snow pack ~~$HR_{\mathrm{tot}}(z)$~~ for three single layers and one multi-layer case assuming ACLOUD conditions. (b) Corresponding daily mean total heating rate profiles within a snow pack $HR_{\mathrm{tot}}(z)$. (c) Corresponding effective BC heating rate profiles $HR_{\mathrm{BC}}(z)$.

Not only the snow albedo, but also the transmission of radiation through the snow pack is affected by BC particles. For the atmospheric boundary conditions of the ACLOUD case, the radiative transfer in the snow pack was simulated and analyzed for different single layer and multi-layer scenarios as introduced in Tab. 3. The transmissivity was calculated from the ratio of the downward irradiance in the snow layer to the in downward irradiance at the top of the snow layer. Figure 8a shows the transmissivity profiles of solar radiation within the snow pack. The homogeneous single layer reference case without BC particles (SSA = $20\,\mathrm{m^2\,kg^{-1}}$) illustrates the general decrease of transmissivity, which is reduced to 0.3 in 20 cm snow depth. Adding a typical Arctic BC concentration of $5\,\mathrm{ng\,g^{-1}}$ reduces the transmissivity to almost 0.2. This obviously may have an impact on the radiative processes below the snow pack, in and below the sea ice as discussed by, e.g., Tuzet et al. (2019) and Marks and King (2014). The inhomogeneous multi-layer case shows in general lower transmissivities due to the enhanced reflection of the smaller snow grains at top of the layer (SSA = $60\,\mathrm{m^2\,kg^{-1}}$ down to 5 cm depth) but also indicates a significant dimming effect of the BC particles.

To access, in which layers of the snow pack the strongest absorption of solar radiation and, therefore, a potential enhancement of the snow metamorphism is located, profiles of the heating rates within the snow pack $HR_{\mathrm{tot}}(z)$ were calculated. To quantify, how BC particles deposited in snow may change these heating profiles, the effective BC heating rates $HR_{\mathrm{BC}}(z)$ were ~~calculated for different single layer and multi-layer scenarios as introduced in Tab. 3. For the atmospheric boundary conditions, the ACLOUD case was chosen~~derived in a second step. Figure 8 shows $HR_{\mathrm{tot}}(z)$ (panel a) and $HR_{\mathrm{BC}}(z)$ (panel b) for all cases in the first 20 cm of the snow pack. ~~Below, the transmittance is less than 1 % and all heating rates become negligible, which is in agreement with .~~ For all cases, the total heating rate ~~quickly decreases with~~ rapidly decreases by one magnitude within the first 10 cm of depth. The simulation for the single layer (solid lines) snow pack shows the maximum values of $HR_{\mathrm{tot}}(z)$ which ~~are~~ were located in the top most layers and reach values up to $6.6\,\mathrm{K\,day^{-1}}$ (note, the scale break in Fig. 8a).

Assuming different BC mass ~~fractions~~ concentrations in the single layer case, slightly increases $HR_{\mathrm{tot}}(z)$ in the entire column. In the multi-layer case, this increase is limited to the upper part of the profile. This contribution of BC particles to the total radiative heating ~~is~~ was quantified by $HR_{\mathrm{BC}}(z)$ and shown in Figure 8b. Largest $HR_{\mathrm{BC}}(z)$ ~~are~~ were observed for the most polluted single layer case with a BC mass ~~fraction~~ concentration of 20 ng g^{-1}. For this case, the contribution by BC particles amounts to almost 0.9 K day^{-1} in the top most layer dropping down to a value of less than 0.1 K day^{-1} in 20 cm snow depth. Compared to the total radiative effect, $HR_{\mathrm{BC}}(z)$ contributes with about 15 % to the heating rate at the top snow layer and 40 % to the heating in the base layer. For the typical Arctic BC mass ~~fraction~~ concentration of 5 ng g^{-1}, this contribution of BC particles is significantly lower ranging between 3 % and 20 %.

The multi-layer cases is characterized by smaller snow grains in the top layer (~~$SSA = 40$~~SSA = 40 m^2 kg^{-1}) compared to the single-layer cases (~~$SSA = 20$~~SSA = 20 m^2 kg^{-1}) and, therefore, shows reduced values of $HR_{\mathrm{tot}}(z)$. According to the structure of the snow pack, $HR_{\mathrm{BC}}(z)$ is largest in the layer of the highest BC mass ~~fraction~~concentration. Beneath this layer ($z < 15$ cm) the heating rates for the pristine and polluted case are almost similar (~~$HR_{\mathrm{BC}}(z) = 0$~~$HR_{\mathrm{BC}}(z) \approx 0$ K day^{-1}). In this base layer, the largest snow grains are assumed (lowest ~~SSA~~SSA) which increases the absorption of radiation by the snow ice water.

Based on these results, it becomes evident that the absorption of solar radiation by the ice water of the snow grains dominates the total heating rate in the snow pack, especially at the top layer, where most radiation is absorbed. Therefore, in Arctic conditions the snow grain size typically plays a larger role than the concentration of BC particles ~~suspended~~ embedded in snow. ~~This illustrates, that~~ To estimate if BC particles can accelerate the snow metamorphism~~is a self amplifying effect and can only slightly be accelerated by BC particles~~, coupled snow physical models need to be applied (e.g., Tuzet et al., 2017). However, compared to the results reported by Tuzet et al. (2017) who studied alpine snow with at least a magnitude higher BC mass concentrations, for Arctic conditions it is likely, that the self-amplification of the snow metamorphism is dominated the reduction of the surface albedo.

Simulations in cloudy conditions (not shown here), ~~result~~ resulted in a reduced $HR_{\mathrm{tot}}(z)$ and $HR_{\mathrm{BC}}(z)$ because the clouds reduce the incoming solar radiation. Similarly, a change of the solar zenith angle affects the results by changing the available solar radiation. Therefore, the ACLOUD case used in the simulations presented in this section represents the maximum radiative effects compared to ARCTAS and PAMARCMiP conditions. In general, it can be concluded that the solar heating by BC particles ~~suspended~~ embedded in the snow pack is most effective for low SZA (spring and summer conditions with high amount of available incoming radiation), decreasing ~~SSA~~ SSA (aged snow in conditions near melting temperature), and increasing BC particle mass ~~fractions~~ concentrations (accumulated BC particles caused by melting). Such conditions are mostly linked to late spring and summer, when the Sun is high, snow is close to melting and BC has accumulated. This suggests that the maximum heating rates due to atmospheric BC and BC ~~suspended~~ embedded in the snow pack typically occur in different periods of the year, early spring and early summer, respectively.

4 Summary and conclusions

This study analyzed the ~~direct~~ instantaneous solar radiative effect at the surface of Arctic BC particles (suspended in the atmosphere and embedded in the snow pack) ~~under~~ over the sea ice covered Arctic Ocean. The difference of the BC effects in cloudless and cloudy conditions was compared. For this purpose, an atmospheric and a snow radiative transfer model were iteratively coupled to account for the radiative interactions between both compartments (atmosphere and snow layer). Typical atmospheric BC vertical profiles and BC particle mass ~~fractions~~ concentrations in the snow pack, derived from three field campaigns in the ~~American and~~ North American and the North Atlantic Arctic, ACLOUD, ARCTAS, and PAMARCMiP, were used ~~to set up~~ in the simulations. These locations typically are not affected by local pollution, but by long-range transport of BC particles. The BC radiative effects were quantified by the surface radiative ~~effect~~ forcing and profiles of heating rates in the atmosphere and the snow, which were presented on the basis of daily averages. For the surface radiative ~~effect~~forcing, the contribution by atmospheric and snow BC particles was separated. For the heating rate profiles, the effective contribution of BC particles to the total heating rates was derived and compared to ~~other parameters~~ further atmospheric and snow parameters also leading to a warming or cooling (e.g., water ~~vapour~~vapor, clouds, snow grain size).

The magnitude of the atmospheric BC radiative forcing at the surface derived in this study (up to -0.2 W m^{-2}) agrees quite well with findings from Wendling et al. (1985). They reported a BC induced solar cooling in the range of 0.0 to -0.5 W m^{-2} for spring measurements in the Svalbard area. Further, the solar surface radiative forcing due to BC embedded in snow showed solar warming between 0.05 and 0.7 W m^{-2} depending on the BC mass concentration and incident solar irradiance. For comparison, Dou and Cun-De (2016) deduced an averaged solar warming over Svalbard in spring of 0.54 W m^{-2} based on a BC mass concentration of 5 ng g^{-1} in snow.

The simulations suggest, that ~~the local radiative effects of BC are~~ for the specific Arctic cases investigated in our study, the radiative forcing of BC is small compared to the radiative impact of ~~these other parameters . Therefore, the~~ other parameters (water vapor, clouds, snow grain size). The significance of the BC radiative effects ~~show~~ shows a strong seasonal dependence. In cloudless conditions, the ~~atmospheric water vapour is~~ absorption by atmospheric water vapor shows a much stronger ~~driver of~~ contribution to the atmospheric heating rates ~~as compared to~~ than the radiative effect of BC particles. In summer (ACLOUD) and in lower latitudes (ARCTAS), the Arctic shows the most humid conditions, where absorption of water vapor dominates over the BC radiative effects. Similarly, the available incident solar radiation limits the magnitude of the BC radiative effects. Despite the more polluted atmosphere, the low solar zenith angle of the cases of PAMARCMiP (high latitude) and ARCTAS (early spring season) did show lower BC radiative effects than the ACLOUD case. Thus, ~~in the Arctic~~over the sea ice covered Arctic Ocean, the BC radiative effect is about a magnitude lower than observed in lower and tropical latitudes, where also the pollution level is typically higher. For example, studies ~~conducted for~~ investigating strong pollution conditions in northern India or China reported on BC heating rates in the atmosphere larger than 2 K day^{-1}~~. Compared to the maximum heating rate of 0.1 K day⁻¹ derived in this study for Arctic conditions, such high values can~~ , which may significantly influence the lapse rate ~~feedback~~ and the atmospheric stability ~~. However, for~~ (Tripathi et al., 2007; Wendisch et al., 2008). For the rather pristine Arctic, ~~the calculated~~ this study showed significantly lower daily mean BC heating rates ~~derived in this study are small, and, thus, BC particles can not~~ of maximum 0.1 K day^{-1}, which have not the potential to significantly modify the atmospheric stability. However, in other Arctic regions characterized by ~~a~~ higher atmospheric BC particle concentrations due to local fires, e.g., northern Siberia, a stronger impact can be expected.

Similarly, the mass ~~fraction~~ concentration of BC particles ~~suspended~~ embedded in the Arctic snow pack is ~~by magnitudes~~ far lower than observed in alpine snow in lower latitudes. Accordingly, the absorption of radiation by the snow water itself dominates the radiative warming in the snow pack. For typical conditions of the central Arctic, the absorption due to BC particles contributes only with 3 % to the total heating rate in the uppermost snow layer. These results indicate, that the microphysical properties of the snow pack (mainly snow grain sizes) are ~~a~~ more important drivers for the degree/strength of the snow metamorphism. It needs to be considered, that this picture might change if the accumulation of BC particles is more efficient than it is over the snow covered Arctic sea ice, where the sea ice and snow pack does not last more than one to three years. Accumulation of BC on e.g. the Greenlandic glaciers will amplify the radiative forcing on a local scale. Furthermore, BC particles are not the only light absorbing impurities, which are transported into the Arctic. The relevance of dust particles and micro-organisms is currently subject of the scientific discussion and may exceed the effect of BC particles (Kylling et al., 2018; Skiles et al., 2018).

~~These dependencies of the~~ However, the changing relevance of the BC radiative effects suggests that the maximum heating rates due to atmospheric BC and BC ~~suspended~~ embedded in the snow pack typically occur in different periods of the year. While atmospheric BC ~~shows the largest effect~~ particles reveal the largest radiative effects in early spring (high concentration of atmospheric BC, medium high Sun, low water vapour), the BC particles ~~suspended~~ embedded in snow warm more ~~effective~~ effectively in early summer (accumulation of BC particles in snow, high Sun, large snow grain size).

~~Radiative transfer simulations assuming cloudless and cloudy conditions were compared to~~ To estimate the role of clouds on the surface warming/cooling by BC particles and the BC heating rates, radiative transfer simulations assuming cloudless and cloudy conditions were compared. Clouds reflect the ~~incoming~~ incident solar radiation and, therefore, reduce the available radiation reaching the ~~lower altitudes~~surface. This reduces the potential of the warming effect by BC particles ~~suspended~~ embedded in the snow. Similarly, the cooling effect by atmospheric BC on the surface radiative budget is weakened in the presence of clouds. The competition of these ~~both~~ two cloud effects depends on the BC concentrations in the snow and atmosphere and is affected by the increased broadband surface albedo and the multiple scattering in presence of a cloud layer. The profiles of the effective BC heating ~~rate~~ rates are mainly affected by the ice cloud in higher altitude. Within and above the cloud, the radiation reflected by the cloud enhances the local radiative heating by BC. Contrarily, a low liquid water cloud reduces the available incoming radiation, such that the effective BC radiative effect is lower for the cloudy case compared to the cloudless case. For the same reason, the presence of clouds reduces the radiative heating rates within the snow pack.

For the ~~Atlantic Arctic , based on the presented study, we therefore~~ sea ice covered Arctic Ocean, we conclude that: (i) the warming effect of BC ~~suspended~~ embedded in the snow ~~overcompensated~~ overcompensates the atmospheric BC cooling effect at the surface, (ii) the impact of clouds ~~shows an opposite direction for atmospheric BC cooling and snow BC warming~~ reduces both, the surface cooling by atmospheric BC particles and the warming by BC particles embedded in snow, and (iii) the BC radiative effect is of minor importance compared to other ~~drivers~~absorbers. However, for the expected increase of BC particle mass concentrations in the future, the relative importance of BC particles ~~will~~ might need to be ~~reevaluated~~re-evaluated. Additionally, ongoing research, e.g.~~by using results expected from the currently ongoing MOSAiC drift experimentand associated flight measurements, will allow~~ , triggered by the current MOSAiC (Multidisciplinary drifting Observatory for the Study of

Arctic Climate) experiment, will enable to quantify the ~~effective radiative effect~~ radiative effects of BC also in the Eastern and Central Arctic using the methods proposed here.

Data availability. Atmospheric BC mass concentrations for the ARCTAS campaign are available on https://www-air.larc.nasa.gov/cgi-bin/ArcView/arctas, last access: 22 January 2020 (PI: Yutaka Kondo). PAMARCMiP and ACLOUD data are available on PANGAEA (https://doi.pangaea.de/10.1594/PANGAEA.899508 and https://doi.org/10.1594/PANGAEA.899937, respectively)

Author contributions. All authors contributed to the editing of the manuscript and to the discussion of the results. MW, AE, and AH designed this study. TD drafted the manuscript, performed the radiative transfer simulations and prepared the figures. AE, EJ, and BH contributed to the interpretation of the radiative transfer simulations. MZ processed the SP2 data. JS compiled the atmospheric BC profiles.

Competing interests. The authors declare that they have no conflict of interest.

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References

Anderson, G., Clough, S., Kneizys, F., Chetwynd, J., and Shettle, E.: AFGL Atmospheric Constituent Profiles (0–120 km), Tech. Rep. AFGL-TR-86-0110, AFGL (OPI), Hanscom AFB, MA 01736, 1986.

Arnold, S., Law, K., Thomas, J., Starckweather, S., von Salzen, K., Stohl, A., Sharma, S., Lund, M., Flanner, M., Petäjä, T., Tanimoto, H., Gamble, J., Dibb, J., Melamad, M., Johnson, N., Fidel, M., Tynkkynen, V.-P., Baklanov, A., Eckhardt, S., Monks, S., Browse, J., and Bozem, H.: Arctic air pollution: Challenges and opportunities for the next decade, ELEMENTA – Science of the Antropocene, 4, 1–17, https://doi.org/10.12952/journal.elementa.000104, 2016.

Bierwirth, E., Ehrlich, A., Wendisch, M., Gayet, J.-F., Gourbeyre, C., Dupuy, R., Herber, A., Neuber, R., and Lampert, A.: Optical thickness and effective radius of Arctic boundary-layer clouds retrieved from airborne nadir and imaging spectrometry, Atmos. Meas. Tech., 6, 1189–1200, https://doi.org/10.5194/amt-6-1189-2013, 2013.

Blanchard, Y., Royer, A., O'Neill, N. T., Turner, D. D., and Eloranta, E. W.: Thin ice clouds in the Arctic: cloud optical depth and particle size retrieved from ground-based thermal infrared radiometry, Atmospheric Measurement Techniques, 10, 2129–2147, https://doi.org/10.5194/amt-10-2129-2017, 2017.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res., 118, 5380–5552, https://doi.org/10.1002/jgrd.50171, 2013.

Choudhury, B. J. and Chang, A. T. C.: The albedo of snow for partially cloudy skies, Bound.-Layer Meteorol., 20, 371–389, https://doi.org/10.1007/BF00121380, 1981.

Corbett, J. J., Lack, D. A., Winebrake, J. J., Harder, S., Silberman, J. A., and Gold, M.: Arctic shipping emissions inventories and future scenarios, Atmos. Chem. Phys., 10, 9689–9704, https://doi.org/10.5194/acp-10-9689-2010, 2010.

Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, A. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, Atmos. Chem. Phys., 10, 11 647–11 680, https://doi.org/10.5194/acp-10-11647-2010, 2010.

Dou, T. and Cun-De, X.: An overview of black carbon deposition and its radiative forcing over the Arctic, Adv. Clim. Change Res., 7, https://doi.org/10.1016/j.accre.2016.10.003, 2016.

Dou, T., Xiao, C., Du, Z., Schauer, J., Ren, H., Ge, B., Xie, A., Tan, J., Fu, P., and Zhang, Y.: Sources, evolution and impacts of EC and OC in snow on sea ice: a measurement study in Barrow, Alaska, Science Bulletin, 62, 1547 – 1554, https://doi.org/https://doi.org/10.1016/j.scib.2017.10.014, http://www.sciencedirect.com/science/article/pii/S2095927317305315, 2017.

Ehrlich, A., M., W., Lüpkes, C., Buschmann, M., Bozem, H., Chechin, D., C. H.-C., Dupuy, R., Eppers, O., Hartmann, J., Herber, A., Jäkel, E., Järvinen, E., Jourdan, O., Kästner, U., Kliesch, L.-L., Köllner, F., Mech, M., Mertes, S., Neuber, R., Ruiz-Donoso, E., Schnaiter, M., Schneider, J., Stapf, J., and Zanatta, M.: A comprehensive in situ and remote sensing data set from the Arctic CLoud Observations Using airborne measurements during polar Day (ACLOUD) campaign, Earth Syst. Sci. Data Discuss., https://doi.org/10.5194/essd-2019-96, 2019.

Emde, C., Buras-Schnell, R., Kylling, A., Mayer, B., Gasteiger, J., Hamann, U., Kylling, J., Richter, B., Pause, C., Dowling, T., and Bugliaro, L.: The libRadtran software package for radiative transfer calculations (version 2.0.1), Geosci. Model Dev., 9, 1647–1672, https://doi.org/10.5194/gmd-9-1647-2016, 2016.

~~Evans, K.: The spherical harmonics discrete ordinate method for three-dimensional atmospheric radiative transfer, J. Atmos. Sci., 55, 429–446, , 1998.~~

Flanner, M., S. Zender, C., T. Randerson, J., and Rasch, P.: Present-day climate forcing and response from black carbon in snow, J. Geophys. Res, 112, https://doi.org/10.1029/2006JD008003, 2007.

Flanner, M. G.: Arctic climate sensitivity to local black carbon, J. Geophys. Res. Atmos., 118, 1840–1851, https://doi.org/10.1002/jgrd.50176, 2013.

Forsström, S., Isaksson, E., Skeie, R., Ström, J., Pedersen, C., Hudson, S., Berntsen, T., Lihavainen, H., Godtliebsen, F., and Gerland, S.: Elemental carbon measurements in European Arctic snow packs, J. Geophys. Res. Atmos., 118, https://doi.org/10.1002/2013JD019886, 2013.

Fu, Q.: A New Parameterization of an Asymmetry Factor of Cirrus Clouds for Climate Models, J. Atmos. Sci., 64, 4140—-4150, https://doi.org/10.1175/2007JAS2289.1, 2007.

Gardner, A. S. and Sharp, M. J.: A review of snow and ice albedo and the development of a new physically based broadband albedo parameterization, J. Geophys. Res. Earth Surf., 115, https://doi.org/10.1029/2009JF001444, 2010.

Gilgen, A., Huang, W. T. K., Ickes, L., Neubauer, D., and Lohmann, U.: How important are future marine and shipping aerosol emissions in a warming Arctic summer and autumn?, Atmos. Chem. Phys., 18, 10 521–10 555, https://doi.org/10.5194/acp-18-10521-2018, 2018.

Gueymard, C. A.: The sun's total and spectral irradiance for solar energy applications and solar radiation models, Sol. Energy, 76, 423–453, https://doi.org/10.1016/j.solener.2003.08.039, 2004.

Herber, Haas, C., Stone, R., Bottenheim, J., Liu, P., Li, S.-M., and Staebler, R.: Regular airborne surveys of Arctic sea ice and atmosphere, EOS Transactions American Geophysical Union, https://doi.org/10.1029/2012EO040001, 2012.

Herber, A.: Meteorological observations during POLAR 5 campaign PAMARCMIP 2018, https://doi.org/10.1594/PANGAEA.898788, https://doi.org/10.1594/PANGAEA.898788, 2019.

Hess, M., Koepke, P., and Schult, I.: Optical properties of aerosols and clouds: the software package OPAC, Bull. Amer. Meteorol. Soc., 79, 831–844, https://doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2, 1998.

Jacob, D. J., Crawford, J. H., Maring, H., Clarke, A. D., Dibb, J. E., Emmons, L. K., Ferrare, R. A., Hostetler, C. A., Russell, P. B., Singh, H. B., Thompson, A. M., Shaw, G. E., McCauley, E., Pederson, J. R., and Fisher, J. A.: The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results, Atmos. Chem. Phys., 10, 5191–5212, https://doi.org/10.5194/acp-10-5191-2010, 2010.

~~Järvinen, O.and Leppäranta, M.: Solar radiation transfer in the surface snow layer in Dronning Maud Land, Antarctica, Polar Sci.~~

Joseph, J., ~~7, 1–17, , 2013.~~ Wiscombe, W., and Weinman, J.: The Delta-Eddington Approximation for Radiative Flux Transfer, Journal of the Atmospheric Sciences, 33(12), 2452–2459, https://doi.org/10.1175/1520-0469(1976)033<2452:TDEAFR>2.0.CO;2, 1977.

Kokhanovsky, A. A. and Zege, E. P.: Scattering optics of snow, Appl. Opt., 43, 1589–1602, https://doi.org/10.1364/AO.43.001589, 2004.

Kylling, A., Groot Zwaaftink, C. D., and Stohl, A.: Mineral dust instantaneous radiative forcing in the Arctic, Geophys. Res. Lett., 45, 4290–4298, https://doi.org/10.1029/2018GL077346, 2018.

Law, K. S., Stohl, A., Quinn, P. K., Brock, C. A., Burkhart, J. F., Paris, J.-D., Ancellet, G., Singh, H. B., Roiger, A., Schlager, H., Dibb, J., Jacob, D. J., Arnold, S. R., Pelon, J., and Thomas, J. L.: Arctic Air Pollution: New Insights from POLARCAT-IPY, Bull. Am. Meteorol. Soc., 95, 1873–1895, https://doi.org/10.1175/BAMS-D-13-00017.1, 2014.

Leaitch, W. R., Korolev, A., Aliabadi, A. A., Burkart, J., Willis, M. D., Abbatt, J. P. D., Bozem, H., Hoor, P., Köllner, F., Schneider, J., Herber, A., Konrad, C., and Brauner, R.: Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic, Atmos. Chem. Phys., 16, 11 107–11 124, https://doi.org/10.5194/acp-16-11107-2016, 2016.

Libois, Q., Picard, G., France, J. L., Arnaud, L., Dumont, M., Carmagnola, C. M., and King, M. D.: Influence of grain shape on light penetration in snow, Cryosphere, 7, 1803–1818, https://doi.org/10.5194/tc-7-1803-2013, 2013.

Libois, Q., Picard, G., Dumont, M., Arnaud, L., Sergent, C., Pougatch, E., Sudul, M., and Vial, D.: Experimental determination of the absorption enhancement parameter of snow, J. Glaciol., 60, 714–724, https://doi.org/10.3189/2014JoG14J015, 2014.

Liu, J., Fan, S., Horowitz, L. W., and Levy, II, H.: Evaluation of factors controlling long-range transport of black carbon to the Arctic, J. Geophys. Res. Atmos., 116, D04307, https://doi.org/10.1029/2010JD015145, 2011.

Luebke, A. E., Avallone, L. M., Schiller, C., Meyer, J., Rolf, C., and Krämer, M.: Ice water content of Arctic, midlatitude, and tropical cirrus – Part 2: Extension of the database and new statistical analysis, Atmos. Chem. Phys., 13, 6447–6459, https://doi.org/10.5194/acp-13-6447-2013, https://www.atmos-chem-phys.net/13/6447/2013/, 2013.

~~Macke~~

Marks, A.~~and Flores, H~~ A. and King, M. D.: The ~~expeditions PS106~~effect of snow/~~1 and 2 of the research vessel POLARSTERN to the Arctic ocean in 2017, Reports on polar and marine research, Bremerhaven, Alfred Wegener Institute for Polar and Marine Research, 719, , 2018.~~ sea ice type on the response of albedo and light penetration depth (e-folding depth) to increasing black carbon, Cryosphere, 8, 1625—-1638, https://doi.org/10.5194/tc-8-1625-2014, 2014.

Matsui, H., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L. K., Zhao, Y., Fuelberg, H. E., Sessions, W. R., Diskin, G., Blake, D. R., Wisthaler, A., and Koike, M.: Seasonal variation of the transport of black carbon aerosol from the Asian continent to the Arctic during the ARCTAS aircraft campaign RID F-4366-2011, J. Geophys. Res., 116, D05 202, https://doi.org/10.1029/2010JD015067, 2011.

Mayer, B. and Kylling, A.: Technical note: The *libRadtran* software package for radiative transfer calculations - description and examples of use, Atmos. Chem. Phys., 5, 1855–1877, https://doi.org/10.5194/acp-5-1855-2005, 2005.

Moteki, N. and Kondo, Y.: Effects of Mixing State on Black Carbon Measurements by Laser-Induced Incandescence, Aerosol Sci. Technol., 41, 398–417, https://doi.org/10.1080/02786820701199728, 2007.

Namazi, M., von Salzen, K., and Cole, J. N. S.: Simulation of black carbon in snow and its climate impact in the Canadian Global Climate Model, Atmos. Chem. Phys., 15, 10 887–10 904, https://doi.org/10.5194/acp-15-10887-2015, 2015.

Pedersen, C., Gallet, J.-C., Ström, J., Gerland, S., Hudson, S., Forsström, S., Isaksson, E., and Berntsen, T.: In-situ observations of black carbon in snow and the corresponding spectral surface albedo reduction, J. Geophys. Res. Atmos., 120, https://doi.org/10.1002/2014JD022407, 2015.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S.-M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., and Zhang, X.-Y.: Recommendations for reporting "black carbon" measurements, Atmospheric Chemistry and Physics, 13, 8365–8379, https://doi.org/10.5194/acp-13-8365-2013, 2013.

Samset, B. H., Myhre, G., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Penner, J. E., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., and Zhang, K.: Black carbon vertical profiles strongly affect its radiative forcing uncertainty, Atmos. Chem. Phys., 13, 2423–2434, https://doi.org/10.5194/acp-13-2423-2013, 2013.

Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S.-M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., and Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, Atmos. Chem. Phys., 14, 12 465–12 477, https://doi.org/10.5194/acp-14-12465-2014, 2014.

Sand, M., Berntsen, T., Seland, Ø., and Kristjánsson, J.: Arctic surface temperature change to emissions of black carbon within Arctic or midlatitudes, J. Geophys. Res. Atmos., 118, 7788–7798, https://doi.org/10.1002/jgrd.50613, 2013.

Sand, M., Samset, B. H., Balkanski, Y., Bauer, S., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Luo, G., Myhre, G., Noije, T. V., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Yu, F., Zhang, K., and Zhang, H.: Aerosols at the poles: an AeroCom Phase II multi-model evaluation, Atmos. Chem. Phys., 17, 12 197–12 218, https://doi.org/10.5194/acp-17-12197-2017, 2017.

Schacht, J., Heinold, B., Quaas, J., Backman, J., Cherian, R., Ehrlich, A., Herber, A., Huang, W. T. K., Kondo, Y., Massling, A., Sinha, P. R., Weinzierl, B., Zanatta, M., and Tegen, I.: The importance of the representation of air pollution emissions for the modeled distribution and radiative effects of black carbon in the Arctic, Atmos. Chem. Phys., 19, 11 159–11 183, https://doi.org/10.5194/acp-19-11159-2019, 2019.

Schulz, H., Zanatta, M., Bozem, H., Leaitch, W. R., Herber, A. B., Burkart, J., Willis, M. D., Kunkel, D., Hoor, P. M., Abbatt, J. P. D., and Gerdes, R.: High Arctic aircraft measurements characterising black carbon vertical variability in spring and summer, Atmos. Chem. Phys., 19, 2361–2384, https://doi.org/10.5194/acp-19-2361-2019, 2019.

Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and Maksyutov, S.: 16-year simulation of Arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition, J. Geophys. Res. Atmos., 118, 943–964, https://doi.org/10.1029/2012JD017774, 2013.

~~Stachlewska, I. S., Neuber, R., Lampert, A., Ritter, C~~

Skiles, S. M., Flanner, M., Cook, J., Dumont, M., and ~~Wehrle, G.: AMALi the Airborne Mobile Aerosol Lidar for Arctic research, Atmos. Chem. Phys., 10, 2947–2963, , 2010.~~

~~Stamnes, K., Tsay, S., Wiscombe, W., and Jayaweera, K.: Numerically stable algorithm for discrete-ordinate-method radiative transfer in multiple scattering and emitting layered media, Appl. Opt.~~ Painter, T. H.: Radiative forcing by light-absorbing particles in snow, Nature Clim. Change, 8, 964–971, https://doi.org/10.1038/s41558-018-0296-5, ~~27, 2502–9, 1988.~~ 2018.

Stamnes, K., Tsay, S.-C., Wiscombe, W., and Laszlo, I.: DISORT, A General-Purpose Fortran Program for Discrete-Ordinate-Method Radiative Transfer in Scattering and Emitting Layered Media: Documentation of Methodology, Tech. rep., Dept. of Physics and Engineering Physics, Stevens Institute of Technology, Hoboken, NJ 07030, 2000.

Stapf, J., Ehrlich, A., Jäkel, E., Lüpkes, C., and Wendisch, M.: Reassessment of the common concept to derive the surface cloud radiative forcing in the Arctic: Consideration of surface albedo – cloud interactions, Atmospheric Chemistry and Physics Discussions, 2019, 1–24, https://doi.org/10.5194/acp-2019-534, https://www.atmos-chem-phys-discuss.net/acp-2019-534/, 2019.

Stohl, A., Klimont, Z., Eckhardt, S., Kupiainen, K., Shevchenko, V. P., Kopeikin, V. M., and Novigatsky, A. N.: Black carbon in the Arctic: the underestimated role of gas flaring and residential combustion emissions, Atmos. Chem. Phys., 13, 8833–8855, https://doi.org/10.5194/acp-13-8833-2013, 2013.

Stone, R., Herber, A., Vitale, V., Mazzola, M., Lupi, A., Schnell, R., Dutton, E., Liu, P., Li, S.-M., Dethloff, K., Lampert, A., Ritter, C., Stock, M., Neuber, R., and Maturilli, M.: A three-dimensional characterization of Arctic aerosol particles from airborne Sun photometer observations: PAN-ARCMIP, April 2009, J. Geophys. Res., D13203, doi:10.1029/2009JD013 605, 2010.

Tripathi, S., Srivastava, A., Dey, S., Satheesh, S., and Moorthy, K.: The vertical profile of atmospheric heating rate of black carbon aerosols at Kanpur in northern India, Atmos. Environ., 41, 6909–6915, https://doi.org/10.1016/j.atmosenv.2007.06.032, 2007.

Tuzet, F., Dumont, M., Lafaysse, M., Picard, G., Arnaud, L., Voisin, D., Lejeune, Y., Charrois, L., Nabat, P., and Morin, S.: A multilayer physically based snowpack model simulating direct and indirect radiative impacts of light-absorbing impurities in snow, Cryosphere, 11, 2633—2653, https://doi.org/10.5194/tc-11-2633-2017, 2017.

Tuzet, F., Dumont, M., Arnaud, L., Voisin, D., Lamare, M., Larue, F., Revuelto, J., and Picard, G.: Influence of light-absorbing particles on snow spectral irradiance profiles, Cryosphere, 13, 2169–2187, https://doi.org/10.5194/tc-13-2169-2019, 2019.

Warren, S. and Wiscombe, W.: A Model for the Spectral Albedo of Snow. II: Snow containing atmospheric aerosols, J. Atmos. Sci., 37, 2734–2745, https://doi.org/10.1175/1520-0469(1980)037<2734:AMFTSA>2.0.CO;2, 1980.

Warren, S. G.: Optical properties of snow, Rev. Geophys., 20, 67–89, https://doi.org/10.1029/RG020i001p00067, 1982.

Warren, S. G.: Can black carbon in snow be detected by remote sensing?, J. Geophys. Res., 118, 779–786, https://doi.org/10.1029/2012JD018476, 2013.

Wendisch, M., Hellmuth, O., Ansmann, A., J. Heintzenberg, J., Engelmann, R., Althausen, D., Eichler, H., Müller, D., Hu, M., Zhang, Y., and Mao, J.: Radiative and dynamic effects of absorbing aerosol particles over the Pearl River Delta, China, Atmos. Environ., 42, 6405 – 6416, https://doi.org/10.1016/j.atmosenv.2008.02.033s, 2008.

Wendisch, M., Brückner, M., Burrows, J. P., Crewell, S., Dethloff, K., Ebell, K., Lüpkes, C., Macke, A., Notholt, J., Quaas, J., Rinke, A., and Tegen, I.: Understanding causes and effects of rapid warming in the Arctic, EOS, 98, 22–26, https://doi.org/10.1029/2017EO064803, 2017.

Wendisch, M., Macke, A., Ehrlich, A., Lüpkes, C., Mech, M., Chechin, D., Dethloff, K., Velasco, C. B., Bozem, H., Brückner, M., Clemen, H.-C., Crewell, S., Donth, T., Dupuy, R., Ebell, K., Egerer, U., Engelmann, R., Engler, C., Eppers, O., Gehrmann, M., Gong, X., Gottschalk, M., Gourbeyre, C., Griesche, H., Hartmann, J., Hartmann, M., Heinold, B., Herber, A., Herrmann, H., Heygster, G., Hoor, P., Jafariserajehlou, S., Jäkel, E., Järvinen, E., Jourdan, O., Kästner, U., Kecorius, S., Knudsen, E. M., Köllner, F., Kretzschmar, J., Lelli, L., Leroy, D., Maturilli, M., Mei, L., Mertes, S., Mioche, G., Neuber, R., Nicolaus, M., Nomokonova, T., Notholt, J., Palm, M., van Pinxteren, M., Quaas, J., Richter, P., Ruiz-Donoso, E., Schäfer, M., Schmieder, K., Schnaiter, M., Schneider, J., Schwarzenböck, A., Seifert, P., Shupe, M. D., Siebert, H., Spreen, G., Stapf, J., Stratmann, F., Vogl, T., Welti, A., Wex, H., Wiedensohler, A., Zanatta, M., and Zeppenfeld, S.: The Arctic Cloud Puzzle: Using ACLOUD/PASCAL Multiplatform Observations to Unravel the Role of Clouds and Aerosol Particles in Arctic Amplification, Bull. Am. Meteorol. Soc., 100, 841–871, https://doi.org/10.1175/BAMS-D-18-0072.1, 2019.

Wendling, P., Wendling, R., Renger, W., Covert, D., Heintzenberg, J., and Moerl, P.: Calculated radiative effects of Arctic haze during a pollution episode in Spring 1983 based on ground-based and airborne measurements, Atmospheric Environment (1967), 19, 2181 – 2193, https://doi.org/https://doi.org/10.1016/0004-6981(85)90127-1, http://www.sciencedirect.com/science/article/pii/0004698185901271, 1985.

Wiscombe, W. J. and Warren, S. G.: A Model for the Spectral Albedo of Snow. I: Pure Snow, Journal of the Atmospheric Sciences, 37, 2712–2733, https://doi.org/10.1175/1520-0469(1980)037<2712:AMFTSA>2.0.CO;2, 1980.

Wyser, K.: The Effective Radius in Ice Clouds, Journal of Climate, 11, 1793–1802, https://doi.org/10.1175/1520-0442(1998)011<1793:TERIIC>2.0.CO;2, 1998.