Co-Editor Decision: Reconsider after major revisions (23 Oct 2018) by Alma Hodzic Comments to the Author: Dear Authors,

I have not received your response to the report of the 3rd reviewer. Could you please address the reviewer's concerns:

RESPONSE: Dear Editor, you claim that you did not receive a response to the report of the 3rd reviewer; however, we never received any notification for any additional review. Please have a look at the interactive discussions of the manuscript (https://www.atmos-chem-phys-discuss.net/acp-2018-94/#discussion). I suppose that this is not our mistake. Nevertheless, we answer the question below.

This manuscript approaches a very interesting topic, increasing fire frequency in high latitude peatlands and the influence of peat fire atmospheric emissions on radiative forcing and climate change through deposition on high albedo surfaces such as snow and ice. However, I believe that the manuscript takes a totally misguided approach by focusing on BC emissions because peat fires burn almost exclusively with smoldering combustion that does not emit BC but large quantities of OC (as shown in the cited emission factors of Agaki et al., 2011 with OC emission factors being a factor of 30 larger than those of BC). Therefore, it is general knowledge that aerosol light absorption by smoke from peat fires is largely caused by BrC, not by BC. Therefore, I suggest that the authors refocus their manuscript on BrC and examine its radiative forcing and compare it to that of BC. Below, I've given a number of relevant publications to help the authors get started with this. In addition, the modification of the spectral reflectivity of snow through the deposition of peat combustion smoke has recently been discussed (Beres and Moosmuller, 2018).

RESPONSE: Although the topic of the paper is to study BC emitted from the 2017 fires in Greenland, we have corrected the whole manuscript and made several additional simulations to include OC and BrC as a sign of good will in order to finally complete this initially intended fast-track publication that was first submitted 1 year ago.

As you may notice from the results, including impact of BrC does not change a lot the impact of the emitted substances to the Greenland Ice Sheet.

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Anonymous Referee #1

Review of "Open fires in Greenland: an unusual event and its impact on the albedo of the Greenland Ice Sheet" by N. Evangeliou and co-authors. General comments.

N. Evangeliou and co-authors present a paper dealing with the atmospheric emission of black carbon by peat fires in Greenland during an extreme event in August 2017. They estimate the total amount of BC released in the atmosphere and its impact on the atmospheric radiative balance and snow albedo. The authors conclude that none of those impact are really significant. I found the paper lacking a focused scientific objective and finally it will have a limited interest for the scientific community. The methodology is sound but many of the assumptions must be clarify. The validation exercise is too qualitative while the dataset can be used for quantitative assessment. The conclusion that peat fire in Greenland could be of a significant importance for climate is not really supported by the findings of this paper.

Response: We agree that the methodology and parts of the discussion needed lots of improvement and we have made substantial effort with numerous changes in all parts of the manuscript according to both reviewers' suggestions (please see manuscript with Track Changes).

However, we do not agree with this description of our work. The validation is qualitative because no direct measurements of BC concentrations exist from this event occurring in a particularly data-sparse region, and also few satellite data document the event. The only data we found are Lidar data from CALIPSO that confirmed the presence of the plume where our model predicted it. Could the reviewer suggest, in concrete terms, which dataset could be used for quantitative assessment?

The reviewer says, "The conclusion that peat fire in Greenland could be of a significant importance for climate is not really supported by the findings of this paper." This is NOT a conclusion, but a logical probability, considering that 25% of Greenland's surface is permafrost that is rich in peat. We now show this more clearly in the updated version of our manuscript. In addition, NASA's satellites show an increasing trend of fires in thawed permafrost over Greenland (see new supplementary figure S1 or attached Fig. R1) and our simulations showed that 30% of the emissions were deposited in the Greenland Ice Sheet (Lines 388-391).

We disagree with the comment of the reviewer that this paper will have a limited interest for the scientific community. We present some statistics from the ACP Discussions website.

In the Discussions page of the journal (https://www.atmos-chem-phys-discuss.net/discussion_papers.html), at the time that we started writing this response (22-05-2018), there are 30 papers in open discussion (ACPD) that were published the same time as ours (March 2018). If we calculate the average views and downloads we get 302±100 (min: 199 – max: 570), while our paper's visibility is 293.

Furthermore, although media coverage does not converge with scientific quality, the present study was selected for a press conference on "Shape of things to come? The 2017 wildfire season" during the EGU 2018 conference (https://client.cntv.at/egu2018/pc5).

Specific comments.

Abstract

Line 43. Your conclusion doesn't support this fact and it's not scientifically based.

Response: Line 43 states "If the expected further warming of Greenland produces much larger fires in the future, this could indeed cause substantial albedo changes and thus lead to accelerated melting of the Greenland Ice Sheet." This sentence is NOT a conclusion, but a logical hypothesis (if). We have slightly rephrased the sentence, so it now reads: "If the expected future warming of the Arctic produces more fires in Greenland, this could indeed cause albedo changes and thus contribute to accelerated melting of the Greenland Ice Sheet." Finally, in order to prove that this is not pure speculation but a solid hypothesis, we support it with references (see last paragraph in conclusions).

Introduction.

The introduction is missing a comprehensive literature review on Arctic peat ecosystem and fire occurrence to better understand why those particular fires have been studied. **Response**: We have focused our introduction on peatlands and fires in Greenland and think that a more comprehensive literature review on Arctic peat ecosystems in general is out of scope of this paper. After all, this paper studies the impact of fires in Greenland on BC concentration and deposition in Greenland, not on future scenarios of fire occurrence, permafrost melt or such.

Line 83-84. Provide evidence of the significance of this event compared to other events. **Response**: Our statement that "... the fires ..., probably represent the largest fires that have occurred on Greenland in modern times.", is now supported by a new plot of the number of MODIS active fire detections (MODIS MCD14DL) over Greenland (see new supplementary Figure S1 or attached Fig.R1).

Method

L89-118. This section is very important as it is the starting point for the estimation of the BC amount released by fires. However the methodology used (eg. which sensor, when, spatial resolution, who and how has done the estimation, . . .) is unclear. On Line 241, we can read that the burnt surface area comes from GlobeCover 2009. So finally, what is your point?

Response: Line 241 has been corrected. We appreciate the reviewer for this constructive comment.

As regards to the methodology, we have done a few corrections to explain better what has been done, also giving specifications of the products we used (lines 97-99). In our opinion, detailed explanations on the calculations are not needed, since the method has been already published in the relevant literature and used in many other previous cases. As we explain in the manuscript, the burned area was mapped using severity levels of dNBR index. The methodology of its application is described in details in Lutes et al. (2006) (pp. 201-270), which is **attached**. There is another paper describing how dNBR was calibrated in field - Escuin et all, 2008 (see reference in the manuscript). Since the index is sensitive to any disturbances, we applied a manually delineated fire perimeter to increase the accuracy of mapping.

You should rewrite this section with a detailed comment of Table 1 and explain how it compares to active fire mapping. Line 118 needs clarification based on quantitative information.

Response: Line 94 explicitly says that the location of the active fires were downloaded from NASA's website. So, what is shown in Table 1 has been confirmed with NASA's active fires (also shown in supplements' Figure S1 and attached Fig.R1). Regarding to the severity levels (Line 118), qualitative information is given in Key and Benson (2006) together with all the details of the methodology used. The same methodology has been used to map the Chernobyl fires (see: Evangeliou et al., 2014; 2015; 2016)

The comment to confirm Line 118 based on quantitative information is too generic and we do not really understand what the reviewer wants us to do.

L155 Explain how you get this number and provide a range of possible values **Response**: Line 155 says "In contrast, tropical peatlands can have deep burn depths of 40-50 cm and release an average of 300-450 t C ha⁻¹ (Page et al., 2015; Reddy et al., 2015)."

It should be obvious that this range of values was reported by Page et al. (2015) and Reddy et al. (2015).

If the reviewer means the average amount of organic fuel available for combustion that we used for the Greenland fires (100 t C ha^{-1}), it has been taken from Smirnov et al. (2015). In this paper, it was assumed that for peat-bog fires, the average amount of fuel available for combustion (including the soil organic matter) is up to 120 t/ha supported from measurements from IPCC (2006).

L180. Provide reference for BC density and size distribution. Peat fires emits large amount of organic carbon. The possible impact of the mixing state of BC and POM on aerosol size distribution, optical properties and residence time should be discuss in this paper.

Response: We agree that fires also emit large quantities of organic carbon (OC). However, the impact of OC on the albedo of the ice sheet is probably small, although it probably enhances the BC effect, since OC can also be slightly absorbing (e.g., brown carbon). But given the lack of information on the optical properties of the emitted OC, we think an additional analysis of OC would not be very meaningful.

With respect to BC density and size distribution, a reference was added in Line 214 of the updated manuscript. We have now also performed a sensitivity study on the impact of different particle size distributions on the deposition of BC over Greenland's Ice Sheet and discuss it in section 3.2. A detailed analysis of residence times of BC has been already presented by Grythe et al. (2017) [reference in the manuscript] and in Evangeliou et al. (2018) [reference under editorial check in ACP Discussions].

L200 and discussion section 3.3

The apportionment between emission from peat fires and other sources remains unclear for me. The methodology is not same as the one use for assessing impact of peat fire. The figure 4 is not really useful while other figures are in the supplement material. **Response**: Lagrangian models such as the one used in our work (FLEXPARTv10) can run forward in time (like CTMs or climate models) using specific emissions that can be taken from an existing inventory (for example ECLIPSE, see:

http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html). Moreover, Lagrangian models have the advantage that they can also run backward in time, from a specific point or region for which the user wants to calculate concentrations. What is produced then is the footprint emission sensitivity (or footprint), which is simply the residence time of the computational particles (in sec) in each grid-cell of the model. Then, by multiplying this footprint with a given emission inventory (e.g. ECLIPSE) given in kg/m²/s and dividing with the altitude of the lowest vertical level in the model, one obtains surface concentrations again. Notice that forward and backward calculations are equivalent, so the methodology is not different. However, depending on the setup, the computational efficiency can be much higher in backward mode, and that is also the reason we used it to assess the impact of emissions outside Greenland.

For FLEXPART that we used in this study, a comparison between forward and backward simulations can be found in Seibert and Frank (2004).

We calculate average concentrations of surface BC in four compartments of Greenland based on ECLIPSE emissions. ECLIPSE includes all anthropogenic sources, while we calculate biomass burning emissions using global MODIS-satellite hot spot data (Giglio et al., 2016) and GFAS (references in the manuscript). Everything is well documented in the associated references.

Figure 4 has been replaced by Figure S4 as suggested.

L204 and section 4.2

The methodology and the discussion section on RF computation must be improved and clearly states how you deal with both surface albedo and atmospheric effect of BC on the radiative balance. Figure 7 is confusing as it deals with both BOA, TOA, time series and geographical distribution as the same time.

Response: We have re-written and re-structured the whole chapter, both in the Methodology and analysis of the Results (see manuscript with Truck Changes). Our perception is not to present in detail methods that have been documented in previous publications. For RF calculations we used the uvspec model from the libRadtran radiative transfer software package (http://www.libradtran.org/doku.php) (see references in the manuscript: Emde et al., 2016; Mayer and Kylling, 2005). Snow albedo was calculated with the SNICAR model (http://snow.engin.umich.edu/info.html) in a two-layer configuration (see references in the manuscript: Flanner et al., 2007, 2009). These are open source codes that have been used by many groups worldwide. Figure 7 has been improved as suggested.

L218 and section 4.1 along with Figures 5 and 6. The validation exercise is really too qualitative and based on visual inspection of satellite data that are not really used scientifically. AERONET data can provide detailed information on aerosol optical properties and radiative forcing. CALIOP data products give aerosol extinction profiles which can be used in the RF computations.

Response: The reason the validation exercise is so qualitative is that we have no clear observations of the Greenland fire plume. The AERONET data show impacts of the forest fires burning outside Greenland. Only at one site, the AERONET data show an AOD increase that is partly (but not exclusively) due to the Greenland fires.

L466 Your last bullet point is rather speculative and not supported by the findings of the paper.

Response: This is true and we have now corrected it. The last bullet is NOT ALL OF IT a conclusion, but rather a comment and therefore, we now show it as a comment (not bulleted) below the bullet. We further support what we say in the sentence with references.

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Anonymous Referee #2

General comments:

This work investigates the quantification of emissions of black carbon (BC) from intense fires on peat lands in Western Greenland during summer 2017 and their impacts on albedo reduction and radiative forcing. The authors conclude that those impacts of BC deposition of the Greenland Ice Sheet are almost negligible, which turns out to be a scientific result for the community. This study is interesting and sound for ACP. I have nevertheless several criticisms requiring a careful and revision and in-depth improvements both in the methodology, often unclear, and in the discussion of the results before the paper is suitable for publication in ACP.

Response: We acknowledge the reviewer's comments and his effort to improve this manuscript. We have tried to follow his suggestions to correct the manuscript and have basically re-written parts of the manuscript (please see manuscript with Track Changes).

Specific comments:

L1-2: The title seems to indicate that the main focus of the paper is the quantification of the reduction in albedo due to open fires in Greenland. Only ten lines in the paper really focus on the modification of the albedo due to BC deposition. The title should reflect the main findings of the paper: quantification of BC emissions of this unusual event, transport of the plume, deposition.

Response: We agree; we have changed the title to "Open fires in Greenland in summer 2017: transport and deposition of BC and impact on the Greenland Ice Sheet"

L41-44 and L496-500: I find a bit strange to conclude both abstract and conclusion by something purely speculative and that does not match the main results of the paper. **Response**: We admit that this is probably an extreme formulation and we have changed it to a weaker statement. We would like to draw attention that this statement is not a conclusion, but a logical hypothesis. To further show that it's not a conclusion, we now support the paragraph with references (see last paragraph in conclusions).

L83-84: "the largest fires". Give maybe statistics or cite a climatological study to support this assertion.

Response: We have plotted the annual number of active fires from NASA's MODIS product in supplements' Figure S1 (or Fig.R1) starting from first year that satellite data were available (2001).

L111 : The authors should give more details about the procedure applied on the data. "Additional classification" is too vague.

Response: The statement has been removed!

L130 : "assuming a 6h persistence". How is this hypothesis justified ? Is it confirmed b observations or by other studies ?

Response: Well, this is confirmed by previous studies (Kaiser et al., 2012 – reference in the paper). We chose a persistence model similar to what is done in Kaiser et al. (2012) and used a time of the same order of magnitude with the mean return time of MODIS in the afternoon (peak time of fire) \sim 4h. For a description of the persistence model that was used, please see line 8 - page 9852 of Paugam et al. (2015).

L161: Say clearly that the only variable computed in this study from measurements is the burned area A. The other factors are based on assumptions or provided by previous studies.

Response: Corrected. This is now explicitly mentioned in Line 184.

L181: Those values suggest that aerosols are not only composed of BC (which is a reasonable assumption). How do the authors justify this size distribution? It has indeed a huge influence on the deposition efficiencies (both sedimentation and wet removal) and on the calculation of aerosol optical properties. Both the radiative forcings and reduction of albedo on snow surfaces will be sensitive to this assumption on the size distribution. I suggest that the authors perform a sensitivity study on the influence of those parameters.

Response: After rapid coagulation, more than 90% of the mass of BC after fires is present in sizes between 0.1 – 1 μm in the atmosphere. This has been highlighted by many experiments/measurements and is now well justified in section 3.2.

However, we have followed the suggestion of the current reviewer and performed a sensitivity study using different size distribution of the BC particles produced from the 2017 fires in Greenland and we calculated the uncertainty on the deposited mass of BC due to different size distribution. We present and discuss the results at the end of section 3.2. The effect of different size distribution on residence times has been already studied by Grythe et al. (2017) [reference in the manuscript] and the different deposition coefficients in Evangeliou et al. (2018) [reference under editorial check in ACP Discussions].

The calculated uncertainty from this sensitivity test ranges from 10%–30% in 86% of the Sheet's surface to up to 50% in the rest of the Sheet's surface.

L200 : "a simple emission scheme". What does it mean? Why don't the authors use the same methodology for all fires?

Response: We appreciate reviewer's comment here. This was a typo error and we have updated this part of the methodology.

L200-201: Those emission factors should depend on the type of soil and vegetation. Which maps have been used here? Which values for emission factors have been finally chosen? The reader should be able to reproduce the results of this study; without such assumptions, it is impossible.

Response: Corrected; See previous comment.

Sect. 2.4: Do the authors calculate radiative forcing assuming refractive index of BC only? The choice of the refractive index should be done in accordance with the size distribution (L181), which probably reflects an internal mixture of aerosols.

Response: The radiative forcing was calculated using the refractive index of BC only. We agree with the reviewer that BC was likely present as an internal mixture with other aerosol components (especially OC). However, we did not simulate OC and therefore used only the refractive index of BC. This will lead to an underestimation of the atmospheric effects of BC, since internal mixing with OC will likely enhance the BC absorption, and there may also be other absorbing components in the aerosol. However, we think that as an order of magnitude estimation of the atmospheric effects, our

assumption should be sufficient. Furthermore, the more important impact of BC on the albedo is less (or not) sensitive to the mixing state of the aerosols.

L226: "we display": where?

Response: We substituted 'display' with 'used'.

L292: "a small portion of the emitted BC". Please quantify it.

Response: We have quantified the portion that lifted up in this particular day (\approx 516 kg).

L334: "due to the generally dry weather when the fires were burning". It can be also ascribed to the fact that dry deposition mostly occurs in the quasi-laminar sublayer close to the surface. Aerosols are quickly deposited close to the sources before being injected at higher altitudes and being transported away from sources.

Response: Thanks for this comment. We have included it in the manuscript.

L365: "the anthropogenic contribution is larger". For the sake of clarity, the authors might write that the anthropogenic is relatively larger in Southern Greenland in contrast to Northern Greenland but remains lower than the biomass burning contribution.

Response: Comment was added to the manuscript.

L367: "the BC concentrations that are calculated here for the studied fire period are relatively high compared to those reported previously". I am not sure this is always true. The authors should also quote more recent studies, e.g. Polashenski et al. (2015), Legrand et al. (2016) or Thomas et al. (2017), who have reported higher events of biomass burning BC deposition over Greenland. If the BC deposited on snow/ice surfaces is much larger in those studies, it also suggests higher surface BC concentrations

Response: We thank the reviewer for providing the references and have added them to the previous section. Please see Line 425-426 for Polashenski et al (2015) and Legrand et al. papers.

However, the Thomas et al. paper is using another unit (g/m2) and without knowing the density of the samples no conversion to ng/g (units used in the present) can be applied.

L378 and L389: "dosages". Do you mean concentrations / mixing ratios?

Response: They are dosages of concentrations. It is now explained in the last paragraph of section 3.3 and in the caption of the respective Figure.

L397-398: BC particles are probably not the main contributors to AOD in this region for two reasons: the BC loadings are rather low in comparison to other aerosol compounds and the diameter of BC-containing particles is much smaller than the wave-length (0.5 um). A better proxy of the temporal evolution of the integrated BC would be the absorbing AOD (AAOD), which is also often provided at AERONET stations. The AAOD/AOD would be also a good indicator of the contribution of BC to the total AOD (even if BC is not the only absorbing component). This should be shown on Fig. 5.

Response: The reviewer has a very good point here and we tried to retrieve AAOD data as he suggested.

Though in Kangerlussuaq and Thule no AAOD Level 2 data are available for July-September 2017, while in Narsarsuaq AAOD Level 2 data are available for 2 September 2017 (when the fires had been already extinguished).

In Andrews et al. (2017) paper is stated that "One obvious limitation of the AERONET inversion retrievals is that the uncertainty of the derived SSA becomes very large at low values of AOD (Dubovik et al., 2000). To minimize the effects of this uncertainty, the AERONET Level 2 data invalidate all absorption-related values if the AOD at wavelength 440 nm (AOD440) is below 0.4 (Dubovik et al., 2000, 2002; Holben et al., 2006)." In page 6043 of the same paper it is stated that "It should be noted that AERONET does not recommend the use of absorption-related parameters (e.g., SSA, AAOD and complex index of refraction) at AOD440 below 0.4." In our case, except for the characteristic peak of AOD that is attributed to the N. American fires, all the other AOD values were below 0.4.

Sometimes researchers use AAOD LEV 1.5 data, but these get high uncertainty (see Andrews et al. 2017). In page 6051 of the Andrews et al. (2017) paper is also stated that "Using the sum-of-squares propagation of errors to calculate the uncertainty in AAOD for both high and low AAOD cases results in an AAOD uncertainty of approximately 0.015 for both high- and low-AOD cases ... An AAOD uncertainty value of 0.015 suggests an uncertainty of about 60% in AAOD for AOD440 D0.5 and more than 140% uncertainty in AAOD for AOD440 < 0.2."

Therefore, we do not think that these uncertain LEV1.5 AAOD measurements should be plotted instead of AOD here. However, if the reviewer or the editor disagree, we have retrieved them and we could use them in a next step. Besides, we only used AOD as an indicator for the presence of the plume, and for that purpose it should be sufficient.

L401-407: How do the authors explain the significant AOD enhancement at the beginning of September observed at Narsarsuag station?

Response: As the reviewer can see, in the attached **Fig.R2** we present the biomass burning BC from GFAS (upper panel) in the beginning of September and the footprint emission sensitivity from the Narsarsuaq station (bottom panel) on September 3rd. We observe that the highest footprint emission sensitivity is located exactly at the place where GFAS emissions are the highest (Canada). Therefore, we have a clear indication that the increase that the reviewer mentioned is due to the Canadian wildfires.

L422: "was not studied". Does it mean that the transport of those North American fire plumes was not correctly captured by FLEXPART? It is indeed impossible to see on Fig. 6d as the vertical scale is not appropriate.

Response: Here, we wanted to state that the existence of the N. American fires in the attenuated backscatter measurements that we get from CALIOP was not further studied. The study of the N. American fires is beyond the scope of this paper. We have used a better formulation in the manuscript now.

Sect. 4.2: The authors should remind that they calculated only the forcing due to the Greenland fires, which is itself small compared to the North American or Eurasian fires. It should also be said explicitly that the calculated radiative forcing values does not include semi-direct nor indirect effects, which may be dominant here.

Response: We have rewritten the first part of the section to include the information requested by the referee.

L436: "cloudless conditions". I do not understand the purpose of this. It is only an ideal simulation, which is not commented in the paper afterwards. What does it bring to the discussion?

Response: The IRF for cloudless conditions is compared against IRF including clouds in the subsequent lines. IRF for cloudless conditions was included, as they show the potential maximum effect of the forcing. The results presented show that the clouds reduce both the TOA and BOA IRF.

L440-442: It is not clear if the given values refer to the total radiative forcing of BC. What are the relative contributions of the direct radiative forcing of BC and of the radiative forcing of BC deposited on snow surfaces? The authors also give the values without any uncertainty, but a lot of assumptions have been done to retrieve the BC emissions, the BC size distribution, the BC optical properties. Each of those hypothesis would lead to a range of values of IRF.

Response: The given values refer to the total instantaneous radiative forcing, that is including both the effect of atmospheric BC and BC deposited on the snow. The latter dominates the IRF contributing between 85 to 99 % depending on BC amount. This has been clarified in the manuscript.

The composition of the BC from the fire is not known. Hence average BC optical properties were adopted. We have subsequently performed an uncertainty analysis using realistic variations in BC optical properties. This uncertainty analysis is included in the supplementary material and referred to in the manuscript.

We have also performed a sensitivity study and estimated the uncertainty of the BC deposition over Greenland due to the use of different size distribution of BC particles (see answer to previous comments).

L 442: "Fig 7c depicts the temporal behaviour..." Does it represent calculations in cloudy conditions?

Response: It is the cloudy conditions that are shown. This information has been added both in the text and the figure caption. The temporal behaviour is shown in Fig 7d. This typo has been corrected as well.

L443-444: I don't see how this information (blue line) can be useful. The location of the pixel where the maximum IRF is found likely varies with time. Besides the analysis of this figure is not done in text. I recommend to remove it.

Response: We have removed the blue line from the plot. The idea of plotting the TOA max IRF was to show that the single pixel maximum and area averaged RFs peak at different times. However, we agree with the reviewer that this information was perhaps not so useful.

L448-455: If the authors want to be able to compare their results to global studies, as it is done here, they need to multiply the value of RF by the area of the simulation domain to obtain a forcing value in watts, and then divide it by the surface area of the Earth to obtain an equivalent global radiative effect in mW/m2 that could be compared to results for global studies.

Response: The cited value from Skeie et al. (2011) is not a global value, but a value representative for the Greenland ice sheet (Fig 17 of Skeie et al., 2011). It is this value we are comparing against. The values from Myhre et al (2013) are included in order to give the reader a global value to compare against. This has been clarified in the text. It is clear that, on a global scale, the obtained RF values are negligible.

L453-455: What about the impact of North American and Eurasian fires, whose plumes reach Greenland during the studied period?

Response: These plumes are not the focus of the present study. Similar plumes have been studied before, so we don't think focusing on these plumes would provide a lot of new information beyond what has been published before. More technically, we only estimate the impact of these plumes using backward calculations, whereas RF calculations would require forward calculations. We think this is out of scope of the present paper.

What we have done, instead, is to calculate the impact of the N. American fires in the surface concentrations of BC over Greenland (see section 3.3). This proved that the BC concentrations from the N. American fires in August 2017 are more than 1 order of magnitude higher compared with those produced from the Greenland fires of August 2017 (see updated Figure 4).

L456-457: What is the albedo reduction due to BC deposition that can be ascribed to Greenland fires / to fires outside Greenland / to anthropogenic sources? If the goal of the paper is indeed to focus on the impact of the Greenland fires, quantifying this effect and comparing it to the relative contribution of the different sources would be really valuable for the paper. The authors should also compare their albedo reduction values to previous studies, e.g. Polashenski et al. (2015).

Response: We now compare our results to those of Polashenski et al. (2015) in section 4.2. The detailed study of the fires outside Greenland and from anthropogenic sources is beyond the scope of this paper. Notice that the albedo effects can't be done on the basis of the backward calculations done for the other sources and would require totally new forward simulations. However, giving the range of surface BC concentrations over Greenland (section 3.3 and Figure 4) is already enough to conclude that the event that we studied in the current paper has minor effects on the albedo or RF compared to BC from the N. American fires or from anthropogenic sources simply because of the different magnitude and duration of these fires.

Sect. 5: The conclusions may be more quantitative.

For example : L478-479 : the ratio of BC deposition from the different sources can be given

Response: We have not quantified how big the deposition from anthropogenic and biomass burning sources is, and we have removed this sentence from the manuscript.

L481-483: the AOD enhancement can be precised

Response: Corrected.

L488: "albedo change due to the BC deposition". Which sources have been considered? **Response**: Corrected. It is the albedo change due to BC deposition from the Greenland fire of 2017.

L496-500 : Remove this purely speculative sentence. The opposite could also be said, given the findings of the paper.

Response: These lines state that "The very large fraction of the BC emissions deposited on the Greenland Ice Sheet (30% of the emissions) makes these fires very efficient climate forcers on a per unit emission basis. If the expected future warming of the Arctic (IPCC, 2013) produces more fires in Greenland in the future (Keegan et al., 2014), this

could indeed cause substantial albedo changes and thus contribute to accelerated melting of the Greenland Ice Sheet."

We do not understand why this is speculative. A fraction of 30% deposition on the Greenland ice sheet is substantial, much higher than from any other source type and source region, so – on a per unit mass basis – the forcing due to albedo change is efficient, even if it is small overall. The second sentence can perhaps be considered somewhat speculative, but we have now reformulated it and, moreover, we support it with references. Furthermore, it is not presented as a conclusion, so it should be very clear to the reader to what extent this sentence is speculative. We nevertheless consider it important enough to keep it.

The choice of the figures kept in the manuscript is rather strange. Most useful figures relevant for the discussion have been displaced to the Supplementary Material. I recommend to move them to the main paper.

Response: We have moved the figure with the calculated dosages to the manuscript (Fig. 4), as the dosages are discussed more in the text. We have now placed back to the supplements (Fig. S5) the figure of the footprint emission sensitivities that are not the main focus of the paper. We are willing to put more figures in the manuscript in a next step, if the reviewer point into this direction. The only reason for using limited number of figures was that this paper was intended to be short.

Fig. 2a: Are those values averaged over the simulation domain? over Greenland? I had hard time to figure out how those values could be realistic. I think there is either a issue with the unit or a mistake in the calculation. Shouldn't it be ng/m3 or ng/kg instead of ug/m3? The total concentrations of BC in the domain should be calculated as the volume average of the grid cell concentrations, not the sum over all grid cells in the domain...

Response: We thank the reviewer for this comment that we have now corrected. We now present the average vertical concentrations over Greenland from the 2017 fires in pg/m3 in the updated Figure 2.

Fig 2b: Here again, there is an issue with the unit. The color bar indicates ug/m2 (which is probably right), but the caption says ng/m2. Which one is correct?

Response: We also appreciate reviewer's help to correct this mistake. The error was in the legend and it has now been updated.

Fig. 4: It is extremely difficult to see the colored grid cells an read their values. Please improve the quality of this figure.

Response: Quality of the figures has been set to 300 dpi. This should solve the problem.

Fig. 5 : Does the altitude represent agl or amsl? The orography in Greenland is not flat. **Response**: It is agl altitude and we now clarify it in the legend.

Fig 5: Why do you keep the contribution of fires burning outside Greenland but exclude the BC contribution of anthropogenic sources? According to Fig. 4, their contribution is absolutely not negligible and they might modify the time series of column-integrated BC in Greenland.

Response: We thank the reviewer for this comment. We tried to put also anthropogenic BC in these time-series. Column-ntegrated anthropogenic BC is very low and a stacked

line does not show anything in the time-series and that's the reason that we decided not to present it. We have added a small comment in the legend.

Fig. 6: it would be better to use the same scale for longitude and altitude on panels (b) and (d).

Response: The reason that we did not use the same scale for longitude and altitude in these two figures is due to the small aerosol structure at high altitudes seen in the CALIOP data. We thought that this is likely due to the N. American fires that were burning at the same time with the Greenland ones. This is visible from the AOD measurements at many of the Greenland stations where large increases in AOD were observed.

In a previous comment for the AOD increase in the Narsarsuaq station at the beginning of September, we provided relevant footprint emission sensitivities and biomass burning emissions from CAMS_GFAS (see Figure R2). They explicitly show that the largest footprint was found in Canada in areas with large biomass burning emissions. However, since we do not study the impact of the N. American fires in detail, the sentences about the presence of N. American fire plumes at high altitudes in section 4.1 are rather speculative and we have removed them. We have also corrected Figure 6, as the reviewer suggested and for this, we acknowledge him.

Fig. 7c: Is the snow albedo reduction plotted for 31 August or for the full period? **Response**: The snow albedo reduction due to BC deposition from the beginning of the fires until 31 August is plotted in Figure 7c (please see last paragraph of section 4). Legend has also been updated.

Table 1: This table is not commented nor anlyzed in text. We can notice changes in the sources of RS data at different periods, which should be detailed in the methodology section.

Response: In line 105 we state that we used different RS data to better delineate fire perimeters and define burn severity. Which day each RS tool was used is shown by pointing to Table 1. In addition, discussion of the results presented in Table 1 is presented in section 3.1 and 3.2.

Legrand, M., et al. (2016), Boreal in Are records in Northern Hemisphere ice cores: A review, Clim. Past, 12(10), 2033–2059.

Polashenski, C. M., J. E. Dibb, M. G. Flanner, J. Y. Chen, Z. R. Courville, A. M. Lai, J. J. Schauer, M. M. Shafer, and M. Bergin (2015), Neither dust nor black carbon causing apparent albedo decline in Greenland's dry snow zone: Implications for MODIS C5 surfacereïn C'ectance, Geophys. Res. Lett., 42,9319–9327, doi:10.1002/2015GL065912. Thomas, J. L., et al. (2017), Quantifying black carbon deposition over the Green-land ice sheet from forest fires in Canada, Geophys. Res. Lett., 44, 7965–7974, doi:10.1002/2017GL073701.

Technical comments:

L350: "adopted". Do you mean "adapted"?

Response: In this sentence we think that "adopted" fits better. We just used active fires from MODIS; we did not adapt anything.

L394: Replace "for validating" by "to validate".

Response: Corrected.

L485: Replace "attenuation" by "attenuated"

Response: Corrected.

L512: Please write "Brent Holben" in two words.

Response: Corrected.

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- Holben, B. N., Eck, T. F., Slutsker, I., Smirnov, A., Sinyuk, A., Schafer, J., Giles, D., and Dubovik O.: AERONET's Version 2.0 quality assurance criteria, http://aeronet.gsfc.nasa.gov/new_web/Documents/AERONETcriteria_final1.pdf, 2006.
- Andrews, E., Ogren, J. A., Kinne, S., and Samset, B.: Comparison of AOD, AAOD and column single scattering albedo from AERONET retrievals and in situ profiling measurements, Atmos. Chem. Phys., 17, 6041-6072, https://doi.org/10.5194/acp-17-6041-2017, 2017.
- Dubovik, O., Smirnov, A., Holben, B. N., King, M. D., Kaufman, Y. J., Eck, T. F., and Slutsker, I.: Accuracy assessment of aerosol optical properties retrieval from AERONET sun and sky radiance measurements, J. Geophys. Res., 105, 9791–9806, 2000.
- Paugam, R., Wooster, M., Atherton, J., Freitas, S. R., Schultz, M. G., and Kaiser, J. W.: Development and optimization of a wildfire plume rise model based on remote sensing data inputs Part 2, Atmos. Chem. Phys. Discuss., 15, 9815-9895, https://doi.org/10.5194/acpd-15-9815-2015, 2015.

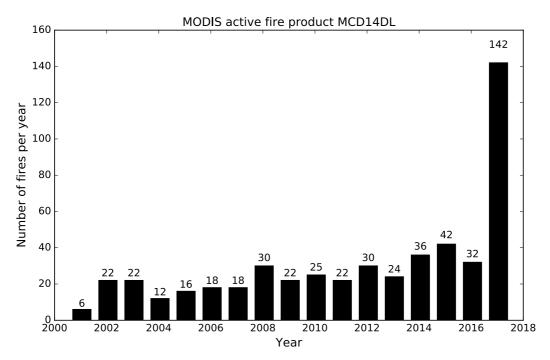


Fig. R 1. Annual number of active fires over Greenland during the last 17 years as seen from NASA's MODIS satellite (product MSC14DL).

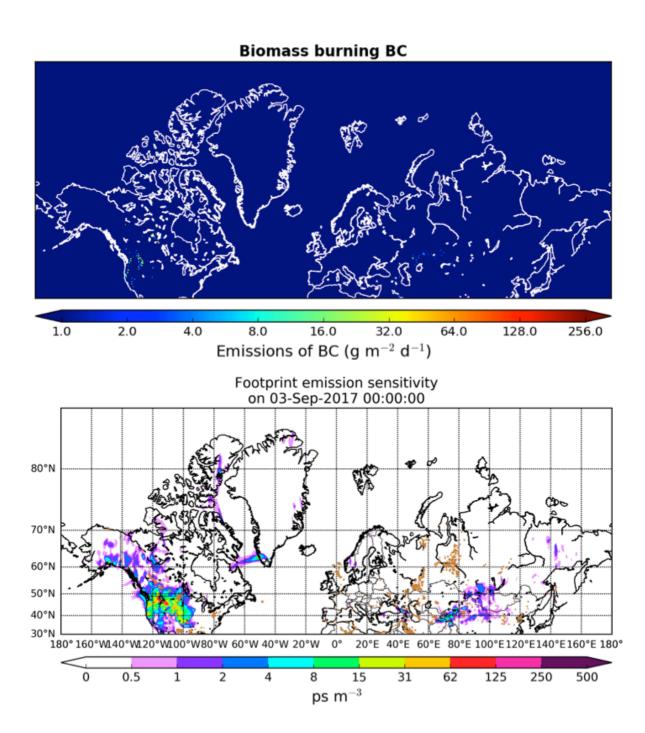


Fig. R 2. Biomass burning emissions of BC from GFAS (upper panel) in the beginning of September and the footprint emission sensitivity from the Narsarsuaq station (bottom panel) on September 3rd. The highest emission probability fits exactly to the place where the highest emissions occurred.

Open fires in Greenland in summer 2017: transport, deposition and radiative effects of BC, OC and BrC emissions,

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Nikolaos Evangeliou^{1,*}, Arve Kylling¹, Sabine Eckhardt¹, Viktor Myroniuk², 5 Kerstin Stebel¹, Ronan Paugam³, Sergiy Zibtsev², Andreas Stohl¹ 6

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- 9 Research (ATMOS), Kjeller, Norway.
- ²National University of Life and Environmental Sciences of Ukraine, Kiev, Ukraine. 10
- ³King's College London, London, United Kingdom. 11

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Abstract

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Highly unusual open fires burned in Western Greenland between 31 July and 21 August 2017, after a period of warm, dry and sunny weather. The fires burned on peat lands that became vulnerable to fires by permafrost thawing. We used several satellite data sets to estimate that the total area burned was about 2345 hectares. Based on assumptions of typical burn depths and emission factors for peat fires, we estimate that the fires consumed a fuel amount of about 117 kt C and emitted about 23.5 t of black carbon (BC) and 731 t of organic carbon (OC) including 141 t of brown carbon (BrC). We used a Lagrangian particle dispersion model to simulate the atmospheric transport and deposition of these species. We find that the smoke plumes were often pushed towards the Greenland Ice Sheet by westerly winds and thus a large fraction of the emissions (30%) was deposited on snow or ice covered surfaces. The calculated deposition was small compared to the deposition from global sources, but not entirely negligible. Analysis of aerosol optical depth data from three sites in Western Greenland in August 2017 showed strong influence of forest fire plumes from Canada, but little impact of the Greenland fires. Nevertheless, CALIOP lidar data showed that our model captured the presence and structure of the plume from the Greenland fires. The albedo changes and instantaneous surface radiative forcing in Greenland due to the fire emissions were estimated with the SNICAR model and the uvspec model from the libRadtran radiative transfer software package. We estimate that the maximum albedo change due to the BC and BrC deposition was about 0,007, too small to be measured. The average instantaneous surface radiative forcing over Greenland at noon on 31 August was 0.03-0.04 W m⁻², with locally occurring maxima of 0.63-0.77 W m⁻² (depending on the studied scenario). The average value is up to an order of magnitude smaller than the radiative forcing from other sources. Overall, the fires burning in Greenland in summer of 2017 had little impact on the Greenland Ice Sheet, causing a small extra radiative forcing. This was due to the – in a global context – still rather small size of the fires. However, the very large fraction of the emissions deposited on the Greenland Ice Sheet makes these fires very efficient climate forcers on a per unit emission basis. If the expected future warming of the Arctic produces more severe fires in Greenland, this could indeed cause albedo changes and thus contribute to accelerated melting of the Greenland Ice Sheet. The fires burning in 2017 may be a harbinger of such future events.

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1 Introduction

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In August 2017 public media reported unprecedented fire events in Western Greenland (BBC News, 2017; New Scientist Magazine, 2017). These events were documented with airborne photographs (SERMITSIAQ, 2017) and satellite images (NASA, 2017b) and raised public concerns about the effects of climate change and possible impacts of soot emissions on ice melting. Historically, wildfires have occurred infrequently on Greenland, because threequarters of the island is covered by a permanent ice sheet and permafrost is found on most of the ice-free land (Abdalati and Steffen, 2001). Permafrost, or permanently frozen soil, lies under a several meters thick "active" soil layer that thaws seasonally. But in certain areas, where the permafrost layer starts melting, it can expose peat, a material consisting of only partially decomposed vegetation that forms in wetlands over the course of hundreds of years or longer. Peatlands, also known as bogs and moors, are the earliest stage in the formation of coal. Globally, the amount of carbon stored in peat exceeds that stored in vegetation and is similar in size to the current atmospheric carbon pool (Turetsky et al., 2014). When peatlands dry, they are often affected by fires burning into the peat layers. Peat fires are difficult to extinguish and they often burn until all the organic matter is consumed. Smoldering peat fires already are the largest fires on Earth in terms of their carbon footprint (Turetsky et al., 2014). For Greenland, it has been suggested that degradation of peat will accelerate towards 2080 (Daanen et al., 2011) and that the area affected by the fires in August 2017 is particularly vulnerable to permafrost thawing (Daanen et al., 2011).

Fires in the high northern latitudes release significant amounts of CO₂, CH₄, N₂O₂, black carbon (BC), and organic carbon (OC) and their emissions are often transported into Arctic regions (Cofer III et al., 1991; Hao et al., 2016; Hao and Ward, 1993; Shi et al., 2015). While BC is the most strongly light-absorbing component of the atmospheric aerosol (Bond et al., 2013), a portion of OC compounds has shown strong absorption towards shorter wavelengths of the electromagnetic spectrum (UV), therefore defined as brown carbon (BrC) (Andreae and Gelencsér, 2006; Chakrabarty et al., 2010). BC is formed by the incomplete combustion of fossil fuels, biofuels, and biomass (Bond et al., 2013). BrC is emitted from smoldering fires or solid fuel combustion (Bond, 2001), from pyrolysis of biomass (Mukai and Ambe, 1986) and from biogenic emissions of humic substances (Limbeck et al., 2003). Due to their particulate nature, both BC and OC are important for human health (Lelieveld et al., 2015) and climate impacts (Myhre et al., 2013), BC has an atmospheric lifetime of 3–11 days (Bond et al., 2013), while BrC lifetimes are estimated at 5–7 days (Jo et al., 2016), thus facilitating

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transport over long distances (Forster et al., 2001; Stohl et al., 2006). BC, OC and BrC from mid-latitude sources can thus reach remote areas such as the Arctic. They absorb, solar radiation in the atmosphere (Feng et al., 2013; Hansen and Nazarenko, 2004), have, a significant impact on cloud formation and also decrease surface albedo when deposited on ice and snow and can accelerate melting processes (Hansen and Nazarenko, 2004; Wu et al., 2016). This raises particular concerns about the effect of fires burning in the immediate vicinity of the Greenland Ice Sheet. If a large fraction of the BC emitted by such fires is deposited on the ice, these fires may be extremely effective in further enhancing the already accelerating melting of the Greenland Ice Sheet (AMAP, 2017). BC, OC and BrC emissions from such high latitude fires may also have a substantial effect on the albedo of sea ice.

Here we study transport and deposition of BC, OC and BrC over the Greenland Ice Sheet from the fires that occurred in Western Greenland in August 2017, which <u>likely</u> represent the largest fires that have occurred on Greenland in modern times (Figure S 1). Since the fires occurred in an area entirely lacking ground-based observations, we use satellite data and a Lagrangian atmospheric dispersion model for our study. Finally, we evaluate the changes in the albedo of the Greenland Ice Sheet from the respective deposition of BC and BrC and present instantaneous radiative forcing calculations for these two atmospheric constituents released from the 2007 fires in Greenland.

2 Methods

2.1 Definition of burned area

Remote sensing has been useful for delineating fire perimeters, characterizing burn severity and planning post-fire restoration activities in different regions. The use of satellite imaging is particularly important for fire monitoring in remote areas due to difficult ground access. The method that is presented in this section has been already used to calculate burned area in the highly-contaminated radioactive forests of Chernobyl (Evangeliou et al., 2014, 2015, 2016). Coordinates of fire locations (hot spots) were downloaded from FIRMS (Fire Information for Resource Management System) (NASA, 2017a). For the mapping of the burned area, Sentinel 2A images were used. To delineate fire perimeters and define burn severity precisely, we used Landsat 8 Operational Land Imager (OLI) (resolution: 30×30 m) together with Sentinel 1A (resolution: 30×30 m) and Sentinel 2A images (resolution: 30×30 m) (see Table 1) by applying the differenced Normalized Burn Ratio (dNBR) (Key and Benson, 2006):

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 $dNBR = NBR_{pre-fire} - NBR_{post-fire}$ (Eq. 1)

Normalized burn ratios for pre- $(NBR_{pre-fire})$ and postfire $(NBR_{post-fire})$ images from Sentinel 2A can be calculated using radiances for near- and shortwave infrared bands (bands 8

178 (NIR) and 12 (SWIR2) at 0.835 μm and 2.202 μm, respectively):

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$$NBR = \frac{1000 \cdot (NIR - SWIR2)}{NIR + SWIR2}$$
 (Eq. 2)

The methodology of applying a dNBR index to assess the impact of fires has been used in forests of the Northern and Western USA (French et al., 2008; Key and Benson, 2006) and elsewhere (Escuin et al., 2008; Sunderman and Weisberg, 2011).

The burned severity mosaics were created using Sentinel 2A images corrected for atmospheric scattering (see Chavez, 1988). Pre– and post–fire images were used to create cloudless mosaics for the area where the Greenland fires burned. A Maximum Value Composite (MVC) procedure (Holben, 1986) was used to select pixels from each band that were not cloud covered and have a high value of Normalized Difference Vegetation Index (NDVI). To avoid spurious burn severity values, manually delineated fire perimeters were applied and all areas outside were classified as unburned. We have used common dNBR severity levels (Key and Benson, 2006) that are presented in Figure 1, The occasionally dense cloud cover was the main obstacle in reconstructing fire dynamics. As an independent source of information, active fires from MODIS satellite product MCD14DL (Giglio et al., 2003) are plotted in Supplemental Information (SI) Figure S 2.

2.2 Injection altitudes, assumptions on biomass consumption and emissions factors

Injection heights into the atmosphere of the emitted smoke were simulated with version 2 of the Plume Rise Model (PRM) (Paugam et al., 2015) which is implemented in the Global Fire Assimilation System (GFAS) emission inventory (Rémy et al., 2017). The model (hereafter referred to as PRMv2) is a further development of PRM (Freitas et al., 2006, 2010) and has already been used in previous studies of fire events (Evangeliou et al., 2015, 2016). The model simulates a profile of smoke detrainment for every single fire, from which two metrics are extracted: (i) a detrainment layer (i.e. where the detrainment rate is > 50% of its global maximum) and (ii) an injection height (InjH, the top of the detrainment layer). Instead of using the GFAS product, which uses the same statistics as in the PRMv2 InjH calculation, we ran the model for every detected fire assuming a 6 h persistence and using the same conversion factor as Kaiser et al. (2012) to estimate the biomass consumption. PRMv2 mass

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detrainment profiles are then time integrated and extracted at 1°×1° spatial resolution with a 500 m vertical mesh to estimate the 3D distribution of biomass burning smoke injection into the atmosphere. Figure S 3, (SI) shows for all fires recorded in the MODIS fire product (Justice et al., 2002) during the fire period (31 July – 21 August 2017) the horizontal distribution of the median height of the emitted smoke and its integration over the longitude (right panel). Fires in Greenland showed a maximum injection height of around 2 km, but according to PRMv2 the majority of the emissions (90%) remained below 800 m. Low injection heights mostly inside the daytime planetary boundary layer are quite typical for smoldering fires including peat fires (Ferguson et al., 2003) such as those burning in Greenland (see below). For modeling the dispersion of BC, OC and BrC released from the Greenland fires, the emission profiles from PRMv2 were ingested into the Lagrangian particle dispersion model FLEXPART (see section 2.3).

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Wildfires in boreal peatlands in the Canadian Arctic and in Alaska typically have (shallow) burn depths of 1–10 cm and consume 20–30 t C ha⁻¹ (Benscoter and Wieder, 2003; Shetler et al., 2008). The consumed carbon is often re-sequestered in 60-140 years after the fire (Turetsky et al., 2011; Wieder et al., 2009). Given that fire return intervals can be as short as 100-150 years in sub-humid continental peatlands (Wieder et al., 2009), and may exceed 2000 years in humid climates (Lavoie and Pellerin, 2007), northern peatlands are generally resilient to wildfire (Magnan et al., 2012). For example, in peatlands of Northern Russia, organic matter available for combustion has been estimated to be 121.8 t C ha⁻¹ for forested lands and 21.3 t C ha⁻¹ for non-forested lands (Smirnov et al., 2015). Accordingly, a severe wildfire that burned within an afforested peatland in the Scottish Highlands during the summer of 2006 had a mean depth of burn of 17.5±2.0 cm (range: 1-54 cm) and a carbon loss of 96±15 t C ha⁻¹ (Davies et al., 2013). In contrast, tropical peatlands can have deep burn depths of 40-50 cm and release an average of 300-450 t C ha⁻¹ (Page et al., 2015; Reddy et al., 2015). In the present study, we assume an average amount of organic fuel available for combustion for the Greenland peat fires of August 2017 of 100 t C ha⁻¹, guided by values suggested in Smirnov et al. (2015).

Estimation of the emissions of BC, OC and BrC, $E_{BC,OC,BrC}$ (kg), was based on the following formula (Seiler and Crutzen, 1980; Urbanski et al., 2011) using the calculated burned area A (ha) and a number of assumptions:

 $E_{BC,OC,BrC} = A \times FL \times \alpha \times EF$ Eq. 1

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Here, FL is the mass of the fuel available for combustion (kg C ha⁻¹); α is the dimensionless combustion completeness, which was adopted from Hao et al. (2016) for litter and duff fuels (50%), EF is the emission factor (kg kg⁻¹), which was assumed to be 0.20 g kg⁻¹ for BC and 6.23 g kg⁻¹ for OC for peatland fires (Akagi et al., 2011), Emission factors for BrC are rarely reported, as BrC is only a fraction of OC. To our knowledge, the only reported emission factors in the literature for BrC are from forest fires in the United States (Aurell and Gullett, 2013) estimated to be 1.0–1.4 g kg⁻¹ (value used here: 1.2 g kg⁻¹). Fuel consumption is calculated as the product of burned area, fuel loading and combustion completeness $(A \times FL \times \alpha)$.

2.3 Atmospheric modeling

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The emissions of BC, OC and BrC obtained from Eq. 1 were fed to the Lagrangian particle dispersion model FLEXPART version 10.2 (Stohl et al., 2005) to simulate transport and deposition. This model was originally developed for calculating the dispersion of radioactive material from nuclear emergencies, but since then it has been used for many other applications (e.g., Fang et al., 2014; Stohl et al., 2011, 2013). The model has a detailed description of particle dispersion in the boundary layer and a convection scheme to simulate particle transport in clouds (Forster et al., 2007). The model was driven by hourly 0.5°×0.5° operational analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF). Concentration and deposition fields were recorded in a global domain of 1°×1° spatial resolution with three hourly outputs. To capture the spatiotemporal variability of BC, OC and BrC over the Greenland Ice Sheet, a nested domain with 0.05°×0.05° resolution was used. The simulations accounted for wet and dry deposition, assuming a particle density of 1500 kg m⁻³ and a logarithmic size distribution with an aerodynamic mean diameter of 0.25 µm and a standard deviation of 0.3 (Hu et al., 2018; Long et al., 2013). The wet deposition scheme considers below-cloud and in-cloud scavenging separately based on cloud liquid water and cloud ice content, precipitation rate and cloud depth from ECMWF, as described in Grythe et al. (2017).

To compare BC and OC concentrations in Greenland due to the emissions of the Greenland fires to those due to emissions occurring elsewhere, we used the so-called "retroplume" mode of FLEXPART for determining the influence of other sources. For only a few receptor points, this mode is computationally more efficient than forward simulations. Computational particles were tracked 30 days back in time from four receptor regions; Northwestern (-62°E to -42°E, 72°N to 83°N), Southwestern (-62°E to -42°E, 61°N to 72°N),

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Northeastern (-42°E to -17°E, 72°N to 83°N) and Southeastern Greenland (-42°E to -17°E, 61°N to 72°N). The retroplume mode allowed identification of the origin of BC and OC through calculated footprint emission sensitivities (often also called source-receptor relationships) that express the sensitivity of the BC and OC surface concentrations at the receptor to emissions on the model output grid. If these emissions are known, BC and OC concentrations at the receptor can be calculated as the product of the emission flux and the emission sensitivity. Also, detailed source contribution maps can be calculated, showing which regions contributed to the simulated concentration. For the anthropogenic emissions, we used the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants) version 5 (Klimont et al., 2017) emission data set. For the biomass burning emissions outside Greenland, we used operational CAMS GFAS emissions (Kaiser et al., 2012). To our knowledge, actual gridded emissions of BrC are not yet available.

2.4 Instantaneous radiative forcing (IRF) calculations

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The JRF of the emitted substances of interest were calculated using the uvspec model from the libRadtran radiative transfer software package (http://www.libradtran.org/doku.php) (Emde et al., 2016; Mayer and Kylling, 2005). The radiative transfer equation was solved in the independent pixel approximation using the DISORT model in pseudo-spherical geometry with improved treatment of peaked phase functions (Buras et al., 2011; Dahlback and Stamnes, 1991; Stamnes et al., 1988). Radiation absorption by gases was taken from the Kato et al. (1999) parameterization modified as described in the libRadtran documentation and Wandji Nyamsi et al. (2015). External mixture of aerosols was assumed, i.e. BC and BrC were treated in isolation of other aerosol types that may also have been present in the plume. This assumption likely leads to underestimates of the radiative impacts, at least for BC (Jacobson, 2001), in the atmosphere as coating, for example, can enhance its radiative effects, However, these assumptions should have little impact on the more important albedo calculations (see below). For snow-covered surfaces, deposited BC and BrC were assumed to reside in the uppermost 5 mm. Below 5 mm the snow was assumed to be without any impurities. The albedo of the snow was calculated with the SNICAR model (http://snow.engin.umich.edu/info.html) in a two-layer configuration (Flanner et al., 2007, 2009).

The IRF was calculated for three scenarios: (a) BC only, (b) BC and BrC and (c) BC and BrC, where all OC is considered to be BrC. The BC only scenario demonstrates the impact of BC alone, while the two other scenarios provide an estimate of the additional impact of BrC

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in the plume, with the last scenario considered to be a maximum estimate. We calculated both the bottom of the atmosphere (BOA) and top of atmosphere (TOA) instantaneous radiative forcing (IRF) due to the Greenland fires at 1°×1° resolution. The IRF includes both the effects of BC and BrC in the atmosphere and deposited in snow. Note that the IRF does not include any semi-direct nor indirect effects. We show IRF for cloudy conditions, which represents the possible radiative effects of BC and BrC due to the 2017 fires with respect to the actual meteorological situation, Liquid and ice water clouds were adopted from ECMWF.

2.5 Remote sensing of the smoke plume

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To confirm the presence of the emitted substances from the Greenland fires and elsewhere in the atmosphere over Greenland, we used the AERONET (AErosol RObotic NETwork) data (Holben et al., 1998). AERONET provides globally distributed observations of spectral aerosol optical depth (AOD), inversion products, and precipitable water in diverse aerosol regimes. We chose data from three stations that were close to the 2017 fires and for which cloud-free data exist for most of the simulated period, namely Kangerlussuaq (50.62°W–66.99°N), Narsarsuaq (45.52°W–61.16°N) and Thule (68.77°W–76.51°N). Their locations are shown in Figure S 2, We used Level 2.0 AOD data (fine and coarse mode AOD at 500 nm and total AOD at 400 nm) from the AERONET version 3 direct-sun spectral deconvolution algorithm (SDA version 4.1) product (downloaded on 20 July 2018) for the simulated period (31 July to 31 August 2017).

To examine in particular the vertical depth of the smoke, we used data from the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) lidar on the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) platform (Winker et al., 2009). CALIOP provides profiles of backscatter at 532 nm and 1064 nm, as well as the degree of the linear polarization of the 532 nm signal. For altitudes below 8.3 km lidar profiles at 532 nm are available with a vertical resolution of 30 m. We have utilized the level 1 data products (version 3.40) of total attenuated backscatter at 532 nm. This signal responds to aerosols (like BC, OC and BrC) as well as water and ice clouds, which in most cases can be distinguished based on their differences in optical properties. The data were downloaded from the ICARE Data and Services Center (http://www.icare.univ-lille1.fr/).

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3 Results

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3.1 Indications of early permafrost degradation and fuel availability

Table 1, reports burned areas in August 2017 calculated for Greenland. In total, 2345 hectares burned between 31 July and 21 August 2017 (Figure 1). We estimate that about 117 kt of carbon were consumed by these fires. The area burned is not large compared to the global area burned each year (464 million hectares), or the areas burned in boreal North America (2.6 million hectares) or boreal Asia (9.8 million hectares) (Randerson et al., 2012), but still highly unusual for Greenland.

It is not yet known how these fires started. Fires on carbon-rich soils can be initiated by an external source, e.g. lightning, flaming wildfire and firebrand, or self-heating. The fires burned relatively close to the town of Sisimut, so it is quite possible that humans started the fires. Self-heating is another possibility as porous solid fuels can undergo spontaneous exothermic reactions in oxidative atmospheres at low temperatures (Drysdale, 2011; Restuccia et al., 2017b). This process starts by slow exothermic oxidation at ambient temperature, causing a temperature increase, which is determined by the imbalance between the rate of heat generation and the rate of heat losses (Drysdale, 2011). Fire initiated by selfheating ignition is a well-known hazard for many natural materials (Fernandez Anez et al., 2015; Restuccia et al., 2017a; Wu et al., 2015) and can also occur in natural soils (Restuccia et al., 2017b). Southwestern Greenland was under anticyclonic influence during the last week of July and according to the MODIS ESDIS worldview tool, direct sunshine occurred for eight consecutive days before the fires started at the end of July 2017. It might be possible that this long period of almost continuous insolation at these latitudes in July heated the soil enough to self-ignite. In any case, the continuous sunshine had dried the soil, making it susceptible to fire.

The fact that these fires were burning for about three weeks but spread relatively slowly compared to above-ground vegetation fires indicates that the main fuel was probably peat. The predominant vegetation in Western Greenland varies from carbon-rich Salix glauca low shrubs (mean canopy height: 95 cm), mainly at low altitude south-facing slopes with deep soils and ample moisture, to dwarf-shrubs and thermophilous graminoid vegetation (Arctic steppe) at higher altitudes (Jedrzejek et al., 2013). In addition, the observed smoke was nearly white, indicating damp fuel, such as freshly thawed permafrost, which produces smoke rich in OC aerosol (Stockwell et al., 2016).

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Literally no fires should be expected in Greenland, since there is little available fuel as it has been suggested by global models and validated by observations (Daanen et al., 2011; Stendel et al., 2008); the only way to provide substantial amounts of fuel in Greenland is permafrost degradation. However, it has been suggested that significant permafrost loss in Greenland may occur only by the end of the 21st century (Daanen et al., 2011; Stendel et al., 2008). The fires in 2017 might indicate that significant permafrost degradation has occurred sooner than expected.

3.2 Transport and deposition of BC in Greenland

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We estimate that about 23 t of BC and 731 t of OC including 141 t of BrC, were released from the Greenland fires in August 2017 (Table 1). According to the FLEXPART model simulations, these emissions were transported and deposited as shown in Figure 2. Due to the low injection altitude of the releases within the boundary layer, transport was relatively slow and thus the emitted substances initially remained quite close to their source. Slow transport was also favored by mostly anticyclonic influence during the first half of August. It seems that even though katabatic winds from the Greenland Ice Sheet occasionally transported the plume westwards, most of the time the large-scale circulation pushed the plume back towards Greenland (see SI animations). Consequently, a large fraction of the emitted substances were deposited in Southwestern Greenland. On 3 August a small portion of the emitted BC, OC and BrC (0.5 t, 16.1 t and 3.1 t, respectively) were lifted higher into the atmosphere and were transported to the east and deposited in the middle of the Ice Sheet over the course of the following two days (4 and 5 August). From 5 to 8 August, when the fires were particularly intense, the emitted aerosols were transported to the south, where they were deposited at the southern part of the Ice Sheet and close to the coastline. At the same time, another branch of the plume was moving to the north depositing BC, OC and BrC over Greenland's western coastline up to 80°N, Around 10 August, the plume circulated north- and then eastwards in the northwestern sector of the anti-cyclone and the emitted aerosols were deposited to the northern part of the Ice Sheet until 13 August. From around 16 August, a cyclone approached from the northwest and the smoke was briefly transported directly eastwards along the southern edge of the cyclone (see SI animations). Strong rain associated with the cyclone's frontal system appears to have largely extinguished the fire by 17 or 18 August, although smaller patches may have continued smoldering for a few more days before they also died out. The exact fire behavior after 16 August is difficult to determine because of

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frequent dense cloud cover. However, satellite imagery on 21 August shows no smoke anymore in the area where the fires had burned.

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The total deposition of BC, OC and BrC from the fires in Greenland was estimated to be 9 t, 280 t and 54 t, respectively, or about 39% of the total emissions. About 7 t of BC, 218 t of OC and 42 t of BrC were deposited on snow or ice covered surfaces, which is equivalent to 30% of the total emissions. Most of the rest was deposited in the Baffin Bay between Greenland and Canada and in the Atlantic Ocean. With 30% of the emissions deposited on snow or ice surfaces, Greenland fires may have a relatively large efficiency for causing albedo changes on the Greenland Ice Sheet.

By comparison, the respective BC deposition on snow and ice surfaces over Greenland from global emissions of BC (from ECLIPSEv5) was only 0.4% (39 kt) of the total emissions. Even the total deposition of BC in the Arctic (>67°N) was only about 3% (215 kt). This indicates the high relative potential of Greenland fires to pollute the cryosphere (on a per unit emission basis), likely also giving them a particularly high radiative forcing efficiency. Considering that the projected rise of Greenland temperatures is expected to result in further degradation of the permafrost (Daanen et al., 2011) and, hence, likely resulting in more and larger peat fires on Greenland, this constitutes a potentially important climate feedback which could accelerate melting of the glaciers and ice sheet of Greenland and enhance Arctic warming.

We also calculated the concentration of the deposited <u>carbon aerosols</u> in Greenland snow (Figure 3) by taking the ratio of deposited <u>quantities</u> and the amount of water deposited by rain or <u>snowfall</u> during the same time period (31 July to 31 August 2017). As expected, snow concentrations show the same general patterns as the simulated deposition with the highest concentrations obtained close to the source <u>(western side of Greenland)</u>. High <u>snow</u> concentrations were also computed in some regions of the Ice Sheet due to relatively intense precipitation events. By contrast, dry deposition <u>(example for BC)</u> over the Ice Sheets was low (Figure S 4). Dry deposition was responsible for a major fraction of the deposition only in regions where the plume was transported during dry weather, and in most of these regions total deposition was low. A notable exception is the region close to the fires, where dry deposition was relatively important due to the generally dry weather when the fires were burning. It can be also ascribed to the fact that dry deposition occurs in the quasi-laminar sublayer close to the surface. A fraction of the aerosols can be quickly deposited close to the sources before they are transported to higher altitudes and away from the sources (Bellouin

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and Haywood, 2014). The average calculated <u>snow</u> concentration of BC on the Ice Sheet was estimated to be <1 ng g⁻¹, but in some areas snow concentrations reached up to 3 ng g⁻¹. These higher values are substantial considering that measured concentrations of BC in snow typically range up to 16 ng g⁻¹ in most of Greenland (Doherty et al., 2010) or from 1 – 17 ng g⁻¹ in summer 2012 and 3–43 ng g⁻¹ in summer 2013 (Polashenski et al., 2015) and up to 15 ppb C (ng g⁻¹) during preindustrial times (from 1740 to 1870) on average (Legrand et al., 2016). OC concentrations in snow were 2 ng g⁻¹ (ppb C), on average, with local maxima of 10 ng g⁻¹. They are lower than those measured in snow over several places in Antarctica (23–928 ppb C) (Antony et al., 2011; Grannas et al., 2004; Legrand et al., 2013; Lyons et al., 2007), in Greenland (400–580 ppb C) (Grannas et al., 2004) or in the Alps (70–304 ppb C) (Legrand et al., 2013). Snow BrC was estimated to be even less; though, to our knowledge, no available measurements exist in the relevant literature so far.

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It has been reported that the size of rapidly coagulated aerosol particles produced by different types of fires ranges between 0.1 to 10 µm, but more than 90% of the mass lies between 0.1 and 1 μm (e.g., Conny and Slater, 2002; Long et al., 2013; Zhuravleva et al., 2017 and many others). Therefore, we simulated the Greenland fires with an aerodynamic mean diameter of 0.25 µm for BC, OC and BrC and a logarithmic standard deviation of 0.3 (see section 2.3), because all these substances have more or less the same lifetimes (Bond et al., 2013; Jo et al., 2016; Lim et al., 2003). To examine the sensitivity of deposition in the Greenland Ice Sheet from the Greenland fires of 2017 to the particle size distribution used in the model, we simulated the same event for particles with aerodynamic mean diameters of 0.1, 0.25, 0.5, 1, 2, 4 and 8 µm and calculated the relative standard deviation of deposition normalized against the aerodynamic mean diameter of 0.25 µm that was our basic assumption. The results are shown in Figure S 5 for BC. The use of different size distributions for the BC particles produced from the 2017 fires created a relative uncertainty on the deposited mass of BC in the Greenland Ice Sheet, which ranges from 10%-30% in 86% of the Sheet's surface to up to 50% in the rest of the Sheet's surface. As expected, the calculated uncertainty is sensitive to the use of larger particles for BC; though BC particles larger than 1 μm are rather rare in peat fires (Hosseini et al., 2010; Leino et al., 2014).

3.3 Impact from other emissions in the Northern Hemisphere

In summertime 2017, intense wildfires were reported in British Columbia, Western Canada (NASA, 2017c), and fires also burned at mid latitudes in Eurasia, as is typical during spring and summer (Hao et al., 2016). Previous studies of wildfires have shown that the

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produced energy can be sufficient to loft smoke above the boundary layer by supercell convection (Fromm et al., 2005) even up to stratospheric altitudes (Leung et al., 2007). As a result, emitted aerosols can become subject to long-range transport over long distances (Forster et al., 2001; Stohl et al., 2007). To examine the impact of these fires in Greenland, average footprint emission sensitivities were calculated for four compartments of Greenland (Northwestern, Southwestern, Northeastern and Southeastern Greenland) for the period 31 July to 31 August 2017 and the results are shown in Figure S 6 together with the active fires in the Northern Hemisphere from 10 July to 31 August 2017 adopted from the MODIS satellite product (MCD14DL) (Giglio et al., 2003). As can be seen in Figure S 6, fires in Alaska and in Western Canada might have affected BC, OC and BrC concentrations in Greenland, as the corresponding emission sensitivities are the highest in North America. On the contrary, emissions from fires in Eurasia seem to have affected Greenland less.

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Using gridded emissions for BC and OC, the contribution of both biomass burning and anthropogenic sources to surface concentrations in the four different regions over Greenland (Northwestern, Northeastern, Southwestern and Southeastern Greenland, Figure S 7) was calculated (see section 2.3). Fires affected the northern part of Greenland more than the southern part with an average BC concentration of about 30 ng m⁻³, almost twice the respective average for Southern Greenland (≈16 ng m⁻³). OC simulated concentrations were much higher that those of BC with an average concentration of 945 ng m⁻³ in North Greenland, while the respective concentrations in the southern part were about 490 ng m⁻³. About one third of BC and OC originated from wildfires in Eurasia and the rest from North America where the year 2017 appears to have been a particularly high fire year. The anthropogenic contribution to surface concentrations of BC and OC, over Greenland was between 14% to 50% of the total contribution from all biomass burning sources (Figure S 7), similar to what has been suggested previously for the Arctic in summer (Winiger et al., 2017). The anthropogenic contribution is larger in Southern Greenland, than in Northern Greenland, due to the shorter distance from the main emission areas of North America and Western Europe, but it remains much lower than the biomass burning contribution. The concentrations of BC and OC that are calculated for the studied fire period (31 July to 31 August 2017) are relatively high compared to those reported previously. For instance, von Schneidemesser et al. (2009) observed an annual average BC concentration of 20 ng m⁻³ at Summit (Greenland) in 2006, while Massling et al. (2015) reported a summer average BC concentration of 11 ng m⁻³ at station Nord (Greenland) between May 2011 and August 2013. As regards to OC, average

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concentrations of its water soluble part were measured in 2006 between 194 and 730 ng m⁻³ in Summit, Greenland (Anderson et al., 2008) showing a large decreasing trend compared to previous years (Dibb et al., 2002). We attribute this difference in the calculated concentrations to more active fires during 2017 in Greenland than in previous years (see Figure S 1).

As an example of the importance of Northern Hemispheric biomass burning emissions for the air over Greenland, we present time-series of surface BC concentrations in Northwestern, Northeastern, Southwestern and Southeastern Greenland from the fires in Greenland and from all the other wildfire emission sources occurring outside Greenland (North Hemisphere) for the same period of time (Figure 4). The calculated dosages (concentrations summed over a specific time period) for the same time period were also computed. The fires in Greenland affected mainly its western part with concentrations that reached up to 4.8 ng m⁻³ (Southwestern Greenland on 10 August) and 4.4 ng m⁻³ (Northwestern Greenland on 12 August), while BC concentrations in the eastern part remained significantly lower (Figure 4). These concentrations are substantial considering that the observed surface BC concentrations in Greenland in summer are usually below 20 ng m⁻³ (Massling et al., 2015). Surface BC due to wildfires occurring outside Greenland was also low most of the time in the studied period (up to 10 ng m⁻³ at maximum) except for a large peak between 19 and 23 August that mainly affected Northern Greenland (Figure 4). The concentrations during this episodic peak were as high as 27 ng m⁻³. During the same period, the contribution from anthropogenic emissions was also a few ng m⁻³ (Figure 4). BC dosages for the simulation period (31 July - 10 August 2017) in Western Greenland due to the Greenland fires were about one order of magnitude smaller than dosages from fires elsewhere but of the same order of magnitude as BC originating from anthropogenic emissions.

4 Discussion

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4.1 A validation attempt

There are few observations available that can be used to validate our model results. We use the AERONET and CALIOP data for some qualitative comparisons. We present only BC here, but similar plots can be generated for OC, considering that we used the same scavenging coefficients as for BC to represent the similar lifetimes of BC and OC (Bond et al., 2013; Jo et al., 2016; Lim et al., 2003). Contours of simulated vertical distribution of BC and column-integrated simulated BC from fires inside and outside Greenland are plotted together with time-series of measured AOD (fine and coarse mode AOD at 500 nm and total AOD at 400

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nm) for the AERONET stations Kangerlussuaq, Narsarsuaq and Thule (Figure 5). It can be seen that observed AOD variations were in very good agreement with the variation of simulated column-integrated BC from fires outside Greenland (mainly in Canada), confirming that the transport of these fire plumes was well captured by FLEXPART. Good examples are the peaks at Kangerlussuaq on 24 August, at Narsarsuaq on 19 August and at Thule on 21 August (Figure 5) that are attributed to the Canadian fires. The simulated contribution of the Greenland fires to simulated BC burdens was negligible by comparison, except at Kangerlussuaq in the beginning of August when the Greenland fire emissions were the highest. This station is less than 100 km away from where the fires burned, but not in the main direction of the BC plume transport. It seems the period of simulated fire influence corresponds to a small increase of the observed AOD values of up to 20% (Figure 5).

To validate the smoke plume's vertical extent, we used the CALIOP data. These data

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were only available from 5 August 2017 onward and frequent dense cloud cover inhibited lidar observations at the altitudes below the clouds. High aerosol backscatter was only found in the close vicinity of the fires. Figure 6a shows NASA's ESDIS view of the plume on 14 2017 6 UTC (available: https://worldview.earthdata.nasa.gov/?p=ge ographic&l=MODIS Aqua CorrectedReflectance TrueColor(hidden),MODIS Terra Correc tedReflectance TrueColor, MODIS Fires Terra, MODIS Fires Aqua, Reference Labels (hidd en), Reference Features, Coastlines & t=2017-08-14 & z=3 & v=-54.13349998138993,66.35888052399868,-50.32103113049877,69.08420005412792), where a clear smoke signal was recorded. A CALIOP overpass through the edge of the plume allows studying its vertical structure. Increased attenuated backscatter is found below ~1.5 km above sea level between 52°E and 51°E (Figure 6b; black line denotes the orography). Figure 6c (red line), shows that the CALIOP overpass transects directly the simulated plume of the Greenland fires. Notice that the simulated plume also agrees very well with the smoke as seen in NASA's ESDIS picture (Figure 6a). The vertical distribution of simulated BC as a function of longitude is illustrated in Figure 6d. It corresponds very well to the vertical distribution of aerosols observed by CALIOP (Figure 6b). In particular, the smoke resides at altitudes below 1.5 km and at exactly the same location both in the simulations and observations.

4.2 **Instantaneous radiative forcing and albedo effects**

BOA IRF due to (a) BC only, (b) BC and BrC and (c) BC and BrC when all OC was assumed to be BrC (extreme scenario) for noon on 31 August 2017 is depicted in Figure 7a-c. This day is shown because almost all the aerosols emitted by the fires had been deposited.

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thus giving a high IRF via albedo reduction due to snow contamination. The IRF is the largest over ice close to the fire site and at locations where relatively large amounts of BC and BrC were deposited. For BC only, the maximum BOA (TOA) IRF is 0.63 W m⁻² (0.59 W m⁻²), and the average 0.03 W m⁻² (0.03 W m⁻²). Including BrC slightly increases the maximum BOA (TOA) IRF to 0.65 W m⁻² (0.61 W m⁻²), while the change in the average IRF values is negligible. For the extreme BrC scenario, the maximum BOA (TOA) IRF is 0.77 W m⁻² (0.71 W m⁻²) and the average 0.04 W m⁻² (0.06 W m⁻²). So, including BrC in our analysis increases BOA IRF by only 20% even for the extreme scenario.

The IRF depends on the optical properties of the smoke from the fire, which are not known. Hence, a sensitivity analysis was performed where the single scattering albedo (SSA) was perturbed in contrast to a "medium case" (Figure S 8a) that was adopted from the SNICAR model (Flanner et al., 2007, 2009) and has been used for the discussion in the previous paragraph. To estimate the uncertainty due to the choice of BC optical properties, additional calculations were made by scaling the SSA (red solid lines in Figure S 8a). The choices of these scaled SSA values were based on the SSA reported for various modified combustion efficiencies (MCE) by Pokhrel et al. (2016). Pokhrel et al. (2016) reported an MCE of 0.9 for peat land. As such, our adopted SSA may be considered low (compare black solid line and red line with upward triangles). Figure S 8b shows the IRF as BC is deposited for the three cases. It suggests that the IRF ranges between 40% and 130% of our aboveassumed medium-case values for realistic variation of the aerosol optical properties.

Figure 7d depicts the temporal behaviour of the cloudy TOA IRF averaged over Greenland (daily averages) for BC only (red line), for BC and BrC (blue line) and for BC and BrC, when all OC is assumed to be BrC (black line, extreme case scenario), The daily averaged IRF is seen to increase as the plume from the fires spreads out and starts to decline after the fires were extinguished at the end of the month. The fact that the reduction towards end of August is relatively slow is caused by the effect of the albedo reduction, which persists until clean snow covers the polluted snow. Overall, albedo reduction dominates the total IRF averaged over Greenland for the period of study contributing between 85% (in the beginning of the study period) to 99% (at the end of the study period) and increasing in relative importance with time as atmospheric BC and BrC are removed. The largest IRF differences between the BC only case IRF and the two BC+BrC cases occur when there is still smoke in the air and the lowest IRF differences occur after August 15th. This indicates that BrC is most important for the IRF when it is airborne, even in the extreme scenario. However, for the

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latter, the impact is also large after August 15th due to a further albedo decrease of about 0.001 compared to the case where only BC was considered.

According to Hansen et al. (2005) the TOA IRF of BC approximates the adjusted RF as reported by Myhre et al. (2013). In their Table 8.4, Myhre et al. (2013) estimated the global averaged RF due to BC between the years 1750 and 2011 to be +0.40 (+0.05 to +0.80) W m⁻². Skeie et al. (2011) estimated a global mean radiative forcing of 0.35 W m⁻² due to fossil fuel and biofuel increases between 1750 and 2000. For Greenland, Skeie et al. (2011) found the RF to be less than about 0.2 W m⁻². This number may be compared to our area averaged IRF estimate due to the Greenland fire. For cloudy conditions the TOA IRF over Greenland due to the Greenland fires is about a factor 4 to 10 smaller compared with the RF over Greenland due to BC from all global anthropogenic sources reported in Skeie et al. (2011).

The albedo reduction at 550 nm for the three scenarios (BC only, BC+BrC and BC+BrC extreme) is shown in Figure 7g-g. The maximum albedo change is about 0.006 when only BC was considered. Adding BrC from the most extreme scenario, the maximum albedo change was calculated as 0.007 This albedo change has an impact on IRF, but it is too small to be measured by satellites. For example, MODIS albedo estimates have been compared to in situ albedo measurements in Greenland by Stroeve et al. (2005). They found that the root mean square error between MODIS and in situ albedo values was ±0.04 for high quality flagged MODIS albedo retrievals. Unmanned Aerial Vehicle (UAV) measurements over Greenland made by Burkhart et al. (2017) have uncertainties of similar magnitude. Also, Polashenski et al. (2015) reported that the albedo reduction due to aerosol impurities on the Greenland Ice Sheet in 2012–2014 period is relatively small (mean 0.003), though episodic aerosol deposition events can reduce albedo by 0.01–0.02. The albedo changes due to BC and BrC from the Greenland fires are generally an order of magnitude smaller (Figure 7g-g) and thus too small to be detected by present UAV and satellite instruments and retrieval methods (Warren, 2013).

5 Conclusions

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We studied atmospheric transport, deposition and impact of BC, BrC and OC emitted as a result of unusual open fires burning in Greenland between 31 July and 21 August 2017. Our conclusions can be summarized below:

The fires burned on peat lands that became vulnerable by permafrost thawing. The region where the fires burned was identified previously as being susceptible to permafrost

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- melting; however, large-scale melting was expected to occur only towards the end of the 21st century. The 2017 fires show that at least in some locations substantial permafrost thawing is already occurring now.
- The total area burned was about 2345 hectares. We estimate that the fires consumed a fuel amount of about 117 kt C and emitted about 23.5 t of BC and 731 t of OC including 141 t of BrC.

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- The Greenland fires were small compared to fires burning at the same time in North America and Eurasia, but a large fraction of <u>BC₂ OC and BrC</u> emissions (30%) was deposited on the Greenland Ice Sheet.
- Measurements of aerosol optical depth at three sites in Western Greenland in August 2017
 were strongly influenced by forest fires in Canada burning at the same time, but the
 Greenland fires had an observable impact doubling the column-integrated BC
 concentrations at the closest station.
 - A comparison of the simulated BC releases in FLEXPART with the vertical cross-section
 of total attenuated, backscatter (at 532 nm) from CALIOP lidar showed that the
 spatiotemporal evolution and particularly the top height of the plume was captured by the
 model.
 - We estimate that the maximum albedo change due to the BC deposition from the Greenland fires was about 0.006, whereas adding deposited BrC increases albedo to 0.007 at maximum, which is too small to be measured. The average instantaneous BOA radiative forcing over Greenland at noon on 31 August was between 0.03–0.04 W m⁻² for the three scenarios (BC only, BC+BrC and BC+BrC extreme), with locally occurring maxima, of 0.63 W m⁻², 0.65 W m⁻² and 0.77 W m⁻², respectively. The average value when only BC was considered is up to an order of magnitude smaller than the radiative forcing due to BC from other sources.
 - We conclude that the fires burning in Greenland in summer of 2017 had small impact on on the Greenland Ice Sheet, causing almost negligible extra radiative forcing. This was due to the in a global context still rather small size of the fires.

The very large fraction of the emissions deposited on the Greenland Ice Sheet (30% of the emissions) makes these fires very efficient climate forcers on a per unit emission basis. Thus, while the fires in 2017 were still relatively small on a global scale, if the expected future warming of the Arctic (IPCC, 2013) produces more and larger fires in Greenland (Keegan et

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al., 2014), this could indeed cause substantial albedo changes and thus <u>contribute</u> to accelerated melting of the Greenland Ice Sheet.

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972 *Data availability.* All data used for the present publication can be obtained from the 973 corresponding author upon request.

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975 Competing financial interests. The authors declare no competing financial interests.

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1400 1401 1402 1403	event over Siberia in summer 2012, Atmos. Meas. Tech., 10(1), 179–198, doi:10.5194/amt-10-179-2017, 2017.
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1404 FIGURE LEGENDS

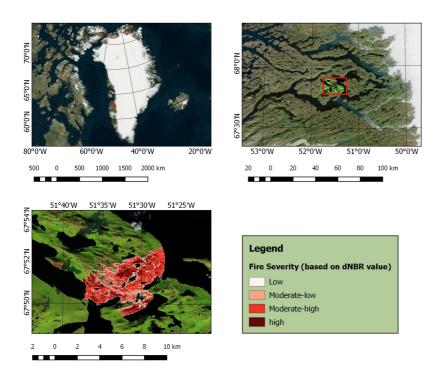


Figure 1. Map of Greenland (upper left) and zoomed map marked with fire location (upper right and burned area classification (bottom) in terms of fire severity according to Sentinel 2A images for fires burning in Greenland in August 2017. To delineate fire perimeters, both Landsat 8 OLI and Sentinel 1A - 2A data were used (Table 1).

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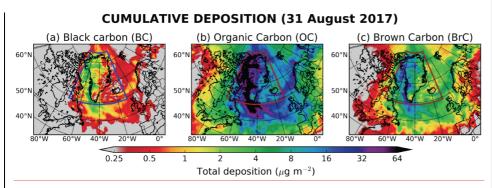


Figure 2. Total (wet and dry) deposition of (a) BC, (b) OC and (c) BrC (in µg m⁻²) from the Greenland fires until 31 August 2017. The colored rectangle depicts the nested high-resolution domain.

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Deleted: (a) Vertical distribution of BC concentrations from the fires in the area of Greenland in summer 2017 as a function of time (h)

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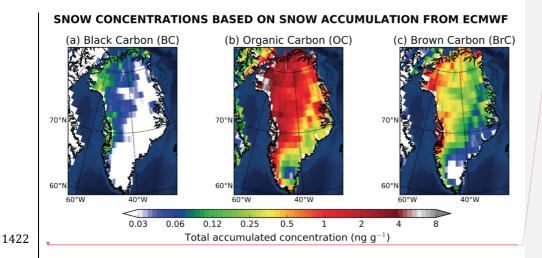
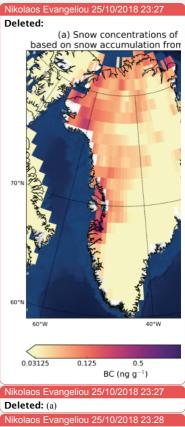


Figure 3. Calculated snow concentrations of (a) BC, (b) OC and (c) BrC over Greenland based on the modeled deposition and the snow precipitation (large scale and convective) adopted from the operational ECMWF data that were used in our simulation (see section 2.3).

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Deleted: (b) Dry to total deposition ratio of BC from the 2017 peat fires over Greenland.

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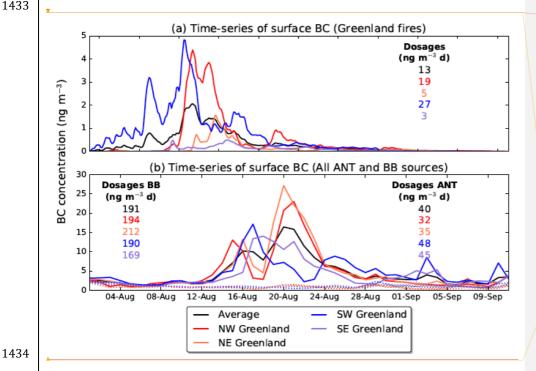


Figure 4. (a) Time-series of surface BC concentrations in Northwestern, Northeastern, Southwestern and Southeastern Greenland from the summer 2017 fires in Western Greenland. (b) Time-series of surface BC concentrations in Northwestern, Northeastern, Southwestern and Southeastern Greenland from global anthropogenic (ANT, dashed lines) and biomass burning (BB, solid lines) emissions for the same period. The numbers represent the respective dosages (time-integrated concentrations) for the time period shown. The color codes are reported in the legend.

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Moved down [1]: Average contribution of biomass burning (upper panels) and anthropogenic emissions (lower panels) to surface concentrations of BC in Northwestern, Northeastern, Southwestern and Southeastern Greenland (in ng m⁻³ per grid cell). Numbers (in red) represent total concentrations in the studied domain, obtained by spatial integration over all source grid cells. Receptor areas in Greenland are highlighted by pink boxes.

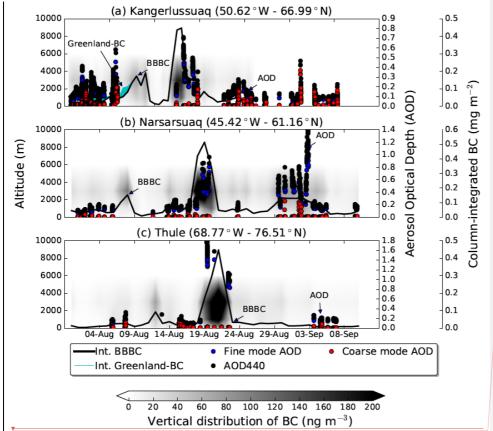
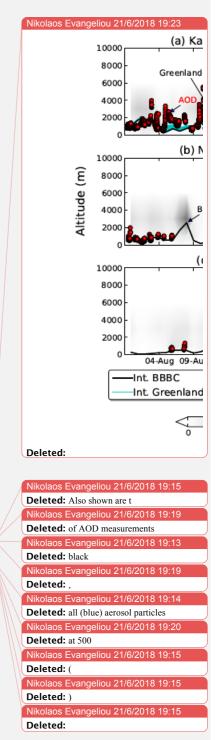


Figure 5. Contour plot of the vertical distribution of simulated BC (altitude <u>a.g.l.</u> shown on left y-axis) as a function of time (x-axis) and time-series of column-integrated simulated BC (extended right axis) from fires burning outside Greenland (black line) and Greenland fires (cyan stacked area). Column-integrated BC from anthropogenic sources was extremely small and it is not plotted here. Time-series for fine mode (blue) and coarse (red) AOD at 500 nm and total AOD at 400 nm (black) correspond to the right y-axis. The three panels show results for stations (a) Kangerlussuaq, (b) Narsarsuaq and (c) Thule (sorted from the closest to the farthest station).



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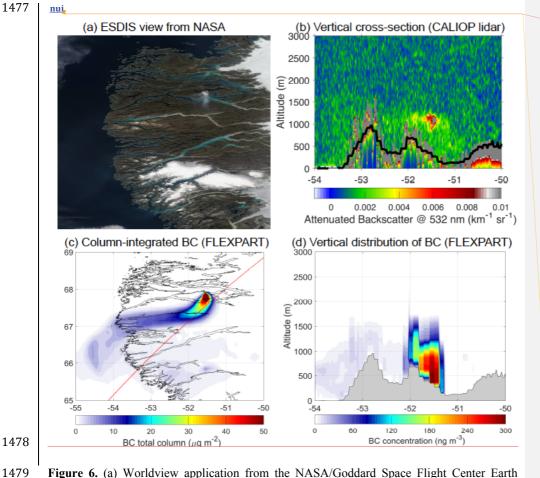
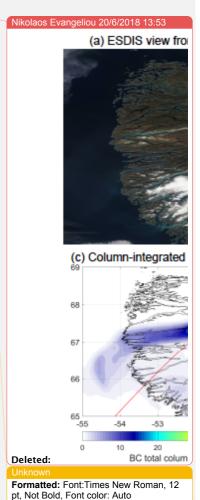


Figure 6. (a) Worldview application from the NASA/Goddard Space Flight Center Earth Science Data and Information System (ESDIS) project on 14 August 2017. (b) Vertical crosssection along satellite's route (red line in c) of total attenuated backscatter at a wavelength of 532 nm obtained from the CALIOP lidar on 14 August 2017 at 6 UTC (black line denotes the orography of the area). (c) Column-integrated BC concentration simulated with FLEXPART (read line shows the path of the satellite). (d) Vertical distribution of BC concentrations with longitude as seen with FLEXPART (grey area denotes the orography of the area).



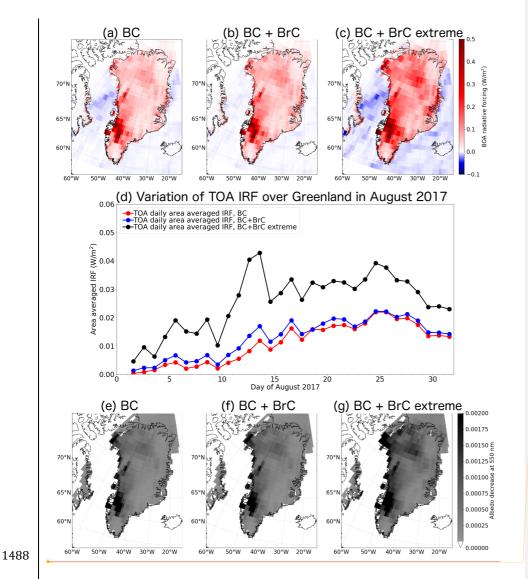


Figure 7. The instantaneous direct BOA RF due to (a) BC only, (b) BC and BrC, and (c) BC and BrC when OC was assumed to be all BrC (extreme case) from the Greenland fire for cloudy conditions on 31 August, 2017. (d) Daily variation of the TOA IRF over Greenland in August 2017 for the three studied scenarios. Albedo reduction at 550 nm due to (e) BC only, (f) BC and BrC, and (g) BC and BrC when OC was assumed to be all BrC (extreme case). Note that the maximum albedo change due to deposited smoke is 0.00585 (BC only), 0.00590 (BC+BrC) and 0.00670 (BC+BrC extreme).

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Deleted: (a) The instantaneous direct BOA RF due to BC from the Greenland fires for cloudless and (b) cloudy conditions on 31 August, and (c) snow albedo reduction. (d) Temporal variation of the TOA IRF over Greenland in August 2017.

1502	SUPPLEMENTARY FIGURE LEGENDS	
1503		
1504	Figure S 1. Annual number of active fires over Greenland during the last 17 years as seen	
1505	from NASA's MODIS satellite (product MSC14DL).	
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1507	Figure S 2. Fire dynamics in Greenland for the August 2017 fires according to MODIS	
1508	(magenta dots show active fire hot spots from the MODIS MCD14DL product). Locations of	Nikolaos Evangeliou 26/10/2018 10:38 Deleted: 21
1509	stations with AOD measurements from AERONET are also shown.	
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1511	Figure S 3, Median injection heights (km above sea level – ASL; left panel) and distribution	
1512	of longitudinally integrated burned biomass (Tg) as a function of injection altitude (right	Nikolaos Evangeliou 26/10/2018 10:38 Deleted: 32
1513	panel) calculated by PRMv2 for the period between 31 July and 21 August 2017.	
1514	Element C. A. Donn to total describer anti- (consult for DO) from the 2017 and for	
1514 1515	Figure S 4. Dry to total deposition ratio (example for BC) from the 2017 peat fires over	
1313	Greenland.	
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1517	Figure S 5, Relative standard deviation of deposited mass (example for BC) for different	Nikolaos Evangeliou 26/10/2018 10:38
1518	assumed size distributions normalized against the results from our reference size distribution	Deleted: 4
1519	with a logarithmic mean diameter of 0.25 μm. Particle size distributions with aerodynamic	
1520	mean diameters of 0.1, 0.25, 0.5, 1, 2, 4, 8 μm and a logarithmic standard deviation of 0.3	
1521	were simulated.	
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1523	Figure S 6 Footprint emissions sensitivities for Northwestern, Northeastern, Southwestern	Nikolaos Evangeliou 26/10/2018 10:38
1524	and Southeastern Greenland for the period 31 July to 31 August 2017. Active fires from	Deleted: 5
1525	NASA's MODIS MCD14DL product are shown with red dots.	Nikolaos Evangeliou 26/10/2018 10:38
1526		Deleted: 64 Nikolaos Evangeliou 6/6/2018 17:46
1527	Figure S 7 Average contribution of biomass burning (upper panels) and anthropogenic	Moved (insertion) [1]
1528	emissions (lower panels) to surface concentrations of (a) BC and (b) OC in Northwestern,	Nikolaos Evangeliou 6/6/2018 17:46 Deleted: (a) Time-series of surface BC
1529	Northeastern, Southwestern and Southeastern Greenland (in ng m ⁻³ per grid cell). Numbers (in	concentrations in Northwestern, Northeastern, Southwestern and Southeastern Greenland
1530	red) represent total concentrations in the studied domain, obtained by spatial integration over	from the summer 2017 fires in Western Greenland. (b) Time-series of surface BC
1531	all source grid cells. Receptor areas in Greenland are highlighted by pink boxes.	concentrations in Northwestern, Northeastern, Southwestern and Southeastern Greenland
1532		from global anthropogenic (ANT) and biomass burning (BB) emissions for the same period.
1334		The numbers represent the respective dosages integrated for the time period shown and each
		color corresponds to the legend.

Figure S & (a) The single scattering albedo (SSA) of BC as a function of wavelength for various modified combustion efficiencies (MCE). The star and dot marked lines are from the parameterization of Pokhrel et al. (2016). (b) The IRF as a function of BC deposited on the Ice Sheet. The calculations were made for cloudless conditions with a snow-covered surface for noon on 31 August 2017 at 65°N.

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Table 1. Start and end date of releases, source of data, type of sensor, burned area and daily increment of burned area, fuel consumption and calculated BC emissions from Eq. 1 during the Greenland fires in 2017. Total numbers for burned area, fuel consumption and BC emissions are highlighted in bold.

Start	End	Source of RS data	Type of sensor	Burned	Increment	Fuel	BC	OC	BrC
				area (ha)	of burned area (ha)	consumption (t C)	emissions (kg)	emissions (kg)	emissions (kg)
				` ′	. ,	` ′			
31/07/17	02/08/17	Sentinel 2A	MSI	304	304	15176	3035	94543	18211
02/08/17	03/08/17	Landsat 8 OLI	MSI	428	125	6247	1249	38916	7496
03/08/17	04/08/17	Sentinel 1A	SAR	588	160	7980	1596	49712	9575
04/08/17	05/08/17	Sentinel 1A	SAR	740	152	7621	1524	47479	9145
05/08/17	07/08/17	Sentinel 2A	MSI	1100	359	17966	3593	111925	21559
07/08/17	08/08/17	Sentinel 2A	MSI	1314	214	10706	2141	66698	12847
08/08/17	12/08/17	Landsat 8 OLI	MSI	1868	554	27714	5543	172658	33257
12/08/17	14/08/17	Sentinel 1A	SAR	2005	136	6817	1363	42470	8180
14/08/17	15/08/17	Sentinel 1A	SAR	2169	165	8244	1649	51363	9893
15/08/17	16/08/17	Sentinel 1A	SAR	2209	40	1998	400	12444	2397
16/08/17	19/08/17	Sentinel 1A	SAR	2254	44	2213	443	13784	2655
19/08/17	21/08/17	Sentinel 2A	MSI	2345	92	4579	916	28530	5495
TOTAL					2345	117259	23452	730524	140711

RS - Remote Sensing

MSI - Multispectral Images SAR - Synthetic Aperture RADAR

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RF was calculated at the top and bottom of the atmosphere at 1°×1° resolution.

Another cloud of enhanced attenuated backscatter is evident at 4–5 km altitude between 50.5°E and 48.5°E. This mid-tropospheric plume was not studied but is likely due to aerosol transport from the North American fires. These large wildfires are eager to lift smoke at stratospheric altitudes as a result of super-cell convection and they have alreadySmoke from these fires was already shown to be present as such altitudes in Greenland during the study period (see Figure 5Figure 5). As shown in

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Effect on snow and ice surfaces

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The bottom of the atmosphere (BOA) instantaneous radiative forcing (IRF) due to the Greenland fires

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at the bottom of the atmosphere (BOA)

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, both for cloudless (Fig. 7a) and cloudy conditions (Fig. 7b)

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For IRF at the top of the atmosphere (TOA), the corresponding values are 0.59 W m⁻² and 0.03 W m⁻².

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In addition the daily averaged IRF is shown (green line).

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The blue line in Figure 7db shows the value for the pixel with maximum IRF.

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IRF or with the BC+BrC IRF in the extreme scenario (all OC is BrC)