We provide detailed responses below in black, with quotation marks showing the changes made in the manuscript and associated line numbers on the final (un-tracked) manuscript.

# Author Response to Reviewer #2

This paper presents the results of a high number of model simulations performed to derive runoff from Greenland Ice Sheet as a function of black carbon deposition. The model is using state-of-the art snow model, which includes interactions of Light Absorbing Impurities (LAI) with solar radiation, as well as scavenging by meltwater. They also provide linear equations that relate the increase of total runoff to black carbon deposition. Their results could be used by climate models, which don't have a sophisticated snow model but would like to estimate total runoff from Greenland for a given black carbon deposition rate.

The paper is well written, their approach valid, and their results interesting. I have only few minor comments.

# Minor comments: All the figures showing time series with one line per month would gain in clarity by reducing to one month per season.

As the reviewer suggested, we tried to make figures with seasonal curves. However, because the seasonal cycle differs among the quantities shown in the different figures, the seasonal plots become confusing. We provide here two seasonal plots for Figure 5 and 6 as examples, which we cannot see clear seasonal patterns for both wet and dry BC deposition fluxes. Thus, we decide not to change the figures in the manuscript.



**Figure R1.** Hydrophilic BC concentration in the top snow layer vs. concentration of BC in precipitation shown at different times (0-11 months, panels a-l) since deposition. The top-snow layer concentrations are averaged over the Greenland region and over ten one-year simulations beginning in years 2006-2015. Different line colors represent different deposition seasons.



**Figure R2.** Same as Figure R1, but showing hydrophobic BC concentration in the top snow layer vs. BC dry deposition flux.

# Are you assuming linearity of surface albedo change by black carbon deposition and metamorphism of snow grains? Should there not be another entry in the kernel for surface temperature?

We agree that surface temperature influences metamorphism of snow grains. However, as the reviewer also pointed out, the main difficulty of climate models is the lack of sophisticated snow model which relates total runoff with black carbon deposition. Therefore, we aimed to find out only the relationship between the amount of black carbon deposition and snow melt in our study. We didn't assume a linear relationship, but based on our simulations and calculations, we found linear regression could appropriately represent this relationship. Thus, we were able to provide linear equations relating the increase of total runoff to black carbon deposition.

It is unclear if the timestep of the snow model is one month, or sub-daily with the kernel data calculated as mean monthly values. If the timestep is monthly then the results for sub-daily timestep may have been quite different. At least an estimated error should be provided in such case.

Another needed precision is about spatial resolution. Line 98 we learned that CRUNCEP input data is 0.5 degree resolution. Is it the same for CLM and the kernel data?

We thank the reviewer for pointing out the confusion of temporal and spatial resolutions. Although the kernel data was calculated as monthly mean values, the timestep of the snow model implemented in our study was actually 0.5 hour. The resolution of CLM is  $0.9^{\circ} \times 1.25^{\circ}$ . And it is the same for the kernel data.

We add an explanation in Line 159 that "The timestep of CLM/SNICAR for our simulations is 0.5 hour, and the spatial resolution is  $0.9^{\circ} \times 1.25^{\circ}$ . The kernel product is calculated as monthly mean values with the same spatial resolution."

# Line 141 "dry" should be "wet"

This was not a typo. We were trying to use the maximum wet BC deposition flux to calculate the maximum dry deposition flux, and then to implement this value to set up a set of dry BC fluxes with logarithmic spacing.

# Line 177: "... as new snowfall dilutes the contaminated snow". Are you mixing layers and not superimposing them? LAI cannot move up in your model, right?

In CLM, snow layers can be combined and divided, as the maximum number of layers is fixed at 5, and both minimum and maximum thicknesses are prescribed for each layer. When layers are combined (e.g., because a layer thickness drops beneath the minimum threshold due to melt), both snow mass and BC mass are added together to establish the properties of the new layer. When the top layer exceeds the maximum thickness (2cm), e.g., due to new snowfall, the excess snow mass and a proportionate amount of BC are 'mixed down' into the layer beneath. We have specified these processes in Line 115, "Deposited particles are assumed to be instantly mixed and homogeneous within the surface snow layer", and "Particle masses are redistributed vertically in each time step proportionately with snow melt through the snow column, scaled by the species-specific melt scavenging efficiency, and snow layer combination and subdivision."

# Lines 249-252: Is this due to an amplification effect by metamorphism of snow?

It could be. I added this hypothesis in Line 254 that "This could also be due to an amplification effect by metamorphism of snow in warmer years."

# Line 262: "runoff... higher temperature" Is this increase of runoff by higher temperature due to darkening by black carbon or larger grain size?

That's a good point. We add the explanation in Line 265 that "The largest perturbation in summer could be caused by immediate melting due to darkening, as well as larger grain size in warmer environment."

Lines 279-280: "higher light absorptivity of hydrophilic BC" seems to contradict the values of single scattering albedo provided for black carbon in Table 3.5 of Oleson et al. (2013). The single scattering albedo of hydrophobic black carbon is lower than hydrophilic black carbon, indicative of stronger absorption by hydrophobic BC.

We agree that the single scatter albedo (SSA) is indeed lower for hydrophobic BC. However, the mass absorption cross section (MAC; product of the mass extinction cross section and co-SSA) is higher for hydrophilic BC. For absorption perturbations within a highly scattering medium like snow, MAC is the more relevant metric of absorptivity than SSA.

# Line 281: "weakly-absorbing sulfate" Sulfate is purely scattering.

The assumed SSA value of sulfate is technically slightly less than 1.0, but we agree with the reviewer's point and have revised this reference to "non-absorbing sulfate" in Line 285.

# Author Response to Reviewer #3

The melting of the Greenland ice sheet is a big concern to global societies. Black carbon and other light absorbing constituents could be one of the most vital factors leading to the melting. The topic of the work is important in science. However, the work itself is purely a computed game based on amounts of presumptions. It is suggested that the investigators going to the field to do some measurements in the surface snow or firn and to know the exact seasonality of black-carbon deposition and then do the simulations back to the lab.

We appreciate the reviewer's perspective that numerical modeling should be guided and constrained by measurements, but we note a couple of clear counter-arguments to the reviewer's suggestion. First, planning and executing a field mission to Greenland that covers a full annual cycle and various locations is well beyond the scope of this study, not to mention beyond the allocated funding on supporting grants for this work and beyond current capabilities of our lab. Second, our study simulates key quantities that are not directly observable, namely runoff perturbations from the entire Greenland Ice Sheet. The absolute runoff flux from Greenland is estimated only through indirect measurements and techniques, such as subtracting calving estimates from gravitational-based estimates of mass balance (themselves uncertain), and hence the absolute annual estimates are uncertain by at least a factor of two. Estimates of Greenland-wide runoff \*perturbations\* from BC deposition, which are small compared with total absolute runoff, would be impossible to directly observe with current observing capabilities. Modeling is therefore required to derive estimates of this nature. We do cite multiple observationally-constrained estimates of BC deposition flux to Greenland (e.g., Polashenski et al., 2015) and place these estimates in the context of our simulated values. Uncertainty in deposition is another reason why we chose to simulate such a huge range of deposition fluxes -- so that future estimates of BC deposition (derived from either measurements or models) can be extrapolated to Greenland-wide melt perturbations. In short, we agree with the reviewer that more observations of BC deposition to Greenland would be useful, but the types of measurements that would provide constraint of the nature suggested by the reviewer are well beyond the scope of this study.

# Author Response to Reviewer #3-2

"This manuscript deals with BC deposition and total melt runoff of the Greenland Ice Sheet (GrIS). The subject is relevant and important. The study is based solely on model simulation. Some comparison to observed values of BC concentration in snow and ice and to runoff of GrIS were made and discussed. These observations were not connected to each other, as it is impossible to make total glacier melting runoff parametrization based on few BC concentration observations in GrIS to total runoff. The paper is mainly clearly written and it proceeds logically. The abstract stands alone and summarizes the content and the main results of the manuscript. However the methodology presented is computationally based and it's use for parametrization in other models without proper validation by experimental observations is somewhat questionable. I would suggest to concentrate on the modelling results, and not only that of BC, and leave the parametrization out from the paper."

We thank the reviewer's suggestions on parameterization. We agree that the simple parameterizations are lack of validation by experimental observations. However, we should note that it is also challenging to perform experimental validation for these relationships. First, as the reviewer pointed out, there are only few and disconnected BC observations in GrIS. Also, the absolute runoff flux from GrIS is estimated only through indirect measurements, e.g., subtracting calving estimates from gravitational-based estimates of mass balance. Therefore, obtaining observational data that can be used for validation is difficult.

We prefer to keep the simple parameterization, as we aim to use it to summarize the modeling results and to serve as one of the key deliverables of the paper. It is valuable because the parameterization allows for simple and rough estimates of how GrIS-wide runoff perturbations relate to deposition fluxes of light-absorbing particles, without the need to run a complex model. Given the large uncertainties that exist in these relationships, even as simulated with complex models, simple parameterizations that include large error bounds (like those presented) are justifiable and will facilitate more analyses of the extended impacts of aerosol deposition to the GrIS. We also add this explanation in Line 392 that "Given the large uncertainties that exist in the bulk relationships, even as simulated with complex models, simple parameterizations for analyses of the extended impacts of aerosol deposition to the GrIS. We also add this explanation in Line 392 that "Given the large uncertainties that exist in the bulk relationships, even as simulated with complex models, simple parameterizations for a simulated with complex models, simple and this explanation in Line 392 that "Given the large uncertainties that exist in the bulk relationships, even as simulated with complex models, simple parameterizations that include large error bounds are justifiable and will facilitate more analyses of the extended impacts of aerosol deposition to the GrIS."

# 1 Investigating the Impact of Aerosol Deposition on Snow

# 2 Melt over the Greenland Ice Sheet Using a Large-Ensemble

### 3 Kernel

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- 8

# 9 Abstract

10 Accelerating surface melt on the Greenland Ice Sheet (GrIS) has led to a doubling of

11 Greenland's contribution to global sea level rise during recent decades. Black

12 carbon (BC), dust, and other light absorbing impurities (LAI) darken the surface and

13 enhance snow melt by boosting the absorption of solar energy. It is therefore

14 important for coupled aerosol-climate and ice sheet models to include snow

15 darkening effects from LAI, and yet most do not. In this study, we conduct several

16 thousand simulations with the Community Land Model (CLM) component of the

17 Community Earth System Model (CESM) to characterize changes in melt runoff due

18 to variations in the amount, timing, and nature (wet or dry) of BC deposition on the

19 GrIS. From this large matrix of simulations, we develop a kernel relating runoff to

20 the location, month, year (from 2006-2015), and magnitudes of BC concentration

- 21 within precipitation and dry deposition flux. BC deposition during June-August
- 22 causes the largest increase in annually-integrated runoff, but winter deposition

- 23 events also exert large (roughly half as great) runoff perturbations due to re-
- 24 exposure of impurities at the snow surface during summer melt. Current BC
- 25 deposition fluxes simulated with the atmosphere component of CESM induce a
- 26 climatological-mean increase in GrIS-wide runoff of ~8 Gt/yr, or +6.8% relative to a
- 27 paired simulation without BC deposition. We also provide linear equations that
- 28 relate the increase in total runoff to GrIS-wide wet and dry BC deposition fluxes. It
- 29 is our hope that the runoff kernel and simple equations provided here can be used
- 30 to extend the utility of state-of-the-art aerosol models.

#### 31 1 Introduction

32 The Greenland Ice Sheet (GrIS) holds the equivalent of about 7 m of sea level 33 rise (Kintisch, 2017). During recent decades, the accelerating decline of the GrIS has 34 doubled Greenland's contribution to global sea level rise to about 0.74 mm per year 35 (Shepherd et al., 2012; Rignot and Kanagaratnam, 2006; van den Broeke et al., 2009; 36 Kintisch, 2017). Mass loss from the GrIS is predicted to raise sea level by more than 37 20 cm by 2100 (Rignot et al., 2011; Dumont et al., 2014), imposing tremendous 38 effects on global society. 39 Ice loss from the GrIS is caused by many physical and biological factors, 40 including: 1) increase of air temperature over the Arctic region, which accelerates 41 surface melting; 2) declining surface albedo, which can be caused by a variety of 42 effects including increased melt area, enhanced snow metamorphism, and accumulation of light-absorbing impurities (LAI) (Tedesco et al., 2016; Box et al., 43 2012; Dumont et al., 2014; Keegan et al., 2014; Shimada et al., 2016; Polashenski et 44 45 al., 2015); and 3) calving of icebergs and submarine melting due to ice dynamics and 46 changes of ocean temperature (Krabill et al., 2004; Zwally et al., 2002; Dumont et al., 47 2014). Since 2005, surface melting has likely contributed more to ice loss than 48 iceberg calving (Kintisch, 2017). Sources of LAI include black carbon (BC), mineral 49 dust, and algae and bacteria growing on the wet surface of the ice sheet, all of which 50 darken the surface, boost the absorption of insolation and enhance snow melt. 51 BC is a major anthropogenic pollutant originating from fossil fuel combustion,

52 open biomass burning, and biofuel use, and is a key LAI because its solar

53	absorptivity is extremely high (e.g., Bond et al., 2013). Previous climate modeling
54	studies simulate annual warming in the Arctic region with the inclusion of BC in
55	snow (Flanner et al., 2007; Flanner et al., 2009; Hansen and Nazarenko, 2004;
56	Hansen et al., 2005; Jacobson, 2004), and find BC/snow forcing induces a global
57	temperature response about three times greater than equal forcing from $\ensuremath{\text{CO}_2}$
58	(Flanner et al., 2007). BC influences snow coverage by warming the atmosphere,
59	reducing surface insolation through "dimming", and reducing snow reflectance
60	through darkening caused by BC deposition to snow surface. Globally, the darkening
61	effect within snow increases solar heating of snowpack, and exceeds the loss of
62	absorbed energy from dimming, causing a positive net surface forcing and
63	snowpack melting (Flanner et al., 2009). Melting snow also tends to retain BC
64	aerosols, which darken the surface more and increase absorption of insolation (e.g.,
65	Doherty et al., 2013). Therefore, it is important for coupled aerosol-climate and ice
66	sheet models to include BC darkening effects, and yet most do not.
67	Global BC emissions from fossil fuel and biofuel combustion have increased
68	dramatically during the industrial era. Since the early $20^{\mathrm{th}}$ century, BC emissions
69	also shifted spatially, decreasing in North American and Europe but increasing in
70	Asia (Bond et al., 2007; Bond et al., 2013). Both the spatial pattern of emissions and
71	the circulation features that are coincident with emissions can strongly effect the
72	amount of BC reaching the Arctic and depositing to the GrIS (Doherty et al., 2010;
73	Thomas et al., 2017; Jiao et al., 2014).
74	In this study, we develop a BC deposition-snow melt kernel using the

75 Community Earth System Model (CESM) to investigate changes in snow melt and

76 surface runoff due to variations in the amount and timing of aerosol deposition on 77 the GrIS. More than 5000 simulations are conducted with the Community Land 78 Model (CLM) component of CESM, driven with a large range of wet and dry BC 79 deposition fluxes to determine relationships between snow melt perturbation and 80 deposition amount occurring in different months. The final kernel product is 81 resolved by type of BC deposition (wet or dry), deposition amount, deposition 82 month, and deposition year ranging from 2006-2015. It is our hope that this kernel 83 will benefit regional and global aerosol modeling communities by allowing them to 84 estimate GrIS snow melt and surface runoff perturbations associated with different 85 aerosol deposition fluxes.

#### 86 2 Methods

#### 87 2.1 Simulation design

88 We use the Community Earth System Model (CESM) version 1.2.2, and run the 89 offline Community Land Model (CLM) (Oleson et al., 2013) version 4.5 with 90 prescribed wet or dry BC deposition in the snow sub-model to study the effects of 91 BC deposition on snow melt and runoff from Greenland. Our motivation for using 92 CLM in this offline configuration is that (1) it reduces noise associated with ocean 93 and atmosphere variability, enabling clearer identification of the runoff signal 94 caused by snow perturbations, and (2) the simulations are computationally cheap, 95 allowing us to explore a large parameter space. We drive the simulations using 96 meteorological forcing data from the Climatic Research Unit and National Centers 97 for Environmental Prediction (CRUNCEP) (Viovy, 2012) over the period 2006-2016.

98 CRUNCEP data cover the global land surface at a spatial resolution of a  $0.5^{\circ} \times 0.5^{\circ}$ , 99 and are provided with relatively low latency, allowing us to explore conditions over 100 the recent past. In CLM, the representation of terrestrial snow, including over ice 101 sheets, is based loosely on the SNTHERM model (Jordan, 1991) and is described in 102 detail by Oleson et al., 2013. Snow albedo and solar absorption within each snow 103 layer are simulated with the Snow, Ice, and Aerosol Radiative Model (SNICAR), 104 which accounts for solar zenith angle, albedo of the substrate underlying snow, 105 mass concentrations of LAI (black carbon, dust, organic carbon, and volcanic ash), 106 and ice effective grain size. In general, CLM/SNICAR represents five vertical snow 107 layers, and accounts for variations in LAI concentration due to dry and wet 108 deposition, particle flushing and retention with melt water, snow sublimation, and 109 layer combinations and divisions (Oleson et al., 2013). Dry BC deposition from the 110 atmosphere occurs through gravitational settling and turbulent mixing, primarily 111 transferring hydrophobic BC to the surface, while wet BC deposition occurs via 112 precipitation and only affects hydrophilic BC in the model. CLM maintains mass 113 burdens of both hydrophilic BC and hydrophobic BC within each snow layer, with 114 each species having unique optical properties and meltwater removal efficiencies. 115 Deposited particles are assumed to be instantly mixed and homogeneous within the 116 surface snow layer, which does not exceed 2 cm in thickness. The particles are 117 added after the computation of inter-layer water fluxes, thus preventing particles in 118 the top layer from being washed out immediately before radiative calculations. 119 Particle masses are redistributed vertically in each time step proportionately with 120 snow melt through the snow column, scaled by the species-specific melt scavenging

121	efficiency, and snow layer combination and subdivision. The masses carried out
122	with meltwater drainage through the bottom snow layer are permanently lost from
123	the snowpack, and are not maintained within the model (Oleson et al., 2013).
124	For each simulation used to generate the kernel relating melt runoff to BC
125	deposition, we prescribe uniform concentrations of BC within precipitation or
126	uniform dry deposition fluxes over the ice sheet for a period of one month, and
127	quantify perturbations to snow and melt runoff for one year or more following the
128	period of deposition. 23 unique BC concentrations in precipitation (e.g., snow) are
129	prescribed to provide wet BC flux in the CLM. We use a wide range of wet BC
130	concentrations (from 1 ng g $^{\rm -1}$ $\sim$ 500 ng g $^{\rm -1}$ ) with logarithmic spacing, aiming to
131	cover historically observed BC concentrations over the Greenland region (Figure 1)
132	and potential severe episodes associated (e.g.,) with extreme fire activity. For
133	example, measured values of BC in Greenland snow during 2012-2014, collected
134	from snow pits across a long transect, averaged 2.6 ng g $^{-1}$ (Polashenski et al.,
135	2015). Peak values throughout the depth of the snow pits averaged 4 ng g $^{\rm 1}$ and 15
136	ng g $^{-1}$ in 2012 and 2013, respectively, and the largest single measurement was 43 ng
137	$\mathrm{g}^{-1}$ . Ice core measurements from D4 dating back to 1788 show annual-mean
138	concentrations peaking in the early 20th century at ~12.5 ng g <sup>-1</sup> , mean
139	concentrations of 2.3 ng g $^{\rm 1}$ during 1952-2002, and occasional monthly-mean peaks
140	exceeding 50 ng g $^{-1}$ due to biomass burning events (McConnell et al., 2007).
141	Correspondingly, the maximum dry BC deposition flux is calculated as the product
142	of the maximum wet BC concentration (500 ng $g^{-1}$ ) and the mean monthly snowfall
143	(1.75×10 <sup>-5</sup> mm s <sup>-1</sup> ) over an 11-year (2006-2016) CESM simulation. We prescribe 24

144 unique dry BC fluxes (0.01 ng m<sup>-2</sup> s<sup>-1</sup>-8.80 ng m<sup>-2</sup> s<sup>-1</sup>) with logarithmic spacing in our 145 study (Figure 1). 146 With each prescribed BC deposition value, we perform 12 simulations with 147 specified BC deposition month from January to December in each deposition year. 148 Each simulation starts from January in the deposition year, and extends to one full 149 year after the deposition month in order to study the annually integrated BC effects. 150 We then repeat these simulations for ten years (2006-2015) to generate a 151 climatological product and explore the magnitude of interannual variability in melt 152 perturbations due, for example, to differences in precipitation and near-surface air 153 temperatures. In summary, we perform 2,760 simulations (23 wet BC 154 concentrations × 12 deposition months × 10 deposition years) to study the effects of 155 wet BC deposition, and 2,880 simulations (24 dry BC fluxes × 12 deposition months 156 × 10 deposition years) to study the effects of dry BC deposition. 120 simulations (12 157 deposition months × 10 deposition years) without BC deposition are also performed 158 as parallel control/base runs from which to derive the perturbations induced by BC 159 deposition. The timestep of CLM/SNICAR for our simulations is 0.5 hour, and the 60 spatial resolution is 0.9° x 1.25°. The kernel product is calculated as monthly mean 61 values with the same spatial resolution. 162 2.2 Simulation length evaluation based on top-snow layer BC concentration 163 To evaluate the importance of simulation length for the purposes outlined, we focus on BC concentration in the top snow layer because properties of this layer 164

- 165 dominate the bulk solar radiative properties of the snowpack. In Figure 2, we
- 166 examine the mean distribution of BC concentration in the top snow layer over

167 Greenland after maximum wet and dry BC deposition fluxes occur in January and 168 June. These contours are averaged over ten years to show a climatological state. 169 Top-layer BC concentrations after wet BC deposition show higher values in the 170 center and south margin of Greenland (Figure 2a, b, e, f), which matches the general 171 precipitation pattern over the GrIS, with elevated precipitation amount in the center 172 and south of GrIS. Meanwhile, higher top-layer BC values occur over the north of 173 Greenland after dry BC deposition (Figure 2c, d, g, h), as lower precipitation in these 174 regions enhances the effect of dry deposition. Also, we find lower concentrations 175 induced by the maximum wet BC (Figure 2a, b, e, f) than by the maximum dry BC 176 deposition (Figure 2c, d, g, h). Top-layer BC concentration decreases rapidly only 177 one month after the deposition month for both winter and summer deposition (i.e., 178 from January to February with January deposition in Figure 2a-d, and from June to 179 July with June deposition in Figure 2e-h), as new snowfall dilutes the contaminated 180 snow. The rapid decrease in BC concentration supports the notion that a one-year 181 simulation length should capture most of the time-integrated effect from a 182 deposition event. To verify this, we also perform two 11-year (2006-2016) 183 simulations with the maximum wet BC deposition occurring in April and October of 184 the first year, to evaluate the long-term variation of top-layer BC concentration. In 185 these long-term simulations, we find top-layer BC concentrations decrease rapidly 186 within a one-year period after both spring and fall depositions (Figure 3). We also 187 see, however, that residual BC reappears at the surface in subsequent summers 188 because of snow melting and associated accumulation of impurities at the top of the 189 snowpack (Doherty et al., 2013). In the model, this occurs because melt scavenging

190	ratios for both types of BC are less than 1, meaning that proportionately less BC
191	moves down vertically in the column than meltwater. As we will see, summer re-
192	appearance of impurities at the snow top has a non-negligible impact on the
193	annually-integrated runoff perturbation, but the peaks in subsequent years are less
194	than 1/20 of the BC concentrations in the deposition month, indicating that the one-
195	year simulation setup is reasonable to capture the major portion of the total BC
196	influences.

# 197 3 Results and Discussion

Deposited BC to snow causes a darkening effect and enhances snow melt andGreenland runoff. Here, we first show variations of top-snow layer BC

200 concentrations, and then examine total GrIS runoff perturbation induced by BC

201 deposition.

# 202 **3.1 BC concentration in the top snow layer**

203	We investigate the climatological (i.e., averaged over ten simulations covering
204	2006-2015) 1-year evolution of top-snow layer BC concentration averaged over
205	Greenland for different deposition months. Figure 4 depicts the temporal evolution
206	of top-layer BC concentration for the maximum wet and dry BC fluxes from our
207	matrix of simulations. For all months of deposition, top-layer concentrations
208	decrease rapidly as fresh snow covers the contaminated surface. Top-snow layer
209	concentrations of dry-deposited BC generally decrease more slowly than those of
210	wet-deposited BC. One reason for this behavior during summer months is that dry-
211	deposited BC is assumed to be hydrophobic and have a lower meltwater scavenging

212	efficiency, reducing the rate at which it is carried away by melting water. We also
213	see, however, that some winter-deposited BC can persist in the top layer until, or
214	even re-appear during, the summer melt season, indicating that BC deposition in
215	non-melting seasons can also be important for GrIS melting. Re-exposure of
216	previously deposited BC occurs as overlying snow melts and flushes through the
217	snow, removing some but not all of the underlying impurities and eventually
218	exposing the dirty snow layer at the surface.
219	We also investigate how top-layer BC concentration varies with different BC
220	deposition values, and how this variation evolves over one year following
221	deposition (Figure 5 and Figure 6). For all deposition months, top-snow layer BC
222	concentration increases, as expected, with increasing BC deposition flux. The
223	relationship between top-layer BC concentration and BC deposition value is nearly
224	linear, although we find indications of saturation at later times with dry BC
225	deposition (e.g., as in Figure 6h-l). Figure 7 summarizes top-layer BC concentration
226	variation with deposition amount at different times (0-11 months since deposition)
227	averaged over all the deposition months. With this averaging, we find that top-layer
228	BC concentrations decrease monotonically from month 0 to month 11. Again, we
229	find slower decrease in the dry deposition simulations than in the wet deposition
230	simulations. With wet BC deposition, the rate of decrease slows down after 3-6
231	months, which could be due to the summer peaks resulting from snow melting and
232	BC integration (Figure 4).

- **3.2 Temporal variation of GrIS total runoff**
- 234 We define total runoff in CLM as the summation of surface runoff, sub-surface

235	drainage and runoff from glacier surface. We first examine the seasonal and inter-
236	annual variation of GrIS total runoff, integrated over the entire GrIS, in the base run
237	without BC deposition (Figure 8). Total runoff shows clear summer peaks and some
238	inter-annual variation. Annual-integrated total runoff is ${\sim}120$ Gt per year, with the
239	highest value of $\sim$ 140 Gt in 2014 (Figure 8b). Compared with satellite gravity
240	measurements during 2005-2010 showing Greenland is losing mass at a rate of
241	$\sim$ 229 Gt/yr (169-290 Gt/yr), of which 50-70% is lost through surface melt
242	(Vaughan et al., 2013), the simulated total runoff in the base run in our study is
243	within a reasonable range.
244	We then calculate perturbed total runoff due to BC deposition as the difference
245	in runoff between simulations with BC deposition and paired base simulations
246	without BC deposition. We select three wet BC deposition values and three dry BC
247	deposition values to illustrate the seasonal and inter-annual variations of perturbed
248	total runoff (Figure 9). We note that different scales are used in the panels of Figure
249	9, which shows that higher deposition causes more total runoff. We also find clear
250	inter-annual variations for different deposition amounts and months. The variations
251	of perturbed total runoff in Figure 9 follow the variation of total runoff in the base
252	run (Figure 8) in general, especially for the perturbed simulations with wet BC
253	depositions, indicating that BC deposition could have greater effect in warmer years,
254	when more of the ice sheet is near the melting temperature. <u>This could also be due</u>
255	to an amplification effect by metamorphism of snow in warmer years.
1 256	To remove the effect of yearly variation, we also investigate the climatological
257	(i.e., averaged over ten simulations spanning 2006-2015) 1-year evolution of total

258	runoff increase due to	BC deposition	starting from	different de	position months.
			··· · ·		

- 259 Again, we examine total runoff increase induced by maximum wet and dry BC
- 260 depositions to understand general behavior associated with very large
- 261 perturbations, for which the signal is much larger than noise. As shown in Figure
- 262 10, we find two summer peaks of total runoff increase, with the second peak caused
- 263 by the re-exposure of BC during the summer after deposition. We also find total
- 264 runoff perturbation is the largest in July for all months of deposition, followed by
- 265 August, The largest perturbation in summer could be caused by immediate melting
- 266 <u>due to darkening, as well as larger grain size in warmer environment.</u>

#### 267 3.3 GrIS total runoff variation with deposition amount

268 As represented by Figure 9, higher deposition amount causes larger total runoff

- 269 increase. Here, we examine total runoff perturbation caused by all deposition
- amounts, and how total runoff distributes over Greenland. Figure 11 and Figure 12
- 271 show climatological mean (2006-2015) annually integrated total runoff increase
- 272 caused by June BC deposition via wet and dry processes, respectively. June
- 273 deposition is selected for these figures because of its high potential to impact
- 274 summer melt. Although BC is well distributed over the whole GrIS (Figure 2), total
- 275 runoff perturbations are largely confined to the low elevation margins with both
- 276 wet and dry BC deposition (Figure 11 and Figure 12), as these remain the only areas
- 277 warm enough in the model simulations for substantial melt to occur, which is
- 278 consistent with the Tedesco, 2007 study. We note that the spatial distribution of
- 279 model melt will be sensitive to surface air temperature and insolation, the latter of
- 280 which can vary substantially between re-analysis products. Runoff effects of dry BC

Deleted: , due to higher temperature in summer

282	deposition show similar patterns as impacts from BC in precipitation. We find that	
283	small fluxes of BC via dry deposition are not as effective as similar deposition fluxes	
284	of BC in precipitation, and we attribute this to the higher light absorptivity (and	
285	hence radiative forcing per unit mass) of hydrophilic BC than of hydrophobic BC.	
286	The former is assumed to be coated with <b><u>non</u></b> absorbing sulfate and hence its mass	
1 287	absorption cross-section is about $50\%$ larger than that of uncoated hydrophobic BC	
288	(Flanner et al., 2007).	
289	Figure 13 summarizes the variation of total runoff perturbation versus	
290	deposition amount for different deposition months. The maximum BC deposition	
291	perturbs total runoff by up to $\sim$ 20% (24 Gt) in the wet BC deposition simulations,	
292	although we note that continent-wide fluxes of this magnitude (500 ng $\mathrm{g}^{\text{-1}}$ in	
293	precipitation) are much larger than ever observed on Greenland. With a modest	
294	deposition concentration (e.g., 5 ng/g), the runoff perturbation is $\sim$ 0.4% ( $\sim$ 0.5 Gt).	
295	Different deposition months cause annually integrated total runoff perturbation to	
296	vary by 40-60% (12 Gt/yr in the maximum BC deposition case). Also, we find BC	
297	deposition in June and July induces the largest annually integrated total runoff	
298	increase. To further verify the most influential deposition month, we select five wet	
299	BC and five dry BC deposition values, and plot total runoff increase versus	
300	deposition month, shown in Figure 14. Figure 14a shows annual integrated total	
301	runoff from the base run, starting from different months, serving as a reference for	
302	the perturbations shown in Figure 14b-f. For different deposition amounts from low	
303	to high (Figure 14b-f), BC deposition in summer (i.e., June, July and August) causes	
304	the highest annual total runoff increase. With higher deposition (Figure 14e-f), June	

Deleted: weakly

306	deposition generates the largest annual total runoff for both dry and wet BC
307	deposition, whereas with lower deposition amounts the month of maximum impact
308	can be June or July. As noted earlier, however, deposition during non-summer
309	months also causes substantial melt and runoff perturbations, owing to the melt-
310	induced re-surfacing of impurities during summer in the ablation zone.

#### 311 **4** Evaluation and application of the kernel product

312 We turn now to evaluating the kernel product and advising a straightforward 313 way in which it can be applied to realistic aerosol deposition fluxes. We perform ten 314 one-year CLM simulations with spatially and temporally-varying wet or dry BC 315 deposition fluxes occurring in randomly selected years and months, and we evaluate 316 the accuracy of the kernel product using total GrIS BC deposition amount and total 317 runoff from these new simulations (the "evaluating simulations"), compared with 318 total runoff from the kernel for equivalent deposition fluxes. The evaluating 319 simulations use prescribed wet and dry BC deposition fields that were generated 320 from a global aerosol run with CAM (Lamarque et al., 2010). In different evaluating simulations, the prescribed BC deposition fields are multiplied by factors ranging 321 322 from 2-50 for wet BC deposition and 2000-20000 for dry BC deposition to provide 323 different BC deposition amounts within the range of wet or dry deposition in the 324 kernel. High scaling factors are applied to dry/hydrophobic BC deposition 325 simulations because the prescribed dry BC deposition from CAM is very low and 326 within the noise regime. We also perform two ten-year (2006-2015) evaluating 327 simulations with both CAM prescribed wet and dry BC deposition fluxes turned on

328	throughout the whole simulations, to examine short-term, long-term and
329	climatological one-year integrated total runoff effects. BC deposition fluxes in one of
330	the ten-year evaluating simulations are multiplied by a factor of 5 to provide a high
331	deposition event and are left at unperturbed (i.e., realistic) values in the other
332	simulation. Also, different deposition years and months indicate different BC
333	distributions. Therefore, the evaluating simulations provide a variety of scenarios
334	with distinct BC deposition amounts and distributions to evaluate performance of
335	the kernel.
336	Figure 15 shows maximum, mean and minimum one-year integrated total
337	runoff increase versus BC deposition amount from the kernel product, with results
338	from the evaluating simulations overlaid on the kernel lines. With varying BC
339	deposition amount and distribution, including with combined wet and dry BC
340	deposition fluxes, results from the evaluating simulations are mostly within the total
341	runoff ranges of our kernel product for both wet and dry BC deposition. In the
342	evaluating simulations with combined wet and dry BC depositions, hydrophobic BC
343	fluxes are very low (<1/100 of wet BC fluxes), therefore, we treat all BC deposition
344	as wet/hydrophilic BC, and overlay the results on the wet deposition curves. We
345	note CAM simulated BC deposition from the 1 time evaluating run is comparable
346	with measured values of BC in Greenland snow and ice core measurements
347	(Polashenski et al., 2015; McConnell et al., 2007). The increases of one-year
348	integrated total runoff from the ten-year evaluating simulations with realistic
349	deposition exhibit wide ranges, for example, with a minimum of $\sim$ 4 Gt in the first
350	deposition year (2006) to a maximum of $\sim$ 10 Gt, and with a minimum of $\sim$ 14 Gt in

351	the first year to a maximum of ${\sim}40$ Gt in the simulation with 5 times BC fluxes. The	
352	climatological (i.e., averaged over 10 years) total runoff perturbations (~8 Gt/yr and	
353	${\sim}30$ Gt/yr in the default and 5 times simulations, respectively) caused by prescribed	
354	combined wet and dry BC deposition are about double the first-year total runoff	
355	integrations, which is due to continuous month by month depositions in the ten-	
356	year evaluating simulations and the long-term effects of the reappearance of	
357	residual BC at the surface as described in section 2.2.	
358	Because the mean and maximum kernel curves in Figure 15 provide estimates	
359	of BC induced total runoff within a reasonable range, we parameterize these curves	
360	to provide a simple application of our kernel product. GrIS-wide melt perturbations	
361	are relatively linear with BC deposition amount, with linear fits to the mean kernel	
362	curves for wet and dry BC deposition fluxes of, respectively:	
363	$\Delta TOTALRUNOFF = 4.498e5 * \Delta wetBC, $ (1)	
364	$\Delta TOTALRUNOFF = 3.062e5 * \Delta dryBC,$ (2)	
365	and with linear fits to the maximum kernel curves of:	
366	$\Delta TOTALRUNOFF_MAX = 9.322e5 * \Delta wetBC,$ (3)	
367	$\Delta TOTALRUNOFF_MAX = 6.348e5 * \Delta dryBC,$ (4)	
368	where $\Delta$ TOTALRUNOFF is the mean increase in one-year integrated total runoff (kg	
369	yr-1), $\Delta$ TOTALRUNOFF_MAX is the maximum increase in one-year integrated total	
370	runoff (kg yr $^{1}$ ), and $\Delta$ wetBC and $\Delta$ dryBC are total wet and dry BC deposition fluxes	
371	to the GrIS (kg yr <sup>.1</sup> ). Unique relationships for wet and dry deposition arise, again,	
372	because of differences in optical properties and melt-induced removal efficiencies of	
373	hydrophilic and hydrophobic BC. Linearity in the relationship between runoff and	

374	BC deposition is encouraging because we have neglected coincident darkening from
375	other types of LAI (e.g., dust, algae). In environments where darkening from other
376	LAI is not too great, the runoff–BC relationships derived here should be valid,
377	though we acknowledge that the incremental effect of BC will be lower in snow that
378	is heavily laden with other impurities.
379	The bulk relationships shown above represent the simplest application of our
380	kernel product, allowing a rough estimation of runoff perturbation caused by
381	Greenland-wide wet and dry BC deposition fluxes. Based on the evaluations in
382	Figure 15, the mean curves (i.e., equations (1) and (2)) from the kernel product tend
383	to provide a conservative estimate of total runoff, whereas the maximum curves
384	(i.e., equations (3) and (4)) represent a more realistic approximation of total runoff
385	induced by BC deposition that occurs continuously and with varying spatial
386	distribution. Ideally, gridcell-by-gridcell fluxes should be matched to the kernel to
387	account for spatial differences in melt associated with elevation and other
388	conditions (Figure 11 and Figure 12), but at the low end of deposition flux we see
389	considerable noise at the individual pixel and deposition month level, especially for
390	wet deposition fluxes that can be subject to anomalies associated with low
391	precipitation amounts in a given month. To ameliorate this, for typical present-day
392	deposition fluxes we recommend matching month-specific, but spatially-integrated
393	deposition fluxes or concentrations with the associated Greenland-wide kernel
394	values. Given the large uncertainties that exist in the bulk relationships, even as
395	simulated with complex models, simple parameterizations that include large error
396	bounds are justifiable and will facilitate more analyses of the extended impacts of

# 397 <u>aerosol deposition to the GrIS.</u>

398	Finally, although this product only includes BC, we suggest that it could be
399	extended to include other LAI species through a simple scaling that accounts for the
400	ratio of mass-specific absorption between the species of interest and the BC
401	explored here. Rationale for this is that radiative forcing and melt perturbation will
402	scale roughly linearly with absorptivity, at least for relatively low and moderate
403	burdens of LAI. The mass-specific visible band absorption cross-sections assumed
404	in our study for hydrophilic (via wet deposition) and hydrophobic (via dry
405	deposition) BC are 7.5 and 11.3 $m^2g^{\text{-}1}$ , respectively. The only other species-specific
406	property affecting results from this study is the meltwater scavenging coefficient,
407	which is assumed to be 0.2 for hydrophilic BC and 0.03 for hydrophobic BC (Flanner
408	et al., 2007). Given relatively large uncertainty in this parameter (Doherty et al.,
409	2013; Qian et al., 2014), we suggest applying the hydrophilic (wet deposition)
410	kernel to all hydrophilic species and the hydrophobic (dry deposition) kernel to all
411	hydrophobic species, along with appropriate scaling of optical properties.

#### 412 **5** Conclusions

In this study, ~6,000 simulations are performed to investigate annually
integrated runoff perturbation from BC deposition on snow. The simulation matrix
includes variations in deposition flux, deposition month and year, and nature of the
deposition (wet or dry). From this matrix we produce a large-ensemble kernel that
relates BC deposition fluxes to GrIS runoff perturbations.

418 In the month after deposition, top-snow layer BC concentration decreases

419 rapidly due to fresh snow coverage, but then increases somewhat in the ablation 420 zone during the following summer due to melt-induced re-exposure of the 421 contaminated snow. Accordingly, the total runoff increase induced by BC deposition 422 is substantial for both summer and winter deposition, though with peak impacts 423 associated with June and July deposition. Impacts from winter deposition suggest 424 that winter emissions, associated for example with biomass heating use, should not 425 be neglected as potential contributors to increased summer melt. Also, we find most 426 of the runoff increase occurs along the margins, and especially on the southern 427 margin of Greenland, with little sign of melting in the center of the GrIS where it is 428 rarely warm enough. Inter-annual variations in total runoff in the base and BC 429 perturbed simulations indicate that BC deposition can generate more impact in 430 warmer years, when more of the ice surface is near the melting temperature. 431 In summary, higher BC deposition amount leads to higher total runoff. We do 432 not find a clear sign of runoff saturation caused by high BC deposition values in our 433 study. Model-generated deposition fluxes associated with realistic BC emissions 434 induce a climatological-mean (2006-2015) GrIS-wide runoff increase of 8 Gt/yr 435 (+6.8% perturbation), which corresponds to 0.022 mm/year of global sea level rise. 436 We also suggest simple, linear equations that crudely relate GrIS-wide wet and dry 437 BC deposition fluxes to annually-integrated runoff perturbation, and provide links 438 to the full spatially-varying kernel dataset so that users can derive more accurate 439 estimates of melt perturbation (e.g., resolved by seasonal timing) from deposition 440 fluxes. We also suggest that the kernel results can be applied to other LAI species 441 via simple scaling of the mass absorption cross-section. Our hope is that these data

442 will extend the utility of state-of-the-art aerosol models.

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- 448 and are available upon request through e-mail.
- 449

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- 558 559

# 560 Figure Captions





562

563 **Figure 1.** Prescribed values of BC concentrations in precipitation (red) and dry

564 deposition fluxes (blue) applied in the CESM modeling studies. Spacing between

565 values is logarithmic.



568 **Figure 2.** Ten-year (2006-2015) averaged hydrophilic BC concentration associated

569 with wet deposition (a, b, e, f) and hydrophobic BC concentration associated with

570 dry deposition (c, d, g, h) in the top snow layer. The contour maps show

571 concentrations caused by January deposition (a-d) and June deposition (e-h),

572 associated with the maximum wet and dry deposition scenarios depicted in Figure

573 1.





576 **Figure 3.** Temporal evolution of hydrophilic BC concentration in the top snow layer

577 averaged over the Greenland region from two 11-year (2006-2016) simulations

578 with wet BC deposition occurring in April and October of the first year. The BC

579 concentration in precipitation is the maximum value shown in Figure 1. Dashed

580 lines represent the ends of one year since different deposition months.



**Figure 4.** Temporal evolution of hydrophilic BC concentration (a) and hydrophobic

584 BC concentration (b) in the top snow layer averaged over the Greenland region from

 $\,$  the simulations with the maximum wet and dry BC deposition, respectively. Each

586 line starts from the BC deposition month, extends one year, and represents the

mean time series from 10 simulations that start in each year from 2006-2015.





590 **Figure 5.** Hydrophilic BC concentration in the top snow layer vs. concentration of

- 591 BC in precipitation shown at different times (0-11 months, panels a-l) since
- by deposition. The top-snow layer concentrations are averaged over the Greenland
- region and over ten one-year simulations beginning in years 2006-2015. Different
- 594 line colors represent different deposition months.





**Figure 6.** Same as Figure 5, but showing hydrophobic BC concentration in the top

598 snow layer vs. BC dry deposition flux.



605 Figure 7. Hydrophilic BC concentration in the top snow layer vs. concentration of

BC in precipitation (a), and hydrophobic BC concentration in the top snow layer vs.
BC dry deposition flux (b) shown at different times (0-11 months) since deposition.
The top-snow layer concentrations are averaged over the Greenland region, over all
the deposition months and over ten one-year simulations beginning in years 20062015.





614 Greenland in the Base run without BC deposition.



616

617 Figure 9. Monthly timeseries of the increase in total Greenland runoff resulting

618 from BC deposition. Values of BC concentrations in precipitation (a, c, e) and dry

619 deposition fluxes (b, d, f) are shown in each plot. Each line starts from the BC

620 deposition month and extends one year. Different line colors represent different

621 deposition months.



624 **Figure 10.** Temporal evolution of ten-year (2006-2015) averaged total runoff

- 625 increase resulting from BC deposition, summed over the entire Greenland region
- from the simulations with the maximum wet (a) and dry (b) BC deposition. Each line
- 627 starts from the BC deposition month and extends one year.



- 630 Figure 11. Ten-year average of annually-integrated total runoff increase resulting
- 631 from different concentrations of BC in precipitation, deposited only during June.
- The average is over ten one-year simulations starting in each year from 2006-2015.



**Figure 12.** Same as Figure 11, but resulting from different dry deposition fluxes of

636 BC in June.





641 **Figure 13.** Increase in total Greenland runoff resulting from BC deposition

642 integrated over one year starting from the month of BC deposition vs. concentration

of BC in precipitation (a) and vs. BC dry deposition flux (b). The runoff values are

644 summed over the Greenland region and averaged over ten one-year simulations

645 beginning in years 2006-2015. Different line colors represent different deposition

646 months.

647





Figure 14. Total runoff in the Base run without BC deposition (a) and the increase
in total Greenland runoff resulting from BC deposition (b-f), integrated over one
year starting from the month of BC deposition. The bars show mean values and the
whiskers depict the full range of values over all ten simulations that each start in a
different year from 2005-2016. Values of BC concentrations in precipitation and dry
deposition fluxes are shown in the title of each plot (b-f).



657

**Figure 15.** Increase in total Greenland runoff resulting from BC deposition

659 integrated over one year starting from the month of BC deposition vs. total wet (a)

and dry (b) BC deposition mass flux over the entire GrIS. The runoff values are

661 summed over the Greenland region and averaged over all deposition years and

662 deposition months. Black lines show mean total runoff values, and grey lines show

663 maximum and minimum values from the entire matrix of simulations. Red dots

664 represent explicitly-simulated total runoff from the evaluating simulations with

665 prescribed spatially and temporally-varying wet or dry BC deposition fluxes in

666 randomly selected months and years. Blue hollow circles represent simulated total

667 runoff from the evaluating simulations with prescribed combined BC deposition

668 fluxes integrated over each year of the ten-year simulations. Blue dots show

669 averages of the hollow circles.