

1 **Multi-decadal trends in aerosol radiative forcing over the Arctic: contribution of**  
2 **changes in anthropogenic aerosol to Arctic warming since 1980**

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4 Thomas J. Breider, Loretta J. Mickley and Daniel J. Jacob, John A. Paulson School of  
5 Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA

6

7 Cui Ge and Jun Wang, Department of Earth and Atmospheric Sciences, University of  
8 Nebraska – Lincoln, NE, USA

9

10 Melissa Payer Sulprizio, John A. Paulson School of Engineering and Applied Sciences,  
11 Harvard University, Cambridge, MA, USA

12

13 Betty Croft, Department of Physics and Atmospheric Science, Dalhousie University,  
14 Halifax, NS, Canada

15

16 David A. Ridley, Department of Civil and Environmental Engineering, Massachusetts  
17 Institute of Technology, Cambridge, MA, USA

18

19 Joseph R. McConnell, Desert Research Institute, Reno, NV, USA

20

21 Sangeeta Sharma, Environment and Climate Change Canada, Atmospheric Science and  
22 Technology Directorate, Climate Research Division, 4905 Dufferin Street, Toronto, ON,  
23 Canada

24

25 Liaquat Husain and Vincent A. Dutkiewicz, Atmospheric Sciences Research Center,  
26 State University of New York at Albany, Albany, NY USA

27

28

29 Kostantinos Eleftheriadis, Environmental Radioactivity Laboratory, INRaSTES, NCSR

30 Demokritos, Athens, Greece

31

32 Henrik Skov, Department of Environmental Science, Aarhus University, 4000, Roskilde,

33 Denmark

34

35 Phillip K. Hopke, Center for Air Resources Engineering and Science, Clarkson

36 University, Box 5708, Potsdam, NY

37

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39

40 Corresponding author: T. J. Breider, School of Engineering and Applied Sciences,

41 Harvard University, 29 Pierce Hall, Cambridge, MA, 02138, USA

42 ([tbreider@seas.harvard.edu](mailto:tbreider@seas.harvard.edu))

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56 **Key Points**

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58 Observed sulfate and BC mass concentrations at Arctic surface sites and Greenland ice  
59 cores show decreases of 2-3% yr<sup>-1</sup> between 1980-2010

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61 Anthropogenic aerosol cools the Arctic troposphere due to stronger cooling from sulfate  
62 in spring (-1.17 W m<sup>-2</sup>) than previously reported

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64 1980-2010 trends in aerosol-radiation interactions over the Arctic and NH mid-latitudes  
65 contributed 0.27 K warming at the Arctic surface

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87 **Abstract**

88 Arctic observations show large decreases in the concentrations of sulfate and black  
89 carbon (BC) aerosols since the early 1980s. These near-term climate-forcing pollutants  
90 perturb the radiative balance of the atmosphere and may have played an important role in  
91 recent Arctic warming. We use the GEOS-Chem global chemical transport model to  
92 construct a 3-D representation of Arctic aerosols that is consistent with observations and  
93 their trends from 1980 to 2010. Observations at Arctic surface sites and Greenland ice  
94 cores show significant decreases in sulfate and BC mass concentrations of 2-3% per year.  
95 Observations of aerosol optical depth at Resolute Bay in Canada from the 1970s until the  
96 present day show a decrease of  $-0.8\% \text{ yr}^{-1}$  averaged over April, May, and June, but no  
97 significant trend in July and August. We find that anthropogenic aerosols cool the Arctic,  
98 with an average 2005-2010 Arctic shortwave radiative forcing of  $-0.19 \text{ W m}^{-2}$  at the top  
99 of the atmosphere (TOA). Anthropogenic sulfate in our study more strongly cools the  
100 Arctic troposphere in spring ( $-1.17 \text{ W m}^{-2}$ ) than previously reported. From 1980 to 2010,  
101 TOA cooling by Arctic aerosol declined, from  $-0.67 \text{ W m}^{-2}$  to  $-0.19 \text{ W m}^{-2}$ , yielding a net  
102 TOA warming of  $+0.48 \text{ W m}^{-2}$ . The warming is due almost entirely to decreases in  
103 anthropogenic sulfate loading over the Arctic. We estimate that 1980-2010 trends in  
104 aerosol-radiation interactions over the Arctic and Northern Hemisphere mid-latitudes  
105 have contributed a net warming at the Arctic surface of  $+0.27 \text{ K}$ , roughly one quarter of  
106 the observed warming.

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114 **1. Introduction**

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116 Arctic surface temperatures have increased by  $\sim 1.2^{\circ}\text{C}$  since 1980, while annual average  
117 Arctic sea ice coverage has declined by 3.6% per decade [Trenberth et al., 2007; NSIDC,  
118 2015]. The factors driving the observed changes in Arctic surface temperatures and sea  
119 ice extent are uncertain. Recent studies suggest that accounting for changes in near-term  
120 (NT) climate forcing agents (aerosol and ozone) might improve global climate model  
121 simulations of Arctic warming and sea ice loss [Mickley et al., 2004; Shindell, 2007;  
122 Quinn et al., 2008; Yang et al., 2014]. Emissions of primary aerosol and aerosol  
123 precursors have decreased in recent decades in Europe, Russia, and North America due to  
124 air quality concerns, and in situ observations in the Arctic show large declines in  
125 concentrations of sulfate and black carbon (BC) aerosol since 1980, consistent with these  
126 emission trends [Gong et al., 2010; Hirdman et al., 2010]. In this study we use the GEOS-  
127 Chem chemical transport model together with a detailed inventory of historical  
128 anthropogenic emissions to quantify decadal changes in Arctic radiative forcing (RF)  
129 from trends in aerosols species from 1980 to 2010.

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131 Aerosols perturb the radiative balance of the atmosphere through scattering and  
132 absorption of solar radiation, reduction of snow albedo, and alteration of cloud properties  
133 [Twomey, 1974; Clarke and Noone, 1985; Charlson et al., 1992]. The radiative impact of  
134 Arctic aerosols is modulated by the large seasonal amplitude in available radiation, which  
135 peaks in summer [Quinn et al., 2008], and in cloud coverage, which peaks in summer-fall  
136 [Shupe et al., 2011]. Aerosol abundance at remote Arctic stations also shows a distinct  
137 seasonal cycle, with a maximum in late-winter and early-spring known as “Arctic Haze”  
138 and a minimum in summer [Mitchell, 1956; Quinn et al., 2007]. Aircraft measurements  
139 reveal that the aerosol seasonal cycle extends into the Arctic mid-upper troposphere  
140 [Fisher et al., 2011; Wang et al., 2011]. The spring peak in aerosol loading arises from  
141 efficient pollution transport along isentropic surfaces from the mid-latitudes to the Arctic  
142 at this time of year [Rahn and McCaffrey, 1979; Radke et al., 1984; Shaw, 1995; Stohl,  
143 2006].

144

145 Observations of sulfate and BC at remote Arctic stations have shown rapid decreases of  
146 1-3% yr<sup>-1</sup> since the 1980s [Sharma et al., 2004, 2013; Eleftheriadis et al., 2009; Quinn et  
147 al., 2009; Hirdman et al., 2010]. Consistent with these trends, ice cores in Greenland and  
148 Svalbard Island indicate that sulfate deposition peaked in the 1970s, followed by decline  
149 [Goto-Azuma and Koerner, 2001; Isaksson et al., 2001; McConnell et al., 2007]. At  
150 Barrow, Alaska, observed background AOD decreased from 1977 to 2002, but increased  
151 slightly between 2002 and 2010 [Tomasi et al., 2012]. Many studies have attributed the  
152 observed decreases in sulfate and BC in the Arctic to reductions in anthropogenic  
153 emissions of primary aerosol and aerosol precursors in Russia and West Eurasia [Sirois  
154 and Barrie, 1999; Gong et al., 2010; Hirdman et al., 2010; Sharma et al., 2013]. Tomasi et  
155 al. [2012] attributed the recent increase in background AOD at Barrow to emissions  
156 increases in developing countries at lower latitudes. Stone et al. [2014] found an increase  
157 of 0.01 per decade in observed background AOD (500nm) at Barrow, Ny-Alesund, and  
158 Alert over the period 2001 to 2012.

159

160 Previous studies have used model simulations to investigate how anthropogenic aerosols  
161 affect Arctic climate through aerosol effects on radiation, clouds, and surface albedo.  
162 Quinn et al. [2008] found that anthropogenic aerosol warms the Arctic atmosphere and  
163 cools the surface in all seasons, with the largest forcing contribution from BC. Shindell  
164 and Faluvegi [2009] found that changes in aerosols from 1976 to 2007 contributed  
165  $1.09 \pm 0.81^\circ\text{C}$  to the observed Arctic surface temperature increase of  $1.48 \pm 0.28^\circ\text{C}$ , while  
166 Koch et al. [2011] found that reductions in BC in the Arctic since 1980 led to cooling at  
167 the Arctic surface. In contrast, Yang et al. [2014] found no significant response in  
168 average Arctic surface temperatures to the 1975-2005 trends in sulfate and BC. Yang et  
169 al. [2014] did find large regional trends, including a warming in the European Arctic of  
170  $0.6 \text{ K decade}^{-1}$  and a cooling of  $0.6 \text{ K decade}^{-1}$  in the Siberian Arctic.

171

172 In this paper we use GEOS-Chem to simulate multi-decadal trends in distributions of  
173 aerosol concentrations in the Arctic. Our aim is first to achieve a satisfactory  
174 representation of observed trends in NT climate forcings over decadal timescales in the  
175 Arctic, and then to quantify the climatic effects of these forcings. Our work builds on the

176 knowledge gained in previous GEOS-Chem studies that focused on the interpretation of  
177 measurements during the International Polar Year of 2008 [Fisher et al., 2011; Wang et  
178 al., 2011; Breider et al., 2014].

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## 181 **2. Model description**

182

183 We use the GEOS-Chem chemical-transport model version v9-01-03 ([http://geos-](http://geos-chem.org)  
184 [chem.org](http://geos-chem.org)) driven by 6-hourly assimilated Modern Era Retrospective Reanalysis  
185 (MERRA) meteorology from the NASA Global Modeling and Analysis Office. The  
186 original MERRA data are at  $0.5^\circ \times 0.67^\circ$  horizontal resolution but are regridded here to  $4^\circ$   
187  $\times 5^\circ$  to enable multi-decadal simulations. There are 47 vertical layers extending up to 0.01  
188 hPa.

189

190 The GEOS-Chem aerosol simulation includes BC, primary organic carbon (OC), sulfate-  
191 nitrate-ammonium, dust, and sea salt, and is described in detail by Breider et al. [2014].

192 The aerosol simulation is coupled to oxidant chemistry through heterogeneous processes,  
193 aerosol effects on photolysis rates, formation of secondary aerosol, and inorganic nitrate  
194 partitioning. The scheme for dust mobilization accounts for sub-grid variability in wind  
195 speeds [Ridley et al., 2013]. The sea-salt aerosol simulation is described by Jaeglé et al.  
196 [2011], and aerosol optical properties are from Drury et al. [2010]. We use the linearized  
197 ozone climatological parameterization for stratospheric ozone [McLinden et al., 2000].

198 We also include updates to aerosol scavenging in convective updrafts and in cold clouds,  
199 as described in Wang et al. [2014].

200

201 In their GEOS-Chem analysis of Arctic aerosols, Breider et al. [2014] reported an  
202 overestimate of Arctic sulfate mass concentrations at the surface in summer. Croft et al.  
203 [2016] also identified an overestimate of summertime Arctic surface-layer accumulation-  
204 mode aerosol-number concentrations in a version of GEOS-Chem with aerosol  
205 microphysics and these authors pointed to problems in model assumptions of cloud  
206 liquid-water content (LWC) in precipitating low-level clouds. The standard GEOS-Chem

207 specifies LWC in such clouds at a constant  $1.5 \times 10^{-6} \text{ g m}^{-3}$ , although observations suggest  
208 that the LWC in summer over the Arctic is an order of magnitude lower [Shupe et al.,  
209 2001]. In our study, we therefore reduce the June-September cloud LWC in GEOS-  
210 Chem to a uniform  $1 \times 10^{-7} \text{ g m}^{-3}$  north of  $65^\circ\text{N}$  from the surface up to 300 m above the  
211 boundary layer, the region of the atmosphere characterized by low-level precipitating  
212 clouds in summer. By decreasing LWC parameter in the Giorgi and Chameides [1986]  
213 wet removal parameterizations, we increase the removal efficiency and areal fraction of  
214 precipitating clouds over the Arctic, and enhance wet deposition of sulfate in the model.

215  
216 We use the Fu-Luo radiative transfer model (RTM) to estimate all-sky aerosol RF due to  
217 aerosol-radiation interactions over the Arctic [Wang et al., 2008, 2013]. Monthly  
218 averaged aerosol fields and meteorology from GEOS-Chem and surface reflectance data  
219 [Koelemeijer et al., 2003] are applied to the RTM. The forcing calculation is conducted  
220 every 6 hours, consistent with input cloud properties. The RTM includes a treatment of  
221 cloud fraction overlap, ice crystal effective size and single scattering properties, and the  
222 effect of hygroscopic growth on sulfate particle size and refractive index [Liou et al.,  
223 2008; Gu et al., 2011].

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## 226 **2.1 Historical emissions**

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228 Anthropogenic emissions of most primary aerosols and trace gases -- including  $\text{NO}_x$ , CO  
229 and speciated non-methane hydrocarbons -- are taken from the Monitoring Atmospheric  
230 Composition and Climate (MACCcity) inventory for 1980 to 2010 [Granier et al., 2011].  
231 Our preliminary simulations show that the MACCcity  $\text{SO}_2$  emissions fail to reproduce the  
232 steep decline in sulfate concentrations in 1988 observed at Kevo and Ny-Ålesund  
233 (Zeppelin), and we therefore update the  $\text{SO}_2$  emissions inventory following Smith et al.  
234 [2011]. While retaining the seasonal and spatial information from MACCcity, we apply  
235 annual scaling factors to the emissions from every country so that they match the annual  
236 national totals from Smith et al. [2011]. After 2005, the last year in the Smith et al.  
237 [2011] inventory, we scale the MACCcity emissions by the mean 2000-2005 ratio of

238 these two inventories for each country.

239

240 Emissions estimates of anthropogenic BC vary by a factor of 2 in the present day [Bond  
241 et al., 2007; Cohen and Wang, 2014], and emissions uncertainties are even larger in  
242 previous decades owing to scant information on fuel use and technologies used. We  
243 update the MACCcity BC emissions following Cohen and Wang [2014]. Again we retain  
244 the seasonal and spatial information from MACCcity, but scale the annual emissions in  
245 the nine regions specified in Cohen and Wang [2014]. Scale factors are calculated as the  
246 mean 2002-2006 ratio of annual emissions in the two inventories, the period analyzed by  
247 Cohen and Wang [2014], and we assume that these factors do not change over time.

248

249 Figure 1 shows the 1980-2010 timeseries of SO<sub>2</sub> and BC anthropogenic emissions used in  
250 this work, aggregated over key regions. We collectively refer to the former Soviet states  
251 (excluding Russia) and eastern bloc countries (excluding East Germany) as  
252 FSS+EBLOC. Total global emissions of anthropogenic SO<sub>2</sub> decrease by 35 Tg SO<sub>2</sub> (-  
253 20%) between 1980 and 2010. The largest decreases in SO<sub>2</sub> emissions during this time  
254 take place in Europe (80%) and North America (70%). In FSS+EBLOC and Russia,  
255 emissions are almost constant between 1980 and 1988, and subsequently decrease by  
256 50% between 1988 and 2010. Over China, anthropogenic SO<sub>2</sub> emissions increase by a  
257 factor of 3 between 1980 and 2006, followed by a decline.

258

259 Global anthropogenic BC emissions in our study increase linearly from 14 Tg C in 1980  
260 to a peak of 17.5 Tg C in 2005 before leveling off. Regionally, BC emissions show much  
261 smaller temporal trends than SO<sub>2</sub> emissions. BC emissions over Europe and North  
262 America decrease by 20% between 1980 and 2010. The smaller decrease in BC emissions  
263 relative to sulfate is in part due to increasing numbers of diesel cars and trucks which  
264 have higher particulate emissions than gasoline engines [Bond et al., 2007]. BC  
265 emissions in Russia and FSS+EBLOC decrease by 60% between 1980 and 2010, while  
266 BC emissions in China increase by a factor of 2.

267

268 SO<sub>2</sub> emissions from erupting and non-erupting volcanoes are from the AeroCom

269 inventory [Diehl, 2009]. Oceanic emissions of dimethylsulfide (DMS) are calculated  
270 using the DMS seawater climatology from Lana et al. [2011] and the sea-air gas transfer  
271 parameterization from Liss and Merlivat, 1986.

272

273 Constructing a continuous biomass burning inventory for 1980-2010 is challenging. We  
274 use biomass burning emissions from Duncan et al. [2003] for 1980-1996 and the Global  
275 Fire Emissions Database version 3 (GFED3) inventory for 1997-2010 [van der Werf et  
276 al., 2006]. In most regions, the seasonality of the Duncan et al. [2003] inventory is based  
277 on area burned data or on retrievals of Aerosol Index from the Total Ozone Mapping  
278 Spectrometer. Elsewhere we apply the 1997-2010 average seasonality from GFED3 to  
279 the Duncan et al. [2003] inventory. To speciate biomass burning emissions prior to 1997,  
280 we use emission factors from Andreae and Merlet [2001]. GFED3 misses most small  
281 fires [Randerson et al., 2012], so we scale these emissions with the mean seasonality  
282 measured by the Global Fire Assimilation System [Kaiser et al., 2012]. Average 1997-  
283 2010 global fire BC emissions in our inventory are 13% higher than during 1980-1996. A  
284 previous inventory also estimated higher fire emissions in the 1990s compared to  
285 previous decades [Schultz et al., 2008]. It is unclear whether this increase reflects the  
286 uncertainty inherent in our inventory or whether it arises from an actual increase in recent  
287 fire activity at northern latitudes [Westerling et al., 2006].

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289

### 290 **3. Observed and modeled trends of Arctic aerosols**

291

292 We focus our study on changes in sulfate and BC aerosol because long-term  
293 observational records are available for these species and both are considered to be  
294 important drivers of Arctic climate forcing [Quinn et al., 2008] We use arithmetic means  
295 throughout our analysis and define the Arctic as the region north of 60°N. To quantify  
296 long-term trends in the data, we use the Mann-Kendall Sens Slope estimator of the  
297 arithmetic seasonal mass mean concentrations as in Hirdman et al. [2010] and Gong et al.  
298 [2010]. We report only those trends significant at the 95% confidence level. The annual  
299 percentage trend is calculated as the overall trend divided by the average of the first five

300 data points in the timeseries.

301

302

### 303 **3.1. Global and Arctic aerosol budgets**

304

305 Table 1 summarizes the global and Arctic aerosol budgets in our simulations for 1980  
306 and 2006. In 2006, the annual-mean global loading of anthropogenic sulfate is 1.49 mg  
307 m<sup>-2</sup>, down 13% from 1980. The annual-mean anthropogenic BC global load is 0.40 mg  
308 m<sup>-2</sup> in 2006, a factor of 3 higher than the average in the Aerosol Comparison between  
309 Observations and Models (AeroCom) project phase II simulations [Myhre et al., 2013].  
310 The higher anthropogenic BC load in our study is due to our use of the Cohen and Wang  
311 [2014] inventory, with BC emissions more than twice those in AeroCom. Using the  
312 updated scavenging scheme in convective updrafts [Wang et al., 2014], we find a global  
313 lifetime of atmospheric BC of 4.4 days. Our estimate is at the lower end of AeroCom  
314 phase II models, which report a global BC lifetime of 3.9 – 11.9 days [Jiao et al., 2014].  
315 Over the Arctic (i.e., north of 60°N), the total sulfate load in our simulations decreases by  
316 42% from 1980 to 2006, even as global SO<sub>x</sub> emissions decrease just 3%. The large  
317 decrease in the annual-mean Arctic sulfate load can be traced to the shift of pollution  
318 sources from the high northern latitudes to lower latitudes, from which transport to the  
319 Arctic is much less efficient [Stohl, 2006]. The modeled annual-mean Arctic BC load  
320 also decreases by 24% from 1980 to 2010, consistent with declining anthropogenic  
321 emissions in the northern mid-latitudes.

322

323 Figure 2 shows the seasonality of the simulated Arctic load of total BC and sulfate,  
324 averaged over the period between 2001 and 2010. The modeled aerosol load is enhanced  
325 in spring compared to summer by a factor of two for sulfate and by 50% for BC. The  
326 figure also presents the simulated annual mean Arctic load of anthropogenic sulfate and  
327 BC at three different altitudes, averaged over two time periods, 1980-1989 and 2001-  
328 2010. By the 2000s, the annual-mean anthropogenic sulfate load decreases by 50% at the  
329 surface, 39% in the middle troposphere (700-500 hPa) and 20% in the upper troposphere  
330 (500-200 hPa), with the largest changes in spring. The anthropogenic annual-mean BC

331 load decreases by 43% at the surface, but changes little aloft. The largest changes in the  
332 Arctic BC load occur in winter.

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### 335 **3.2 Trends in sulfate and black carbon over source regions**

336

337 We next assess the model capability to reproduce observed trends in sulfate and BC over  
338 two key source regions, North America and Europe (Figure 3). Over North America we  
339 focus our analysis on the eastern part of the US ( $< 90^\circ\text{W}$ ), as that region is most relevant  
340 for transport of pollution to sites in southern Greenland. Over Europe, the observed 1980-  
341 2010 trends in sulfate in surface air ( $-2.8\% \text{ yr}^{-1}$ ) and in precipitation ( $-3\% \text{ yr}^{-1}$ ) are  
342 consistent with Tørseth et al. [2012]. Over Canada non-sea salt (nss) sulfate deposition  
343 shows a decreasing trend of  $-2\% \text{ yr}^{-1}$  from 1984 to 2010, while sites in the eastern United  
344 States show decreases in sulfate in surface air of  $-2.9\% \text{ yr}^{-1}$  from 1992 to 2010. The  
345 observed decreases in sulfate in Europe and North America reflect the influence of air  
346 quality regulations. Table 2 summarizes the observed and modeled annual trends in  
347 sulfate in surface air and in precipitation in Europe and North America. The simulated  
348 percentage trends in sulfate and BC are roughly consistent with the observed trends. In  
349 Europe, the overestimate in surface sulfate and the underestimate in nss-sulfate in  
350 precipitation indicate that the aerosol scavenging is too weak in the region.

351

352

### 353 **3.3 Trends in Arctic aerosol optical depth**

354

355 Long-term measurements of AOD help to constrain trends in light extinction by aerosols  
356 through the atmosphere column. At Resolute Bay, Canada, observed AOD ( $\lambda=700\text{nm}$ )  
357 from the 1970s to the present-day decreased about -25 to -30% averaged over April, May  
358 and June (AMJ), yielding a 1975-2010 linear decrease of  $-0.8\% \text{ yr}^{-1}$  during these months  
359 (Figure 4). Present-day (1997-2010) AOD observations averaged over the eight Arctic  
360 sites in the Aerosol Robotic Network (AERONET) reveal the expected springtime  
361 maximum, consistent with enhanced transport to the region at this time of year (Figure 5).

362 Sampled at these eight sites, the model successfully captures the spring peak and  
363 September minimum, but underestimates AOD by 18% in summer. The summertime bias  
364 is largely due to the failure of the model to capture higher Arctic AOD values over  
365 Canada at that time of year.

366

367

### 368 **3.4 Observed and modeled trends in sulfate and BC at remote Arctic surface**

#### 369 **stations**

370

371 Table 2 summarizes the observed seasonal and annual trends in sulfate and BC for the  
372 Arctic surface sites and ice cores analyzed in our study. Figure 6 shows the observed  
373 mean spring (March-May) and summer (June-August) mass concentrations of sulfate  
374 aerosol during 1980-2010 at five Arctic sites [Heidam et al., 2004; Quinn et al., 2007;  
375 Gong et al., 2010; Laing et al., 2013]. All stations show higher sulfate concentrations in  
376 spring than summer due to more efficient meridional transport, increased surface  
377 stratification, and lower rates of removal by precipitation at that time of year [Stohl,  
378 2006]. The highest nss-sulfate concentrations occur in spring at Kevo in the 1980s, with  
379 values about double the concentrations at other Arctic sites, reflecting its closer proximity  
380 to pollution sources in western Eurasia than other sites. Between 1980 and 2010,  
381 observed sulfate mass in spring decreases by 2-3% yr<sup>-1</sup> at all sites with the exception of  
382 St. Nord, where observations began only in 1991. Taken together, these trends are  
383 consistent with observed trends in sulfate over source regions (Figure 3), indicating the  
384 influence of more stringent restrictions on air quality. In summer, the more remote Arctic  
385 sites (Alert, St. Nord, and Barrow) show no significant trend in observed surface sulfate  
386 mass, which suggests that these sites are not influenced by anthropogenic sources at this  
387 time of year.

388

389 Observed mean sulfate mass concentrations at Arctic surface sites are overestimated in  
390 GEOS-Chem by 4% in spring, while summer mean sulfate is underestimated by 26%. At  
391 Barrow the observations are filtered to remove pollution from local sources, and this may  
392 explain the model overestimate at that site. GEOS-Chem reproduces the observed annual

393 mean trends in nss-sulfate to within 30% at Kevo and Alert. At all sites except Kevo,  
394 natural sources of sulfate from oceanic emissions of dimethylsulfide (DMS) and  
395 volcanoes contribute 28% of the total nss-sulfate mass in spring and over 60% in summer  
396 during 2001-2010. The natural source contribution at Kevo is smaller, 18% in spring and  
397 40% in summer. Observations of SO<sub>2</sub>, the major precursor to sulfate aerosol are  
398 reproduced to within 35% at St. Nord in all seasons except the fall, when the simulation  
399 overestimates the observations by 77%.

400

401 Figure 7 shows the observed mean spring and summer mass concentrations of BC during  
402 1980-2010 at four Arctic sites [Sharma et al., 2004, 2013; Eleftheriadis et al., 2009;  
403 Dutkiewicz et al., 2014]. As with sulfate, BC concentrations are higher in spring than  
404 summer, when anthropogenic BC from northern mid-latitudes is efficiently transported to  
405 the Arctic. The observed BC concentrations during spring at the three sites with long-  
406 term records (> 20 years) show a decreasing trend of between -1.9% yr<sup>-1</sup> and -2.2% yr<sup>-1</sup>,  
407 consistent with the estimated decline in anthropogenic BC emissions in Russia and FSS +  
408 EBLOC (-2.1% yr<sup>-1</sup>). In summer, observed trends in BC are weaker or not significant.  
409 BC concentrations at Kevo are a factor of two higher in spring and a factor of five higher  
410 in summer relative to other Arctic sites, reflecting the close proximity of this site to  
411 pollution sources in western Eurasia. The large drop in BC after 1988 at Kevo has been  
412 attributed to the economic contraction in FSS+EBLOC countries at that time [Dutkiewicz  
413 et al., 2014]. At Kevo, observed trends are underestimated in all seasons except spring,  
414 while at Alert simulated absolute trends are overestimated by 42% in winter (Table 2).  
415 Simulated summertime BC at Barrow shows large interannual variability due to the effect  
416 of fire activity, an effect not seen in the observations. This mismatch may be explained  
417 by the filtering of observations at Barrow, which removes data points when the hourly  
418 average wind direction is between 131° and 359°.

419

420 Ice core records in the Arctic provide a valuable constraint on historical trends in sulfate  
421 and BC deposition [Goto-Azuma and Koerner, 2001; McConnell et al., 2007]. In Figure 8  
422 we compile the observed long-term trends in sulfate and BC in Greenland ice cores and  
423 compare these with the observed trends at Arctic surface sites and monitoring stations in

424 Europe and North America. Long-term observations show decreases in annual mean  
425 sulfate at all sites of between 2 and 3% yr<sup>-1</sup>. The largest observed surface-air trends are  
426 found over Europe (-3.0% yr<sup>-1</sup>), and these are consistent with trends in the remote Arctic  
427 at Alert (-2.9% yr<sup>-1</sup>). Trends in ice core sulfate in southern Greenland are similar to those  
428 observed in surface-air in Acadia National Park, USA and in Ifaross, Iceland, ~2% yr<sup>-1</sup>.  
429 Arctic BC observations show decreasing trends of 1.7-2.8% yr<sup>-1</sup> at all locations, with the  
430 exception of the ice core at D4 in central Greenland where there is no significant trend.

431

432 Consistent with these observations, GEOS-Chem shows 1980-2010 decreases of 2-3% in  
433 both surface concentrations and deposition fluxes of sulfate and BC over much of the  
434 high northern latitudes and the Arctic (Figure 8). The strongest trends occur over Europe,  
435 FSS+EBLOC, and Russia. The model yields no significant trend in BC concentrations  
436 over Canada or Alaska, where increasing emissions from boreal fire activity appear to  
437 cancel out the effect of decreasing anthropogenic emissions. Simulated concentrations of  
438 both sulfate and BC increase by ~1% yr<sup>-1</sup> over the Pacific due to increasing  
439 anthropogenic emissions in East Asia. Simulated AOD also decreases over much of the  
440 Arctic, especially in spring, but at much slower rates than the observations of about 1%  
441 yr<sup>-1</sup>. GEOS-Chem shows a decrease in AMJ AOD at Resolute Bay of -0.6% yr<sup>-1</sup> for  
442 1980-2010 to the observed trend of -0.8% yr<sup>-1</sup> (Figure 5).

443

444 Records of ice-core BC show 1980-2010 decreases of -2.7% yr<sup>-1</sup> in southern Greenland  
445 and -1.6% yr<sup>-1</sup> in northern Greenland, but no significant trend in central Greenland. Using  
446 GEOS-Chem, we find that trends in anthropogenic emissions yield a 1980-2010 decrease  
447 in BC deposition of -1.6% yr<sup>-1</sup>, averaged over the Arctic.

448

449

### 450 **3.5 Observed and modeled changes in aircraft measurements**

451

452 Aircraft observations have provided sporadic measurements of trace species in the Arctic  
453 since the early 1980s. Figure 9 compiles observations of sulfate aerosol over Arctic  
454 Canada and Alaska from the Arctic Boundary Layer Expedition (ABLE-3A, 1988), the

455 Arctic Haze campaign (1995), the Tropospheric Ozone Production about the Spring  
456 Equinox experiment (TOPSE, 2000) and the Arctic Research of the Composition of the  
457 Troposphere from Aircraft and Satellites campaign (ARCTAS, 2008). We compare  
458 GEOS-Chem with ARCTAS observations by sampling the model along the aircraft flight  
459 track. Simulated sulfate concentrations in spring 2008 are captured to within 15% of the  
460 observations at all altitudes, while surface sulfate in summer 2008 is overestimated by a  
461 factor of two.

462

463 Aircraft sulfate observations over Alaska during April show a 30% increase in the middle  
464 troposphere between 1995 and 2008, but little change below 2km. GEOS-Chem attributes  
465 the elevated sulfate over Alaska in 2008 to similar enhancements in both natural and  
466 anthropogenic sulfate. Over Arctic Canada, sulfate remains roughly constant at all levels  
467 between April 2000 and 2008. Below 2km across all time periods, springtime sulfate  
468 concentrations are 20% higher over Alaska than over Arctic Canada, and GEOS-Chem  
469 attributes two-thirds of this enhancement to a larger influence from natural sources in  
470 Alaska.

471

472 In June-July, aircraft observations of sulfate in the lower troposphere show similar values  
473 over Alaska in 1988 and Arctic Canada in 2008, in agreement with surface observations  
474 at Alert and St. Nord. In the middle troposphere, however, summertime mean sulfate  
475 concentrations are 22% lower over Canada in 2008 than in 1988 over Alaska. GEOS-  
476 Chem shows a similar 29% difference in sulfate in the middle troposphere in summer  
477 2008 over Canada, compared to 1988 over Alaska. This difference can be explained in  
478 roughly equal parts by the greater influence of natural sources in Alaska (Volcanoes and  
479 DMS) and by greater anthropogenic SO<sub>2</sub> emissions in 1988.

480

481

### 482 **3.6 Reconciling observed trends in surface and column aerosol**

483 Observed sulfate concentrations in surface air in spring show decreases of 2-3% yr<sup>-1</sup>  
484 throughout the Arctic from 1980-2010, while AOD decreases by only about 0.5% yr<sup>-1</sup>  
485 (Figure 4). To understand the difference in the observed surface and column trends, we

486 use GEOS-Chem to calculate the contributions of regional changes in anthropogenic  
487 emissions to the total change in AOD as a function of altitude for the 1980-2010 time  
488 frame for the Arctic as a whole. For each layer of AOD, we aggregate the changes into  
489 two regional categories: (1) regions where SO<sub>2</sub> emissions have decreased since 1980  
490 (West Eurasia, Russia, and North America) and (2) all other regions including China,  
491 where SO<sub>2</sub> emissions have increased since 1980.

492 We find that the 1980-2010 modeled decrease in total AOD in the Arctic is driven by a  
493 combination of strongly decreasing aerosol load at low altitudes and increasing aerosol  
494 load at high altitudes (Figure 10). Over the 30-year period, emissions changes in West  
495 Eurasia, Russia, and North America yield a ~30% decrease in AOD specific to these  
496 regions in the lower troposphere, with less pronounced decreases aloft. In contrast,  
497 emissions changes in China and elsewhere yield ~20% increases in region-specific AOD  
498 in the upper troposphere, with little effect in the mid- to lower troposphere. The altitude  
499 dependence of these trends reflect the transport pathways of aerosol from source regions,  
500 with the dominant influence from the northern high latitudes at the surface and from  
501 lower latitudes aloft [Stohl, 2006; Fisher et al., 2010]. The net AOD change in each layer  
502 of the troposphere reflects the competition between these two trends, with ~25%  
503 decreases in the lower troposphere and ~15% increases in the upper troposphere. The  
504 absolute change in total AOD is strongly weighted towards changes in the lower  
505 troposphere (Figure 10).

506

#### 507 **4. Arctic aerosol radiative forcing between 1980 and 2010.**

508

509 Applying GEOS-Chem aerosols to the Fu-Luo radiative transfer code, we find that  
510 anthropogenic aerosol cools the Arctic troposphere. Average 2005-2010 top of  
511 atmosphere (TOA) RF is  $-0.19 \text{ W m}^{-2}$  (Figure 11), reflecting the balance between cooling  
512 from sulfate ( $-0.60 \text{ W m}^{-2}$ ) and warming from BC ( $+0.44 \text{ W m}^{-2}$ ). Average TOA forcing  
513 from anthropogenic OC is small,  $-0.03 \text{ W m}^{-2}$ . Surface RF from atmospheric  
514 anthropogenic aerosol averaged over the Arctic during 2005-2010 is  $-1.20 \text{ W m}^{-2}$ , due to  
515 roughly equivalent cooling from sulfate and BC. Our results stand in contrast to Quinn et

516 al. [2008], who found that the interaction of radiation with present-day, anthropogenic  
517 aerosol warms the Arctic by  $0.92 \text{ W m}^{-2}$  in spring. The difference between these two  
518 results can be traced to the smaller sulfate forcings in Quinn et al. [2008], leading to the  
519 dominance of BC in total RF. In the model used by Quinn et al. [2008], the springtime  
520 sulfate burden is enhanced by 40% compared to summer (K. Tsgaraidias, personal  
521 communication), while we find a doubling of sulfate at that time of year (Figure 2).

522

523 To assess the contribution of aerosol trends to recent Arctic climate change, we examine  
524 the net 1980-2010 RF due to anthropogenic aerosol, calculated as the difference between  
525 the mean 2005-2010 forcings and mean 1980-1985 forcings. At TOA, we calculate an  
526 annual mean, net Arctic warming of  $+0.48 \text{ W m}^{-2}$  over this time period (Figure 12),  
527 reflecting the trend from strong aerosol cooling ( $-0.67 \text{ W m}^{-2}$ ) in the 1980s to more  
528 moderate aerosol cooling in the present-day ( $-0.19 \text{ W m}^{-2}$ ). The 1980-2010 decline in  
529 anthropogenic sulfate contributes a net TOA RF of  $+0.51 \text{ W m}^{-2}$ , while the net TOA RF  
530 due to changes in anthropogenic BC is small ( $-0.04 \text{ W m}^{-2}$ ). At the surface, the net 1980-  
531 2010 Arctic RF from anthropogenic aerosol is even larger,  $+0.70 \text{ W m}^{-2}$ . Previous studies  
532 have suggested that Arctic climate may respond not just to local forcings but also to  
533 remote TOA forcing [Shindell, 2007]. For the 1980-2010 period over mid-latitudes (28-  
534  $60^\circ\text{N}$ ), we estimate a net TOA forcing  $+0.31 \text{ W m}^{-2}$  due to anthropogenic sulfate and,  
535  $+0.14 \text{ W m}^{-2}$  due to anthropogenic BC.

536

537 A rough assessment of the contribution of aerosol trends to the observed Arctic warming  
538 since 1980 requires knowledge of the climate sensitivity ( $\lambda$ ) in the Arctic to both local  
539 and remote TOA forcings. Climate sensitivity provides a measure of the response in  
540 surface temperatures (degrees K) to a given change in TOA RF ( $\text{W m}^{-2}$ ). Here we follow  
541 Shindell and Faluvegi [2009], who used a global model to derive climate sensitivities in  
542 the Arctic due to aerosol-radiation interactions for different aerosol species. For local  
543 forcings, Shindell and Faluvegi [2009] estimated sensitivities of  $+0.36 \text{ K (W m}^{-2}\text{)}^{-1}$  for  
544 sulfate and  $-0.08 \text{ K (W m}^{-2}\text{)}^{-1}$  for BC. For mid-latitude (28- $60^\circ\text{N}$ ) forcings, they estimated  
545 sensitivities of  $+0.20 \text{ K (W m}^{-2}\text{)}^{-1}$  for sulfate and  $+0.15 \text{ K (W m}^{-2}\text{)}^{-1}$  for BC.

546

547 We apply the Shindell and Faluvegi [2009] sensitivities to the values reported here for  
548 Arctic and mid-latitude TOA RF from anthropogenic sulfate and BC. We estimate that  
549 between 1980 and 2010, trends in anthropogenic aerosols contribute +0.27 K warming to  
550 average Arctic surface temperatures, or approximately 25% of the observed Arctic  
551 warming during this time period. Changes in Arctic RF contribute +0.19 K of this  
552 warming, while changes in mid-latitude RF provide +0.08 K additional warming. The  
553 temperature response due to changes in sulfate is +0.18 K for Arctic sulfate, and +0.06 K  
554 mid-latitude sulfate. For BC, changes in the mid-latitudes provide an Arctic temperature  
555 response of +0.02 K, while changes in BC in the Arctic contribute a warming of +0.01 K.

556

557

#### 558 **4.6 Increased BC RF efficiency in the Arctic between 1980 and 2010**

559

560 Annual BC TOA RF in the Arctic decreased by just  $0.04 \text{ W m}^{-2}$  between the early 1980s  
561 and the late 2000s, while the annual Arctic BC load declined by 33%, from  $0.42 \text{ mg m}^{-2}$   
562 to  $0.28 \text{ mg m}^{-2}$  over the same period. The larger relative decrease in annual BC load  
563 compared to the TOA RF from BC indicates that the BC forcing efficiency increased  
564 over this period. We find that Arctic BC forcing efficiency increased from  $1.07 \text{ W m}^{-2}$   
565  $(\text{mg m}^{-2})^{-1}$  in the early 1980s, to  $1.5 \text{ W m}^{-2} (\text{mg m}^{-2})^{-1}$  in the late 2000s.

566

567 The increase in Arctic BC forcing efficiency is explained by the shifting influence of  
568 regional sources. High latitude sources, particularly those within the polar dome, are  
569 efficiently transported to the Arctic surface in winter and early spring, while lower  
570 latitude sources in Asia influence the Arctic free troposphere in mid-late spring [Stohl,  
571 2006]. Declining BC emissions at high latitudes have contributed to a lower BC load at  
572 the Arctic surface in winter when incident solar radiation is small, while growing BC  
573 emissions in East Asia have contributed to a higher BC load in mid-late spring when  
574 there is ample incident solar radiation. This re-distribution of BC in the Arctic from  
575 winter to spring and from lower to higher altitudes -- explains the larger BC forcing  
576 efficiency per unit mass in the present day compared to 1980.

577

578 We find no change in the sulfate Arctic TOA RF efficiency. Formation of sulfate aerosol  
579 from high latitude sources is inefficient in winter, due to limited availability of oxidants  
580 [Gong et al., 2010].

581

582

## 583 **5. Discussion and conclusions**

584

585 We have used the chemical transport model GEOS-Chem to construct a 3-D  
586 representation of Arctic aerosols that is consistent with observed distributions and 1980-  
587 2010 aerosol trends. Our goal was to quantify the potential contribution of trends in  
588 anthropogenic aerosols to the rapid Arctic warming ( $> 1$  K) observed over this timeframe.  
589 We focused only on the climate response to aerosol-radiation interactions and used the  
590 Fu-Luo radiative transfer model together with published estimates of the sensitivity of  
591 surface temperatures to regional aerosol radiative forcings [Shindell and Faluvegi, 2009].

592

593 We find that most Arctic surface and ice-core observations show decreases in sulfate and  
594 BC concentrations of  $2\text{-}3\% \text{ yr}^{-1}$ , consistent with estimated emissions reductions in  
595 Eurasia and North America. Observed springtime (April to June) Arctic AOD at Resolute  
596 Bay, Canada decreases over 1975-2010, but the trend is relatively weak ( $0.8\% \text{ yr}^{-1}$ ). The  
597 weakness of this trend can be traced to the competing influences of decreasing aerosol  
598 load in the lower troposphere, driven by emissions trends at high northern latitudes, and  
599 increasing aerosol in the mid- to upper troposphere, driven by trends in China and other  
600 developing countries at lower latitudes.

601

602 Our results show that anthropogenic aerosol cools the Arctic, with a TOA RF of  $-0.19 \text{ W}$   
603  $\text{m}^{-2}$  in the present-day (2005-2010) and even greater forcing ( $-0.67 \text{ Wm}^{-2}$ ) in the early  
604 1980s. We find that the 1980-2010 emission reductions in anthropogenic aerosols in the  
605 developed world may have contributed  $+0.27 \text{ K}$  warming to present-day Arctic  
606 temperatures at the surface, or approximately 25% of the observed Arctic warming.  
607 About two-thirds of the warming can be attributed to forcing from anthropogenic aerosols  
608 over the Arctic, with the rest from forcing over mid-latitudes.

609

610 Our results differ from those of Quinn et al. [2008], who calculated a net warming  
611 influence of present-day aerosols on Arctic climate. The Quinn et al. [2008] model,  
612 however, yields a relatively small enhancement of sulfate load in spring compared to  
613 summer, in contradiction with the seasonality inferred from aircraft and AOD  
614 observations. In their multi-model study, Shindell and Faluvegi [2009] reconstructed the  
615 influence on Arctic climate of the 1976-2007 trends in both local and mid-latitude aerosol  
616 loads. They found that aerosols may have warmed the Arctic surface over this timeframe  
617 by as much as  $1.09 \pm 0.81$  K, or about three times what we calculate for 1980-2010. Their  
618 study however, inferred zonally averaged, mid-latitude aerosol forcings as large as +1 to  
619  $+5 \text{ Wm}^{-2}$  for the 1976-2007 timeframe, values that seem inconsistent with observed  
620 aerosol trends. Finally, Yang et al. [2014] found significant warming over the European  
621 Arctic in response to recent aerosol trends, but no significant trend in surface  
622 temperatures over the Arctic as a whole. Their study, which focused on the 1975-2005  
623 period, estimated similar declines in sulfate AOD over the Arctic to our study, and unlike  
624 our study reported slight increases in Arctic BC AOD during this timeframe.

625

626 By comprehensively testing GEOS-Chem aerosol trends against a large suite of observed  
627 trends, our conclusions gain greater credibility than previous studies. However, our use  
628 of a simple, offline method to estimate the temperature response to TOA forcing yields  
629 only a rough estimate of the influence of aerosol trends on Arctic surface temperatures.  
630 Our approach also focuses only on aerosol-radiation interactions, and does not take into  
631 account aerosol-cloud interactions, the BC-albedo effect, or changes in greenhouse gases.  
632 Within these limitations, our study suggests that air quality measures aimed at reducing  
633 particulate pollution have inadvertently accelerated warming of the Arctic over the past  
634 30 years.

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639

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924 **Figure 1.** Timeseries of total anthropogenic emissions of (top) SO<sub>2</sub> and (bottom) black  
925 carbon (BC), classified by region or country. The plot apportions emissions from the  
926 former Soviet Union and the Eastern Bloc countries into two categories: (1) Russia and  
927 (2) all other former Soviet states (FSS) and eastern bloc countries (excluding East  
928 Germany), which we collectively refer to as FSU+EBLOC.

929

930 **Figure 2.** (Top) Seasonal cycle in the simulated load of sulfate (black) and BC (brown) in  
931 the Arctic (>60°N), averaged over the period 2001-2010 in units of mg m<sup>-2</sup>. Vertical bars  
932 show one standard deviation of the interannual mean. The other two panels compare the  
933 simulated mean Arctic loads of anthropogenic sulfate (middle) and BC (bottom) for  
934 1980-1989 vs. 2001-2010. Loads are binned by altitude and the black horizontal bars  
935 show one standard deviation of the interannual mean in each bin.

936

937 **Figure 3.** Trends in annual mean sulfate concentrations in surface air and in precipitation  
938 at sites in Europe and North America. Observations in all panels are shown as black  
939 circles, with black bars denoting the standard deviation of the annual means. Stacked  
940 contours represent the anthropogenic (blue) and natural (green) contributions to the  
941 modeled values. (a) Trend in total sulfate in surface air, averaged over the 14 EMEP sites  
942 with at least 25 years of data between 1980 and 2010. (b) Trend in total sulfate at the 10  
943 IMPROVE stations on the US East Coast (<90°W) with at least 14 years of data between  
944 1992 and 2010. (c) Trend in nss-sulfate in precipitation at the 19 EMEP stations with at  
945 least 24 years of data between 1980 and 2010. (d) Trend in nss-sulfate in precipitation at  
946 the 14 CAPMON stations in Canada with at least 20 years of data between 1984 and  
947 2010.

948

949 **Figure 4.** Mean seasonality of AOD at 700 nm at Resolute Bay, Canada (75°N, 95°W)  
950 for 1970-1979 (black) and 2004-2014 (red). Vertical bars show one standard deviation of  
951 the mean. Only those monthly averages with at least three data points are shown.  
952 Observations for 1970-1979 are from McGuiffe et al. [1985]. Observations for 2004 to  
953 2014 are from AERONET Version 2 data.

954

955 **Figure 5.** Seasonal variation of AOD, averaged over 1997-2010 at eight AERONET  
956 stations in the Arctic. Black circles denote observed monthly mean AOD, and vertical  
957 bars show one standard deviation of the means. To remove the influence of large volcanic  
958 eruptions in September 2008 and 2009, we exclude these months in the September mean.  
959 Monthly means in the plot also do not include outliers, here defined as those months  
960 when AOD is greater than 2.7σ from the mean. This filtering removes 14% of the 234-  
961 available observations. Stacked contours represent the contributions from different  
962 aerosol types to modeled AOD at 500 nm. White dashed line shows the modeled AOD  
963 contribution from natural sources.

964

965 **Figure 6.** Observed nss-sulfate surface concentrations at five Arctic sites in spring  
966 (March-May) and summer (June-August). Seasonal means are denoted by solid black  
967 circles, medians as short horizontal bars, and the 25<sup>th</sup> to 75<sup>th</sup> percentile ranges as vertical  
968 bars. The observations shown by the solid black circles is provided by the following  
969 sources; Alert (Canadian National Atmospheric Chemistry (NAtChem) Database and  
970 Analysis System); St. Nord and Ny-Alesund-Zeppelin (European Monitoring and  
971 Evaluation Programme (EMEP)); Barrow (NOAA PMEL); and Kevo [Laing et al.,  
972 2013]). Black triangles early in the record at Barrow are from Rahn and MacCaffrey  
973 [1980] and Li and Winchester [1989]. Black triangles at Ny-Ålesund for the period  
974 1979-1981 shows mean observations from Heitzenberg et al. [1983]. Black diamond at  
975 Ny-Alesund in summer shows median nss-sulfate concentration from Maenhaut et al.  
976 [1989]. Open circles in the spring panel for Ny-Ålesund are mean March-April values  
977 [Sirios and Barrie, 1999]. Stacked contours represent the anthropogenic (blue) and  
978 natural (green) contributions to the modeled concentrations. Note that the y-axis range  
979 differs among panels.

980

981 **Figure 7.** Same as Figure 6, but for surface BC at four Arctic sites. The data sources are  
982 as follows: Sharma et al. [2004, 2006, 2013] at Alert, Eleftheriadis et al. [2009] at Ny-  
983 Alesund-Zeppelin, NOAA Earth System Research Lab at Barrow, and Dutkiewicz et al.  
984 [2014] at Kevo.

985

986 **Figure 8.** Trends in annual mean sulfate and BC concentrations in surface air and in  
987 precipitation, as well as in annual mean and April-May-June (AMJ) AOD. Circles  
988 indicate observed trends at a range of sites over different time periods: small white circles  
989 denote trends at sites with greater than 25 years of observations; large white circles show  
990 average long-term trends across multiple sites in Europe and North America; yellow  
991 circle, 1976-2002; and red circles 1990-2009. At Ifaross, Iceland, we remove the  
992 influence of volcanic sulfate as described in Figure 5. Observed AOD at Resolute Bay,  
993 Canada, indicates the mean April-May-June value. GEOS-Chem results are shown as  
994 background contours. White indicates regions where the 1980-2010 trend is statistically  
995 insignificant or is less than  $0.5\% \text{ yr}^{-1}$ .

996

997 **Figure 9.** Aircraft profile observations of sulfate in the Arctic in spring and summer.  
998 Sulfate observations are shown for ARCTAS in 2008 [Fisher et al., 2011], TOPSE in  
999 2000 [Scheuer et al., 2005], Arctic Haze in 1995 [Jaeschke et al., 1999], and ABLE-3A in  
1000 1988 [Talbot et al., 1992]. ARCTAS-A sampled in April, and so only TOPSE April  
1001 flights are used in the comparison. For ARCTAS and TOPSE, we remove outliers that are  
1002 more than 1.5 interquartile ranges below the first quartile or above the third quartile in  
1003 each altitude bin. The filtering removes 6% of the 2656 available observations. The box-  
1004 whisker plots show the minimum, 25<sup>th</sup> percentile, median, 75<sup>th</sup> percentile, and maximum  
1005 values for ARCTAS and TOPSE. The Arctic Haze campaign provides data collected  
1006 during only three flights from Deadhorse, Alaska, in April [Jaeschke et al., 1999]. For the  
1007 Arctic Haze and ABLE-3A campaigns, we show only the mean and  $\pm 1$  standard  
1008 deviation.

1009

1010 **Figure 10.** Simulated 1980-2010 changes in annual mean layer AOD as a function of  
1011 altitude, averaged over the Arctic (>60°N). The panels show percent change (left) and  
1012 absolute change (right) in AOD resulting only from trends in anthropogenic aerosol. The  
1013 AOD layers correspond to model layers, and range in thickness from 125 m at the surface  
1014 to 1000 m in the upper troposphere. Dark blue curves indicate AOD changes due to  
1015 sources in West Eurasia, Russia and North America, and red curves denote changes due  
1016 to sources in China and elsewhere. Light blue curves represent the net change of AOD in  
1017 each layer, and the light blue triangle is the simulated percent change in total Arctic  
1018 AOD.

1019  
1020 **Figure 11.** Seasonal and annual mean top-of-atmosphere radiative forcings (TOA RFs)  
1021 due to aerosol, averaged over the Arctic. The top panel shows RFs due to anthropogenic  
1022 aerosol calculated by this study for 2005-2010 (black) and as reported by Quinn et al.  
1023 [2008] for 2003 (red). The lower panel shows the sulfate and BC contributions to the RFs  
1024 in the top panel, as calculated by this study and by Quinn et al. [2008].

1025  
1026 **Figure 12.** Comparison of the seasonal and annual mean TOA RFs due to anthropogenic  
1027 aerosol during 1980-1985 and 2005-2010, averaged over the Arctic. The bottom panel  
1028 presents the net RFs due to the trends in sulfate and BC anthropogenic aerosol, calculated  
1029 as the difference between the mean 2005-2010 and 1980-1985 forcings.  
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