

1 **Wildfires in Northern Eurasia affect the budget of black**  
2 **carbon in the Arctic. A 12-year retrospective synopsis**  
3 **(2002–2013).**

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1 **Abstract**

2 In recent decades much attention has been given to the Arctic environment, where climate  
3 change is happening rapidly. Black carbon (BC) has been shown to be a major component of  
4 Arctic pollution that also affects the radiative balance. In the present study, we focused on  
5 how vegetation fires that occurred in Northern Eurasia during the period of 2002–2013  
6 influenced the budget of BC in the Arctic. For simulating the transport of fire emissions from  
7 Northern Eurasia to the Arctic, we adopted BC fire emission estimates developed  
8 independently by GFED3 (Global Fire Emissions Database) and FEI-NE (Fire Emission  
9 Inventory - Northern Eurasia). Both datasets were based on fire locations and burned areas  
10 detected by MODIS (MODerate resolution Imaging Spectroradiometer) instruments on  
11 NASA's (National Aeronautics and Space Administration) Terra and Aqua satellites.  
12 Anthropogenic sources of BC were adopted from the MACCity (Monitoring Atmospheric  
13 Composition & Climate / megaCITY - Zoom for the ENvironment) emission inventory.

14 During the 12-year period, an average area of 250,000 km<sup>2</sup> yr<sup>-1</sup> was burned in Northern  
15 Eurasia (FEI-NE) and the global emissions of BC ranged between 8.0 and 9.5 Tg yr<sup>-1</sup> (FEI-  
16 NE+MACCity). For the BC emitted in the Northern Hemisphere (based on FEI-  
17 NE+MACCity), about 70% originated from anthropogenic sources and the rest from biomass  
18 burning (BB). Using the FEI-NE+MACCity inventory, we found that 102±29 kt yr<sup>-1</sup> BC was  
19 deposited in the Arctic (defined here as the area north of 67°N) during the 12 years simulated,  
20 which was twice as much as when using MACCity inventory (56±8 kt yr<sup>-1</sup>). The annual mass  
21 of BC deposited in the Arctic from all sources (FEI-NE in Northern Eurasia, MACCity  
22 elsewhere) is significantly higher by about 37% in 2009 (78 versus 57 kt yr<sup>-1</sup>) to 181% in  
23 2012 (153 versus 54 kt yr<sup>-1</sup>), compared to the BC deposited using just the MACCity emission  
24 inventory. Deposition of BC in the Arctic from BB sources in the Northern Hemisphere thus  
25 represents 68% of the BC deposited from all BC sources (the remaining being due to  
26 anthropogenic sources). Northern Eurasian vegetation fires (FEI-NE) contributed 85% (79–  
27 91%) to the BC deposited over the Arctic from all BB sources in the Northern Hemisphere.

28 We estimate that about 46% of the BC deposited over the Arctic from vegetation fires  
29 in Northern Eurasia originated from Siberia, 6% from Kazakhstan, 5% from Europe, and  
30 about 1% from Mongolia. The remaining 42% originated from other areas in Northern  
31 Eurasia. About 42% of the BC released from Northern Eurasian vegetation fires was  
32 deposited over the Arctic (annual average: 17%) during spring and summer.

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Deleted: Arctic total BC burden showed strong seasonal variations, with highest values during the Arctic Haze season. High winter–spring values of BC burden were caused by transport of BC mainly from anthropogenic sources in Europe, whereas the peak in summer was mainly due to the fire emissions in Northern Eurasia. BC particles emitted from fires in lower latitudes (35°N–40°N) were found to remain the longest in the atmosphere due to the high release altitudes of smoke plumes, exhibit tropospheric transport resulting in a high summer peak of burden, and grow by condensation processes.

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## 1 **1 Introduction**

2 The Arctic environment has experienced rapid modifications (e.g. warming, ice  
3 degradations, etc.) during the last four decades and concerns have been raised that human  
4 activities were the main cause for these transformations. The thinning of Arctic sea ice  
5 (Hansen and Nazarenko, 2004) and the Arctic's rapidly growing human influence (e.g.  
6 transportation, drilling, industry) indicates the need not only for further decrease of  
7 greenhouse emissions, but also a better understanding of aerosol properties, as well as of  
8 aerosol interaction with radiation, clouds, and ecosystems in Polar Regions. The "Arctic  
9 Haze" phenomenon in winter and spring is a major feature of Arctic air pollution. Several  
10 studies have been conducted to determine the sources of Arctic air pollution using trajectory,  
11 regional, and global models (e.g., Hirdman et al., 2010a; Klonecki et al., 2003; Koch and  
12 Hansen, 2005; Law and Stohl, 2007; Stohl, 2006). They all agree that the majority of the  
13 pollution in the high-latitude Arctic, especially near the surface, originates at mid- and high-  
14 latitudes, and that the accumulation of pollution in the Arctic is a consequence of the slow  
15 removal processes in winter and spring (Shaw, 1995). Also, Northern Eurasia (Europe,  
16 Siberia, Kazakhstan, Mongolia, etc.) is the main source of the Arctic BC due to both wildfire  
17 and anthropogenic emissions.

18 Episodic emissions from mid- and high-latitude vegetation fires can affect tropospheric  
19 concentrations of trace gases (e.g. carbon monoxide (CO), ozone (O<sub>3</sub>), volatile organic  
20 compounds (VOC), and aerosols (e.g. BC)) several thousand kilometers away from the  
21 sources (Forster et al., 2001; Wotawa and Trainer, 2000). Additionally, emissions from boreal  
22 fires lifted by convection can substantially alter upper tropospheric and the lowermost  
23 stratospheric radiation balance and chemistry (Waibel et al., 1999; Jost et al., 2004; Fromm et  
24 al., 2005). Aerosols and trace gases are uplifted during transport to the Arctic due to the  
25 upward sloping surfaces of constant potential temperature towards the Arctic (Klonecki et al.,  
26 2003; Stohl, 2006). However, understanding of aerosol transport from mid-latitudes to the  
27 Arctic has been limited because of the lack of quantification of the relevant aerosol sources  
28 and removal processes.

29 Globally, BC contributed to climate warming with recent estimates of radiative forcing  
30 at the top of the atmosphere ranging between 0.17 and 0.71 W m<sup>-2</sup> (Bond et al., 2013; Myhre  
31 et al., 2013; Wang et al., 2014). Snow albedo may be reduced by 1–3% in fresh snow by BC  
32 deposited in the Arctic and by another factor of 3 as snow ages and the BC becomes more  
33 concentrated (Clarke and Noone, 1985). Hansen and Nazarenko (2004) found that the  
34 decreased albedo in Arctic snow and ice since preindustrial times resulted in a hemispheric

1 radiative forcing of  $+0.3 \text{ W m}^{-2}$ , which may have had a substantial impact on the climate in  
2 the Northern Hemisphere, while for Northern Russia it amounts to  $0.2 \text{ W m}^{-2}$  (Lee et al.,  
3 2013a; 2013b). Airborne soot also absorbs incoming solar radiation thus warming the air and  
4 reducing tropical cloudiness (Ackerman et al., 2000).

5 Biomass burning (BB) constitutes a major source of BC, in addition to incomplete  
6 combustion of fossil fuels (primarily coal and diesel) and burning of biofuels. BC is the most  
7 absorbing portion of carbonaceous aerosols, commonly referred to as “soot”. BC deposited on  
8 snow/ice surfaces reduces surface reflectance and can promote faster melting of snow/ice in  
9 the Arctic, which is tightly coupled to climate effects through snow-albedo feedback (Flanner  
10 et al., 2007, 2009; Hansen and Nazarenko, 2004). In addition, high aerosol concentrations in  
11 the Arctic Haze lead to the enhancement of cloud longwave emissivity (Garrett and Zhao,  
12 2006; Lubin and Vogelmann, 2006), leading to surface warming and accelerating the melting  
13 of snow/ice.

14 Model simulations by Stohl (2006) suggested that the contributions from BB to Arctic  
15 BC loadings, particularly from fires in Siberia, exceeded the anthropogenic contributions in  
16 the summer. Moreover, large amounts of BC from Siberia and Kazakhstan have been  
17 observed during aircraft campaigns over Alaska in spring 2008 (Warneke et al., 2009), which  
18 was a year with an unusually early start of the BB season in Northern Eurasia. Warneke et al.  
19 (2010) estimated that BB in Russia may have doubled aerosol concentrations in the Arctic  
20 Haze during the spring. BC has been monitored at several surface stations in the Arctic (e.g.  
21 Alert in Canada, Barrow in Alaska, and Janiskoski in Russia) for many years (e.g. Sirois and  
22 Barrie, 1999; Sharma et al., 2006; Quinn et al., 2008; Eleftheriadis et al., 2009; Gong et al.,  
23 2010; Huang et al., 2010 and many others), showing decreasing trends during the 1980s and  
24 1990s, which have been attributed to reductions in anthropogenic emissions (Sharma et al.,  
25 2013; Hirdman et al., 2010b).

26 In this study, we focused on the transport of BC produced by vegetation fires in  
27 Northern Eurasia to the Arctic from 2002 to 2013. We define Northern Eurasia from  $10^{\circ}\text{W}$  to  
28  $170^{\circ}\text{E}$  and  $35^{\circ}\text{N}$  to  $80^{\circ}\text{N}$ . The Arctic is defined here as the area north of the Arctic Circle  
29 ( $\sim 67^{\circ}\text{N}$ ). Fires were mapped using satellite measurements from MODIS on NASA’s Terra  
30 and Aqua satellites. We investigated the geographic distribution of BC sources contributing to  
31 the Arctic BC budget, which needed to be better understood for developing BC mitigation  
32 policies. Moreover, the transport of BC to the Arctic after vegetation fires was defined as  
33 transport efficiency of BC. Shindell et al. (2008) showed large differences in the calculated  
34 BC concentrations in the Arctic using different General Circulation Models (GCMs). A large

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1 part of these differences was attributed to the different model treatment of BC aging from  
2 hydrophobic to hydrophilic and rainout/washout processes during transport. It indicated the  
3 necessity of a continuously improving description of BC transport in order to better assess the  
4 impact on the Arctic climate despite many recent improvements (e.g. Browse et al., 2012;  
5 Eckhardt et al., 2015).

6 This paper consists of five sections. The methodology (transport model, model set-up,  
7 emission altitude, satellite-derived BC emissions) is discussed in detail in the next section.  
8 The results, with respect to Arctic transport and deposition of BC, are presented in Section 3.  
9 Then, we show how different regions in Northern Eurasia contribute to the Arctic BC,  
10 distinguishing between anthropogenic and BB sources (Section 3.3). In Section 4.1, we  
11 discuss how our modeling results compare to observations of BC using data from five  
12 different Arctic stations for the period of our simulations (2002–2013). Finally, we calculate  
13 and study transport efficiencies of BC to the Arctic from different BB regions (Section 4.2).  
14 The main conclusions are presented in Section 5.

## 15 **2 Methodology**

### 16 **2.1 The LMDz-OR-INCA model**

17 We used the LMDz-OR-INCA global chemistry-aerosol-climate model, which couples  
18 the LMDz (Laboratoire de Météorologie Dynamique) GCM (Hourdin et al., 2006) and the  
19 INCA (INteraction with Chemistry and Aerosols) model (Hauglustaine et al., 2004). The  
20 interaction between the atmosphere and the land surface was ensured through the coupling of  
21 LMDz with the ORCHIDEE (ORganizing Carbon and Hydrology In Dynamic Ecosystems)  
22 dynamical vegetation model (Krinner et al., 2005). In the present configuration, the model  
23 included 39 hybrid vertical levels extending to the stratosphere, and a horizontal resolution of  
24  $1.29^{\circ} \times 0.94^{\circ}$  (280 grid-cells in longitude, 192 in latitude). A more detailed description and an  
25 extended evaluation of the GCM can be found in Hourdin et al. (2006). The large-scale  
26 advection of tracers was calculated based on a monotonic finite-volume second-order scheme  
27 (Hourdin and Armengaud, 1999). Deep convection was parameterized according to the  
28 scheme of Emanuel (1991). The turbulent mixing in the planetary boundary layer (PBL) was  
29 based on a local second-order closure formalism.

30 A comparison made with inert tracers indicated an enhanced vertical transport as the  
31 horizontal resolution of the model was increased from  $144 \times 142$  grid-points to  $280 \times 192$ . [We](#)  
32 [have studied the effect of the model resolution on the robustness of the predicted BC](#)

1 concentration distribution in Wang et al. (2014). The higher model resolution and an updated  
2 emission inventory used in this work allowed improving substantially the correlation between  
3 predicted and measured BC concentrations. The vertical profiles of BC over the Arctic  
4 measured during the ARCTAS campaigns (Arctic Research of the Composition of the  
5 Troposphere from Aircraft and Satellites) were used to assess the ability of such model to  
6 represent the vertical distribution of BC. The comparison showed that BC loads over the  
7 Arctic were slightly underestimated for the time of the ARCTAS flights. The model was also  
8 compared to other models in the recent AeroCom Phase II inter comparison (Myhre et al.,  
9 2013). The global mean anthropogenic all-sky and clear-sky aerosol radiative forcing is  
10 slightly more negative than the median for all the models (-0.36 versus -0.27 W m<sup>-2</sup>) and the  
11 BC load from fossil-fuel, and biofuels emissions is very close to the corresponding median of  
12 the models (0.15 versus 0.14 mg m<sup>-2</sup>).

13 The INCA model simulates the distribution of anthropogenic aerosols such as sulfates,  
14 nitrate (NO<sub>3</sub>), BC, particulate organic matter (POM), as well as natural aerosols such as sea-  
15 salt and dust. The aerosol model keeps track of both the number and the mass of aerosols  
16 using a modal approach to treat the size distribution, which is described by a superposition of  
17 5 log-normal modes (Schulz, 2007), each with a fixed spread. To treat the optically relevant  
18 aerosol size diversity, particle modes were categorized in three ranges: sub-micronic  
19 (diameter < 1 μm) corresponding to the accumulation mode, micronic (diameter 1–10 μm)  
20 corresponding to coarse particles, and super-micronic or super coarse particles (diameter > 10  
21 μm). Compared to a bin-scheme, the treatment of the size distribution with modes was  
22 computationally much more efficient (Schulz et al., 1998). Furthermore, to account for the  
23 diversity in chemical composition, hygroscopicity, and mixing state, we distinguished  
24 between soluble and insoluble modes. In both sub-micron and micron size ranges, soluble and  
25 insoluble aerosols were treated separately. Sea-salt, SO<sub>4</sub>, and methane sulfonic acid (MSA)  
26 were treated as soluble components of the aerosol, dust was treated as insoluble, whereas  
27 nitrate, BC, and POM appeared both in the soluble and insoluble fractions. The aging of  
28 primary insoluble carbonaceous particles transfers insoluble aerosol number and mass to  
29 soluble ones with a half-life of 1.1 days (Chung et al., 2002). The deposition scheme used in  
30 the model is described in detail in Evangeliou et al. (2013).

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## 1 2.2 Model set-up, BC inventories and injection height

2 The simulations lasted from January 1<sup>st</sup>, 2002 to December 31<sup>st</sup>, 2013. The model ran in  
3 a nudged mode using 6-hourly ERA Interim Re-analysis data (ECMWF, 2014) with a  
4 relaxation time of 10 days (Hourdin and Issartel, 2000).

5 To support the IPCC-AR5 (Intergovernmental Panel for Climate Change Assessment  
6 Report 5) and the ACCMIP (Atmospheric Chemistry and Climate - Model Intercomparison  
7 Project), a historical emissions dataset was developed (Lamarque et al, 2010) on a decadal  
8 basis (from 1850 to 2000 for the historical dataset), as well as RCP (Representation  
9 Concentration Pathways) emission scenarios for the period after the year 2000. As part of a  
10 project funded by the European Commission, MACC (Monitoring Atmospheric Composition  
11 & Climate) and CityZen (megaCITY - Zoom for the ENvironment), the ACCMIP and the  
12 RCP emissions dataset were adapted and extended to the year 2013 on a yearly basis. For  
13 anthropogenic emissions, emission data was interpolated on a yearly basis. For BB emissions,  
14 the ACCMIP dataset was extended to yearly and monthly mean calculated from modified  
15 RETRO (REanalysis of the TROposhpheric chemical composition) BC emission data for the  
16 years 1980 to 1996, and from GFEDv3 carbon emission data for the years 1997 to 2013, by  
17 applying a single set of vegetation type specific emission factors, and the predominant  
18 vegetation map used in the GFEDv3 inventory. This extension of the ACCMIP and RCP  
19 emission dataset for the MACC and CityZEN projects is referred to as MACCity  
20 (MACC/CityZen) emission dataset (Granier et al., 2011). As it is explained below, emissions  
21 of MACCity (anthropogenic and BB) were used as the input source of the model worldwide,  
22 in which BB sources were derived from GFEDv3. In addition, we adopted BC emissions from  
23 BB in Northern Eurasia from 2002 to 2013 (FEI-NE) described in the companion paper (Hao  
24 et al., 2016), while MACCity emissions were used for all the sources in other regions and for  
25 anthropogenic sources in Northern Eurasia.

26 Injection height is a key factor that controls transport and in turn deposition of BC  
27 emitted from fires. It is generally accepted that only explosive volcanic eruptions and strong  
28 crown fires (more common in North America than in Northern Eurasia) have the energy to  
29 inject pollutants from the surface into the stratosphere (Jost et al., 2004; Fromm et al., 2005).  
30 In a modeling study of mid-latitude supercell thunderstorms (Wang, 2003), it was reported  
31 that these plumes could induce important transport into the lowermost stratosphere. These  
32 findings suggest that extreme convection, even unassociated with energetic forest fires, may  
33 represent an important pathway for rapid, efficient redistribution of gases and particles from  
34 the lowest levels of the atmosphere to the lower stratosphere or, more commonly, the upper

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1 troposphere. Nedelec et al. (2005) described such a case for a fire happening over Siberia.  
2 However, injection of emissions in the lower troposphere is more common. Recently, Sofiev  
3 et al. (2013) published global maps of emission heights of wildfires that occurred between  
4 2000 and 2012 reporting that about 80% of the smoke is generally injected within the PBL,  
5 while the rest is injected at higher altitudes. Here we follow the same pattern as Sofiev et al.  
6 (2013) for Northern Eurasia, where 90% of the emissions were injected below 1.1 km, while  
7 the rest in heights up to 1.5 km. [Outside Eurasia, a similar injection profile is followed with](#)  
8 [100% of the emissions to occur up to 1 km.](#)

### 9 2.3 BC emissions from FEI-NE and MACCity

10 For the 12-year global simulations, anthropogenic sources of BC were adopted from the  
11 MACCity emission database. As regards to BB emissions, MACCity BB emissions from  
12 GFED3 were applied for all the regions outside Northern Eurasia, while within Northern  
13 Eurasia anthropogenic emissions from MACCity and BB from FEI-NE (Hao et al., 2016)  
14 were adopted. In summary, BC emissions from BB, excluding agricultural fires, in Northern  
15 Eurasia were estimated based on the area burned, fuel loading, percentage of the fuel burned,  
16 and emission factors of BC from different vegetation types (Hao et al., 2016).

17 This combined simulation is hereafter referred to as FEI-NE+MACCity. For  
18 comparison, we carried out the same simulation but using MACCity emissions alone (i.e. BB  
19 emissions in Northern Eurasia were also taken from MACCity) for the same period (2002–  
20 2013) (from now on referred to it as MACCity simulation). The different simulations are  
21 shown in [Table 1](#).

### 22 2.4 Lifetime calculations

23 Several definitions for atmospheric lifetime exist. In any domain of Earth's atmosphere  
24 the mass balance can be expressed as:

$$25 \quad \frac{dB(t)}{dt} = S(t) - \frac{B(t)}{\tau(t)} \quad (\text{Eq. 1})$$

26 where  $B(t)$  is the atmospheric burden,  $S(t)$  is the mass entering or exiting the domain and  
27  $\tau(t)$  is the removal time over a given time-step. If one assumes equilibrium between  $S(t)$  and  
28 deposition (steady state conditions), the mean steady state lifetime will be:

$$29 \quad \tau_{ss} = \frac{\bar{B}}{\bar{D}} \quad (\text{Eq. 2})$$

30 where  $\bar{B}$  and  $\bar{D}$  are the mean atmospheric burden and deposition over a specific period (Croft  
31 et al., 2014).

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## 1 **2.5 Observation data**

2 With different types of instruments, we collected measurements of BC, which may not  
3 always be directly comparable. Following the nomenclature of Petzold et al. (2013), we  
4 referred to measurements based on light absorption as equivalent BC (eBC) and  
5 measurements based on thermal-optical methods as elemental carbon (EC).

6 Aerosol light absorption data was obtained from five sites in different parts of the  
7 Arctic: Alert, Canada (62.3°W, 82.5°N; 210 m above sea level (a.s.l.)), Barrow, Alaska  
8 (156.6°W, 71.3°N; 11 m a.s.l.), Zeppelin/Ny Ålesund, Spitsbergen, Norway (11.9°E, 78.9°N;  
9 478 m a.s.l.), and Tiksi, Russia (128.9°E, 15 71.6°N; 1 m a.s.l.). Different types of particle  
10 soot absorption photometers (PSAPs) were used for the measurements at Barrow and  
11 Zeppelin, and an aethalometer was used at Alert and Tiksi. All these instruments measured  
12 the particle light absorption coefficient  $\sigma_{ap}$ , each at its own specific wavelength (typically at  
13 around 530–550 nm), and for different size fractions of the aerosol (typically particles smaller  
14 than 1, 2.5, or 10  $\mu\text{m}$  are sampled at different humidities). Conversion of  $\sigma_{ap}$  to eBC mass  
15 concentrations is not straightforward and requires certain assumptions (Petzold et al., 2013).  
16 The mass absorption efficiency used for conversion can be specific to a site and is uncertain  
17 by at least a factor of two. For Tiksi, the conversion was done internally by the aethalometer.  
18 For the other sites, a mass absorption efficiency of  $10 \text{ m}^2 \text{ g}^{-1}$ , typical of aged BC aerosol  
19 (Bond and Bergstrom, 2006), was used. Sharma et al. (2013) used an even higher value of 19  
20  $\text{m}^2 \text{ g}^{-1}$  for Barrow and  $10 \text{ m}^2 \text{ g}^{-1}$  for Alert data.

21 At Villum Research Station, Station Nord, Greenland, thermo-optical measurements  
22 were available. Weekly aerosol samples were analyzed with a thermal-optical Lab OC/EC  
23 instrument from Sunset Laboratory Inc (Tigard, OR, USA). Punches of  $2.5 \text{ cm}^2$  were cut from  
24 the filters sampled at Villum and analyzed according to the EUSAAR-2 protocol (Cavalli et  
25 al., 2010).

26 At Alert, eBC data was available for the years 2006–2013, at Barrow for 2002–2013, at  
27 Tiksi for 2009–2013, and at Zeppelin for 2002–2013. At Villum station, EC data was  
28 available for 2008–2011. The EC and eBC data were directly compared with modeled BC  
29 concentrations for the same locations and periods. Tiksi data was not filtered for a clean air  
30 sector and may have been affected by local pollution events. Barrow and Alert data were  
31 routinely subject to data cleaning, which removed the influence from local sources. Zeppelin  
32 generally was not strongly influenced by local emissions; however, summer values were  
33 enhanced by some 11% due to local cruise ship emissions (Eckhardt et al., 2013).

## 1 3 Results

### 2 3.1 Emission, transport and deposition of BC

3 Northern Eurasia encompasses diverse ecosystems including forest, shrubland,  
4 cropland, grassland, and savanna (Friedl et al., 2002). The total burned areas (excluding  
5 agricultural fires) during the period of 2002–2013 were estimated to be 250,000 km<sup>2</sup> yr<sup>-1</sup> (n =  
6 12) (Hao et al., 2016), which consisted of 61% of grassland and 27% of forest. Grassland fires  
7 occurred predominantly over Central and Western Asia, and forest fires over Siberia in  
8 Russia. The years 2003, 2006, and 2008 showed 96%, 40%, and 30%, respectively, more burn  
9 scars than the annual mean from 2002 to 2013 (Figure S 1). The unusual high fire activity in  
10 2003, 2006, and 2008 was a result of extensive grassland fires over Central and Western Asia,  
11 and forest and grassland fires over Russia.

12  
13 Table 2, presents the yearly mean atmospheric emissions of BC from anthropogenic and  
14 BB sources for the period 2002–2013 from FEI-NE+MACCity and MACCity. Annual global  
15 BC emissions varied in a range from 8.02 Tg yr<sup>-1</sup> in 2007 to 9.48 Tg yr<sup>-1</sup> in 2003 with an  
16 average: 8.42±0.43 Tg yr<sup>-1</sup> over the period 2002 to 2013 according to FEI-NE+MACCity.  
17 These values compared well with other published results. For instance, Wang et al. (2014)  
18 reported that, according to PKU-BC-2007 (Peking University BC Inventory for 2007)  
19 inventory of global BC emissions (both anthropogenic and biomass burning sources), a total  
20 amount of 8.9 Tg yr<sup>-1</sup> was emitted in 2007. The ECLIPSE inventory estimated for 2010 by  
21 Eckhardt et al. (2015) was 8.32 Tg yr<sup>-1</sup>, only slightly higher than our estimations of 8.02 Tg  
22 yr<sup>-1</sup>. In contrast, the ACCMIP BC emissions for 2005 (Lamarque et al., 2010) was 15% lower  
23 (e.g. ~7.82 Tg yr<sup>-1</sup> compared to 8.13 Tg yr<sup>-1</sup>). BC emissions from vegetation fires in Northern  
24 Eurasia ranged between 0.45 and 2.19 Tg yr<sup>-1</sup> for the period 2002–2013 representing 20% to  
25 49% of the total BC emissions over this region. In comparison, the emissions over the same  
26 region based on the MACCity inventory ranged between 0.13 and 0.43 Tg yr<sup>-1</sup> and accounted  
27 for 5% to 16% of total BC emissions. The BC emissions in the Northern Hemisphere (FEI-  
28 NE+MACCity) were classified as 70% from anthropogenic sources and 30% from BB.  
29 Northern Eurasian vegetation fires accounted for 56% of the BB BC emissions in the  
30 Northern Hemisphere.

31 BC emissions from FEI-NE were relatively constant in time except for four (2003,  
32 2006, 2008, 2012) intense years (Figure S 2). Similarly, the GFED3 of MACCity showed  
33 relatively high emissions in some of these years (2003 and 2006), but the relative emission

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1 | increase was significantly lower for these years (Table 2). The fire episodes that occurred in  
2 | Northern Eurasia during these extreme fire years were particularly intense. In 2003, fire  
3 | events in the Transbaikal region (Russian provinces Chita and Buryatia) caused severe smoke  
4 | pollution in the Far East of the Russian Federation (IFFN, 2004), while in spring 2006, smoke  
5 | from peat and forest fires in the western Russian Federation was noted as far as the United  
6 | Kingdom (Hao et al., 2009) and in the Arctic (Stohl et al., 2007). In summer 2006, smoke  
7 | from vegetation fires in the Russian Federation persisted for weeks over Finland (GFMC,  
8 | 2006).

9 | Figure 1 depicts deposition anomalies of BC for the period 2002–2013. A remarkable  
10 | feature was that the highest anomalies were observed in the northernmost part of Asia and in  
11 | the Arctic in 2003, 2006, 2008, and 2012. Taking into account emissions fluxes from these  
12 | years (Figure S 2), one can note that the largest amounts of BC were deposited over Arctic  
13 | regions as a result of large fire events in Siberia, Western Russia, and Kazakhstan. The annual  
14 | amount of BC deposited over the Arctic from all possible global sources (including BB)  
15 | during the 12-year period ranged from 65 kt yr<sup>-1</sup> to 152 kt yr<sup>-1</sup> (average: 102±29 kt yr<sup>-1</sup>)  
16 | representing about 0.9–18% of total global emissions. The total annual deposition of BC to  
17 | the Arctic from vegetation fires in Northern Eurasia (FEI-NE) during the same period ranged  
18 | between 29–120 kt yr<sup>-1</sup> (average: 65±28 kt yr<sup>-1</sup>) or about 0.3–14% of total global BB  
19 | emissions (Table 2). Hence, more than half of the total BC that deposited over the Arctic  
20 | originated from BB in Northern Eurasia, which underlines the importance of the Northern  
21 | Eurasian vegetation fires on the Arctic BC budget comparing with contribution of other  
22 | sources.

23 | Figure 2 illustrates BC deposition from FEI-NE detected vegetation fires only,  
24 | excluding anthropogenic sources, while Figure S 3 shows the deposition from all sources  
25 | (FEI-NE+MACCcity). For the most intense fire years, 2003, 2006, 2008, and 2012, an annual  
26 | amount of 142, 129, 117, and 152 kt yr<sup>-1</sup> of BC, respectively, was deposited over the Arctic  
27 | from Northern Eurasian vegetation fires (Table 2). It amounts to 3.2 to 4.1 times the 37±5 kt  
28 | yr<sup>-1</sup> average (2002 to 2013) deposition flux for anthropogenic sources over Northern Eurasia.  
29 | Finally, when using MACCcity emissions, both from anthropogenic and BB sources, the  
30 | estimated average deposition of BC over the Arctic was 56±8 kt yr<sup>-1</sup> for the period 2002–  
31 | 2013. Consequently, Arctic deposition was lower by 45% compared to FEI-NE, when BB  
32 | emissions of BC from GFED (MACCcity) were used.

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### 1 3.2 Aerosol lifetime and seasonality of BC

2 | [Figure 3](#), depicts the global mean aerosol lifetimes for anthropogenic and BB BC from  
3 the combined FEI-NE+MACCcity simulation. The results are given in Box & Whisker plots of  
4 daily lifetimes for the 12-year period. Mean aerosol lifetimes for anthropogenic BC were  
5 stable for all the studied years with an annual average value of  $5.6 \pm 0.2$  d. Mean aerosol  
6 lifetimes of BC from FEI-NE+MACCcity simulation exceeded these lifetimes by 1.2 days  
7 ( $6.8 \pm 1.0$  d). The pronounced variability of the mean lifetime of BB BC was attributed to the  
8 variability in regions and injection height. According to the injection scheme used (Sofiev et  
9 al., 2013), continuous injection close to the PBL (around 65% of the mass of BC is emitted up  
10 to 0.8 km, 90% up to 1.1 km) results in accumulation of BC in the troposphere, where longer  
11 lifetime occurs compared to the PBL. This was likely the main reason for transport of BC into  
12 the Arctic. For any soluble species emitted in the PBL, the lifetime is controlled by the  
13 removal, which happens within a few days if it is not transported to the free troposphere. Two  
14 weeks after being injected in the atmosphere, most of the tropospheric aerosol has been  
15 scavenged through wet deposition, whereas aerosols that have been transported into the high  
16 troposphere/lower stratosphere persist, given the absence of wet scavenging at these heights.  
17 The total aerosol mass and the lifetimes are then dominated by the stratospheric loading  
18 (Cassiani et al., 2013).

19 Mean aerosol lifetimes from global models are typically in the range of 3–7 days  
20 (Benkovitz et al., 2004; Textor et al., 2006), very similar to our estimations for BC, but more  
21 variable in this study due to the higher injection heights of BC in smoke plumes. As stated, in  
22 the present case of wildfires in Northern Eurasia, the lifetimes and the behaviour of BC was  
23 strongly affected by the fact that it was directly emitted aloft. Although 80–90% of BC was  
24 emitted inside the PBL, nearly 65% of the BC was emitted near the PBL height, while 10%  
25 was injected above (according to Sofiev et al., 2013).

26 In this study, we analyzed monthly values of both the Arctic BC burden and the mass  
27 deposited in the Arctic for all BC sources (FEI-NE+MACCcity) and for BC produced from BB  
28 sources only over Northern Eurasia (FEI-NE). A strong seasonal variation can be seen in both  
29 Arctic burden and deposition ([Figure 4a, b, c and d](#)). Relatively high BC burden from the  
30 combined emissions of the FEI-NE+MACCcity run occurred in winter (December–January,  
31 [Figure 4b](#)), while two peaks were found in late spring and in summer for intense fire years.  
32 The latest were clearly caused by spring and summer fire events over Northern Eurasia  
33 ([Figure 4](#)). The higher winter values corresponded to the meridional transport of BC, mostly

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1 emitted from anthropogenic sources (Figure 4a). These large increases in the burden were not  
2 proportional among the different aerosol modes (hydrophilic and hydrophobic) and affected  
3 mostly hydrophilic BC aerosol from vegetation fires. This indicated that wet scavenging was  
4 less efficient than the rest of the year along the transport path between mid- and high-  
5 latitudes. A spring maximum was also simulated for BC deposition over the Arctic. It was  
6 caused by a combination of anthropogenic BC in the Northern Hemisphere and fires in  
7 Northern Eurasia (Figure 4a and c), the majority of which occurred between March and May.  
8 The annually deposited mass of BC over the Arctic exceeded the annual average for the  
9 period 2002–2013 by 52% in 2003 and 86% in 2012.

10 There were three noteworthy findings. First, the year 2012 appeared to be a year when  
11 BC transport to the Arctic was particularly efficient. Although the year itself (2012) was not  
12 an extreme fire year (Figure S 1) in terms of burn scars, it appeared that the prevailing winds  
13 and the lack of scavenging in mid-latitudes during June and July when fires were most  
14 intense, favored transport and subsequent deposition to high-latitudes (120 kt from vegetation  
15 fires only and 152 kt from all possible sources). Second, we simulated a higher relative  
16 contribution of wet to total deposition in the Arctic (90%) than at mid-latitudes (69%). Third,  
17 the annual mean lifetime of anthropogenic BC particles from BB was longer (6.8 d) than for  
18 BC from combustion (5.6 d). These values are within the range of other published results  
19 (e.g., 5.8 days from Park et al. (2005) and 7.3 days from Koch and Hansen (2005)).

### 20 3.3 Geographic distribution of sources contributing to the Arctic BC

21 In this section, we compare the contributions from different source regions and from  
22 different emission types to the deposition of BC in the Arctic region. Several simulations with  
23 BC tracers tagged by source region were carried out to isolate the different contributions. We  
24 selected the following regions (Figure 5a): Europe, Asia, Siberia, Kazakhstan, and Mongolia,  
25 as well as from the latitude bands 35°N to 40°N, 40°N to 50°N, 50°N to 60°N, above 60°N  
26 (60°N–90°N), and also the entirety of Northern Eurasia. We discuss the results of these  
27 separate simulations and compare them to our FEI-NE+MACCity run (Figure 5). Each  
28 simulation covered the whole period from 2002 to 2013.

29 Quinn et al. (2007) were the first to report that for BC over the Arctic, anthropogenic  
30 sources dominated during winter and early spring Arctic Haze conditions. We estimated that  
31 anthropogenic emissions accounted for 70% of the Arctic BC burden from all sources in the  
32 North Hemisphere during winter and fall and for 40% during spring and summer when  
33 vegetation fires are most frequent.

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1 Vegetation fires from Northern Eurasia contributed 68% of the annual BC deposition  
2 over the Arctic coming from that same region (Figure 5d), whereas anthropogenic emissions  
3 constituted a lesser share (32%). Northern Eurasian vegetation fires were the most numerous  
4 ones over the Northern Hemisphere; they contributed for 81% of the Arctic deposition of BC  
5 from North Hemispheric fires (Figure 5d), while the rest came from other sources (e.g. BB in  
6 North America).

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7 Of the Northern Eurasian BB BC deposition in the Arctic, 95% was from Asia, while  
8 only 5% came from Europe. On a more regional basis, Siberia contributed 46% of the  
9 Northern Eurasian BB BC deposited in the Arctic, whereas Kazakhstan contributed 6%, and  
10 Mongolia only 1%. The rest was shared between fire events in Europe (5%) and elsewhere in  
11 Asia (42%) from areas that were not masked.

12 The relative contributions of fires at different latitudes to the Arctic BB BC deposition  
13 were distributed as follows: fires from the 35°-40°N latitudinal band over Northern Eurasia  
14 contributed only 7%, from 40°-50°N the contribution was 21%, 40% of this deposition came  
15 from fires at 50°-60°N, and 32% from fires above 60°N (Figure 5c).

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16 Moreover, we examined the vertical distribution of BC over the Arctic for the different  
17 source regions (Figure 6). When emitted from Europe, BC over the Arctic was found mostly  
18 below 5 km (either in the PBL or the low free troposphere), while BC emitted from Asia was  
19 found in higher layers that extended up to the mid- to high- free troposphere (Figure 6).  
20 Similar vertical distribution for aerosols have been reported by Stohl et al. (2002), who  
21 estimated that aerosol originating from Asia was mixed throughout the entire troposphere  
22 within a few days.

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23 These findings can be discussed in light of the ones reported by other authors. For  
24 instance, our results agree well with those of Hirdman et al. (2010a), who reported that the  
25 Northern Eurasian region (Europe and Russia) was the main contributor to the Arctic surface  
26 concentrations of BC and sulfate. The present study shows the importance of all the regions  
27 north of 50°N. Stohl (2006) reported that Asia contributed 10 times less than Europe to the  
28 Arctic BC surface concentrations, which was not supported by our findings for total BC. Our  
29 results agreed with the conclusions of Koch and Hansen (2005), who reported that Europe  
30 contributed 10% to the Arctic deposition of BC (anthropogenic and BB). In any case, all  
31 surface measurements of BC over the Arctic indicated that the main contributors to the Arctic  
32 BC during the summer were high-latitude sources (Hirdman et al., 2010a; Sharma et al.,  
33 2013).

## 1 4 Discussion

### 2 4.1 Observed and simulated BC concentrations at Arctic surface stations

3 There are contradictory results in the literature about where geographically the Arctic  
4 BC pollution originates. For example, Koch and Hansen (2005) estimated that Southeast Asia,  
5 Europe and Russia contributed each about 20–25% of the Arctic pollution during January to  
6 March period, whereas Stohl (2006) reported that Europe was the main contributor for the  
7 Arctic BC concentration at the surface. In addition, Huang et al. (2010) stated that Russia  
8 contributed 67% to the surface Arctic pollution, whereas Europe and North America were  
9 18% and 15%, respectively (16-year observations from Alert). Northern Eurasia appeared to  
10 be the main contributor in terms of Arctic BC during most seasons (Hirdman et al., 2010a).  
11 More recently, Stohl et al. (2007) and Warneke et al. (2009) reported that boreal and  
12 agricultural fires in Eastern Europe and in Siberia might be strong contributors to Arctic BC  
13 especially in the spring. Furthermore, Stohl et al. (2013) highlighted gas-flaring emissions in  
14 high latitudes as a major contributor to the Arctic BC.

15 [Figure 8](#) compares the simulated surface BC concentrations from this study with in-situ  
16 eBC and EC measurements from monitoring stations at Alert (Canada), Barrow (Alaska,  
17 USA), Villum (Greenland, Denmark), Zeppelin (Ny-Ålesund, Svalbard, Norway), and Tiksi  
18 (Russian Federation). The observations, when available, were represented for the entire period  
19 of our simulations (2002–2013). At Zeppelin and Barrow stations, the measurements were  
20 available between 2002 and 2013, at Alert from 2005 to 2013, at Villum for 2008 to 2011,  
21 and at Tiksi station for 2009 to 2013. [Figure 7](#) compares the simulated versus observed daily  
22 surface concentrations by a Box and Whisker plot at the five Arctic stations (Alert, Barrow,  
23 Villum, Tiksi, Zeppelin) for the period 2002 to 2013.

24 At Alert, there was a systematic underestimation in winter and early spring and an  
25 overestimation in late spring and summer, when the station did not record any enhanced eBC  
26 concentrations that would be attributed to vegetation fires in Northern Eurasia ([Figure 7](#) and  
27 [Figure S 2](#)). At Barrow, the model not only accurately estimated surface concentrations  
28 during winter, when the Arctic Haze is important, but also in spring and summer, when  
29 vegetation fires persisted ([Figure 7](#) and [Figure S 2](#)). At Villum, the measured seasonality was  
30 reproduced quite well by the model and also individual episodes of elevated BC surface  
31 concentrations in spring and summer were captured and attributed to vegetation fires. At the  
32 Tiksi station, the comparison showed a notable deviation from the measurements. Although  
33 some peaks in spring and summer were captured, the model missed completely the high

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1 concentrations of eBC in winter and early spring. Finally, at Zeppelin, it appeared that the  
2 anthropogenic BC contribution was slightly underestimated (spring), while vegetation fires in  
3 Northern Eurasia elevated modeled surface concentration of BC.

4 Looking at other inventories and results from the Lagrangian particle dispersion model  
5 FLEXPART, we are convinced that local anthropogenic sources play an important role in the  
6 apparition of these peaks (Eckhardt et al., 2015). It was already noted that our model  
7 underestimated surface concentrations at Alert and Villum stations during the Arctic Haze  
8 period. This is likely the consequence of an underestimation of Arctic transport in the model  
9 or a misleading emission inventory used for anthropogenic sources (MACCity). To verify it,  
10 we estimated surface concentrations of BC in the same longitude as the five Arctic stations,  
11 but five and ten degrees south in latitude (Figure S 4). In all stations except Tiksi, surface  
12 concentrations increased to the south confirming that the underestimation by our model over  
13 the Arctic could be attributed to a too weak transport simulated towards the Arctic.

14 It has been reported that most models underestimated BC in the Arctic during winter  
15 and early spring (Eckhardt et al., 2015), likely due to an improper representation of the  
16 scavenging processes (lack of below-cloud scavenging for solid phase water), the different  
17 emission profiles used for BC, and underestimated emission inventories used as input to the  
18 models (e.g. Koch and Hansen, 2005; Liu et al, 2011; Jiao et al., 2014). Using FEI-  
19 NE+MACCity inventory, BC concentrations over the Arctic are reasonable compared to  
20 observations, with a tendency to underestimate winter concentrations but capturing some  
21 summer peaks, Figure 7 also shows that using MACCity emissions globally, spring and  
22 summer concentrations are rather underestimated (e.g., in Villum, Tiksi and Zeppelin). In  
23 both cases, misleading anthropogenic emissions appear to be the main problem during winter.  
24 The largest deviations occur at the station Tiksi, Russia. The station is located only a few  
25 kilometers away from Tiksi town and local pollution is likely to affect the measurements, as  
26 the town has both a small airport and a harbor. Despite these drawbacks at Tiksi station, the  
27 data from the station has been used to estimate the sources in Northern Eurasia (Cheng,  
28 2014). Eckhardt et al. (2015) compared both surface- and aircraft measurements of sulfate and  
29 BC in the Arctic to model output from eleven different models. They found that the models  
30 generally underestimated the surface concentrations of BC and sulfate in winter/spring,  
31 whereas concentrations in summer were overestimated. They also found a strong correlation  
32 between surface measured sulfate and BC concentrations in winter/spring (anthropogenic  
33 impact), which indicated that the sources contributing to sulfate and BC were similar  
34 throughout the Arctic and that the aerosols were internally mixed and undergo similar

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1 removal. Neither Eckhardt et al. (2015) nor Samset et al. (2013) could isolate the reason to  
2 explain why some models performed better than the others.

3 In the present model configuration, we included emission inventory from FEI-NE and  
4 MACCity's anthropogenic BC inside Northern Eurasia and MACCity (BB and  
5 anthropogenic) outside Northern Eurasia (FEI-NE+MACCity), respectively, to evaluate if it  
6 produced reliable results with respect to observations. The question that stemmed from this  
7 comparison was whether or not existing datasets included all possible sources of BC  
8 emission. This was examined by comparing FEI-NE+MACCity inventory with MACCity,  
9 which included lower BC global emissions. [Figure 8](#) depicts the difference of the average  
10 atmospheric burden of BC between FEI-NE+MACCity and MACCity runs, while [Figure S 5](#),  
11 and [Figure S 6](#) show the same comparison for emissions and Arctic deposition of BC. It is  
12 apparent that the difference in average atmospheric burden between FEI-NE+MACCity and  
13 MACCity simulations ([Figure 8](#)) is positive over [Northern Eurasia and over](#) the Arctic  
14 showing that vegetation fires [in](#) Northern Eurasia have a direct impact on the Arctic budget,  
15 especially during the most intense fire years (2003, 2006, 2008, and 2012). The  
16 aforementioned impact extends up to North America and may affect the BC concentrations  
17 there as well. Subsequently, the deviation of the deposition of BC from Northern Eurasian  
18 vegetation fires relative to FEI-NE is shown to be large over the Arctic ([Figure S 6](#)).

19 We also analyzed the influence of [anthropogenic emissions, as well as](#) BB emissions  
20 from the regions (defined in [Table 1](#)) to the average surface concentration of the Arctic  
21 stations ([Figure 9](#)). As expected, the predominant contributor to the surface concentrations of  
22 the Arctic stations was Northern Hemisphere anthropogenic emissions (29–55%) (e.g. Shaw  
23 et al., 2010). The explanation is twofold. On one hand, transport of BC from the southern  
24 latitudes to the Arctic takes place as the air-masses follow the trajectories of potential  
25 difference in temperature (a dome effect), they are lifted up leaving the surface, especially  
26 from North America and Asia (China). On the other hand, [transport from](#) Russia/Siberia  
27 during the winter/spring is closer to the surface due to large anthropogenic emissions that are  
28 also effectively transferred from Europe. In addition, transport of BC from Russia/Siberia is  
29 less efficient during summer due to pressure systems that block the BC transport. Fires from  
30 Northern Eurasia contributed less BC to Barrow, Zeppelin, and Villum, while this pattern  
31 changed for Alert and Tiksi stations ([Figure 9](#)). This shows that emissions from Northern  
32 Eurasia may extend up to the American Arctic (Barrie, 1986). The region marked as “other”  
33 in [Figure 9](#), stands for all BB emissions occurring over the North Hemisphere, excluding  
34 Northern Eurasia, and shows that 4–42% of the surface concentrations may be due to fires in

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1 North America and other BB sources. In all cases, fires in regions north of 50°N contributed  
2 the most, especially to Tiksi station. Fires in the Asian part of Northern Eurasia contributed to  
3 the surface concentrations of the stations by 13% to 57%, with the maximum at Tiksi station.  
4 This was expected since this station is in the middle of the Northern Eurasian Arctic region  
5 and receives a lot of BC emitted from BB in Siberia. The respective portion for fires  
6 occurring in Europe was estimated to be 1–2% only. Similarly, BC emitted from Siberia  
7 contributed 9–43% to the simulated surface concentrations at the stations (with a maximum in  
8 Tiksi station). We estimated that the total of all vegetation fires in Northern Eurasia  
9 contributed around 56 ng m<sup>-3</sup>, on average, to the Alert station (during spring and summer  
10 months), which is close to 89 ng m<sup>-3</sup> that Huang et al. (2010) estimated for USSR and  
11 European Union, although with different geographic definitions than those assumed here, and  
12 also including anthropogenic emissions. Gong et al. (2010) and Shindell et al. (2008)  
13 estimated that the Northern Eurasian contribution to Alert varied between 80–90%, while our  
14 runs suggested a lower BB contribution (29%). Nevertheless, considering our comparison of  
15 modelling results with surface observations for BC from the five Arctic stations, it should be  
16 noted that the calculated contribution constituted an upper bound, especially when  
17 considering Asian and Siberian regions.

#### 18 4.2 Transport efficiency of BC to the Arctic

19 In this section, we examine the relative roles of different regions to emitting BC that  
20 will ultimately be deposited over the Arctic. To do so, we computed the probability of BC  
21 emitted from different regions to reach the Arctic (based on FEI-NE). We defined the  
22 transport efficiency to the Arctic as the ratio between the mass of BC deposited in the Arctic  
23 to the total mass of BC emitted from a given region. These estimates were obtained by  
24 masking the same geographical regions as in section 3.3 (anthropogenic sources in the  
25 Northern Hemisphere, vegetation fires in Europe, Asia, between 35°N–40°N, 40°N–50°N,  
26 50°N–60°N and above 60°N) and simulating fires that occurred inside the masked areas for  
27 the period 2012–2013 (Table 1).

28 [Figure 10](#) depicts the transport efficiency with which BC from vegetation fires reached  
29 the Arctic for the different geographic regions. The results clearly show that anthropogenic  
30 BC from large emitting regions in southeastern Asia and other Asian regions were not  
31 transported efficiently to the Arctic. The main source contributing to Arctic deposition was  
32 BB in Northern Eurasia with a transport efficiency of 10–32% during spring and summer, and  
33 1–6% in autumn and winter. Overall, the transport and subsequent deposition of BC from

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1 | Asia was more effective than from Europe (12% of the BB emissions from Asia were  
2 | deposited in the Arctic, whereas only 5% of the European BB emissions reach the Arctic),  
3 | which was attributed to the fact that European BC tends to remain close to the PBL (Figure  
4 | 10), whereas the Asian BC mixes up rapidly into the free troposphere (Stohl et al., 2002).  
5 | Therefore, European BC was much more affected by removal processes since its transport to  
6 | the Arctic is much less efficient.

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7 | In contrast, Siberian BC was deposited very efficiently to the Arctic in summer and  
8 | autumn similar to fires above 60°N (besides, Siberia covers the Asian part of the 60°N–90°N  
9 | area). However, it was evident from our results, that in our model the most efficient regional  
10 | transport of BC to the Arctic occurred in the summer months and was attributed to vegetation  
11 | fires in Kazakhstan and Mongolia (apart from Siberia, Figure 10). To summarize these  
12 | results, the highest transport efficiencies in our model occurred in the spring and summer for  
13 | all Northern Eurasia. This may be a result of (i) extreme fire events, (ii) the relatively weak  
14 | removal processes occurring in mid- and high-latitudes which favor transport without removal  
15 | of BC, and (iii) the imposed fixed injection profiles used in these simulations.

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## 16 | 5 Conclusions

17 | The present study focused on the impact of vegetation fires occurring in Northern  
18 | Eurasia on BC deposition in the Arctic. For this reason, a three dimensional global transport  
19 | model (LMDz-OR-INCA) was used to simulate fire events that took place during 2002–2013.  
20 | Anthropogenic emissions were adopted from MACCity inventory, while BB emissions within  
21 | Northern Eurasia were from FEI-NE and beyond Northern Eurasia from MACCity's GFEDv3  
22 | database.

23 | A total of  $3.0 \times 10^6$  km<sup>2</sup> was burned during the 12-year period in Northern Eurasia with  
24 | the majority to be grassland and forest fires. Total global emissions of BC ranged from 8.02  
25 | Tg yr<sup>-1</sup> to 9.48 Tg yr<sup>-1</sup> (average:  $8.42 \pm 0.43$  Tg yr<sup>-1</sup>) with the highest ones recorded for the  
26 | years 2003, 2006, 2008, and 2012. The annual emissions from vegetation fires in Northern  
27 | Eurasia were estimated to be between 0.45 and 2.19 Tg yr<sup>-1</sup> (average:  $0.86 \pm 0.51$  Tg yr<sup>-1</sup>).  
28 | Comparing to the MACCity emission inventory, our simulations suggested that 10–17%  
29 | (average: 8%) more BC was emitted by FEI-NE+MACCity, while FEI-NE biomass burning  
30 | emissions were 3.5 times higher than GFED3.

31 | The annual mean deposition of BC in the Arctic from vegetation fires in Northern  
32 | Eurasia was found to be  $65 \pm 28$  kt yr<sup>-1</sup> for the 12-year period, which represents 45–78% of the  
33 | BC deposited from all possible sources and origins. The combined run (FEI-NE+MACCity)

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1 brought around 55% (1218 versus 675 kt in total for the 12-year period) more BC to deposit  
2 over the Arctic environment comparing to the conventional MACCity emission inventory.

3 Arctic burden showed a strong seasonal variation, which peaks during late winter and  
4 early spring in the presence of the Arctic Haze. The peak in winter depicted the latitudinal  
5 transport of BC mainly from anthropogenic sources in Europe, whereas the peak in spring and  
6 summer clearly stemmed from the fire episodes in Northern Eurasia. The annual mass of BC  
7 deposited over the Arctic increased during the most intense fire years (37% in 2009 to 181%  
8 in 2012) in comparison to the annual average for the period of 2002–2013.

9 Fires occurring in the Northern Hemisphere contributed 68% to the simulated  
10 deposition of BC in the Arctic, while the rest originated from anthropogenic sources. The  
11 majority of the vegetation fires in the Northern Hemisphere was attributed to Northern  
12 Eurasian vegetation fires (85%), of which Asia contributed 81% and Europe only 4%. These  
13 results were consistent with what other researchers have reported, as Asian BC experienced  
14 fast elevation to the free troposphere and hence long-range transport.

15 The present results were compared to surface observations from five stations (Alert,  
16 Barrow, Villum, Tiksi, and Zeppelin) showing relatively good results and in most stations  
17 capturing the trend in surface BC concentrations (a notable deviation was observed at Alert  
18 Alert). We estimated that vegetation fires in Northern Eurasia contributed 14% to 57% to the  
19 surface environment of these stations, mostly affected by fires that took place in Siberia. This  
20 showed the importance of fires occurring over Northern Eurasia in the Arctic BC budget.  
21 However, anthropogenic sources also remain essential contributing 29% to 54% to the surface  
22 of the Arctic stations.

23

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32

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1 **TABLE CAPTIONS**

2 **Table 1.** List of simulations from this study to characterize the transport and origin of BC.

<b>Name</b>	<b>Anthropogenic sources</b>	<b>BB sources</b>	<b>Purpose</b>
FEI-NE+MACCity	MACCity	FEI-NE	Study of BC transport to the Arctic
MACCity	MACCity	MACCity	Comparison with the combined FEI-NE+MACCity run
Europe	–	FEI-NE	Study of BC originating from Europe by masking this region
Asia	–	FEI-NE	Study of BC originating from Asia by masking this region
Siberia	–	FEI-NE	Study of BC originating from Siberia by masking this region
Kazakhstan	–	FEI-NE	Study of BC originating from Kazakhstan by masking this region
Mongolia	–	FEI-NE	Study of BC originating from Mongolia by masking this region
35°N–40°N (10°W–170°E)	–	FEI-NE	Study of BC originating from latitudes 30°N–40°N in Eurasia by masking this region
40°N–50°N (10°W–170°E)	–	FEI-NE	Study of BC originating from latitudes 40°N–50°N in Eurasia by masking this region
50°N–60°N (10°W–170°E)	–	FEI-NE	Study of BC originating from latitudes 50°N–60°N in Eurasia by masking this region
60°N–90°N (10°W–170°E)	–	FEI-NE	Study of BC originating from latitudes >60°N in Eurasia by masking this region

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4

1 **Table 2.** Comparison between annual BC emissions (Tg<sub>yr<sup>-1</sup></sub>) for the period 2002–2013 (FEI-NE+MACCity) and MACCity emissions for the  
 2 same period. The deposition of BC (ktons<sub>yr<sup>-1</sup></sub> or kt<sub>yr<sup>-1</sup></sub>) from vegetation fires over the Arctic is also compared to those from the MACCity  
 3 inventory.

	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	Range
Anthropogenic sources (Tg <sub>yr<sup>-1</sup></sub> )	5.22	5.26	5.30	5.34	5.31	5.28	5.25	5.23	5.20	5.17	5.15	5.15	5.15–5.34
Anthropogenic sources in Eurasia (Tg <sub>yr<sup>-1</sup></sub> )	2.26	2.27	2.27	2.28	2.28	2.27	2.26	2.25	2.24	2.22	2.20	2.20	2.20–2.28
BB sources (FEI-NE+MACCity) (Tg <sub>yr<sup>-1</sup></sub> )	2.83	4.22	2.98	2.89	3.29	2.74	3.47	3.03	2.82	3.15	3.55	2.90	2.74–4.22
<b>FEI-NE fires in Eurasia (Tg<sub>yr<sup>-1</sup></sub>)</b>	<b>0.62</b>	<b>2.19</b>	<b>0.57</b>	<b>0.47</b>	<b>0.90</b>	<b>0.57</b>	<b>1.39</b>	<b>0.69</b>	<b>0.45</b>	<b>0.77</b>	<b>1.17</b>	<b>0.53</b>	<b>0.45–2.19</b>
<b>FEI-NE+MACCity total (Tg<sub>yr<sup>-1</sup></sub>)</b>	<b>8.05</b>	<b>9.48</b>	<b>8.28</b>	<b>8.23</b>	<b>8.60</b>	<b>8.02</b>	<b>8.72</b>	<b>8.26</b>	<b>8.02</b>	<b>8.32</b>	<b>8.70</b>	<b>8.05</b>	<b>8.02–9.48</b>
Arctic deposition from fires in Eurasia (kt <sub>yr<sup>-1</sup></sub> )	84	98	43	36	88	29	79	42	49	58	120	51	29–120
Arctic deposition from <u>all sources outside Eurasia</u> (kt <sub>yr<sup>-1</sup></sub> )	38	44	38	42	41	36	38	36	33	36	32	28	28–44
<b>Total deposition over the Arctic (kt<sub>yr<sup>-1</sup></sub>)</b>	<b>122</b>	<b>142</b>	<b>81</b>	<b>78</b>	<b>129</b>	<b>65</b>	<b>117</b>	<b>78</b>	<b>82</b>	<b>94</b>	<b>152</b>	<b>79</b>	<b>65–152</b>
MACCity anthropogenic sources (Tg <sub>yr<sup>-1</sup></sub> )	5.22	5.26	5.30	5.34	5.31	5.28	5.25	5.23	5.20	5.17	5.15	5.15	5.15–5.34
MACCity anthropogenic sources in Eurasia (Tg <sub>yr<sup>-1</sup></sub> )	2.26	2.27	2.27	2.28	2.28	2.27	2.26	2.25	2.24	2.22	2.20	2.20	2.20–2.28
MACCity BB sources globally (Tg <sub>yr<sup>-1</sup></sub> )	2.51	2.47	2.55	2.57	2.34	2.64	2.04	2.62	2.62	2.62	2.62	2.62	2.04–2.62
<b>MACCity BB (GFED3) in Eurasia (Tg<sub>yr<sup>-1</sup></sub>)</b>	<b>0.30</b>	<b>0.43</b>	<b>0.13</b>	<b>0.13</b>	<b>0.21</b>	<b>0.14</b>	<b>0.25</b>	<b>0.24</b>	<b>0.24</b>	<b>0.24</b>	<b>0.24</b>	<b>0.24</b>	<b>0.13–0.43</b>
<b>MACCity total (Tg<sub>yr<sup>-1</sup></sub>)</b>	<b>7.73</b>	<b>7.73</b>	<b>7.85</b>	<b>7.91</b>	<b>7.65</b>	<b>7.92</b>	<b>7.29</b>	<b>7.85</b>	<b>7.82</b>	<b>7.79</b>	<b>7.77</b>	<b>7.76</b>	<b>7.29–7.92</b>
Wet deposition over the Arctic (kt <sub>yr<sup>-1</sup></sub> )	64	65	50	49	54	40	52	52	51	51	50	42	42–65
Dry deposition over the Arctic (kt <sub>yr<sup>-1</sup></sub> )	5	6	6	6	4	3	3	5	5	5	5	4	3–6
<b>Total deposition over the Arctic (kt<sub>yr<sup>-1</sup></sub>)</b>	<b>69</b>	<b>71</b>	<b>56</b>	<b>55</b>	<b>58</b>	<b>43</b>	<b>55</b>	<b>57</b>	<b>56</b>	<b>56</b>	<b>55</b>	<b>46</b>	<b>43–71</b>

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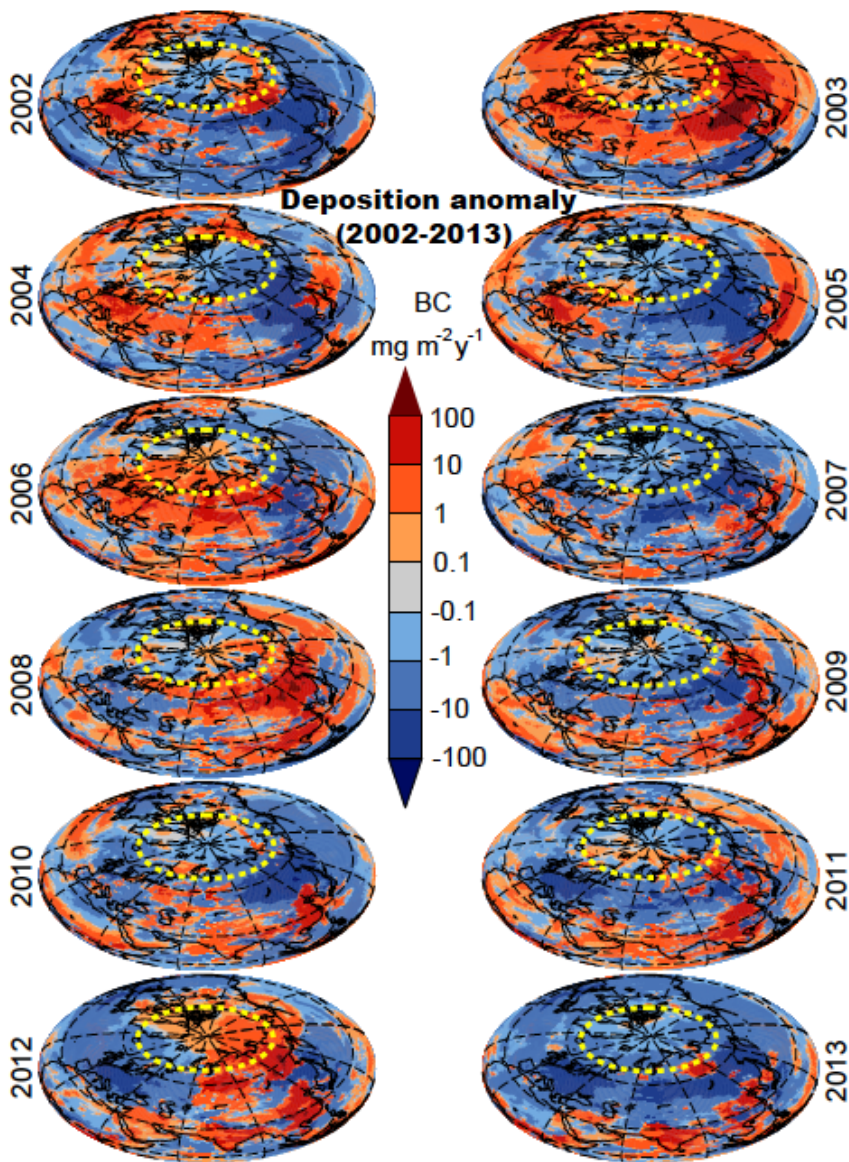
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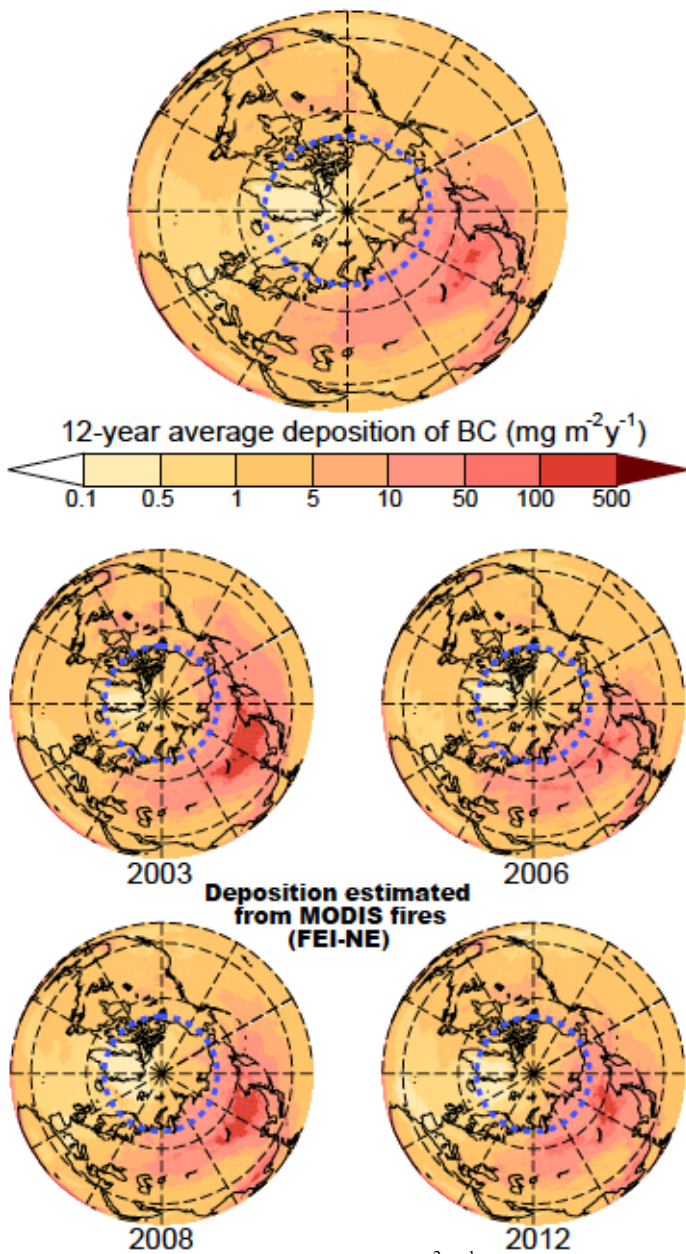


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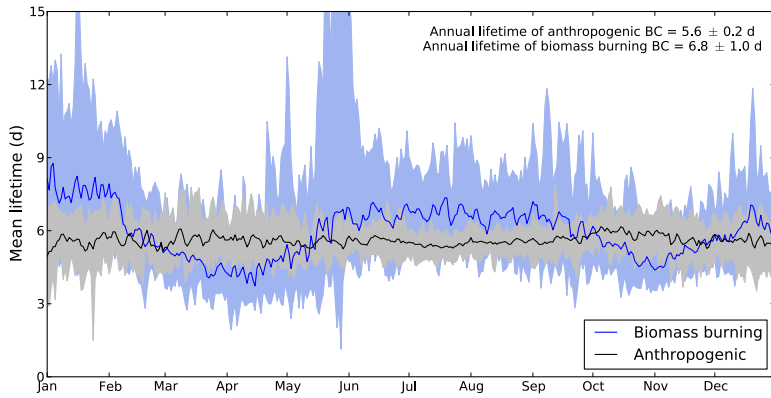
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4 **Figure 1.** Deposition anomalies of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) in the North Hemisphere for the period  
5 2002–2013 from our combined simulation (FEI-NE+MACCity) in Northern Eurasia. The  
6 dashed yellow line represents the border of the Arctic ( $\sim 67^\circ\text{N}$ ). Note that the most intense  
7 years were estimated to be 2003, 2006, 2008, and 2012.



1  
 2 **Figure 2.** Arctic deposition of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) from BB emission according to the FEI-NE  
 3 inventory. The upper panel depicts the 12-year average deposition, while the lower 4 panels  
 4 show the most intense fire years (2003, 2006, 2008, and 2012). The dashed blue line  
 5 represents the border of the Arctic ( $\sim 67^\circ\text{N}$ ).



1  
2 **Figure 3.** Annual mean global lifetime ( $\pm$  standard deviation) of the global anthropogenic and  
3 BB BC from our combined FEI-NE+MACCcity simulation. The results show minimum,  
4 average, maximum daily lifetimes of BC (both for anthropogenic and BB) for the period  
5 2002–2013.

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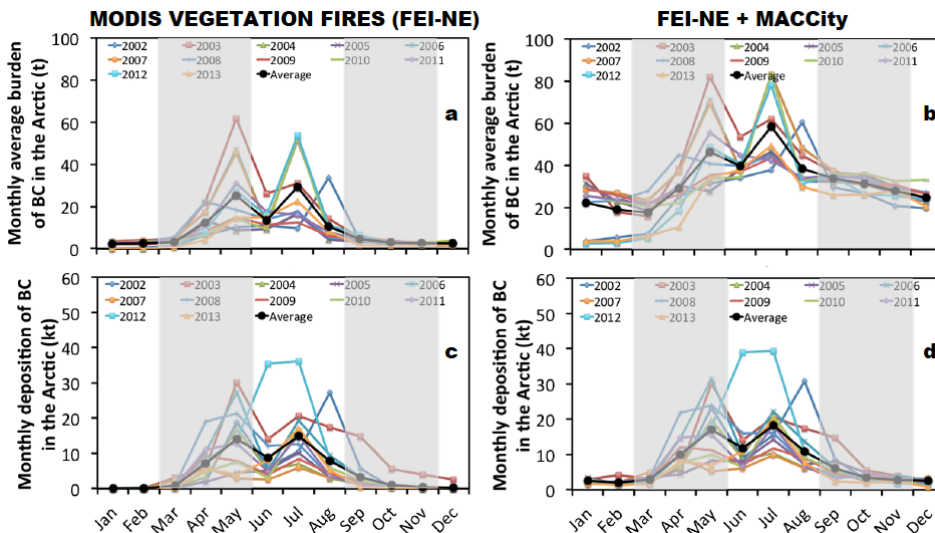
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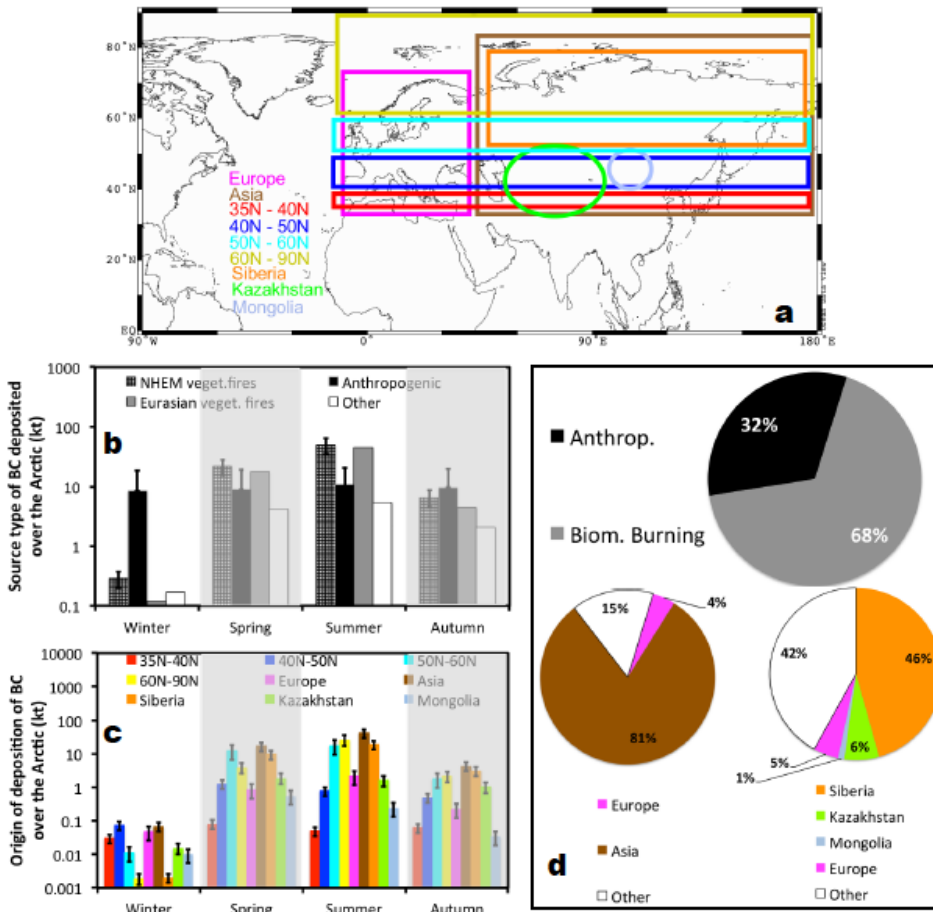
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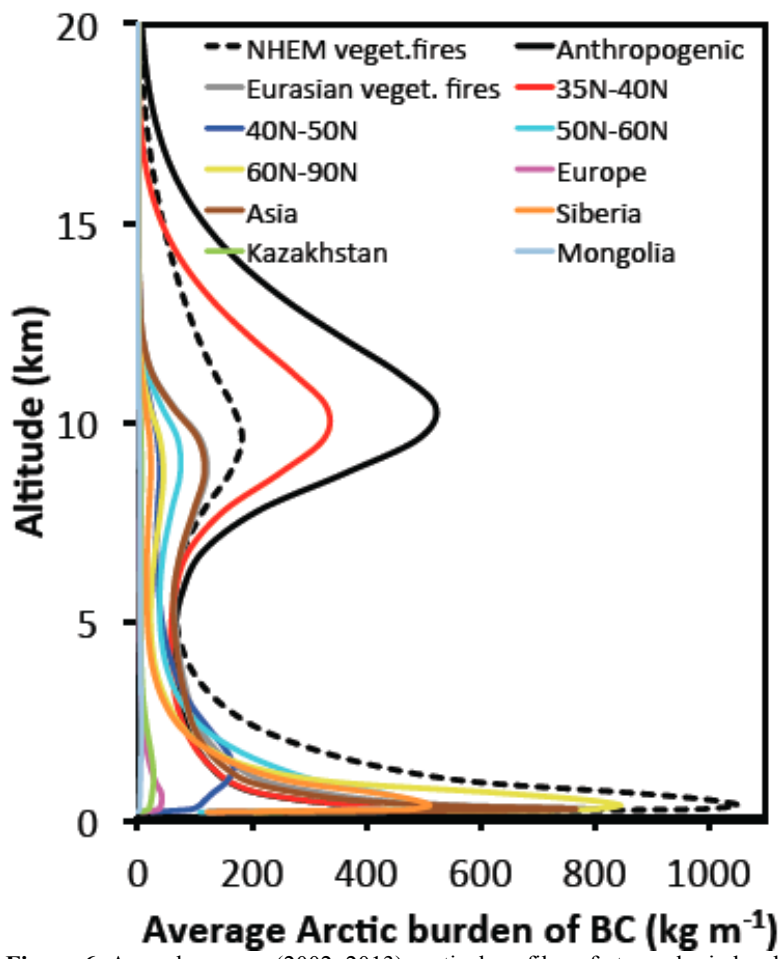
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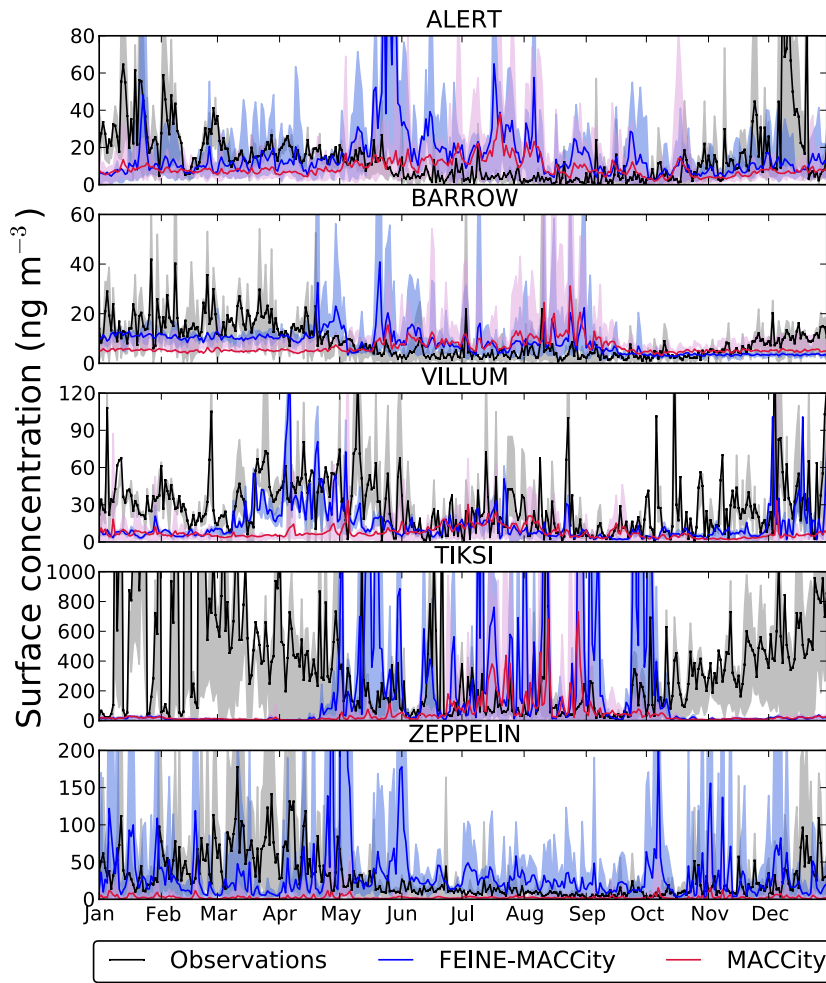
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7 **Figure 4.** Monthly atmospheric burden of BC (t) in the Arctic with each year between 2002  
8 and 2013 represented by a different colored line: **(a)** from vegetation fires (FEI-NE) only and  
9 **(b)** from all BC emissions (FEI-NE+MACCcity). Panel c and d shows the same, but for the  
10 Arctic deposition of BC (kt).



1  
2 **Figure 5.** (a) The nine geographic regions of BC emission in Northern Eurasia defined for  
3 this study. (b) Seasonal values of BC emitted from all fires in Northern Hemisphere (NHEM),  
4 anthropogenic sources (black) and Northern Eurasian vegetation fires (grey) deposited in the  
5 Arctic for the period of 2002–2013. (c) Contribution of several geographic regions to the  
6 Arctic BC deposition. Colors are used according to the ones used in panel a. Red stands for  
7 regions within 35°N–40°N, dark blue for 40°N–50°N, turquoise for 50°N–60°N, yellow for  
8 regions located above 60°N (60°N–90°N), magenta for Europe, brown for Asia, orange  
9 denotes Siberia, green for Kazakhstan, and light blue for Mongolia. (d) Pie-charts showing  
10 the origin of BC deposition in the Arctic from [anthropogenic sources \(MACCCity\)](#), [vegetation](#)  
11 [fires in Northern Eurasia \(FEI-NE\)](#) and [BB outside Northern Eurasia](#).



1  
 2 **Figure 6.** Annual average (2002–2013) vertical profiles of atmospheric burden of BC ( $\text{kg m}^{-1}$ )  
 3 <sup>1</sup>) in the Arctic originating from different regions.



1  
 2 **Figure 7.** Modeled versus measured surface concentrations of BC ( $\text{ng m}^{-3}$ ) for the FEI-  
 3 NE+MACCity and MACCity simulations in the Arctic stations Alert, Barrow, Villum, Tiksi,  
 4 and Zeppelin. Due to the high variability of the surface concentrations, the results are  
 5 presented as minimum, average, maximum modeled (FEI-NE+MACCity and MACCity) and  
 6 measured surface daily concentrations of BC for the period 2002–2013.

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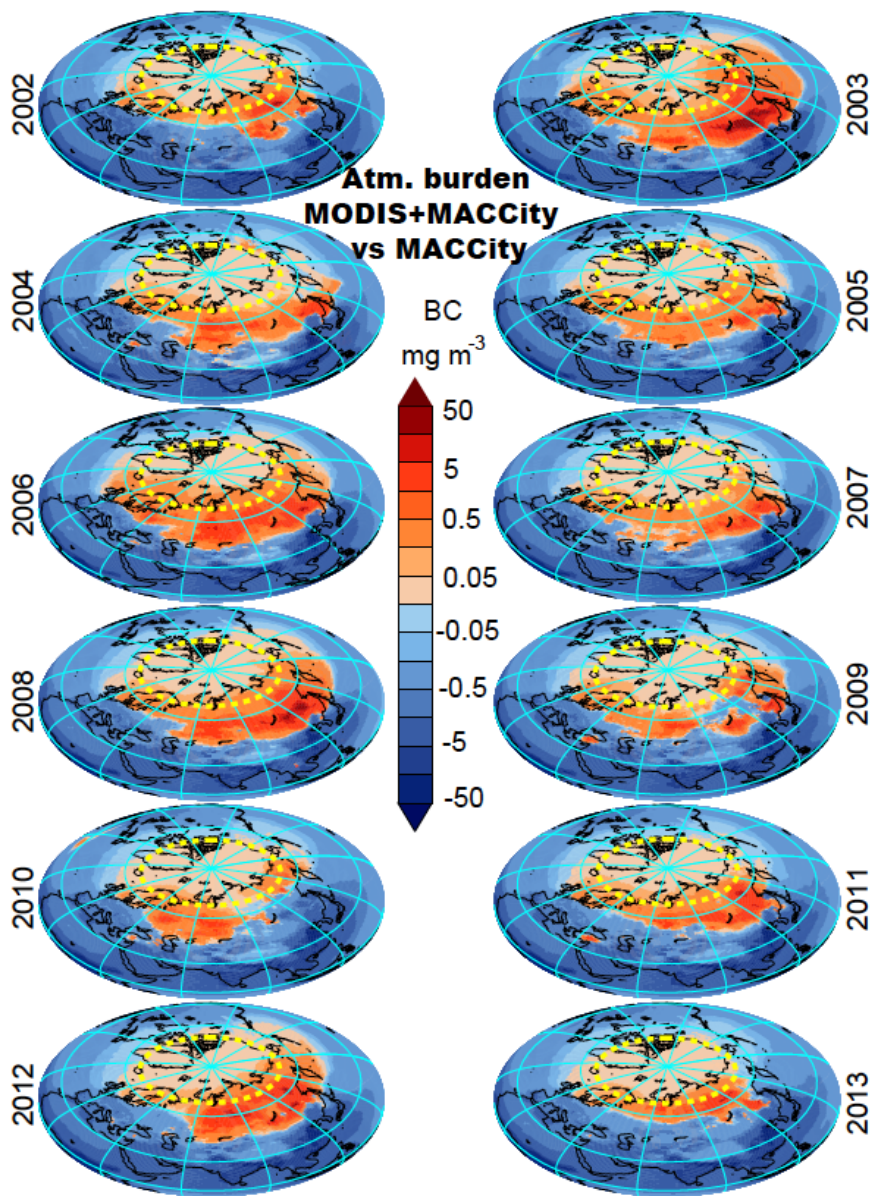
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1  
 2 **Figure 8.** Difference in BC atmospheric burden ( $\text{mg m}^{-3}$ ) between our simulation that  
 3 combines emission inventories (FEI-NE+MACCity) and MACCity. The dashed yellow line  
 4 represents the limit of the Arctic ( $\sim 67^\circ\text{N}$ ). The BC burden was estimated by summing all the  
 5 vertical layers and averaging with time (365 days) for each of the years between 2002 and  
 6 2013.

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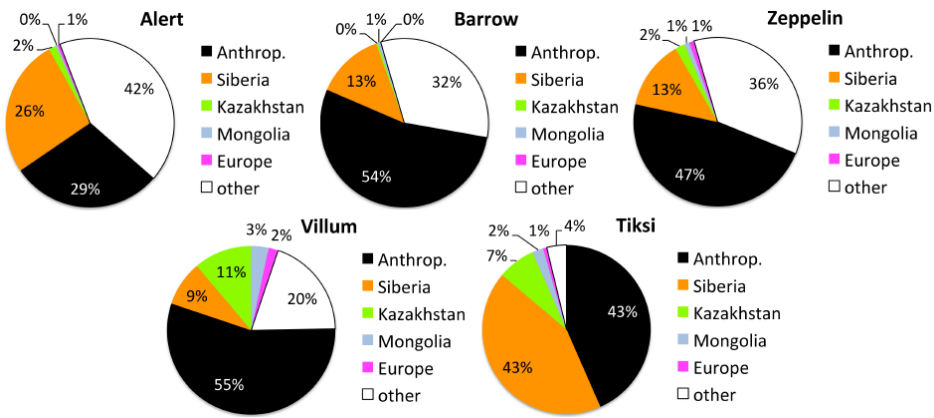
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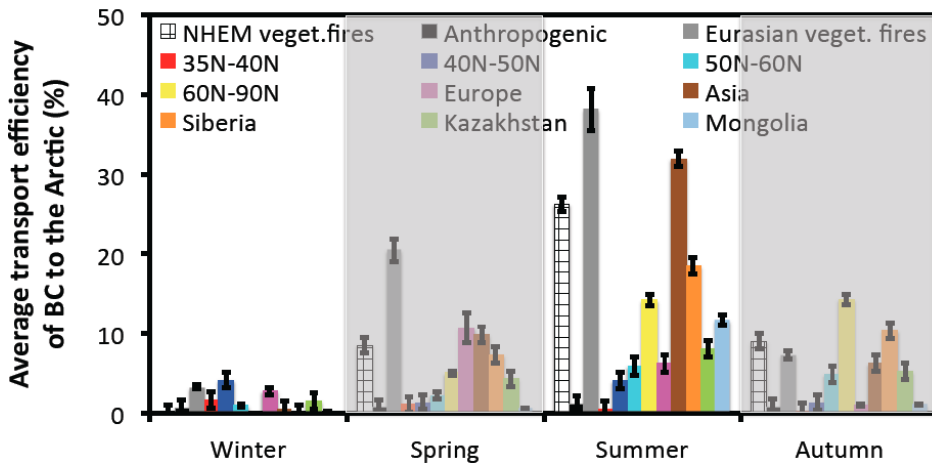
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2 **Figure 9.** Multi-year average (2002–2013) contribution of global anthropogenic and BB BC  
3 from the respective geographic regions to surface concentrations at the Arctic stations (Alert,  
4 Barrow, Zeppelin, Villum, and Tiksi). Region “other” stands for other locations where BB  
5 sources were not accounted for.

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7 **Figure 10.** Relative transport efficiency of BC from vegetation fires from different  
8 geographic regions across the Arctic. The same colors were used as in [Figure 5](#).

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1  
2 **FIGURE CAPTION FOR SUPPLEMENTS**

3  
4 **Figure S 1.** Percentage variation of MODIS [burn scars](#) from the 12-year (2002–2013)  
5 average. The most intense fire years are characterized by positive values comparing to the  
6 average for the period 2002–2013 ( $1,075,208 \pm 378,399$  [burn scars](#)).

7 **Figure S 2.** Emission anomalies of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) in Northern Eurasia for the period 2002–  
8 2013 from our combined simulation (FEI-NE+MACCcity). The dashed yellow line represents  
9 the border of the Arctic ( $\sim 67^\circ\text{N}$ ).

10 **Figure S 3.** Arctic deposition of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) over the Arctic (FEI-NE+MACCcity  
11 simulation). The upper panel depicts the 11-year average deposition, while the lower 4 panels  
12 the most intense fire years (2003, 2006, 2008, and 2012). The dashed blue line represents the  
13 border of the Arctic ( $\sim 67^\circ\text{N}$ ).

14 **Figure S 4.** Surface modelled concentrations of BC for the FEI-NE+MACCcity simulation in  
15 the Arctic stations  $5^\circ$  and  $10^\circ$  south of Alert, Barrow, Villum, Tiksi, and Zeppelin. The results  
16 are presented as Box & Whisker plots of surface daily concentrations of BC for the period  
17 2002–2013. The plots show the minimum value, the 25<sup>th</sup> percentile, which holds 25% of the  
18 values at or below it. The median is the 50<sup>th</sup> percentile, the third quartile is the 75<sup>th</sup> percentile  
19 and the maximum is the 100<sup>th</sup> percentile (i.e., 100% of the values are at or below it).

20 **Figure S 5.** Difference in atmospheric emissions of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) between our combined  
21 simulation (FEI-NE+MACCcity) and MACCcity. The dashed yellow line represents the border  
22 of the Arctic ( $\sim 67^\circ\text{N}$ ). Emissions were estimated by summing all the vertical layers for 365  
23 days of each of the years (2002–2013). Negative values were only observed in latitudes above  
24  $35^\circ\text{N}$  over Northern Eurasia, as a result of the difference in the emissions between FEI-  
25 NE+MACCcity and MACCcity simulations.

26 **Figure S 6.** Difference in Arctic deposition of BC ( $\text{mg m}^{-2} \text{y}^{-1}$ ) between our combined  
27 simulation (FEI-NE+MACCcity) and MACCcity. The dashed yellow line represents the border  
28 of the Arctic ( $\sim 67^\circ\text{N}$ ). Deposition was estimated by summing all the 365 days of each of the  
29 years (2002–2013).

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Table 2