

**A one year time series at the Arctic observatory Zeppelin**

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# Quantifying black carbon from biomass burning by means of levoglucosan – a one year time series at the Arctic observatory Zeppelin

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## Abstract

Levoglucosan, a highly specific tracer of particulate matter from biomass burning, has been used to study the influence of residential wood burning, agricultural waste burning and boreal forest fire emissions on the Arctic atmosphere black carbon (BC) concentration. A one year time series from March 2008 to March 2009 of levoglucosan has been established at the Zeppelin Observatory in the European Arctic. Elevated concentrations of levoglucosan in winter (Mean:  $1.02 \text{ ng m}^{-3}$ ) compared to summer (Mean:  $0.13 \text{ ng m}^{-3}$ ) were observed, resembling the seasonal variation seen for e.g. sulphate and BC. The mean concentration in the winter period was two to three orders of magnitude lower than typical values reported for European urban areas in winter, and one to two orders of magnitude lower than European rural background concentrations. Episodes of elevated levoglucosan concentration were more frequent in winter than in summer and peak values were higher, exceeding  $10 \text{ ng m}^{-3}$  at the most.

Concentrations of elemental carbon from biomass burning ( $\text{EC}_{\text{bb}}$ ) were obtained by combining measured concentrations of levoglucosan and emission ratios of levoglucosan and EC for wild/agricultural fires and for residential wood burning. Neglecting chemical degradation by OH provides minimum levoglucosan concentrations, corresponding to a mean  $\text{EC}_{\text{bb}}$  concentration of  $3.7 \pm 1.2 \text{ ng m}^{-3}$  in winter (October–April) and  $0.8 \pm 0.3 \text{ ng m}^{-3}$  in summer (May–September) or  $8.8 \pm 4.5\%$  of the measured equivalent black carbon (EBC) concentration in winter and  $6.1 \pm 3.4\%$  in summer. When accounting for chemical degradation of levoglucosan by OH, an upper estimate of 31–45% of EBC could be attributed to  $\text{EC}_{\text{bb}}^*$  ( $\text{EC}_{\text{bb}}$  adjusted for chemical degradation) in winter and  $< 65\%$  in summer. Hence, fossil fuel sources appear to dominate the European Arctic BC concentrations in winter, whereas the very wide range obtained for summer does not allow us to conclude upon this for the warm season.

Calculations using the Lagrangian particle dispersion model FLEXPART show that the seasonal variation of the modelled  $\text{EC}_{\text{bb}}$  ( $\text{EC}_{\text{bb}, \text{m}}$ ) concentration compared relatively well with observationally derived  $\text{EC}_{\text{bb}}$  from agricultural/wild fires during summer,

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and residential wood burning in winter. The model overestimates by a factor of 2.2 in winter and 4.4 in summer when compared to the observationally derived mean  $EC_{bb}$  concentration, which provides the minimum estimate, whereas it underestimates by a factor of 2.3–3.3 in winter and a factor of 4.5 in summer when compared to  $EC_{bb}^*$ , which provides the upper estimate. There are indications of too low emissions of residential wood burning in Northern Russia, a region of great importance with respect to observed concentrations of BC in the European Arctic.

## 1 Introduction

The presence of ambient air pollutants in the Arctic has been demonstrated in several studies (e.g. Shaw, 1995). These studies have had a particular focus on the Arctic Haze phenomenon (Quinn et al., 2007), caused by long range transport of pollutants from the mid-latitudes, leading to elevated concentrations of air pollutants during late winter and early spring. Long-term trend studies show a downward trend in black carbon and sulphate aerosol at the three Arctic sites Alert, Barrow and Zeppelin over the last decade (Hirdman et al., 2010a), which is in line with observed aerosol trends at mid-latitudes during the last decades (Murphy et al., 2011; Hand et al., 2012; Torseth et al., 2012). Radiative effects of aerosols, both direct and indirect are complex (Myhre et al., 2013), and can be quite different in the Arctic compared to elsewhere. Due to the high surface albedo of snow and ice, even weakly absorbing aerosol layers in the Arctic can heat the Earth/atmosphere system (Pueschel and Kinne, 1995). Infrared emissions from the haze can heat the surface during the polar night, and during spring when the solar zenith angle is still large (MacCracken et al., 1986). Furthermore, absorbing aerosols deposited in the polar region can affect the snow and ice albedo and increase the amount of solar radiation absorbed, enforcing melting of snow on tundra and sea ice (Clarke and Noone, 1985; Hansen and Nazarenko, 2004). Consequently, it can be argued that the largest uncertainties when studying the aerosols' impact on climate

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in the Arctic are attributed to the highly absorbing species black carbon (BC)<sup>1</sup>, which is generated by incomplete combustion of fossil fuel and biomass. Due to its short atmospheric lifetime compared to CO<sub>2</sub> it has been suggested that reducing emissions of BC is an effective strategy, complementing CO<sub>2</sub> mitigation, to slow global warming in the near future (Bond, 2007; Hansen et al., 2000; Jacobson, 2002; Shindell et al., 2009). It is thus essential to increase current knowledge of the relative strengths of different BC sources in order to implement effective mitigation steps.

Some recent studies have argued that boreal wild fires with their high emissions and proximity to the Arctic could be the most important source of Arctic BC in seasons of high wild fire activity (Stohl, 2006; Stohl et al., 2007; Warneke et al., 2009). Furthermore, an increased frequency of boreal forest fires has been postulated as a consequence of global warming, augmenting its present source strength. Also agricultural fires were identified as a strong episodic source of BC for the Arctic (Stohl et al., 2007; Warneke et al., 2009). Warneke et al. (2009) argued that in spring biomass burning (BB) plumes on average contribute at least 80 % as much BC to the Arctic atmospheric burden as is otherwise present as an Arctic background from all sources. However, Brock et al. (2011) have shown that the BB layers were present only above the sea-ice inversion layers and thus may not regularly be seen at Arctic surface sites.

<sup>1</sup>Here, the term black carbon (BC) is used according to the recommendations of Petzold et al. (2013). BC is defined as a substance with the following properties: (1) consists of mainly sp<sup>2</sup>-bonded, graphitic carbon; (2) consists of aggregates of carbon spherules of 10–50 nm diameter; (3) refractory, volatile at near 4000 K, volatilization by oxidation from 613 K; (4) insoluble in organic solvents and water; (5) strongly absorbing for visible light. There exists no measurement method that is sensitive to all these properties at the same time, i.e. no method for uniquely measuring BC, and BC only. Therefore, the term BC is used in a qualitative manner. Only certain BC properties can be quantified, e.g. the absorption coefficient  $\sigma_{ap}$ , the equivalent black carbon (EBC) concentration, obtained by multiplying  $\sigma_{ap}$  with an assumed mass absorption coefficient, or the elemental carbon (EC) concentration, obtained by thermal-optical analysis.

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Countries bordering the Arctic, i.e. the Scandinavian countries, Canada, USA, and Russia are known to use substantial amounts of wood for residential heating; e.g. residential wood burning accounts for more than 60 % (<https://www.ssb.no>) of the Norwegian PM<sub>2.5</sub> emissions of which BC is likely to constitute 20–30 %. Due to the proximity to the Arctic, BC emissions from these countries are disproportionally important with respect to the Arctic BC contribution compared to emission sources situated further south (Hirdman et al., 2010b). Also, there is a growing number of papers demonstrating that residential wood burning is more widespread and contributes more to the ambient PM, and BC, level in continental Europe than previously assumed (e.g. Sillanpää et al., 2006; Gelencsér et al., 2007; Puxbaum et al., 2007; Lanz et al., 2010; Maenhaut et al., 2012; Genberg et al., 2013). Projections made by the International Atomic Energy Agency (2008) show that the consumption of biomass is likely to increase by approximately 200 % between 2000 and 2020, in order to compensate for reductions in the use of fossil fuels.

Levoglucosan is a thermal degradation product of cellulose and a unique tracer of particulate matter emissions from biomass burning. The component possesses features which make it suitable to trace biomass burning aerosols in the ambient atmosphere; this includes in particular a high emission factor and a low vapour pressure (Locker, 1988; Simoneit et al., 1999; Oja and Sudberg, 1999). The studies by Locker (1988) and Fraser and Lakshmanan (2000) long provided the basis for levoglucosan being stable in the atmosphere for a time period extending up to 10 days. However, more recent indications of degradation of levoglucosan have been inferred from ambient measurements (e.g. Gao et al., 2003; Saarikoski et al., 2007), as well as demonstrated in a few recent laboratory studies (Hennigan et al., 2010; Hoffmann et al., 2010). In particular, aqueous phase reaction with OH radical in deliquescent particles appears to be the most efficient pathway causing depletion of levoglucosan in the atmosphere. E.g., Hoffmann et al. (2010) found  $\tau_{1/2}$  values (the time until half of the levoglucosan has been degraded) for levoglucosan ranging from 12.7 to 83.2 h in a model study representative of polluted continental plume conditions, where the variability in  $\tau_{1/2}$

reflected variations in photochemical activity and OH concentrations, being a function of temperature and season. However, confirming the range of  $\tau_{1/2}$  values reported by Hoffmann et al. (2010) for wood burning particles in the atmosphere is still missing.

Although levoglucosan appears to be best suited to trace biomass burning emissions in winter and on a local to regional scale, conservative estimates of the biomass burning aerosol concentration can still be provided for the remote environment. At present there are only a very few studies reporting levoglucosan concentrations for the Arctic environment (Stohl et al., 2006, 2007; Fu et al., 2009; von Schneidmesser et al., 2009; Zangrando et al., 2013) and typically for a short duration, days to a few months, and with a low time resolution (48 to 168 h). Thus, a time series of levoglucosan including all seasons and with an improved time resolution is needed to get an overview of the current biomass burning aerosol concentration in the Arctic. The presence of levoglucosan in the Arctic has been associated with boreal forest fires (Stohl et al., 2006), agricultural waste burning (Stohl et al., 2007), and closed combustion of wood (Hegg et al., 2009), but no attempt has been made to convert the ambient aerosol levels of levoglucosan into levels of carbonaceous aerosol, e.g. BC, from these respective sources.

The current study presents the first one-year time series of levoglucosan in the Arctic environment, and allows for a quantification of the BC fraction of the aerosol in the Arctic originating from agricultural and wild fires during summer, and from residential wood burning during winter.

## 2 Experimental

### 2.1 Site description and measurement period

Aerosol filter samples were collected at the Zeppelin atmospheric observatory (78°54' N, 11°53' E, 478 m a.s.l.) (Fig. 1) outside Ny-Ålesund at the west coast of the Spitsbergen Island in the Svalbard Archipelago (Norway). The samples were collected

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during the one year period from the 12 March 2008 to the 7 March 2009. The Zeppelin observatory is an important site in various global, regional and national monitoring networks, including the European Evaluation and Monitoring Programme (EMEP), the Global Atmospheric Watch (GAW), and the Arctic Monitoring and Assessment Programme (AMAP). The site is also included in the EU infrastructure project ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network).

## 2.2 Aerosol filter sampling for analysis of levoglucosan

Ambient aerosol filter samples subjected to levoglucosan analysis were obtained using a Digital high volume sampler equipped with a PM<sub>10</sub> inlet, collecting aerosols on prefired (850 °C; 3 h) quartz fibre filters (Munktel MK360; 150 mm in diameter, batch number 11415138). The sampler was operated at a flow rate of 20 m<sup>3</sup> h<sup>-1</sup>. The sampling time was 24 h. The sampling inlet was installed approximately 2 m above the roof level of the observatory and 5 m above the ground level.

For certain days throughout the sampling period the Digital sampler was not operational, hence filter samples were obtained from a co-located high volume sampler with a PM<sub>2.5</sub> inlet collecting aerosols on 8'' × 10'' cellulose filters (Whatman 41) according to a 2+2+3 days weekly sampling scheme. The sampler was operated at a flow rate of 1.1 m<sup>3</sup> h<sup>-1</sup>. Post exposure and prior to analysis the filter samples were wrapped in aluminium foil, placed in polyethylene zip lock bags, and stored in a freezer (-18 °C). One field blank was assigned to each month of sampling and was treated in exactly the same manner regarding preparation, handling, transport and storage, as the filters being exposed, except that they were not inserted in the sampler.

## 2.3 Measurement of levoglucosan, mannosan and galactosan

Quantification of the monosaccharide anhydrides (MAs) levoglucosan, mannosan and galactosan were performed according to the method described by Dye and Yttri (2005), and is only briefly described here. 50–100 % (77–154 cm<sup>2</sup>) of the quartz fibre filter were

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soaked in tetrahydrofuran (10 mL) and subjected to ultrasonic agitation (30 min). The filter extract was filtered through a syringe filter (0.45  $\mu\text{m}$ ) to remove PM and filter parts. Each filter was extracted twice. The extracted volumes were pooled and evaporated to a total volume of 1 mL in an  $\text{N}_2$  atmosphere. Before analysis the sample solvent elution strength was adapted to the mobile phase by adding Milli-Q water (0.8 mL). The concentrations of MAs were determined using High Performance Liquid Chromatography (HPLC) in combination with HRMS-TOF (High Resolution Mass spectrometry Time-of-Flight) operated in the negative electrospray mode. The compound separation was performed with two series-connected reversed-phase C18 columns (Atlantis dC18, Waters). Levoglucosan, mannosan, and galactosan were identified on the basis of retention time and mass spectra of authentic standards. Quantification was performed using isotope labeled standards of levoglucosan and galactosan.

## 2.4 Measurement of black carbon (BC)

The concentration of light absorbing particles was measured using a particle soot absorption photometer (PSAP). In this instrument, light (522 nm wavelength) illuminates two 3 mm diameter spots on a single filter substrate, on one of which particles are collected from ambient air flushed through the filter, while the other is flushed with the same amount of particle free air for reference. The particle light absorption coefficient  $\sigma_{\text{ap}}$  is derived from the change in light transmittance across the filter. The influence of particle scattering has been corrected according to Bond et al. (1999). Assuming that all the light absorption measured is due to BC and that all BC has the same light absorption efficiency,  $\sigma_{\text{ap}}$  values can be converted to equivalent BC (EBC) mass concentrations when divided by the mass absorption cross section ( $\alpha$ ). For the  $\alpha$  value used to calculate EBC in the present study, see Sect. 2.7.



## 2.5 Measurements of elemental, organic and total carbon

Total carbon (TC), elemental carbon (EC), and organic carbon (OC) were quantified using the thermal-optical transmittance (TOT) instrument from Sunset laboratories Inc., which corrects for charring of OC during analysis (Birch and Cary, 1996). The instrument was operating according to the EUSAAR\_2 protocol (Cavalli et al., 2010).

## 2.6 Emission ratios used to calculate EC from biomass burning ( $EC_{bb}$ )

For residential combustion, emission ratios for levoglucosan have been reported, or can be derived, for a wide range of softwood and hardwood trees, see e.g. Fine et al. (2001, 2002a, b, 2004), Mazzoleni et al. (2007), and Simoneit et al. (1999), but are typically for US tree types, using US appliances. There are only few stove measurements for European tree types, e.g., Puxbaum et al. (2007) and Schmidl et al. (2008). Studies with single appliances show a large variability, owing to different combustion conditions (e.g. Schmidl et al., 2008), different technologies or type of wood used. Studies using ambient data could provide a good alternative, as they reflect the weighted mean of the all the factors mentioned above, but results will vary from region to region (Yttri et al., 2009).

Obviously, the choice of emission ratio will be critical for the EC derived from biomass burning ( $EC_{bb}$ ) emissions. Thus, in order to account for the fact that the emission ratio is likely to differ as a function of e.g. wood type, type of appliance, and flaming vs. smoldering combustion, and consequently between geographical regions, we have allowed for a rather wide range in our calculations and adopted the range and approach reported by Yttri et al. (2011a) (see Table 1). As a consequence, the analytical method used to determine levoglucosan is self-consistent, as the method by Dye and Yttri (2005) was applied both in the present study as well as in that of Yttri et al. (2009), from which the emission ratio range listed in Yttri et al. (2011a) was derived. Thus, one does not employ any assumptions about the equivalence of the ambient data with those of an emission inventory.

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Emission ratios for wild and agricultural fires likely have a similarly wide range as those reported for residential wood burning, as the vegetation type being burnt can vary widely. In the present study we derived emission ratios of levoglucosan from the study of Saarikoski et al. (2007), which reported observed concentrations of levoglucosan, EC and OC at a rural background site in Helsinki during influence of emissions from the major wild and agricultural fires taking place in Eastern Europe in Spring, 2006 (see Table 2). A main argument for deriving emission ratios from Saarikoski et al. (2007) is the proximity of Helsinki with respect to the area burnt. Hence, the transport time of the air masses passing over Helsinki from the area burnt causes deposition as well as potential depletion of levoglucosan by OH still to be moderate. Also the area burnt is considered to be in the Sub-Boreal region and thus ought to be representative for wild/agricultural fire emissions impacting the European Arctic. Indeed, emissions from this exact fire event have been reported to reach the European Arctic, causing record high levels of air pollutants (Stohl et al., 2007). Further, the LC–MS method used for levoglucosan analysis by Saarikoski et al. (2007) was similar to the method of Dye and Yttri (2005), which was used in the present study, and which also is the basis for the levoglucosan emission ratio used for emissions from residential wood burning.

$EC_{bb}$  was calculated using Eqs. (1)–(3) (see Table 1), using the emission ratios listed in Table 2. Obviously, a mix of emissions from residential wood burning and wild/agricultural fires can occur (see Fig. 2d), particularly during the transient periods in spring and fall. In the present study we have used weighted emission ratios according to the relative contribution of BC from wild/agricultural fires vs. BC from residential wood burning, as predicted by the FLEXPART model. See Sect. 2.8 for further details on the model runs, and Fig. 2d for the time series of BC associated with wild/agricultural fires and residential wood burning as predicted by the FLEXPART model.

### 2.7 $\alpha$ values for calculation of equivalent black carbon (EBC)

A challenge when providing a whole-year time series of the relative content of  $EC_{bb}$  in aerosol at Zeppelin lays in the fact that  $EC_{bb}$  can be calculated using only the lev-

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oglucosan concentration as proxy, while the (total) EC concentration as reference is available only in winter. Summer time EC concentrations are below the detection limit of the thermal-optical analysis at Zeppelin. To overcome this challenge, the relative  $EC_{bb}$  concentration is referenced not to the EC concentration, but to the equivalent black carbon (EBC) concentration derived from the aerosol absorption coefficient  $\sigma_{ap}$  by employing a mass absorption cross-section  $\alpha$ . The calculation is tied to the EC concentrations measured at Zeppelin by calculating a site specific  $\alpha$  using the concurrent measurements of  $\sigma_{ap}$  and EC in winter:

$$\alpha = \sigma_{ap}/EC \quad (4)$$

Deriving the relative  $EC_{bb}$  concentration,  $EC_{bb, rel}$ , is then straightforward:

$$EC_{bb, rel} = EC_{bb}/EBC = [LG](TC/LG)_{bb}(1 - (OC/TC)_{bb})/\sigma_{ap}/\alpha \quad (5)$$

where [LG] denotes the levoglucosan concentration, and the emission ratios  $(TC/LG)_{bb}$  and  $(OC/TC)_{bb}$  the source specific emission ratios taken from Yttri et al. (2011a) in winter, and Saarikoski et al. (2007) in summer (see Table 2). We obtain a site and season specific  $\alpha$  value of  $5.7 \pm 2.3 \text{ m}^2 \text{ g}^{-1}$  for the Zeppelin Observatory in winter. The number of samples ( $n = 35$ ) from which the  $\alpha$  value was derived was limited by the low ambient EC levels and the detection limit of the thermal-optical instrument (see Sect. 3). In the absence of any other more specific information, the same  $\alpha$  value is used for summer as well.

The  $\alpha$  value obtained and applied in the current study is substantially lower than that used for Arctic sites by Sharma et al. (2013), but consistent with that applied by Hegg et al. (2011) for Svalbard. Various instruments, operating at different wavelengths likely explain most of the variability.

### 2.8 FLEXPART model simulations

BC concentrations and source receptor relationships were simulated with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005; Stohl and

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Thomson, 1999) using three-hourly operational meteorological analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF) with 91 model levels and a horizontal resolution of  $1^\circ \times 1^\circ$ . For each levoglucosan measurement carried out at Zeppelin 50 000 particles were released and followed 30 days backward in time.

5 A  $BC_{bb}$  tracer was used that is subject to dry and wet deposition. The model set-up and the parameters used for the  $BC_{bb}$  tracer were identical to the BC tracer simulations described by Stohl et al. (2013). However, for the  $BC_{bb}$  tracer we only considered BC emissions from residential wood burning, wildfires and agricultural fires (see below).

10 The experimental determination of  $EC_{bb}$  according to Eqs. (1)–(3) assumes that levoglucosan is not chemically degraded in the atmosphere. However, in reality the levoglucosan chemical half life time  $\tau_{1/2}$  may range from 12.7 to 83.2 h according to Hoffmann et al. (2010). To explore the impact of such degradation on modelled  $BC_{bb}$  concentrations, chemical decay following a number of different half life times (1/2, 1, 2, 3, 4 and 5 days) was superimposed on the other BC loss processes (i.e. dry deposition and wet scavenging) (see Table 3). The differences to simulated  $BC_{bb}$  without decay allow quantifying the impact of this process.

20 The estimate of emissions from residential wood burning and open burning of agricultural residues used in this study have been developed with the GAINS (Greenhouse gas – Air pollution Interactions and Synergies) model (Amann, 2011, see <http://gains.iiasa.ac.at>) within the ECLIPSE (Evaluating the CLimate and Air Quality Impacts of ShortlivEd Pollutants) project. More detailed description is provided in Klimont et al. (2013) and Kupiainen and Klimont (2007) and gridded emission data sets are available through the ECLIPSE project website (<http://eclipse.nilu.no>) upon request. Open biomass burning (forest and savannah fires) emissions originate from the Global Fire Emissions Database (GFED) version 3.1 (van der Werf et al., 2010).

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### 3 Quality assurance

5 Sampling was conducted for a time period of 352 days of which 321 days were covered by the sampling. The mismatch between the two numbers was typically attributed to instrument malfunction. In total 289 filter samples were collected. For some filters the sampling time extended beyond 24 h because poor weather conditions prevented the engineers to access the station. Certain filter samples were considered unacceptable for analyses because they were soaking wet from snow entering the inlet during harsh weather conditions. Moreover, during occasions when the Digital sampler was not operational due to e.g. maintenance, filter samples were obtained from a co-located high volume sampler operating at a 2 + 2 + 3 days weekly sampling scheme. In total 106 samples were collected during summer time (May–September) sampling, whereas the corresponding number for winter (October–April) was 183.

10 Aerosol particles emitted from biomass burning are found to reside in the fine particle size fraction (e.g. Kleeman et al., 1999; Hedberg et al., 2002; Yttri et al., 2005). Hence, we regard the PM<sub>2.5</sub> cut-off size of the co-located high-volume sampler, which was used when the Digital high volume sampler (PM<sub>10</sub> inlet) was out of order due to maintenance, as a minor uncertainty.

15 The HPLC/HRMS-TOF methodological limit of detection (LOD) for levoglucosan, mannosan and galactosan at a signal-to-noise ratio of three was 7.5 pgm<sup>-3</sup>. 97 % of the samples contained levoglucosan at levels exceeding the LOD. Concurrent data of the isomeric compounds mannosan and galactosan were available for only 30 % of the samples, due to their low observed levels. Levoglucosan was the most abundant isomer accounting for 79 ± 6.4 % of the sum of the three isomers, which is comparable to what has been reported in the literature for areas impacted by biomass burning (e.g. Zdrahal et al., 2002; Yttri et al., 2011b). When calculating mean concentrations, values below the detection limit were accounted for by using a value corresponding to 1/2 of the LOD. The precision was found to be satisfactory with a relative standard deviation below 5 %. The quartz fiber filters field blanks did not contain monosaccharide

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anhydrides, while the cellulose filters had a 30 pg content of levoglucosan per filter, which was accounted for when performing the calculations.

Thermal-optical analysis (TOA) of EC, OC and TC was conducted for a selection of samples only ( $n = 73$ ); i.e. for the filter samples experiencing the highest concentration of levoglucosan and/or filters which showed visual sign of colouring. 12 of the filter samples subjected to TOA were picked from the period 20 March to the 25 April 2008, whereas the remaining filter samples were from the period 18 November 2008 to the 7 March 2009. 10% of the filter samples had TC concentrations below the TOA analytical detection of  $0.2 \mu\text{g C m}^{-2}$ , whereas the corresponding percentage for EC was 45%. Field blanks ( $n = 2$ ) showed remarkably low levels of carbon concentrations; i.e.  $< 0.2 \mu\text{g C cm}^{-2}$ , being the analytical LOD of the TOA instrument. The sampling approach did not allow for addressing positive and negative artefacts associated with sampling of OC.

## 4 Results and discussion

### 4.1 Levels of levoglucosan

Ambient concentrations of levoglucosan observed at the Zeppelin Observatory during the period 12 March 2008–7 March 2009 are shown in Fig. 2a. The annual mean concentration of levoglucosan was  $0.68 \text{ ng m}^{-3}$ . Elevated concentrations in winter (October–April) (mean  $1.02 \text{ ng m}^{-3}$ ) compared to summer (May–September) (mean  $0.13 \text{ ng m}^{-3}$ ) were observed, resembling the well known seasonal pattern seen for e.g. sulphate and BC caused by the Arctic haze phenomenon (Shaw, 1995). The winter time mean concentration of levoglucosan at the Zeppelin observatory was two to three orders of magnitude less than typical concentrations reported for European urban areas in winter (Szidat et al., 2009), and one to two orders of magnitude less than that of the European rural background environment (Puxbaum et al., 2007; Yttri et al., 2013). The very few studies reporting levoglucosan concentrations for the Arctic environment,

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i.e. Stohl et al. (2006, 2007) and Zangrando et al. (2013) for the Zeppelin observatory, Fu et al. (2009) for Alert (Canada), Schneidemesser et al. (2009) for Summit (Greenland) and Qu et al. (2013) for the Beaufort Sea (Canada), have found levels within the concentration range reported in the present study. The daily maximum concentration of  $10.4 \text{ ng m}^{-3}$  was observed on the 17–18 January 2009 and is more than three times higher than the concentration reported by Stohl et al. (2007) ( $3.2 \text{ ng m}^{-3}$ ) during the severe air pollution episode caused by agricultural fires in Eastern Europe affecting the European Arctic in spring 2006.

The highest monthly mean concentration of levoglucosan was observed for January ( $3.2 \text{ ng m}^{-3}$ ), which along with February ( $2.0 \text{ ng m}^{-3}$ ) were the only two months for which the mean concentration exceeded  $1.0 \text{ ng m}^{-3}$ . The lowest monthly mean was observed for September ( $0.07 \text{ ng m}^{-3}$ ).

The ambient concentration of levoglucosan (Fig. 2a) showed minor variability during summer time sampling with only two episodes of elevated levoglucosan levels. The maximum 24 h concentrations increased by a factor of 7–11 compared to the summer time mean ( $0.13 \text{ ng m}^{-3}$ ) during these episodes. During winter, episodes of elevated levoglucosan concentration were more frequent than in summer. Peaks of similar magnitude as seen during summer were observed regularly from the middle of October on. From the start of December and until the end of the sampling period (7 March), the levoglucosan time series was characterized by several episodes and periods of considerably elevated concentrations, one of which extended up to two and a half weeks duration in January. The 24 h maximum levoglucosan concentration for the entire year of sampling was observed during this period, exceeding  $10 \text{ ng m}^{-3}$ , hence being one order of magnitude higher than the winter time mean ( $1.02 \text{ ng m}^{-3}$ ). The winter time sampling period also included the months March and April in 2008, and a major peak in the transition between March and April is evident.

## 4.2 Main source regions contributing to elevated levoglucosan concentrations at the Zeppelin Observatory

To identify the main source regions of the observed levoglucosan concentrations at the Zeppelin Observatory we have used the FLEXPART model (see Sect. 2.8) to calculate the emission sensitivity of BC tracer aerosols for the summer and winter months. Figure 3 shows the overall sensitivity of measurements at the Zeppelin Observatory to surface emissions during the last 30 days of air transport to the site for summer (panel a) and winter (panel b), respectively. High values show where the air mass has been in contact with the surface for a long time (unit:  $\text{ns m}^{-3}$ ), and consequently has the greatest potential to take up emissions. Hence, the plots indicate where potential surface sources influence the measurements. There is a clear seasonal variation in the BC emission sensitivity. During winter (panel b), relatively high values extend towards northern Eurasia, and as far south as approximately  $50^\circ \text{N}$ , covering large parts of Russia, Scandinavia and Eastern Europe. This is expected to be the most important source region for BC during winter time at Zeppelin. During summer (panel a), high values are limited to the Arctic Ocean basin and the emission sensitivity decreases distinctly from the Northern continental coast line and southward. This indicates that BC aerosol particles emitted at the relatively warmer land masses at lower latitudes are less likely to reach the Zeppelin Observatory on a 30 day timescale during the summer season. Consequently, sources located in the central European and Eurasian region are less likely to have a direct impact on the measurements during summer than during winter. This is consistent with the current understanding of atmospheric transport patterns in the Arctic (Hirdman et al., 2010b and references therein).

## 4.3 Levels of equivalent carbon (EBC) and EC from biomass burning ( $\text{EC}_{\text{bb}}$ )

Ambient concentrations of equivalent black carbon (EBC) and elemental carbon from biomass burning ( $\text{EC}_{\text{bb}}$ ) at the Zeppelin Observatory during the one year sampling period are shown in Fig. 2b and c, respectively. The annual mean concentration of

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cept for  $\text{SO}_4^{2-}$  ( $R^2 = 0.74$ ). Consequently, we cannot point to either of the two suggested reasons explaining the high correlation observed between levoglucosan and  $\sigma_{\text{ap}}$ .

#### 4.4 Comparison of modelled and observed $\text{EC}_{\text{bb}}$

The FLEXPART modelled  $\text{EC}_{\text{bb}}$  (here:  $\text{EC}_{\text{bb}, \text{m}}$ ) from wild fires and agricultural fires (green curve) and residential wood burning (red curve) are shown in Fig. 2d. The model captures the seasonal pattern of the levoglucosan time series with elevated levels in winter compared to summer. Also the mean seasonal concentration is in relatively good agreement, although the deviation during summer is considerably larger than during the winter period; i.e. the mean modelled winter and summer concentration of  $\text{EC}_{\text{bb}, \text{m}}$  was  $8.5$  and  $4.0 \text{ ng m}^{-3}$ , respectively, compared to  $3.9$  (winter) and  $0.9 \text{ ng m}^{-3}$  (summer) obtained on the basis of the levoglucosan observations. Note that the observational based means are slightly different from those presented in Sect. 4.3, as the population of samples compared consists of samples which have concurrent measurements of EBC as well.

We consider OH degradation of levoglucosan and uncertainties in the emissions to be the most important reasons for the observed deficit between the modeled and observational derived mean  $\text{EC}_{\text{bb}}$ . Uncertainties in emissions from biomass burning have been estimated to range from  $-50\%$  to  $+200\%$  for both contained (stoves, boilers) and open burning sources (Bond et al., 2004), and substantial differences between countries can appear. The perhaps most important criterion for a tracer to be used in a quantitative manner is that its mass is conserved from emission source to receptor point or its degradation well known. Hence, in the current study, any selective depletion of levoglucosan following from reaction with the OH radical, as shown by e.g. Hoffmann et al. (2010), will cause an underestimation of  $\text{EC}_{\text{bb}}$ . Hence, the levoglucosan concentration observed at the Zeppelin Observatory will provide minimum estimates of the biomass burning EC aerosol concentration. To explore the possible magnitude of OH degradation, we have re-calculated the  $\text{EC}_{\text{bb}, \text{m}}$  model tracer with assumed chemical

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half life times of 1/2, 1, 2, 3, 4 and 5 days (see Table 3), spanning the range of half life times of 12.7–83.2 h given by Hoffman et al. (2010), being shorter in summer compared to winter. The ratio between the modelled concentration of  $EC_{bb, m}$ , assuming dry and wet deposition only, and that of  $EC_{bb, m}$  including chemical degradation in addition to wet and dry deposition would then give the relative underestimation due to ignoring the chemical degradation. These ratios can in turn be used to provide chemical life time corrected concentrations of  $EC_{bb}$ , denoted  $EC_{bb}^*$  (see Table 3). Chemical half-life times of levoglucosan ranging from 3–4 days include the  $\tau_{1/2}$  values suggested for winter by Hoffmann et al. (2010), and suggest that  $EC_{bb}$  is degraded by a factor of 5–7, which corresponds to an  $EC_{bb}^*$  concentration ranging from 20–28  $ng\ m^{-3}$ . Chemical half life times ranging from 1/2–2 days includes the  $\tau_{1/2}$  values suggested for levoglucosan by Hoffmann et al. (2010) for summer. Compared to winter, degradation in summer appears much higher (i.e. a factor of 130–4000). This in turn corresponds to an  $EC_{bb}^*$  concentration range which far exceeds that of the summer time mean EBC concentration, and thus is unlikely. A chemical life time of 4 days, which is in line with that suggested for winter (Hoffmann et al., 2010), provides an  $EC_{bb}^*$  concentration of 18  $ng\ m^{-3}$  for summer. This finding indicates that the chemical life time of levoglucosan during the Arctic summer is longer than that suggested by Hoffmann et al. (2010). Given the lower concentrations of OH in winter compared to summer, chemical life times for levoglucosan extending beyond the range suggested by Hoffmann et al. (2010) could also be assumed during the Arctic winter. Indeed, the chemical life times suggested by Hoffmann et al. (2010) are best estimates for typical tropospheric aqueous phase oxidation processes under polluted continental plume conditions and thus their use under other atmospheric conditions should be performed with caution. Nevertheless, Hoffmann et al. (2010) is the most comprehensive study published on this topic yet and thus is a natural choice for the current study.

Episodes of elevated concentrations of levoglucosan are in many cases nicely reflected in the modelled time series, however sometimes the timing and duration of the episodes are not captured well. During the spring and summer months, the model pre-

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dicts two prolonged periods influenced by wild and agricultural fires at Zeppelin, both extending about one month in time; i.e. from 20 April to 20 May 2008 and from the 1 August to the 25 August 2008. The first of the two periods corresponds with an increase in the observed level of levoglucosan ( $1.4 \text{ ng m}^{-3}$ ). However, the levoglucosan level does not continue with high values as long as predicted by the model. For the second part of the period the observed level of levoglucosan is below the summertime mean of  $0.13 \text{ ng m}^{-3}$ . Unfortunately, during the August episode only a few levoglucosan measurements were available. Observations of levoglucosan exist only for the first four days of August, for which the level increased to approximately  $1 \text{ ng m}^{-3}$  and hence corroborates to the increased levels from wild and agricultural fires predicted by the model.

The winter period is characterized by much higher background values and a number of peaks. The model predicts a period of increased emissions from residential wood burning from mid November 2008 and until the end of the sampling period in the beginning of March 2009. This is in line with the observational based time series. For two of the most substantial periods, extending up to 13 (9–22 December 2008) and 18 days (13–31 January 2009) in length, the model matches well with the observed time series of levoglucosan, both with respect to onset and duration. The maximum of the first period was reached on 15 December when the air was transported from Central Europe. The maximum of the second period was observed on 17 January when the source region was confined to the northern parts of Central Europe, as well as for a region extending to the East and the West of the Ural mountain range, being centralised around the major Russian cities Perm and Sverdlovsk.

During the winter time period there were two distinct episodes with a rapid increase in the levoglucosan and  $\text{EC}_{\text{bb}}$  concentration, both indicated with an orange asterisk in Fig. 2c, that are not captured well by the model. One is the major episode observed during the transition of March to April (29 March to 6 April 2008) with a maximum levoglucosan concentration of  $5.6 \text{ ng m}^{-3}$ . The model predicts an influence by both agricultural/wild fires and residential wood burning during the period, but neither of the two sources, nor the sum of them, seems to reconstruct the evolution of the episode. The

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second episode is characterized by a sharp peak rising from 2 to 3 February 2009, reaching a peak levoglucosan concentration of  $7.7 \text{ ng m}^{-3}$ . One plausible explanation of these observed deviations is the distribution and strength of the emissions used in the model. The days 29 March to 2 April during the 29 March to 6 April episode in 2008 is not captured well by the model, while the smaller peak on the 6 April is reproduced. From 27 March to 2 April, the air transport shifted from air masses originating mainly from the Arctic Ocean basin to increasingly include more of air masses from Northern Russia. For the last part of the period, extending towards 6 April, the air masses were almost exclusively originating from the North-American continent, passing over the Arctic Ocean. This is illustrated in Fig. 4; Panel a shows the emission flux of BC from the GFED and GAINS residential wood burning emissions for the month March 2008 [unit:  $\text{g month}^{-1} \text{ m}^{-2}$ ], whereas the footprint sensitivities for the days 27 and 31 March and 6 April 2008 are shown in panel b–d.

A similar analysis was performed for the episode early in February, for which a rapid increase in the levoglucosan concentration was observed; i.e. it increased by a factor of 9 from 2 to 3 February. A corresponding increase was observed for EBC. Only a modest increase, starting earlier, is predicted by the model for these days, peaking at the 4 to 5 February. For this episode there was a rapid shift in transport to the Zeppelin Observatory from the Atlantic Ocean and Northern-Europe to air masses from the Northern Russian region. These two examples indicate that there might be an underestimation in the emissions strength from residential wood burning in Russia, while such emissions from the other regions seem to fit relatively well with the observations. A more systemic study of the distribution of the emissions from residential wood burning in Russia is necessary to assess its importance for the  $\text{EC}_{\text{bb}}$  level in the Arctic region.

### 4.5 Relative contribution of $\text{EC}_{\text{bb}}$ to EBC

The monthly distribution of the relative contribution of  $\text{EC}_{\text{bb}}$  to EBC is shown in Fig. 5. For the winter time period  $\text{EC}_{\text{bb}}$  was found to constitute  $8.8 \pm 4.5\%$  of EBC on average,

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whereas during summer  $EC_{bb}$  constituted  $6.1 \pm 3.4\%$  of EBC on average. These are lower estimates as chemical degradation of levoglucosan has not been accounted for. The mean relative contribution of  $EC_{bb}$  to EBC does not differ substantially between the summer and the winter time periods, despite totally different regimes with elevated  $EC_{bb}$  and EBC concentrations in winter compared to those in summer. As an example, the mean relative contribution of  $EC_{bb}$  to EBC for September (14 %) was close to that observed for January (16 %), despite that September recorded the lowest monthly mean level of levoglucosan and EBC and January the highest ones.

By means of the chemical life time corrected  $EC_{bb}$  concentrations ( $EC_{bb}^*$ ) presented in Table 3, an upper estimate for the relative contribution of  $EC_{bb}$  to EBC can be provided. During winter a chemical half life time of levoglucosan ranging from 3–4 days corresponds to a mean relative contribution of  $EC_{bb}^*$  to EBC of 31–45 %. For life times ranging from 1/2–2 days the relative contribution of  $EC_{bb}^*$  to EBC becomes unrealistically high for summer; i.e. in excess of 100 %. In summer, only a life time extending to 4 days or more provide a realistic mean contribution of  $EC_{bb}^*$  to EBC, i.e.  $< 65\%$ .

A recent study suggests that BC emissions from gas flaring dominate the BC emissions in the Arctic at latitudes greater than  $66^\circ$  N and that flaring emissions contribute 42 % to the annual mean BC surface concentrations in the Arctic (Stohl et al., 2013). The current study could support the conclusion by Stohl et al. (2013), at least in winter, as we find that  $EC_{bb}^*$  account for 31–45 % of EBC at most; hence, fossil fuel sources of BC appear to be most important in the Arctic near the surface. The rather high maximum relative contribution estimate of  $EC_{bb}^*$  to EBC in summer  $< 65\%$  does not allow for a support of fossil fuel sources being dominant.

When discussing the relative contribution of biomass and fossil fuel sources to the Arctic EBC loading, one should recall that the summer time concentration of EBC is very low. Thus, even if the chemical degradation is expected to be higher in summer, and consequently also the impact and uncertainty of our estimated summer time  $EC_{bb}$  and  $EC_{bb}$  to EBC values, the Arctic absorbing aerosol loading is dominated by the winter time sources; hence, for a successful mitigation, focus should be to understand

these in more detail. From a radiative forcing (climate) perspective, however, winter values are not as important as summer values, although they do lead to BC deposition, which subsequently may have a radiative impact persisting into spring/summer.

## 5 Conclusions

5 In the present study we have demonstrated the presence of biomass burning aerosol particles in the European Arctic based on measurements of levoglucosan at the Zeppelin Observatory at Svalbard. To our knowledge this is the first time series of levoglucosan in the Arctic covering a full year.

10 Elevated concentrations of levoglucosan in winter ( $1.02 \text{ ng m}^{-3}$ ) compared to summer ( $0.13 \text{ ng m}^{-3}$ ) were observed, resembling the seasonal variation seen for e.g. sulphate and BC caused by the Arctic haze phenomenon. The winter time mean concentration of levoglucosan was two to three orders of magnitude less than that reported for European urban areas and one to two orders of magnitude less than that of the European rural background environment.

15 Neglecting chemical degradation by OH provides minimum levoglucosan concentrations, corresponding to a mean  $\text{EC}_{\text{bb}}$  concentration of  $3.7 \pm 1.2 \text{ ng m}^{-3}$  in winter (October–April) and  $0.8 \pm 0.3 \text{ ng m}^{-3}$  in summer (May–September) or  $8.8 \pm 4.5 \%$  of the measured equivalent black carbon (EBC) concentration in winter and  $6.1 \pm 3.4 \%$  in summer. When accounting for OH degradation of levoglucosan, 31–45 % of EBC could be attributed to  $\text{EC}_{\text{bb}}^*$  ( $\text{EC}_{\text{bb}}$  adjusted for chemical degradation) in winter and < 65 % in summer. Hence, fossil fuel sources appear to dominate the European Arctic BC concentrations in winter, whereas we cannot conclude upon this for the warm season. We recommend that complementary, observational based approaches, such as e.g. radio-carbon measurements of EC, should be undertaken in the near future to narrow the range reported for  $\text{EC}_{\text{bb}}$  to EBC in the current study. Note though, that the Arctic EBC loading is totally dominated by the winter time sources, thus focus should be to understand these in more detail for a successful mitigation. On the other hand winter values

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are not as important as summer values from a radiative forcing (climate) perspective, although they do lead to BC deposition, which subsequently may have a radiative impact persisting into spring/summer.

Results from the current study indicate that the chemical lifetime of levoglucosan in the Arctic atmosphere is longer than that suggested by Hoffmann et al. (2010) for polluted continental plume conditions.

Calculations using the Lagrangian particle dispersion model FLEXPART show that the seasonal variation of the modelled  $EC_{bb}$  concentration compared relatively well with observationally derived  $EC_{bb}$  from agricultural/wild fires during summer, and residential wood burning in winter. The model overestimates by a factor of 2.2 in winter and 4.4 in summer when compared to the observationally derived mean  $EC_{bb}$  concentration, which provides the minimum estimate, whereas it underestimates by a factor of 2.3–3.3 in winter and a factor of 4.5 in summer when compared to  $EC_{bb}^*$ , which provides the upper estimate.

The current study indicates that there might be too low emission of residential wood burning in Northern Russia, a region of great importance for the BC concentrations observed in the European Arctic. This topic needs further investigation.

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**Table 1.** Equations used to calculate concentrations of EC from biomass burning ( $EC_{bb}$ ).

$$TC_{bb} = [LG] \times (TC/LG)_{bb} \quad (1)$$

$$OC_{bb} = TC_{bb} \times (OC/TC)_{bb} \quad (2)$$

$$EC_{bb} = TC_{bb} - OC_{bb} \quad (3)$$

Notes: Square brackets denote measured quantities. Parentheses denote emission ratios (see Table 2).

Notation: TC = Total Carbon;  
 LG = Levoglucosan; OC = Organic Carbon;  
 EC = Elemental Carbon;  
 subscript bb = biomass burning.



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**Table 2.** Emission ratios (Mean  $\pm$  SD) used to calculate concentrations of  $EC_{bb}$  according to Eqs. (1)–(3).

	–SD	Mean	+SD
Res wood burning <sup>1</sup>			
$(TC/LG)_{bb}$	10.9	14.7	18.4
$(OC/TC)_{bb}$	0.73	0.78	0.82
Wild/Agricultural fires <sup>2</sup>			
$(TC/LG)_{bb}$	38	48	58
$(OC/TC)_{bb}$	0.80	0.85	0.90

<sup>1</sup> Yttri et al. (2009, 2011a)

<sup>2</sup> Saarikoski et al. (2007)

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**Table 3.** Modeled ( $EC_{bb,m}$ ), observationally derived ( $EC_{bb}$ ) and chemical life time corrected ( $EC_{bb}^*$ ) mean concentrations of EC assuming chemical half life times ( $\tau_{1/2}$ ) of levoglucosan ranging from 12 h to 5 days and relative contribution of  $EC_{bb}^*$  to EBC.

	$EC_{bb}(\text{ng m}^{-3})$	$EC_{bb}^*(\text{ng m}^{-3})$	$EC_{bb}^*/\text{EBC}(\%)$
<b>Winter</b>			
$EC_{bb}$ ([Levo]* ER) Observed	3.9		
$EC_{bb,m}$ Mod (Dry/Wet dep.)	8.5		
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 1/2	0.03	1203	1908
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 1	0.18	181	286
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 2	0.67	50	72
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 3	1.2	28	45
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 4	1.7	20	31
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 5	2.1	15	25
<b>Summer</b>			
$EC_{bb}$ ([levo]* ER) Observed	0.91		
$EC_{bb,m}$ Mod (Dry/Wet dep.)	4.0		
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 1/2	0.001	4672	16 556
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 1	0.004	802	2843
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 2	0.03	123	436
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 3	0.09	39	139
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 4	0.20	18	65
$EC_{bb,m}$ Mod (Dry/Wet dep.) Life t. 5	0.33	11	39

$EC_{bb}^*$  =  $EC_{bb}$  adjusted with respect to lifetime/degradation as predicted by the FLEXPART model.

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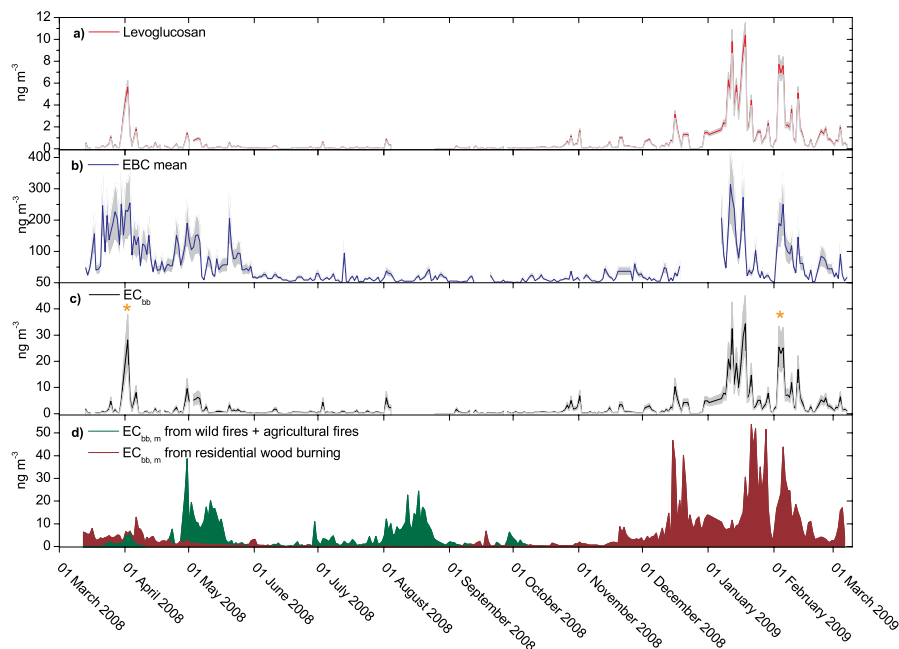


**Fig. 1.** The Zeppelin Observatory located at the Zeppelin Mountain (478 m a.s.l.) close to Ny-Ålesund (Svalbard) (78°54' N, 11°53' E).

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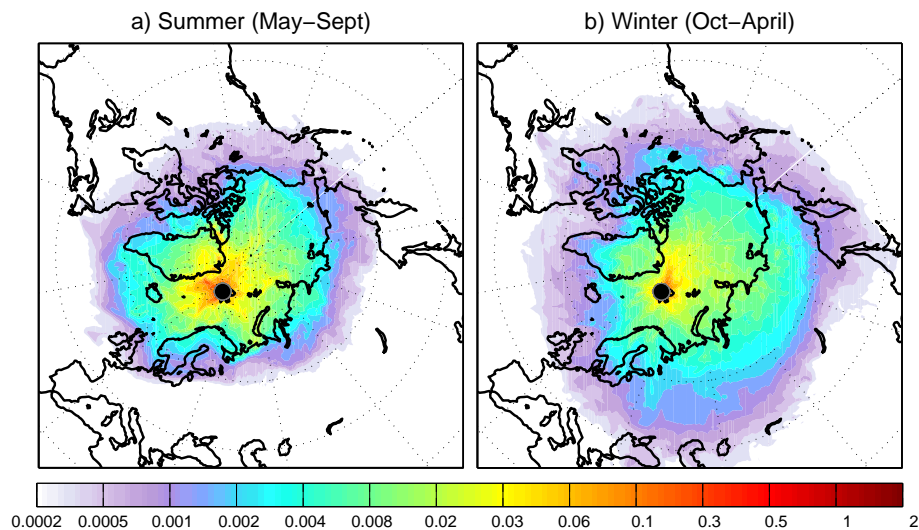
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**Fig. 2.** Ambient aerosol concentrations of levoglucosan **(a)**, equivalent black carbon (EBC) **(b)**, and elemental carbon from biomass burning ( $EC_{bb}$ ) **(c)**, presented as 24 h mean concentrations for the Zeppelin Observatory during the period 12 March 2008–7 March 2009. **(d)** shows modeled concentrations of  $EC_{bb}$  ( $EC_{bb,m}$ ), as obtained by the FLEXPART model. The green curve is the sum of EC from wildfires and agricultural fires, whereas the red curve is EC from residential wood burning. The two orange asterisks in **(c)** indicates episodes with a rapid increase in the  $EC_{bb}$  concentration that are not captured well by the model and which are discussed in further detail in Sect. 4.4.

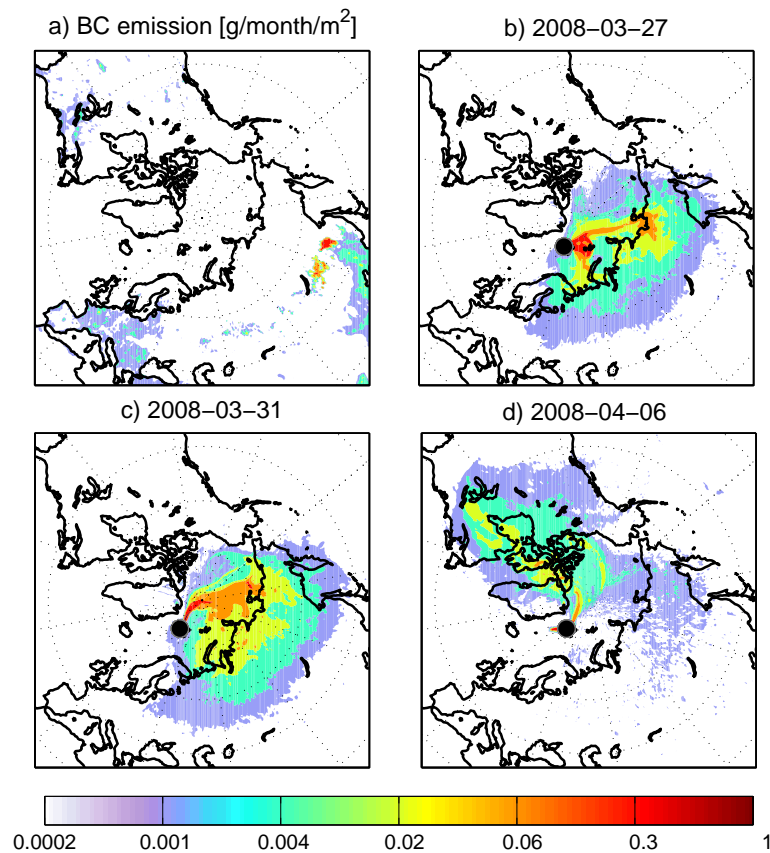
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**Fig. 3.** The overall emission sensitivity of measurements at the Zeppelin Observatory to surface emissions during the last 30 days of air transport to the site for summer (here: Mean of June–August 2008) **(a)** and winter (here: Mean of January–March 2009) **(b)** respectively. Unit:  $\text{ns m}^{-3}$ .

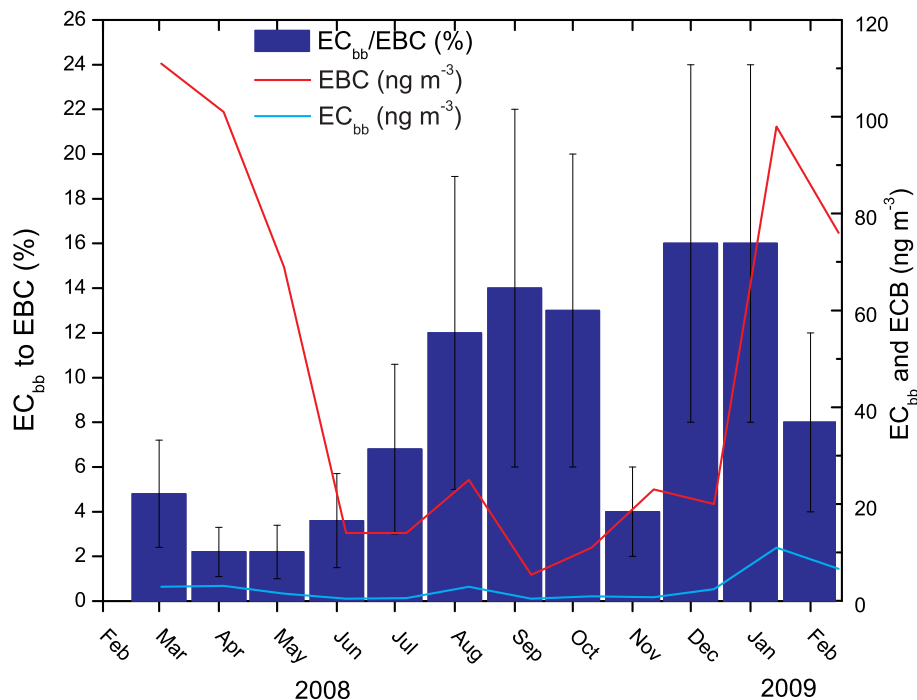
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**Fig. 4.** (a) shows the geographical distribution of the total emission flux of BC from GFED and GAINS biofuel emission for March 2008 [kg<sup>s</sup><sup>-1</sup>]. (b-d) shows the footprint sensitivities for the days 27 and 31 March and 6 April 2008.

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**Fig. 5.** The blue bars show the monthly mean relative contribution of elemental carbon from biomass burning ( $EC_{bb}$ ) to total equivalent black carbon (EBC) at Zeppelin for the time period March 2008–February 2009 (left axis). Standard deviations are included. The light blue curve is the monthly mean ambient concentration of elemental carbon from biomass burning ( $EC_{bb}$ ), whereas the red curve is that of equivalent black carbon (EBC) for the same period (right axis).