Dear Editor,

We thank the three referees for their useful comments and suggestions which have helped us to improve the manuscript. Our responses are below. The referees' comments are in *Italic* and our responses are in normal font. We also identify the locations of each correction in the revised manuscript using green shading, and the page numbers in our responses refer to the revised manuscript.

Referee 1:
The paper presents an analysis of the contribution of various source regions and sectors to the BC burden observed in the Arctic troposphere during spring of 2013. Attribution of the Arctic BC burden to different source regions and sectors is conducted through use of FLEXPART trajectory analysis. Precipitation amount based on HYSPLIT trajectory analysis is used to determine the fraction of BC that was scavenged en route to the Arctic. Overall, this paper contributes to our understanding of the sources that impact the BC burden in the Arctic and the altitude dependence of these impacts. One aspect that was lacking was an assessment of the significance of flaring in northern Russia on low altitude BC burdens in the Arctic as suggested by Stohl et al., ACP (2013). The paper should be publishable in ACP once this concern and those listed below have been addressed.

We thank the referee for their comments. Our reply to the comment regarding flaring is dealt with below in our response to the specific point raised. The revised manuscript includes an analysis of flaring emissions obtained from ECLIPSE global emission inventory in section 4.2 and Table S1.

Abstract: Define ng s mˆ-3.
Added on Page2 line17.

p. 14846, line 15: Is there a reference for this statement ("visibly reddish-brown in color")
We have removed this sentence.

p. 14846, line 26: Change to ": : :intensively conducted AT various: : :"
p. 14847, line 2: Change to " : : :transported TO and built up: : :."
p. 14847, last paragraph: change to "THE IMPACT OF open biomass burning sources ON BC CONCENTRATIONS IN THE ARCTIC WAS OBSERVED during the recent ARCTAS: : :."
p. 14847, line 26: Change to "forest fires in Siberia largely contributed: : :"
All of these have been corrected as suggested on Page 3 and marked as green shading.
p. 14848, line 4 - 7: This sentence is confusing. Please edit for clarity.
This has been clarified on Page 4, line 93-94.
p. 14848, Lines 8 – 9: What is the topic of considerable debate – the poor representation by models or the concentration of BC in the Arctic?
This sentence has been removed.
p.14848, line 21: What is meant by "microphysical system"?
This has been changed as "removal system" on line 107.
p. 14848, line 25: "an explicit determination of BC scavenging efficiency" is a tall order. How would this be done?
This has been changed as "and improved understanding of BC scavenging efficiency" on line 111.

p. 14849, line 22: Corrected for ambient what? Aerosol?
This is corrected on Page 5, line 137.

p.14850, line 5-6: What was the range of the applied collection efficiencies? Please give quantitative information on the degree of agreement between the AMS-measured mass and the SMPS volume concentration.
This point is corrected in Page5. Line 154-157.
The texts are added: "An AMS-SMPS volume comparison could not be performed as we have found that the SMPS concentrations at altitude are incorrect, possibly due to an inaccuracy in the assumed charge distribution during inversion".

p. 14850, line 11: "Data ARE missing: : :"
Figure 2: Define PES and FPES in the figure caption.
Figure 3: Define "NA", "CL", etc. in the figure caption.
Fig.2 and Fig.3 are revised.

p. 14853, lines 16 - 19: Are flaring emissions included in the anthropogenic emission inventory? Stohl et al., ACP (2013) suggest that flaring in northern Russia could be a large source of low altitude Arctic BC. If flaring is not included (or the emission inventory of Stohl et al. is not used), then the SI region may incorrectly be assumed to be a minor contribution to ground sources.
p. 14854, lines 5 – 14: Again, were flaring emissions included? Based on what inventory?
p. 14854, lines16 – 17: The statement that "SI emissions are relatively low during that time of year" suggests that only OBB and not flaring emissions are considered to be significant for this region.

It should be noted that the flaring emission sources at high latitude mainly advect into the Arctic at low altitude within the boundary layer, however the plumes observed in this study were within free troposphere. As Fig. S2 shows, the FLEXPART footprints do not significantly fall within the SI region for any of the observed plumes, which implies that flaring was not an important source, irrespective of the inventory data. However, to quantify the extent of the contribution, we have performed the analysis using a flaring inventory.

After the analysis, the following statement has been added: "In particular, flaring sources make a negligible contribution to the BC and CO loadings (of the order of 10^2 or 10^3 magnitude lower than the other anthropogenic contributions as shown in Table S1). This is because the plumes in this study were mainly encountered in the middle or upper troposphere and the resulting back trajectories show that there was little land contact over the high-latitude regions where flaring sources are present."

Revised texts are in Page 8 line 260 and line 299-304; a new Fig. S3E is added. The detailed results of flaring source contribution are in the revised Table S1.

p. 14856, lines 4 – 5: It is stated that "NA influences were low at all levels" yet the contribution of NA to lower troposphere SO4 is higher than or equivalent to contributions from the other regions (Figure 8c).
This is corrected on Page 5, line 315 by specifically relating the comment to BC.

p. 14856, lines 18 – 20: Do the precipitation fields from HYSPLIT indicate more precipitation for air masses en route from MA compared to the other regions?

We can't explicitly give the reason why BC size is smaller for NA, so this statement is removed.

p. 14857, lines 7 – 9: Change to "The lower-latitude source REGIONS had potential temperatures: : :."
This is corrected on Page 5, line 346.

Figure 11: Couldn't the lack of a correlation between precipitation and SF_BC also be due to the assumptions in the calculation of SF_BC, i.e., that BC/deltaCO_source is adequately parameterized using emission inventory data?

Though the emission ratio of SF_BC to CO is uncertain, for this effect to change the correlation plot as the referee suggests would imply that the ratio changes in a systematic way as a function of precipitation. We can see no physical reason for this to be the case. In addition, the uncertainty of the inventories is not provided and thus not able to be quantified in this study.

Referee 2:

This paper covers an interesting topic – black carbon in the Arctic. The paper provides good motivation and background for the necessity of studying the sources of black carbon to the Arctic. The findings are relevant to predictions of future climate and confirm that pollution from Asian sources contribute to the black carbon deposition and radiative warming in the Arctic. But the authors need to discuss the relevance of these findings for March 2013 compared to future years. The paper is well written but could be slightly shortened and more focused.

We have included a discussion of the long-term trends of open biomass burning and anthropogenic emissions in this region in the revised manuscript. We have tried to rewrite the text to make it shorter and more focused.

Major Comments.
The choice of "Arctic clean air" seems strange since the measurements here show influences of polluted air to the Arctic. At what point can the Arctic air be considered actually clean when it is influenced from other sources and when pollutants can be trapped? Could this instead be referred to as "Arctic background air" since it may not be perfectly clean? Additionally, is there any evidence that latitudes as far south as Mexico (larger than 30_N) can contribute to pollution in the Arctic and need to be included in the source regions suggested?

The "clean air" is replaced by "background air" throughout the revised version. The Mexico region is included in the predefined North America and has already been included in the emission sensitivity study.

The authors note that the black carbon source regions and concentration from biomass burning in this 2013 study are different from those in 2008. For this reason, is it then relevant to use 2013 as the main marker for the sources of black carbon to the Arctic? It is nice that 2013 was a year with low biomass burning emissions which allowed the

authors to clearly demonstrate the anthropogenic influence to black carbon concentrations. The authors should add some more discussion on how their findings for 2013 are or are not representative of a current and/or future black carbon contributions. Is 2013 relevant for the future or is it an outlier compared to other years? In years with higher biomass burning, is the anthropogenic source of black carbon from Asia still going to be as important, relatively, or will it be masked by biomass burning emissions?

In the revised version, we have performed a 10-year analysis on the fire counts based on FINN open biomass burning inventory; in addition the ECLIPSE anthropogenic emissions are compared between 2005 and 2010. A new Fig.12 is added and corresponding discussions are now included on Page 12, line 465-469: "Annual fire counts across Siberia from the FINN emissions inventory for March-April over the 10 year period from 2004 to 2013 are shown in Fig. 12A. The year 2008 is shown to be anomalously high, compared to the decadal average of 2.8 $x10^4$ by approximately a factor of 2.4. In all other years in the last decade fire counts in the Siberia spring varied between $1.4x10^4$ and $3.7x10^4$. This contrasts with 2013 which showed the lowest number of fire counts ($1.4x10^4$). Therefore, whilst 2008, does indeed show the importance of biomass burning in the Arctic, Asian pollution sources make a substantial contribution to Arctic BC during many spring seasons. Figure 12B shows the changes in anthropogenic emissions of BC between 2005 and 2010 as described in the ECLIPSE database. There is a marked increase in BC emissions over this period of between 20-30% and these increases are projected to continue in the coming decades."

With the mixing state of the black carbon described in this paper, how much of an influence will it make on reducing the albedo of snow and ice when it is deposited in the Arctic? How much is the effect of coated black carbon on the albedo compared to uncoated black carbon?

The referee raises some very important questions, and indeed these motivated our study. However, the paper is observationally based and the questions raised can only be answered through a modelling study that combines transport with deposition and radiative closure. This is far beyond what we can cover in this paper but we very much hope that our data can motivate such a study in future.

How relevant are anthropogenic emission inventories from March 2010 to a study that takes place in March 2013? It does seem that these would remain more consistent than the biomass burning regions over the years, but the authors should comment on this potential difference and whether or not it is important.

The latest year available in the ECLIPSE emissions inventory is for 2010 and we have now noted this in the text on Page 8, line 266-270. Our analysis discussed above shows that the inventory changed by 20-30% in Asia between 2005 and 2010 and less in other regions. It is therefore likely that any differences in absolute amount between 2010 and 2013 are less than this and the relative changes between regions are smaller. The anthropogenic inventory emissions at different years is compared and discussed in the revised manuscript.
Corrected texts are in Page 8, line 266-270.
Please refer to the new Fig. 12. The discussions are in Page 12, line 473-484.

For the FLEXPART and HYSPLIT models, the discussion about their usage and comparison needs some clarifying. Why were different numbers of days back used? Why were those chosen? Can black carbon be transported over that many days (at the given altitudes) without being scavenged? How does the height factor into the FLEX PART output? Figure 9

uses HYSPLIT back trajectories that go back 12 days, but the text states that uncertainties increase after 5 to 6 days.

The back trajectories of FLEXPART and HYSPLIT are both applied over 12 days. While the accuracy of the footprint decreases with time, this was the minimum duration found such that candidate source regions could be identified. Because of the small size and thus settling velocity of the particles, BC can be transported over many days in the free troposphere, where precipitation and mixing are minimal. As stated, the FLEXPART potential source footprints are given where the back trajectories are below 500m.

Minor Comments.

More information could be included in the Introduction about the cause of Arctic haze and the springtime meridional transport of air masses and pollutants from the midlatitudes to the Arctic.

The texts have been added on Page 3, line 76-78:

"The emissions in Eastern/Northern Asia have grown rapidly in the past two decades and many studies have pointed out that this region may have a significant impact on the Arctic BC concentration in late winter and early spring time, especially in the free troposphere (Koch and Hansen, 2005; Shindell et al., 2008; Wang et al., 2011; Shaw et al., 2010; Frossard et al. 2011). The meridional transport to the Arctic during springtime could also be important (Shaw et al., 2010; Marelle et al., 2014; Raatikainen et al., 2015)."

How is "pollution" defined in the abstract? Black carbon only or the mixture of the other species measured?

There is one place of "pollutants" we think need to be replaced by "BC", on other occasions we refer to pollution as we identified plumes by a number of particle characteristics and CO. On Page 2 line14, the term 'pollution' has been corrected to read "BC".

Were any other chemical species measured during this campaign that could be used as tracers for the anthropogenic pollution sources or biomass burning sources?

As the aircraft was primarily equipped for studying cloud microphysics, there were no other tracers available relevant to this work other than those discussed in this paper.

Other studies that have looked at sources of pollution in the Arctic should also be considered and included in the Introduction or Discussion: Rahn 1981 (used metal tracers to show the European sources of aerosol particles in the Arctic); Hole et al. 2006 (contributions from European anthropogenic sources to Arctic haze); Shaw et al. 2010 (anthropogenic and biomass burning sources, as well as shipping); Frossard et al. 2011 (contribution from European and Asian anthropogenic sources); Stohl et al. 2013, which is in the reference list but not discussed in the paper; Raatikainen et al. recently on ACPD (contribution from continental Europe to black carbon at Pallas GAW in Finland); etc.

These references are added in the revised version.

Page 14854, Line 6: Why was an anthropogenic inventory from March 2010 used to compare to a fire inventory from March 2013?

The 2010 anthropogenic emission inventory is the most recent one which can be obtained. See our response above.

Page 14847, Line 2: Transported from where?

Page 14848, Line 23: But if there is less transport of BC to the Arctic during the colder seasons, then the bias in the scavenging estimate should not be as important to the total estimate of BC in the Arcitc?
This is clarified on Page 4, line 110.
The authors could add more discussion about the relative importance of black carbon in the Arctic at the different levels of the atmosphere observed in this study.
More discussions are included Page 3, line 85-88.
"These sources are at high latitude locations and thus their emissions tend to follow low-level quasi-isentropic transport to the Arctic and influence the Arctic lower/middle troposphere (Stohl 2006)."

How was the FLEXPART model calculated at different heights?
The starting point of the retroplumes is specified at the exact horizontal and vertical locations of the observed plumes and is allowed to travel and disperse in three dimensions according to the model.

Page 14850, Line 1: How reliable or relevant are the chosen refractive indices for the core and the coating? Was all of the black carbon coated?

The detailed discussion can be found at Taylor et al., 2015 and Liu et al., 2014, which have been referenced in the texts.

Page 14850, Line 5: Drewnick et al. 2005 should be referenced here for the C-ToFAMS.
The authors need to include more discussion of the calculated collection efficiency, here or in the supplement. How reliable is this CE given that the black carbon core is refractory? Would that fraction of black carbon cause particles to bounce off of the detector? Or would the non-black carbon fraction just volatilize? How relevant is the Middlebrook CE technique when there is black carbon?

The Drewnick et al reference is now included.
Please see our comment above. Note that the AMS measures the total non-refractory mass. Given that the number of particles containing BC is less than 10% of the total number, any change in the CE induced by a BC core will not change the overall CE of the AMS. We cannot of course rule out completely that the material condensed onto the BC particles may be very different from the rest of the submicron non-refractory aerosol, but given that the majority of the material secondary in nature it is unlikely that this is the case.
We have added texts in the revised version to clarify.
The revised texts are in Page 5, line 148-154.

Page 14852, Line 24: Add some numbers to quantify "significantly higher". Is this ratio consistent with previous studies?
Page 14854, Line 17: What makes up the "residential sector"?
Page 14855: The second paragraph seems like just a list of where the data is presented.
These are clarified and corrected in the revised version.
Page 7, line 222; Page 8, line 270. The paragraph in line 301 has been largely shortened.

Page 14856: The coating paragraph seems out of place in this section. Are all of the coating organics on the BC?
The SP2 instrument is not able to determine the coating compositions.

Page 14858, Line 18: How well do the COsource and COmeasured compare?
It must be noted that we do not attempt to compare the absolute concentrations of the measured and modelled CO, as this would necessarily involve uncertainties associated with the initial dispersal of the emissions in the boundary layer, which are not the focus of this investigation.

Page 14860, Line 25: Add some references for "consistent with previous studies"
Added.

Figures and Tables.
Figure 1: Change the flight track color of B759 so that it is not the same color as the land. The outline of the land is very thick and seems unnecessary for this resolution.
Could the flight tracks be shaded by time or height?

Corrected. We have not shaded the fight track to avoid confusion among flights.

Revised Fig. 1.

Figure 5: It is hard to see the details and labels without making the image a full page. These should be bigger. Additionally, the markers that show the days back in the right-hand column are not that useful, especially when the back trajectories overlap in time (i.e. B2 and C2). Instead, could the back trajectories be colored by days back? Or maybe the markers could be numbers instead of open circles? The color scheme of 1 and 2 is the same even though they are showing different things (FPES vs. precipitation). Making these different could improve the clarity of the figure. Panels D2 and E2 don't seem to provide that much information with the lack of clear direction of the back trajectories. What defines the consistency listed in the text?

This figure is revised. For the cases of background and mixed region, the days back are not shown as there is no clear trend however the FLEXPART and HYSPLIT still show high consistency as this figure shows. The same colour schemes have been used throughout the texts but have been clearly labelled with the meaning.
Revised Fig. 5 has been labelled by the number of days back.

Figure 6: Are these both from March 2010? The text states that the OBB is from March 2013.
Figure 7: These two panels should be better explained in the caption. They should either both be time series or both be bar plots. If time resolved data is only available for the BB emissions, then that can be a separate figure. It is difficult to directly compare the two emissions types by region when they are plotted in such different styles. Or, could the biomass burning emissions be just added to the bar plot with a new right axis for scaling?
Figure 8: Change the legend colors to black in (c) for the profiles, since green is already defined as CL.
Technical Corrections. Abstract: Remove the "s" from the middle of the units. Page 14846, Line 21: Add "in the Arctic" Figure 8: There is a "c" in the X-axis of panel B
Page 14862, Line 24: Change "European" to "Europe".

All of above are corrected.

Fig.6, Fig.7 and Fig.8 are revised. The unit sm^{-3} means STP corrected which has been clarified in the revised abstract.

Referee 3:

GENERAL REMARKS
The manuscript presents observation data from a flight campaign in the year 2013, focusing on black carbon (BC) transport into the Arctic, which is a topic of high relevance. Presented data cover BC mass concentrations, properties of BC-containing particles such as coating thickness, and ratios of BC to excess CO which are relevant for the determination of BC scavenging during atmospheric long-range transport. Observation data are combined with FLEXPART studies on source regions and HYSPLIT back trajectory analyses for the determination of meteorological conditions during transport. Both pieces of information are used to determine a scavenging fraction of BC for the long-rang transport into the Arctic air. The topic is well suited for publication in ACP and the study is well conducted whereas the data analysis requires major revisions, in particular the section on the scavenged BC fraction; see comments below. Furthermore, the presentation of the material may benefit from restructuring and focusing. In summary the manuscript is acceptable for publication in ACP after major revisions have been considered.

Many parts of the revised manuscript have been shortened and the manuscript rewritten to be more focused.

SPECIFIC COMMENTS
Concerns raised by referees #1 and #2, mainly related to the representativeness of 2013 observations for long-term considerations, to missing of flaring as another BC source in the Arctic, and to the use of inventories for anthropogenic emissions and OBB emissions from different years, are not discussed here; please refer to reviews #1 and #2.
As replied above for referee 1 and referee 2.
All of the comments have been addressed.

1| Source regions: The classification of source regions uses geographical terms for all cases, except for the "Clean Air" source –region. This nomenclature is strongly confusing since the manuscript's focus is on the point that the Arctic air is not clean. Following Fig. 3 it is recommended to use the term "Arctic air" or Arctic Background" or something similar. Then the terminology of source regions is consistent.
The "clean air" is replaced by "background air" throughout the revised version.

2| Scavenging of BC particles: One core part of the data analysis is focusing on the determination of the fraction of BC which is scavenged during atmospheric transport.
The analysis of the scavenged fraction builds on the determination of the ratio of BC to excess CO and its differences between source characteristics and observations in the far field of atmospheric transport. Here the authors use the ratio of BC mass concentration (reported in ng m-3) divided by the volume mixing ratio of excess CO (reported in ppbv). This data product is also used to build vertical profiles; see Fig.10. Referenced literature data refer to observations in the boundary layer over Europe, south-east Asia and an urban environment like Mexico City. Similar observations from elevated plumes of anthropogenic pollution (Park et al.,2005) and boreal fires (Petzold et al., 2007) are not considered, but should be discussed. Constructing vertical profiles of BC/deltaCO, however, requires the analysis of mass ratios of BC to CO (reported in gBC / g deltaCO) instead of combining volume concentrations and mixing ratios. Values of mass ratios are independent of the reporting altitude, while mass concentrations change with altitude, as long as they are not reported for STP conditions; see Park et al. (2005) and Petzold et al. (2007) for details. Because of this

inconsistency, the analysis of BC/deltaCO needs to be repeated, using mass mixing ratios. Referring to Andreae and Merlet (2001), this parameter can be compared to emission characteristics for specific types of biomass burning. It is therefore strongly recommended to repeat the analysis of BC/deltaCO and the related data interpretation.

We think the referee may misunderstand our data interpretation at this point. As the previous ACPD version clearly stated on page 14850, line 9, the BC volume mixing concentration has been corrected for standard temperature and pressure (STP) at 273.15K and 1013.25mbar, and is presented as ng sm^{-3} which is altitude independent. We have used the unit "sm^{-3}" throughout the texts which stands for "standard volume" to emphasize this point.
The issue raised by the referee regarding the vertical profiles is not a problem.
However, in the revised version, the BC/delta CO will be also given in the units of mg BC/gCO to be comparable with the other studies which use this unit. As the referee suggested, Park et al., 2005 and Petzold et al., 2007 have been added and discussed.

The unit in mgBC/gCO is given in revised Table 3 and revised Fig.10.
The added texts are on Page 10, line 392; Page 11 line 405-406.

Another point of discussion arises from the applied methodology in the determination of BC scavenging. Data shown in Fig. 11 show the scavenging factor as a function of the total precipitation, determined from the integrated precipitation along the back trajectories. The analysis does not show any dependence between the two parameters. Looking at Fig. 9, a large amount of precipitation is associated to trajectory altitudes above 7 km (pressure below 450 hPa) at mid-latitudes higher than 40 deg. north. It would be of interest to see the air temperatures at the considered altitudes. Which information on precipitation was used here?

Additional information such as relative humidity along the back trajectory path is added and discussed. We considered the different phases of precipitation but there is insufficient information to be able to distinguish whether snow or ice has a different scavenging efficiency for BC than rain.
RH information is added in revised Fig. 9. The discussion is in Page10, line 379.

In general, the link of BC scavenging to total precipitation neglects potential scavenging pathways by aerosol-cloud interactions (both liquid and ice clouds). A detailed discussion of this topic is recommended.
We have made the discussions in the ACPD version Page 14861, line 23-27.
"Improved agreement between model and measurement is achieved for many of the models by reducing the scavenging efficiency in ice cloud during the winter and spring time in the Arctic. The hygroscopicity of BC can be increased by acquiring more hygroscopic materials (Liu et al., 2013), such as sulphate, a process that is sometimes parameterised in models by changing its solubility after sometime in the atmosphere. This process may be only efficiently applied to warm clouds. For ice clouds, the water soluble coatings on BC may inhibit its ice nucleation activity (Koehler et al., 2009). The aerosols however may also experience removal by processes other than ice nucleation, for example through impaction onto ice surfaces (Baumgardner et al., 2008), scavenging by convective clouds (Koch 2011), or wash out by below cloud precipitation."

However, because the HYSPLIT precipitation data product is not able to discriminate the different forms of precipitation and the role of aerosol-cloud interactions, we believe further detailed investigation of this is beyond the context of this work and would be speculative.

3| Applying mean and standard deviation to 12 days back-trajectories should at least be checked against median and 25- and 75-percentiles (which is more robust) to justify the application of the averaging procedure. Furthermore, it should be discussed whether 12 days back-trajectories can be interpreted in case air mases have crossed convective systems (which are often associated to precipitation).
We have checked the 25th, 75th percentiles and median values along the back trajectories, and the median values are close to the mean, thus we still use the mean and standard deviation for the statistics. The back trajectories are not accurate enough to capture small-scale convection, but these may not be an issue for the free troposphere, where the majority of the transport took place. The models can capture frontal uplift, which is where the large-scale convection will take place.

The revised Fig. 9 has inserted the median value to show the consistency between the median and mean.

4| Restructuring of chapters: Large parts of sections 4.1 and 4.2 might be shifted into a new section on methodologies of air mass origin determination and plume source determination. Actually, the description of these methods in the results section causes some duplication which might be avoided when inserting a new section on methods in chapter 3. Furthermore, chapters 5 and 6 may be merged into a single chapter on Discussion and Conclusions. Actually there is some redundancy in both chapters.

We thank the referee for pointing our areas where there was some duplication and we have revised the text to shorten it and remove any redundancy. However, we disagree with the reviewer that it would improve the paper to move sections 4.1 and 4.2 to a methods section. These sections describe how we have carried out the analysis of back trajectories and the footprint analysis and these are integral results. The descriptions of the tools were in the methods section and we feel this provides the best clarity for the reader. Likewise, we prefer to present a separate detailed discussion of the paper and its context and a conclusion which summarises the main findings of the paper for easy review by the reader.

The revised texts are on Page 11, line 425-439; page 12, line 456-473; page 13, line 490-493.

MINOR ISSUES
1| The last paragraph of the abstract contains some information already given above, it can be removed.
This is a summary and delivers the key message and no obvious duplication is found.

2| Page 14846, line 11: replace the term "soot" by "black carbon".
3| Page 14846, line 23: the reference to Polissar et al. (1999) may be replaced by another more general reference to the radiative impact of BC, e.g. by the reference Bond et al. (2013) which is already cited in the manuscript.
4| In the manuscript OBB is sometimes referred to as open biomass burning, sometimes as open-fire biomass burning. Consistent terminology is recommended.
All of these are corrected in Page 3.
5| Page 14847, line 4-7: The sentence is confusing and should be rephrased.
6| Page 14850, line 3: typo, it should read Aerodyne C-TOF.
7| Section 3.1: Figure 2 contains similar information as Fig. 5 and might be removed.

Fig. 2 is used as an introduction of PES and FPES therefore we would like to keep it. The others are corrected.

8| Page 14856, line9 ff: Differences of the BC coating thickness are insignificant in a statistical sense, this should be stated clearly.

9| Fig. 9b, lower panel: pressure line should be coloured in red.

All of these are corrected.

Page9, line 321.

The importance of Asia as a source of black carbon to the European Arctic during springtime 2013

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Abstract

Black carbon aerosol (BC) deposited to the Arctic sea ice or present in the free troposphere can significantly affect the Earth's radiation budget at high latitudes yet the BC burden in these regions and the regional source contributions are poorly constrained. Aircraft measurements of aerosol composition in the European Arctic were conducted during the Aerosol–Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign in March 2013. Pollutant plumes were encountered throughout the lower to upper Arctic troposphere featuring enhancements in CO and aerosol mass loadings, which were chemically speciated into BC and non-refractory sulphate and organic matter. FLEXPART-WRF simulations have been performed to evaluate the likely contribution to the pollutants from regional ground sources. By combining up-to-date anthropogenic and open fire biomass burning (OBB) inventories, we have been able to compare the contributions made to the observed pollution layers from the sources of eastern/northern Asia (AS), Europe (EU) and North America (NA). Over 90% of the contribution to the BC was shown to arise from non-OBB anthropogenic sources.

AS sources were found to be the major contributor to the BC burden, increasing background BC loadings by a factor of 3-5 to 100.8±48.4 ng sm^{-3} (in standard air m^3 at 273.15K and 1013.25mbar) and 55.8±22.4 ng sm^{-3} in the middle and upper troposphere respectively. AS plumes close to the tropopause (about 7.5-8km) were also observed, with BC concentrations ranging from 55 to 73 ng sm^{-3}, which will potentially have a significant radiative impact. EU sources influenced the middle troposphere with a BC mean concentration of 70.8±39.1 ng sm^{-3} but made a minor contribution to the upper troposphere due to the relatively high latitude of the source region. The contribution of NA was shown to be much lower at all altitudes with BC mean concentration of 20 ng sm^{-3}. The BC transported to the Arctic is mixed with a non-BC volume fraction representing between 90-95% of the mass, and has a relatively uniform core size distribution with mass median diameter 190-210nm and geometric standard deviation σ_g=1.55-1.65 and this varied little across all source regions. It is estimated that 60-95% of BC is scavenged between emission and receptor based on BC/ΔCO comparisons between source inventories and measurement.

We show that during the springtime of 2013, the anthropogenic pollution particularly from sources in Asia, contributed significantly to BC across the European Arctic free troposphere. In contrast to previous studies, the contribution from open wildfires was minimal. Given that Asian pollution is likely to continue to rise over the coming years, it is likely that the radiative forcing in the Arctic will also continue to increase.

1 Introduction

The Arctic is one of the most sensitive regions in the world to climate change (IPCC 2014). It is warming more rapidly than anywhere else on Earth and is subject to various forcing and feedback processes such as black carbon deposition and albedo reduction (Flanner et al., 2007), increase of low-level atmospheric heating over high albedo surfaces (Pueschel and Kinne, 1995) and the increase of cloud longwave emissivity (Garret and Zhao, 2006). The Arctic Haze phenomenon in springtime has been regularly reported and is associated with high levels of gaseous and aerosol air pollutants (e.g. Greenaway 1950). The low surface temperature and the diabatic cooling in the Arctic region during winter and early spring can lead to a thermally stable stratification - the so called Arctic Dome (Klonecki et al., 2003), where the inversion layer tends to trap the pollutants and form a barrier at the top of the atmospheric surface layer.

Although black carbon (BC) is a relatively small mass fraction of aerosol in the Arctic compared to organic matter and sulphate (Quinn et al., 2002), it affects the radiative balance by efficient absorption of shortwave radiation in the free troposphere (Bond et al., 2013) as well as its deposition on the snow, which decreases the surface albedo and promotes melting (Hansen and Nazarenko, 2004). Measurements of BC in the past decade have been intensively conducted at various Arctic surface stations including many long-term studies (e.g. Polissar et al., 1999; Sharma et al., 2004, 2006; Stohl et al., 2007; Hirdman et al., 2010; Shaw et al., 2010; Yttri et al., 2014). In these studies, BC from polluted regions was consistently observed to be transported from outside of the Arctic and built up in the Arctic boundary layer during winter-spring time. Anthropogenic fossil fuel (FF) sources have long been recognized as sources of BC transported to the Arctic (e.g. Quinn et al., 2002; Koch and Hansen, 2005; Law and Stohl, 2007), but the important contributions of open biomass burning (OBB) sources such as agriculture and boreal forest fires have also been reported (e.g. Reid et al., 2005; Quinn et al 2007 and references therein). The overall relative importance of FF and OBB influences is currently uncertain.

Surface measurements of aerosols are however not necessarily representative of the overall troposphere, and this is particularly the case for the Arctic because of the strong stratification (Klonecki et al., 2003). The BC transported to the Arctic free troposphere may have been transported through a different route compared to the BC in the boundary layer and the high-altitude troposphere is more likely to be influenced by the sources further south (Koch and Hansen, 2005; Stohl 2006; Hirdman et al., 2010). The emissions in Eastern/Northern Asia have grown rapidly in the past two decades and many studies have pointed out that this region may have a significant impact on the Arctic BC concentration in late winter and early spring time, especially in the free troposphere (Koch and Hansen, 2005; Shindell et al., 2008; Wang et al., 2011; Shaw et al., 2010; Frossard et al. 2011). The meridional transport to the Arctic during springtime could also be important (Shaw et al., 2010; Marelle et al., 2014; Raatikainen et al., 2015). These long-range transport may be facilitated by warm conveyor belts (WCBs) (Eckhardt et al., 2004) when air parcels can be vertically uplifted in the free troposphere before being transported to the Arctic (Koch and Hansen 2005; Stohl 2006).

The impact of open biomass burning sources in BC concentrations in the Arctic has been increasingly emphasized during the recent ARCTAS (Jacob et al., 2010) and ARCPAC (Brock et al., 2011) campaigns in springtime 2008. The intensive agricultural burning in Eastern Europe (Stohl et al., 2007) as well as the boreal forest fires in Siberia (e.g. Warneke et al., 2010) largely contributed to the Arctic BC burden in the 2008 Arctic springtime. These sources are at high latitude locations and thus their emissions tend to follow low-level quasi-isentropic transport to the Arctic and influence the Arctic lower/middle troposphere (Stohl

2006). Compared to the lower-latitude sources, these sources had likely undergone less wet scavenging. The OBB plumes observed in these studies were throughout the lower to higher Alaskan Arctic troposphere, featuring with higher loadings of particulate organic matter than sulphate (e.g. Brock et al., 2011; Warneke et al., 2010; Matsui et al., 2011). These studies all consistently reported the BC from the remote Asian fossil fuel sources were significantly scavenged (e.g. Stohl 2006; Matsui et al., 2011), and hereinafter concluded a more significant influence of OBB sources than fossil fuel for that time of the year.

The concentration of BC in the Arctic is poorly represented by models (Quinn et al., 2007; Koch et al., 2009 and references therein). It has been widely reported that the measured vertical BC profiles in the Arctic show large diversity among models but almost all models underestimate BC throughout the lower and middle troposphere (e.g. Lee et al., 2013 and references therein), whereas some of the models overestimate BC in the upper troposphere and lower stratosphere (Koch et al., 2009). Many models show the scavenging parametrization could fundamentally lead to model bias. A seasonal variation in the wet scavenging mechanism (e.g. Liu et al., 2011; Bourgeois and Bey 2011; Browse et al., 2012; Hodnebrog et al., 2014; Myhre and Samset 2015) has shown considerably improved comparison with measurements by extending the lifetime of BC in the spring/wintertime, largely because during the cold season ice clouds result in a less efficient nucleation scavenging efficiency compared to warm clouds. An extended BC lifetime in cold seasons and a seasonally dependent BC removal system are thus suggested for these models. However to extend the BC lifetime may at the same time exacerbate model overestimates of BC mass at higher altitudes (Lund and Berntsen, 2012). The scavenging mechanism of BC is source and meteorologically dependent at different levels of Arctic troposphere and improved understanding of BC scavenging efficiency is desired to evaluate model outputs.

Aircraft measurements of aerosol and gaseous pollutants including BC, sulphate, organic matter and CO were conducted during the Aerosol–Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) campaign in springtime 2013. Plumes from Eastern/Northern Asia, Europe and North America were encountered at different levels in the European Arctic troposphere. Lagrangian dispersion models combined with up-to-date emission inventories are used to evaluate the source origins for the observed plumes. The statistics of plumes and vertical profiles under classified source regions are analysed to determine the influences of different source regions as a function of altitude throughout the Arctic troposphere. Finally, the scavenging efficiency of BC is estimated by comparing the inventory and measured BC/excess CO ratio.

2 Measurements

The flights took place during March-April 2013 as part of the Aerosol–Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) project. The measurements described here were made using the UK Facility for Airborne Atmospheric Measurements (FAAM), a BAe-146 aircraft, in the region between continental Norway and Svalbard. A number of straight level runs (SLRs) and vertical profiles were performed for each flight. Fig. 1 shows the flight tracks during the campaign.

The physical properties of individual refractory BC particles (rBC, as defined by Petzold et al., 2013) were characterised using a single particle soot photometer (SP2) manufactured by DMT Inc (Boulder, CO, USA). The instrument operation and data interpretation procedures of the specific Manchester SP2 instrument have been described elsewhere (Liu et al., 2010; McMeeking et al., 2010). The SP2 incandescence signal was calibrated for BC mass using Aquadag® black carbon particle standards (Aqueous Deflocculated Acheson Graphite, manufactured by Acheson Inc., USA) and corrected for ambient BC with a factor of 0.75 (Baumgardner et al., 2012). An rBC spherical equivalent core diameter (D_c) can be derived from the rBC mass and a coating thickness (the ratio of the particle diameter, D_p, to the core diameter, D_p/D_c) may be estimated for each particle containing BC assuming a complete and concentric covering of the core. The methodology to determine the D_p/D_c is detailed in Taylor et al., (2015) and Liu et al., (2014): the measured scattering cross section of coated BC was derived using a prescribed Mie look up table at the SP2 operational wavelength λ=1064nm using a core refractive index 2.26-1.26i and coating refractive index 1.50+0i, and the uncertainty of the derived D_p/D_c due to particle geometry is <6% (Liu et al., 2015).

The chemical composition of non-refractory PM1 was measured by an Aerodyne C-ToF aerosol mass spectrometer (AMS). A detailed description of the instrument can be found elsewhere (Drewnick et al., 2005). A time and composition dependent collection efficiency (CE) was applied to the data based on the algorithm by Middlebrook et al. (2012). It is noted that most atmospheric BC particles contain non-absorbing material as a result of co-emission of either the particulate matter or its precursors. This additional material will be observed by the AMS. BC particles with little associated non-absorbing material may bounce and result in a reduced CE but the fraction of the total non-refractory mass mixed with BC is small and is therefore unlikely to affect the reported AMS mass loadings. An AMS-SMPS volume comparison could not be performed as we have found that the SMPS concentrations at altitude are incorrect, possibly due to an inaccuracy in the assumed charge distribution during inversion (Sakamoto et al., 2015; López-Yglesiasand and Flagan, 2013). The AMS was calibrated using mono-disperse ammonium nitrate particles. All of the SP2 and AMS measured concentrations are reported as mass concentrations at standard temperature and pressure (STP, 273.15K and 1013.25mbar), denoted by sm^{-3}. Data are missing from some flights due to a malfunction of the logging computer.

Carbon monoxide was measured by an Aero-Laser AL5002 VUV resonance fluorescence gas analyser, and TECO 49 UV photometric ozone instrument measured O_3. In-flight CO calibrations were applied to the raw CO data. The background concentrations of CO were defined as the lowest 5^{th} percentiles (Koike et al., 2003) for all of data collected during the campaign. Because some of the flights in this study had reached high altitudes close to the tropopause, the CO background concentrations were derived as a function of altitude, as shown in supplemental information (Fig. S1). The significantly lower CO background corresponding with an enhancement of O_3 at about 6km may suggest a stratospheric intrusion (Thomas et al., 2013). The altitude-dependent CO background (Fig. S1D) is therefore used to calculate the excess CO relative to the background (ΔCO). This method produces consistent

ΔCO among flights when the sampled air masses were not significantly influenced by recognized source regions (Fig. S4B1). A background concentration of zero was assumed for BC (Matsui et al., 2011).

3 Model simulations

In this study, a Lagrangian particle dispersion model (FLEXPART) is applied to characterise the origin of the sampled air masses up to 12 days backward. In addition, the HYSPLIT backward trajectory model was used to interrogate the meteorological history through the pollutant transportation pathways. The back trajectories will be subject to increased model integration error when backward modelling time exceeds 5-6 days, leading to uncertainties when assigning the meteorological information to the back trajectory path, meaning that sources can only be assigned on continental scales.

3.1 FLEXPART-WRF

The Lagrangian particle dispersion model FLEXPART-WRF (Brioude et al, 2014), adapted from the FLEXPART model (version 6.2, Stohl et al., 2005) was used in a backward mode to characterise the origin of the sampled air masses. FLEXPART-WRF was driven by WRF meteorological forecasts and a new simulation was initialized each time the aircraft position changed by more than 0.20° in latitude/longitude or 250m vertically. For each simulation, 20000 particles were released in a volume 50x50 km (horizontally) and 500 m (vertically), particles were tracked 12 days backward in time. The primary output of FLEXPART backward calculations is the potential emission sensitivity (PES) which expresses the residence time of particles at a given location and is used to characterise the transport pathways of the sampled air masses. A further extraction of the lowest model layer, i.e. integrated on the 0–500 m layer, termed as the footprint PES (FPES), is used to evaluate the air mass potential sensitivity to ground sources. Fig. 2 gives a typical example of the FLEXPART outputs of PES and FPES.

The relative contributions to the sampled air masses from different ground sources can be attributed by multiplying the FPES values with the source-specified inventory emissions (in kg m^{-2} s^{-1}). This study will focus on the BC and CO source evaluations from both anthropogenic and open biomass burning (OBB) sources.

We have classified the major recognized ground source regions in the following way: Asia (R_{AS}), Europe (R_{EU}), North America (R_{NA}), Siberia (R_{SI}) and Background Air (R_{BG}), as shown in Fig. 3. The extent of each region is detailed as following:

i) Asia (R_{AS}, 20-50°N, 30-145°E), including northern/eastern China, Japan and Mongolia.

ii) Europe (R_{EU}, 35-70°N, 10°W-30°E), corresponding to the whole of Europe.

iii) North America (R_{NA}, 20-50°N, 60-160°W), including USA, Canada and Mexico.

iv) Siberia (R_{SI}, 50-70°N, 30-180°E).

v) The Artic Background air (R_{BG}, 70-90°N), which is without significant continental source contact.

3.2 HYSPLIT backward trajectories

The HYSPLIT 4.0 model (Draxler and Hess, 1998) back trajectories were initiated from the latitude, longitude and altitude of the aircraft every 30 seconds along the flight path and calculated 14 days backward in time. Horizontal and vertical wind fields for trajectory calculations were provided by the 1°x1°, 3-hourly GDAS1 reanalysis meteorology (Global Data Assimilation System; NOAA Air Resources Laboratory, Boulder, CO, USA). This also allows retrieval of potential temperature (θ) and precipitation rate along each trajectory path. The reported meteorological information extracted along the trajectory path is averaged over all of the trajectories along the flight track for the targeting time period.

4 Results

4.1 Air mass origins of pollution plumes

Plumes were encountered at various levels throughout the Arctic troposphere from 300-8000m and all plumes were characterized by enhancements over the average, altitude resolved background values of BC, CO, sulphate and organic matter. The sulphate content was found to be significantly higher than the organic matter by a factor of 1.5-4. Fig. 4 shows the measurement results and the FPES derived air mass origins for the flight B763 (identical information for the other flights can be found in Fig. S2). Note that the plumes observed during straight and level runs (SLRs) for each flight are marked with Roman numerals (as shown on the flight altitude track in Fig. 4) and were analysed separately from the vertical profiles sampled during aircraft ascent and descent.

For each FLEXPART-WRF simulation, the FPES was gridded into the defined source regions (Fig. 3) along each flight track (Fig. 4A). For each FPES calculation, the air masses may have passed over multiple source regions (R_{BG}, R_{AS}, R_{EU}, R_{NA} and R_{SI}) as defined in Fig. 3. The air mass origin is classified as BG when the R_{BG} contributes more than 80% of the grid-integrated total FPES. For the rest of the cases when R_{BG} contributes less than 80%, the air mass origin is then classified according to the relative contributions of R_{AS}, R_{EU} and R_{NA}. The air mass will be classified as the specified source region if the relative contribution of any corresponding source region is greater than 60% of the total polluted source region contribution, for example if $R_{AS}/(R_{AS}+R_{EU}+R_{NA})$ is >60%, the air mass is classified as AS. In some cases R_{EU} and R_{NA} both contributed more than 40%, where the air mass was classified as EU+NA. The R_{SI} is excluded from the air mass classification because this region was found to make a minor contribution to the ground sources during the time of the year according to the inventories (section 4.2). This is supported by the observation that for most of the flights the contribution of R_{SI} to total FPES is not important (top panels in Fig. S2).

Fig. 5 shows typical examples of FLEXPART-WRF FPES and the corresponding HYSPLIT backward trajectories for the plumes with each defined air mass origin. Because of the good consistency between FLEXPART and HYSPLIT model outputs, the plume age since emission can be estimated by locating the trajectories on the FPES map: the source emission starts from the backward time when the trajectories for that particular plume passed over the region that is most sensitive to the recognized ground sources, i.e. with the largest FPES values. The number of plumes and vertical profiles for each flight is summarized in Table 1. ~~As figure 5 shows, the AS airmasses had significant continental contact with eastern/northern China and Japan. NA air masses tend to be transported northward towards Greenland. EU air masses were generally at higher latitude and the direction of the air path was very variable. BG air masses circulated within the polar oceanic region without significant continental source contact.~~

4.2 Attribution of Plume Sources

Fig. 6 shows the BC emission inventories as used in this study. The anthropogenic sources are classified to occur from the following sectors (Fig. S3): residential (RE), transport (TR), industry (IN), energy (EN) and Flaring (FL). The RE, TR, IN and EN are taken from the HTAPv2 0.1°×0.1° inventory for March 2010, which is the most recent available anthropogenic inventory, and the FL is from the ECLIPSE (Evaluating the CLimate and Air Quality ImPacts of ShortlivEd Pollutants) global emission inventory (Klimont et al., 2015). Comparison between the emissions inventory in 2010 and 2005 shows the BC emissions increase by less than 30%. It is to be expected that any differences between 2010 and 2013 will be less than this amount.

The open biomass burning (OBB) inventory is from FINN v1 (http://bai.acd.ucar.edu/Data/fire/) (Wiedinmyer et al., 2006) for March 2013. Because the OBB emissions are based on observed fires, these have significant time variation and have been computed at 1 hour time resolution. CO emissions are also taken from the equivalent inventories as BC. ~~Multiplying the source inventories by FPES allows an estimation of the relative contributions of observed BC and CO from the sources of RE, TR, IN, EN and OBB.~~

The total BC emissions from each region are shown in Fig. 7. AS is the largest source region for both anthropogenic and OBB emissions, whereas SI emissions are relatively low during that time of the year. The residential sector, largely domestic solid fuel burning, is the most important contributor to the total anthropogenic emissions. ~~The emission distributions in the SI region contrast with the Arctic study in the same month of 2008 during the ARCPAC and ARCTAS campaigns when forest fires in southern Siberia and agricultural burning in southern Russia were prevalent.~~ OBB sources show significant time variations, with higher OBB emissions from AS dominating in the first half of the month, and those from NA increasing during three short periods with high loadings of enhanced emissions, though these are approximately an order of magnitude smaller than the OBB emissions from NA.

The anthropogenic and OBB source contributions to the BC or CO within an air parcel were determined by multiplying the FPES by the BC or CO emission inventories at the same grid resolution (Quennehen et al., 2011, 2012). The grid-integrated FPES×inventory is interpreted as the total ground source contributions to the observed plume in the 12 days prior to measurement. The modelled source attributions (from RE, TR, IN, EN and OBB) of BC and CO for each plume are summarized in Table S1. The resulting contributing fractions of OBB sources to the total BC and CO are shown in the 4^{th} and 5^{th} column of Table 2. The plumes with sources originating from Eastern AS and NA in the first half of the month had more OBB contributions (Fig. 6E and Fig. 7B) whereas the OBB contribution was weaker when the source origins were located towards higher latitudes. The overall OBB contribution to BC is 0.2-10% and 0.2-16% for CO. Anthropogenic contributions therefore dominate during the experimental period, which is consistent with the observation that the particulate sulphate mass was significantly higher than that of organic matter for all plumes. In particular, flaring sources make a negligible contribution to the BC and CO loadings (of the order of 10^{-2} or 10^{-3} magnitude lower than the other anthropogenic contributions as shown in Table S1). This is because the plumes in this study were mainly encountered in the middle or upper troposphere and the resulting back trajectories show that there was little land contact over the high-latitude regions where flaring sources are present.

Fig. S4 summarizes the ~~concentration of BC, CO, sulphate, organic matter and BC coating thickness for all~~ plume characteristics and vertical profiles classified by air mass origin~~s~~, with the mean value presented in Fig. 8 and Table 2. ~~The mean values for rBC, ΔCO, sulphate and organic matter as a function of altitude are shown in Fig. 8 and classified by the dominating~~

~~source region. The information obtained during each plume sampled during a straight and level run is provided in detail in Table 2, and the statistics of vertical profiles are provided in Table 3.~~ The data from the plumes and vertical profiles compare well for the concentrations of all aerosol and gaseous species ~~as shown in Fig. S4 and Fig. 8~~, indicating a high level of consistency between different flights and aircraft locations.

Three altitude ranges have been used to represent the broad vertical distribution of the pollution layers. These are defined as: lower, middle and upper troposphere (LT, MT, UT) and cover the altitudes 0-2500m (750-1000mbar), 2500-5500m (500-75mbar) and 5500-8000m (350-500mbar (Fig. 8). The height of the Arctic boundary layer (ABL) is determined to be about 200-400m, based on the lowest potential temperature inversion derived from the aircraft profiles. The Arctic tropopause in this study was observed to be around 7500-8000m based on analysis of ozone profiles. Compared to the BG background air (Table 3), AS sources show the largest perturbation to the vertical profiles of BC and ΔCO at all levels. The AS contribution to the BC profile was largest in the MT with a value of 100.8±48.4 ng sm^{-3}; AS derived BC plumes close to the tropopause (at ~7.5-8km) were also observed with concentrations of 55 to 73 ng sm^{-3}. EU air masses significantly influenced the MT, whereas airmasses from the EU region made only a minor contribution to the UT. NA influences on BC were low at all levels. The sulphate concentration was 2-4 times higher than organic matter, which is consistent with the dominance of anthropogenic sources, though there are other possible natural sources contributing the Arctic sulphate burden that may contribute (Fisher et al., 2011).

BC particles show relatively consistent coating thickness (within 10%) for different airmass origins (Table 2) with average D_p/D_c 2.25±0.55, equivalent to 90-95% of the volume of BC containing particles being due to non-refectory material associated with the BC-core. Although during the AS dominated air masses, a slight vertical dependence of the BC coating thickness was observed with reduced coating thicknesses occurring at lower altitudes (Fig. S4). Compared to previous observations of lower D_p/D_c in close proximity of sources (1.28-1.65, Liu et al., 2014; Liu et al., 2011), this suggests that the observed BC has been significantly aged. The BC size distribution is almost uniform for AS, EU, and NA influenced air masses with mass median diameter of 190-210nm, 190-200nm, and 180-190nm with geometric standard deviation σ_g=1.55-1.65. The NA showed a slightly smaller BC MMD.

4.3 The transport mechanisms of air parcels

The transport pathways of pollutants to different Arctic altitudes may vary. To investigate this, HYSPLIT back trajectories from plumes intercepted within the Arctic LT and the UT are investigated separately, as shown in Fig. 9. For each plume, physical properties including altitude, ambient pressure, latitude, precipitation, potential temperature (θ), are averaged over all back trajectories during the plume duration. The light grey shading in Fig. 9 marks the region along the trajectory when trajectories passed over the region with the most sensitivity to the ground sources as derived from the FPES. The total precipitation experienced by an average plume (as given in Table 2) is the accumulated average precipitation along the trajectory pathway from the source emission to when the plume was observed, and the uncertainty is given by the propagation of the uncertainties of average precipitation along the trajectory pathway.

As Fig. 9 and Fig. S5 show, the sources of the UT plumes were located between 30-40°N whereas the sources of LT plumes were at 50-60°N. The lower-latitude source regions have

potential temperatures that are 15-20K higher than those of the plumes from higher-latitude sources. The pollution plumes observed in the Arctic UT had experienced rapid vertical ascent, for example, plumes from AS and EU experienced vertical ascent of 4-5km within 1-2 days; similar ascents rates were also observed for NA influenced air masses but the ascent was typically over a lower altitude (~2km vertically). Ascent of sampled polluted air masses which arrived in the Arctic LT also occurred, however to a much lesser extent and on a longer timescale. Due to the close proximity of the Arctic for the higher-latitude source, the air parcel has been transported on shorter time scale compared to lower-latitude sources.

Back trajectories of air masses from lower-latitude sources arriving as plumes in the UT show an increase of θ at the source during ascent of the air shortly after passing over the source region. This increase of θ is about 5K for all air parcels arriving in the Arctic UT from all source regions, however there are increased uncertainties in determining θ from the model when the trajectory extends further backward in time. The increase of θ along with rapid, eastward and poleward ascent of air masses in the mid-latitudes indicates ascent to be associated with warm conveyor belts (WCBs) (Eckhardt et al., 2004), although the vertical increases in θ in our study are lower than previous work (30-40K, Matsui et al., 2011) investigating mid-latitude air masses transported to the Arctic. In contrast to the transport of UT plume sources, the observed plumes arriving in the Arctic LT arising from high-latitude pollution sources have a lower latitudinal θ gradient and there was no obvious vertical increase of θ observed for these sources. For plumes arriving in both LT and UT, the air mass ascent was followed by a smooth decrease in potential temperature. For EU influenced air masses, which represent the highest latitude anthropogenic sources, the change of θ was minor, suggesting the transport tends to be quasi-isentropic. The rapid ascent of lower-latitude sources was shown to be associated with heavier precipitation and increased relative humidity, compared to air parcels from higher-latitude source regions with lower precipitations.

4.4 The scavenging of BC particles

During transport, particularly during uplift, it can be expected that a significant fraction of particles are removed through scavenging and subsequent precipitation. This is an important process to quantify, as models show high sensitivity to this (e.g. Mann et al., 2014). The extent of this is estimated by comparing the particle concentrations with CO. CO is not removed by precipitation and is not significantly removed by gas phase oxidation on timescales equivalent to transport from the source regions to the Arctic (within 12 days) (Forster et al., 2011). The ratio of BC/ΔCO can therefore be used to estimate the extent to which BC is removed from the air mass between the source region and the receptor (Park et al., 2005). For a specified plume, the scavenged fraction of BC (SF_{BC}) can be estimated according to Equation (1):

$$SF_{BC} = 1 - \frac{BC/\Delta CO_{measured}}{BC/\Delta CO_{source}} \qquad (1)$$

The $BC/\Delta CO_{measured}$ and $BC/\Delta CO_{source}$ represent the ratio as measured at the receptor and determined at the emission source respectively. To obtain the SF_{BC} requires an explicit determination of the $BC/\Delta CO_{source}$. However, values obtained from measurements in the literature are subject to different ageing time of samples, various source characteristics, variations in emissions throughout the year and influences on particular experimental locations, which leads to significant variations in reported BC/ΔCO. For example a BC/ΔCO

(in ng sm^{-3} $ppbv^{-1}$) of 6.5-8.8 was obtained in the south-east Asian boundary layer (Pan et al., 2011), in the European boundary layer BC/ΔCO could range from 0.8-2.3 (McMeeking et al., 2010), whereas Baumgardner et al. (2007) observed a value of 1.4 for the Mexico city urban environment, Spackman et al. (2008) report 6.8 for the Houston region and 3.8-9.4 for boreal fires in North America (Petzold et al., 2007). It is therefore more appropriate to use values of BC and CO from emission inventories as a 'best estimate' to consistently evaluate the scavenging on BC since emission, recognising that there is uncertainty in these values. The FPES×inventory modelled BC and ΔCO attributed to anthropogenic and OBB sources (Table S1) are used to represent the ratio of $BC/\Delta CO_{source}$. The SF_{BC} calculated by Equation (1) can therefore be considered as the integrated scavenged fraction from emission to receptor.

The $BC/\Delta CO_{measured}$ for vertical profiles and plumes is shown in Fig. 10. The average $BC/\Delta CO_{measured}$ ranged 0.4-3 for the plumes (Table 4) and 0.1-4.8 for vertical profiles, and these values are of a comparable magnitude to those reported by Matsui et al. (2011). As shown in Fig. 10, AS air masses significantly increased the BC/ΔCO throughout the Arctic troposphere at all altitudes; NA sources also increased the ratio but to a considerably lesser extent; EU influenced air masses showed a $BC/\Delta CO_{measured}$ that was enhanced by a similar order of magnitude as that in the AS influenced air masses in the mid troposphere, but a lack of available data for LT prevented a similar comparison being made for EU. The plumes in the UT experienced heavier precipitation for both AS and EU influenced plumes (Table 2).

The SF_{BC} was calculated for each intercepted plume and ranges from 0.6-0.95 (Table S2). The dependence of SF_{BC} for each plume on precipitation integrated along the trajectory is shown in Fig. 11. In contrast to the previous observation by Matsui et al. (2011), where the BC removal was observed to be significantly correlated to the precipitation intensity, in this study no obvious correlation between precipitation and SF_{BC} was observed. This may be due to the uncertainties of modelled precipitation along the back trajectories, or the scavenging of BC via precipitation was subject to a complex mechanism not well represented by the total precipitation. The main contrast between this study and Matsui et al. (2011) is the significantly higher BC/ΔCO for air masses influenced by Asia; BC/ΔCO was most elevated (3-8) in the mid troposphere at altitudes of 4-6km, influenced mainly by mid-latitude Asian sources, indicating that the lowest scavenging efficiencies were observed in these regions.

5 Discussion

In this study, ~~the non-open fire anthropogenic sources contributed over 90% of the European Arctic BC burden and over 85% of the ΔCO.~~ Asian sources were observed to influence all levels of the troposphere (400-8000 m): the source origins include higher-latitude Northern China (~40-60°N) and a mid-latitude (~30-40°N) region including Eastern China and Japan. The influences on the Arctic MT/UT were mainly from the mid-latitude sources, for which the air parcel could be rapidly uplifted by WCBs ~~within 1-2 days~~ to the free troposphere and then followed by a long-range northeastward transport to the Arctic MT/UT. These observations are consistent with previous studies that indicate long-range transported fossil fuel sources from northern hemisphere mid-latitude affect the Arctic MT/UT (Stohl 2006; Quinn et al., 2007 and refs therein). ~~Based on the emission inventories, the main contributor to the BC and CO is the residential sector. Additionally, in this study there were also strong plumes with high BC content observed at significantly high altitude, i.e. around 8km close to the Arctic tropopause, which will potentially contribute a large radiative warming to the atmospheric column (Ramanathan and Carmichael 2008; Koch et al., 2009).~~

Air masses influenced by northern European sources may follow various pathways to reach the Arctic (Stohl 2006). Transport of these air masses to high altitudes within the Arctic atmosphere has previously been shown to arise via WCBs (Marelle et al., 2014), however this was only clearly the case for southern European influenced airmasses. In several cases both uplift through WCBs and quasi-isentropic transport may operate in combination during transport to the Arctic region. However, consistent with previous studies (Stohl 2006; Marelle et al., 2014), we show that the influence of European sources on the Arctic UT is low.

Airmasses influenced by North American sources were generally observed to be lifted to a lower altitude, followed by a longer transport time. The influence of NA sources in the Arctic free troposphere in the vicinity of Greenland were previously reported (Quennehen et al., 2011), however most of the previous studies have reported weak or occasional influence of NA sources on the Arctic (e.g. Stohl 2006; McConnell et al., 2007; Brock et al., 2011), which is to be expected as the main industrial sources in NA lie southward of the mean position of the Arctic front, and since advection from these sources to the Arctic involves transport through the meteorologically active North Atlantic region. This study is consistent with those previous results in that the observed NA influences were close to, or only slightly higher than the background conditions.

The main contrast between this study and those conducted previously is that high-latitude OBB sources from Eastern Europe and Siberia~~, identified using the hourly resolved FINN emissions database,~~ did not make a significant contribution to BC in the Arctic atmosphere; whereas in springtime of 2008, OBB sources were prevalent in these regions (e.g. Brock et al., 2011; Warneke et al., 2009). As a result, our results show that during spring 2013, the OBB contribution to the Arctic BC burden was less 10%, a result that is consistent with the high relative sulphate concentrations observed in all sampled plumes. This contrasts with the springtime of 2008 when the aerosols in observed Arctic plumes had a higher organic content than sulphate (e.g. Brock et al., 2011; Warneke et al., 2009; Jacob et al., 2010). Annual fire counts across Siberia from the FINN emissions inventory for March-April over the 10 year period from 2004 to 2013 are shown in Fig. 12A. The year 2008 is shown to be anomalously high, compared to the decadal average of 2.8 $x10^4$ by approximately a factor of 2.4. In all other years in the last decade fire counts in the Siberia spring varied between $1.4x10^4$ and $3.7x10^4$. This contrasts with 2013 which showed the lowest number of fire counts ($1.4x10^4$). Therefore, whilst 2008, does indeed show the importance of biomass burning in the Arctic, Asian pollution sources make a substantial contribution to Arctic BC during many spring seasons. Figure 12B shows the changes in anthropogenic emissions of BC between 2005 and 2010 as described in the ECLIPSE database. There is a marked increase in BC emissions over this period of between 20-30% and these increases are projected to continue in the coming decades.

~~The higher latitude location of these OBB sources led to a considerably more polluted Arctic atmosphere during 2008 and hence a greater impact on the Arctic MT/LT. The organic matter content of Arctic MT/LT in this study is therefore considerably lower than the studies in 2008.~~

A number of studies point out the scavenging efficiency of BC represents a major uncertainty in model prediction of BC in the Arctic (e.g. Liu et al., 2011; Hodnebrog et al., 2014; Myhre and Samset 2015), and these studies have shown that models could underestimate the BC loadings in the Arctic during the cold season by a factor of up to 10 due to a possible underestimation of the BC lifetime. Improved agreement between model and measurement is achieved for many of the models by reducing the scavenging efficiency in ice cloud during the winter and spring time in the Arctic. The hygroscopicity of BC can be increased by acquiring more hygroscopic materials (Liu et al., 2013), such as sulphate, a process that is

sometimes parameterised in models by changing its solubility after sometime in the atmosphere. This process may be only efficiently applied to warm clouds. For ice clouds, the water soluble coatings on BC may inhibit its ice nucleation activity (Koehler et al., 2009). The aerosols however may also experience removal by processes other than ice nucleation, for example through impaction onto ice surfaces (Baumgardner et al., 2008), scavenging by convective clouds (Koch 2011), or wash out by below cloud precipitation. The integrated SF_{BC} of 0.60-0.95 from this study provides a constraint for future model tests.

6 Conclusions

Intercontinental transport of pollutants to the European Arctic was observed during the ACCACIA campaign in springtime 2013. ~~Plumes originating from Eastern/Northern Asia, Europe and North America were all encountered in the Arctic troposphere.~~ FLEXPART-WRF Lagrangian dispersion modelling was performed to evaluate contributions of ground sources to the sampled air masses. When this analysis was combined with up-to-date emission inventories, anthropogenic sources in populous regions were found to contribute over 90% of the BC for all the plumes sampled. In contrast to previous observations, open wildfire contributions from Siberia and Eastern Europe to the observed enhanced BC were small. Our measurements of significantly higher sulphate content compared to organic matter are consistent with this. Within the plumes observed, Asian sources were found to have the most significant influence at all levels throughout the Arctic troposphere, with the maximum BC mass loading arising in the middle troposphere (MT) at concentrations of around 100 ng sm^{-3}. European sources (EU) also have an important influence in the MT are an order of magnitude lower than those from Asia. Due to the high latitude location of the sources, Europe does not significantly influence the Arctic upper troposphere (UT). North American sources (NA) displayed a weak influence at all altitudes largely due to the longer transport time and active meteorological influences.

The transport from Asian pollution sources is likely to be facilitated by warm conveyor belts, through which air parcels are rapidly uplifted on timescales of 1-2 days, followed by long-range transport (8.5-11.5 days). We show that this pathway efficiently transports BC from mid-latitude Asian sources to the high altitude Artic troposphere, raising BC concentrations to between 55 and 73 ng sm^{-3} close to the Arctic tropopause. The scavenged fraction of BC for plumes was estimated by comparing the measured BC abundance relative to the measured excess CO with the same ratio derived from values at source determined from the EDGAR and FINN emission inventories. This showed that about 60-95% of BC is scavenged between the source region and the Arctic atmosphere where the measurements took place. No direct correlation was found between the scavenged fraction and accumulated precipitation.

Compared to previous studies when plumes from open fire sources were intensively observed, this study shows that open fire contributions to BC vary significantly between years and are sporadic, whereas the contribution of Asian sources to pollution in the Arctic atmosphere is substantial and is delivered via a persistent and consistent pathway. Asian outflow will accumulate in the Artic troposphere during the winter season and early spring, being retained north of the polar front in slowly descending cold polar air. The radiation balance of the upper tropospheric Arctic atmosphere has been shown to be very sensitive to absorbing aerosol in spring time and given that Asian pollution is likely to continue to rise over the coming years it is likely that this process will continue to increase in importance when considering regional climate effects.

Data availability

Processed data is available through the ACCACIA project archive at the British Atmospheric Data Centre (http://badc.nerc.ac.uk/browse/badc/accacia). Raw data is archived at the University of Manchester and is available on request.

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Tables and Figures

	Number of profiles	Number of plumes
B759(20/03/2013)	7AS; 1NA	5AS; 1NA
B760(21/03/2013)	3AS; 2NA	2AS; 2NA
B761(22/03/2013)	3EU; 1EU+NA; 1AS	3EU;1EU+NA;1NA
B762(23/03/2013)	6NA	3NA; 1EU
B763(26/03/2013)	2NA	2AS;1NA

Table 1. The number of vertical profiles and plumes observed for each flight.

Plume Index	Source origin	Latitude/Region	% OBB BC model	% OBB CO model	Altitude in m (mbar)	BC in ng sm-3	SO4 in ng sm-3	Org in ng sm-3	ΔCO in ppbv	rBC core MMD in nm(geo sdev)	BC D_p/D_c	plume age in days	Precip in mm
B759 plumes													
i	AS	30-40°N; Japan, E China	4.51	8.07	7787(360)	66.4±30.6			32.3± 19.2	190(σ_g=1.55)	2.25±0.16	9-10.5	23.4±1.6
ii	AS	30-40°N; Japan, E China	4.18	7.19	4640(570)	75.3±25.4			18.3±7.4	193(σ_g=1.57)	2.11±0.13	8-9.5	17.2±1.3
iii	AS	40-60°N; N China	0.82	1.30	2934(720)	115.6±30.5			53.2±12.8	197(σ_g=1.62)	2.09±0.09	10-11.5	3.4±0.1
iv	AS	30-50°N; N and E China, Japan	2.59	3.58	4681(570)	91.5±35.9			27.5±13.5	193(σ_g=1.57)	2.18±0.12	9-10.0	15.7±1.2
v	NA	30-50°N; N US	4.96	5.51	4660(570)	35.5±19.6			18.2±10.8	194(σ_g=1.61)	2.14±0.18	8.5-10	7.9±1.7
vi	AS	20-40°N;S and E China, Japan	9.16	16.28	8110(340)	40.3±22.3			19.7±11.7	194(σ_g=1.58)	2.13±0.25	9.5-10.5	15.9±1.7
B760 plumes													
i	NA	20-40°N; S US, Mexico	8.99	10.41	7772(360)	29.8±15.9				184(σ_g=1.62)	2.08±0.24	10-12.0	16.4±0.9
ii	AS	40-60°N; N and E China, Japan	0.87	1.23	1745(840)	70.2±38.9			26.9±10.4	208(σ_g=1.60)	2.08±0.14	8-10.0	14.8±0.5
iii	AS	40-75°N; N China, E Russia	1.14	1.86	430(1010)	40.3±12.6			18.1±1.6	222(σ_g=1.60)	1.96±0.13	7.5-9	17.5±1.5
iv	NA	20-40°N; South US, Mexico	4.49	5.05	7758(360)	32.1±17.3				180(σ_g=1.61)	2.13±0.22	10.5-12	37.4±3.8
B761 plumes													
i	EU+NA	30-60°N;E US, N Europe	8.94	10.40	2505(750)	51.1±16.3	694±79	188±96	24.9±2.8	188(σ_g=1.62)	2.34±0.14	4-6.0	24.9±2.5
ii	EU	50-80°N; W and N Europe	5.72	10.16	2455(750)	76.4±26.5	768±279	226±110	36.0±9.6	200(σ_g=1.61)	2.39±0.14	6-7.0	27.3±1.6
iii	EU	50-80°N; N Europe	0.34	0.71	1756(810)	37.5±12.6			9.04±2.4	193(σ_g=1.67)	2.33±0.16	6.5-7	26.8±1.0
iv	NA	35-60°N; E and S US	0.74	1.10	7536(360)	8.7±6.1	123±49	108±65	23.6±7.1	195(σ_g=1.65)	2.38±0.44	10.5-11.5	43.6±3.2
v	EU	50-75°N; N Europe	1.06	1.86	7536(360)	9.4±6.1	182±73	113±64	35.8±8.1	188(σ_g=1.62)	2.23±0.34	5-6.5	41.6±2.9
B762 plumes													
i	NA	40-60°N; E and N US	0.36	0.34	2693(720)	6.0±4.4	213±67	60±54	16.7±2.1	192(σ_g=1.62)	2.33±0.32	10-11.5	9.4±1.5
ii	NA	40-60°N; E and N US	1.29	2.31	1435(850)	35.0±17.9	1015±148	331±57	21.9±9.3	202(σ_g=1.60)	2.28±0.19	11.5-13	8.9±1.9
iii	NA	40-60°N; E and N US	0.63	1.40	1452(850)	27.5±16.4	707±200	260±97	22.7±2.0	197(σ_g=1.59)	2.28±0.26	10.5-12	6.7±1.0
iv	EU	45-75°N; N and S Europe	0.39	0.50	7614(360)	12.8±10.2	51±30	68±64	21.8±15.6	190(σ_g=1.63)	2.32±0.38	7.5-8.5	40.3±2.8
B763 plumes													
i	AS	30-60°N; N and E China, Japan	0.66	1.12	7411(360)	73±37	1215±585	447±222	41.7±25.6	193(σ_g=1.67)	2.38±0.24	9-11.0	50.5±2.9

ii	NA	35-50°N; E US	0.27	0.27	6181(430)	20.7±8.9	293±62	126±55	16.6±5.9	187(σ_g=1.61)	2.45±0.27	11-12.0	11.6±1.2
iii	AS	30-55°N; E and N China, Japan	0.41	0.72	7692(370)	55±24.5	1273±557	430±145	28.3±15.8	198(σ_g=1.65)	2.50±0.25	9-10.5	37.2±1.8

Table 2. Summary of data for all plumes sampled during straight and level runs. From left to right: plume index as marked in Fig. 4; source origin; source latitude location and region; BC and CO fraction contributed by OBB sources modelled by FPES×emission rate; plume altitude expressed as height and pressure; mean BC, SO_4, Org and CO concentrations and standard deviation (σ); rBC core mass median diameter (MMD) and the geometric standard deviation of the size distribution (in brackets); BC coating thickness D_p/D_c; plume age as estimated by back trajectories; accumulated precipitation along trajectory pathway.

Troposphere Levels	Asia	Europe	North America	Background
	BC (ng sm^{-3})			
LT (0-2500m; 750-1000hpa)	32.1±26.2		21.8±14.0	13.3±8.5
MT (2500-5500m; 500-750hpa)	100.8±48.4	70.8±39.1	19.9±13.3	
UT(5500-8000m; 350-500hpa)	55.8±22.4	12.6±7.0	23.6±16.7	
	ΔCO (ppbv)			
LT	14.6±12.1		17.0±6.6	10.4±4.3
MT	28.8±17.4	24.3±12.7	16.8±6.9	
UT	32.4±14.7	25.3±9.9	19.8±7.4	
	BC D_p/D_c			
LT	2.03±0.24		2.25±0.24	2.21±0.29
MT	2.16±0.13	2.21±0.20	2.33±0.29	
UT	2.21±0.20	2.28±0.40	2.24±0.30	
	Sulphate (ng sm^{-3})			
LT			870±255	336±173
MT		905±479	445±213	
UT		108±50	280±121	
	Organic (ng sm^{-3})			
LT			235±83	94.5±73.0
MT		210±94	129±92	
UT		97±53	176±130	
	Measured BC/ ΔCO (ng sm^{-3} $ppbv^{-1}$) (mg g^{-1} in branket)			
LT	2.25±0.69 (1.80)		1.16±0.36(0.93)	1.82±0.98(1.46)
MT	4.22±1.50 (3.38)	2.79±1.23(2.23)	1.26±0.30(1.01)	
UT	2.01±1.49(1.61)	1.03±0.08(0.82)	1.33±0.67(1.06)	

Table 3. Average statistics for BC, ΔCO, BC D_p/D_c, sulphate, organic mass, and BC/ ΔCO in the lower (LT), middle (MT), and upper (UT) troposphere during periods when the different source regions dominated. For BC/ΔCO, both units ng sm^{-3} $ppbv^{-1}$ and mg g^{-1} (in branket) are presented.

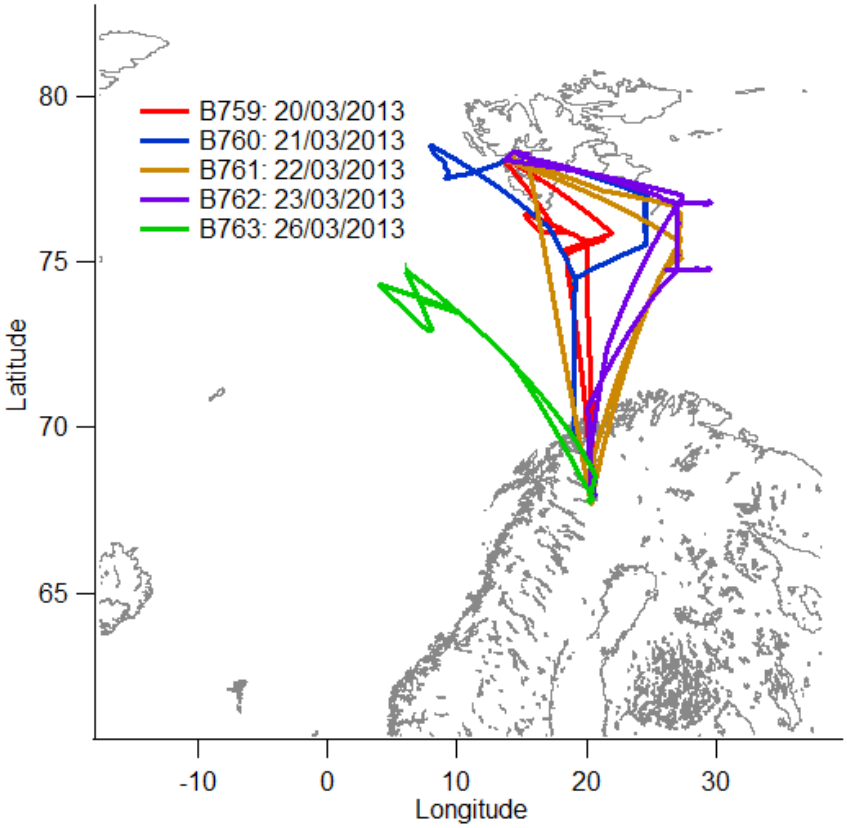


Fig. 1. Flight tracks during ACCACIA spring campaign.

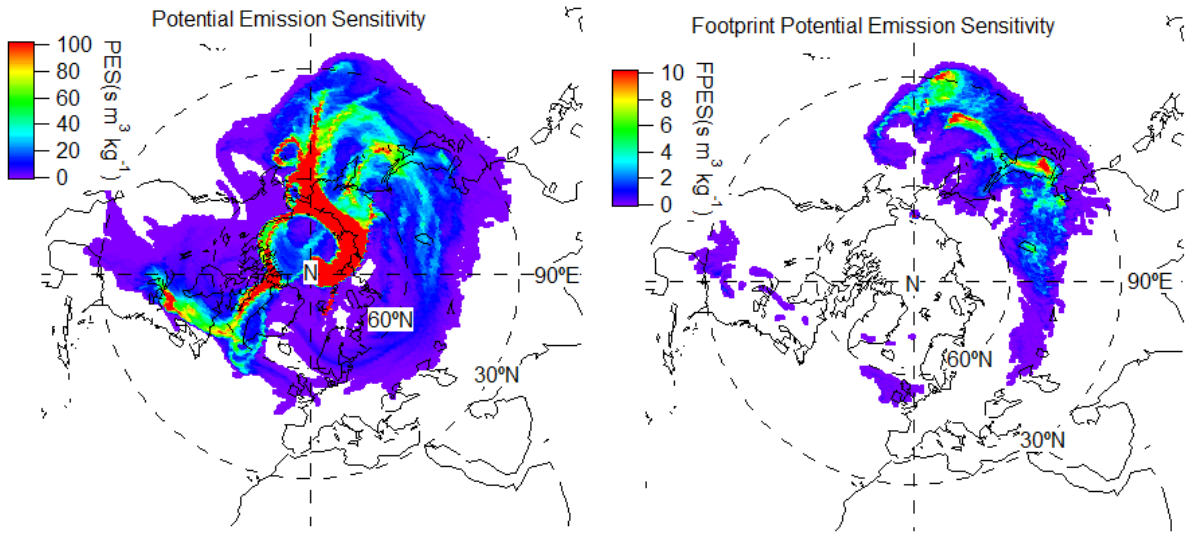


Fig. 2. A typical example of (a) PES and (b) FPES from FLEXPART simulation from flight B759.

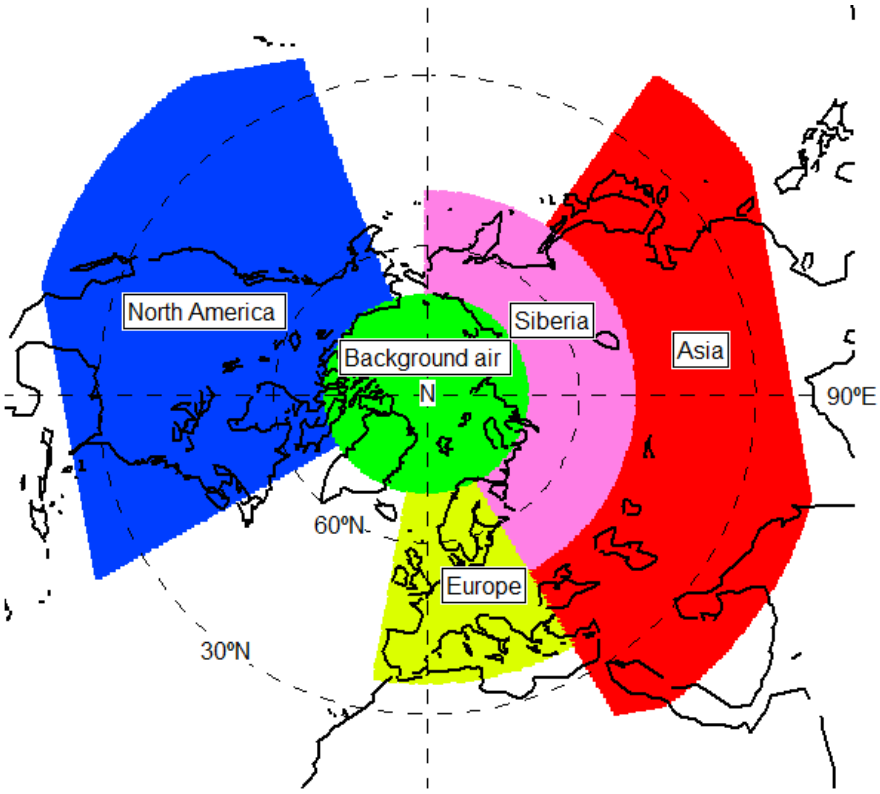


Fig. 3. Source regions defined in this study.

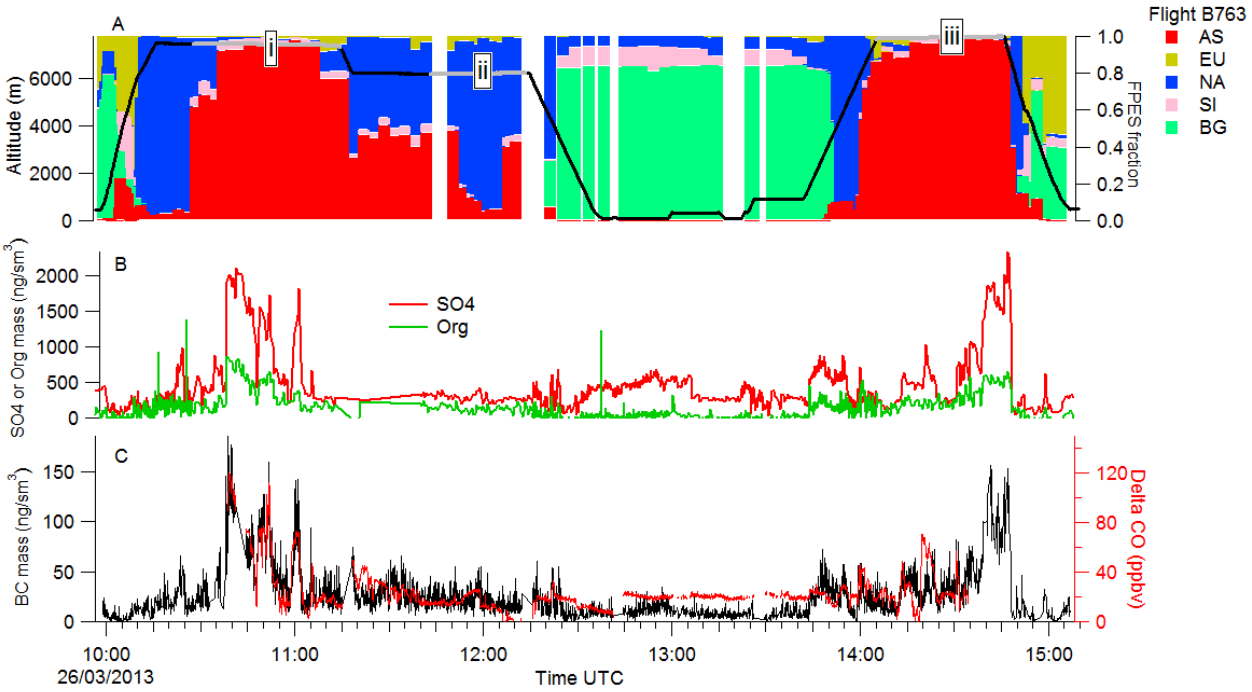
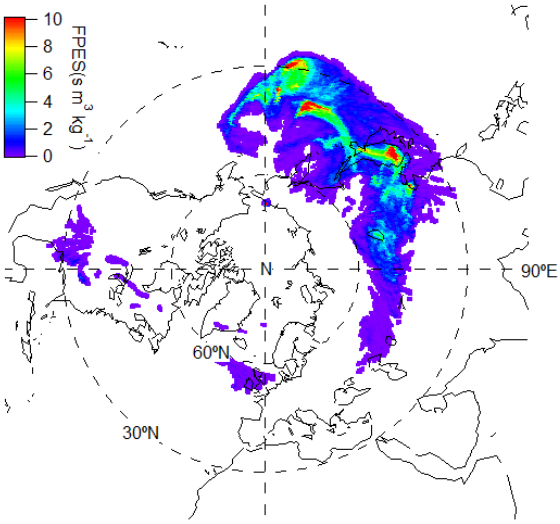


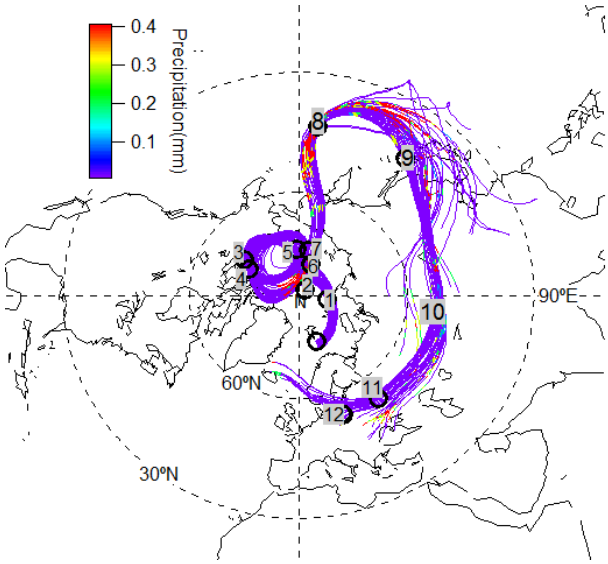
Fig. 4. A) The flight altitude and FLEXPART FPES fractions for each source region during B763, the plume locations during SLR are marked as i, ii and iii and grey lines along the flight altitude track; B) the time series of organic matter and sulphate; C) BC mass and CO concentration.

Asia (AS)

A1

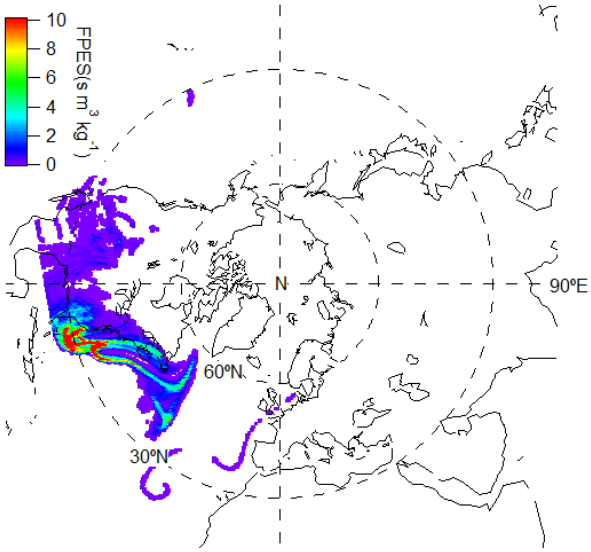


A2

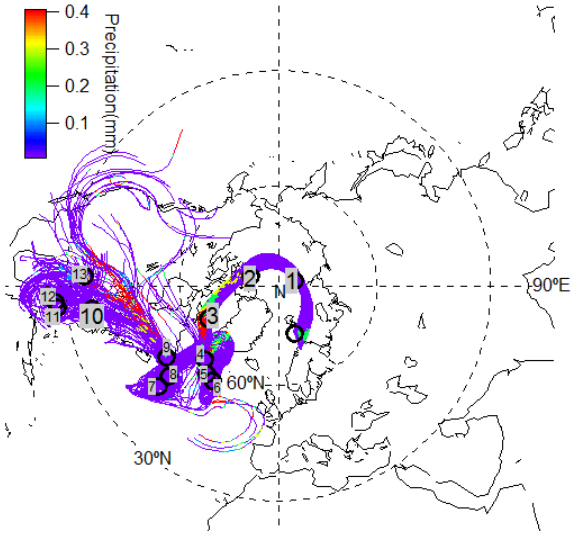


North America (NA)

B1



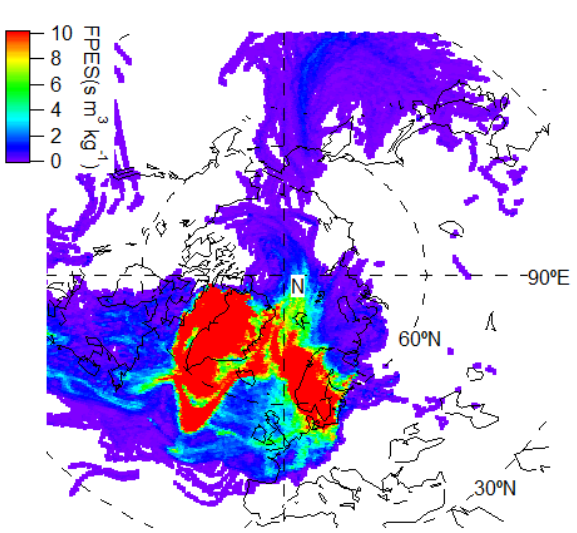
B2

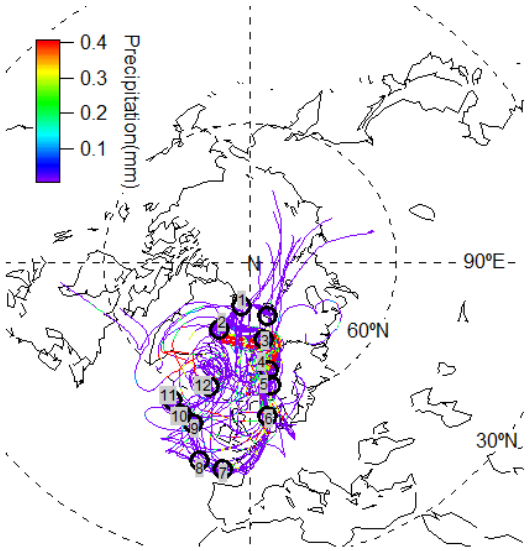


Europe (EU)

C1

C2

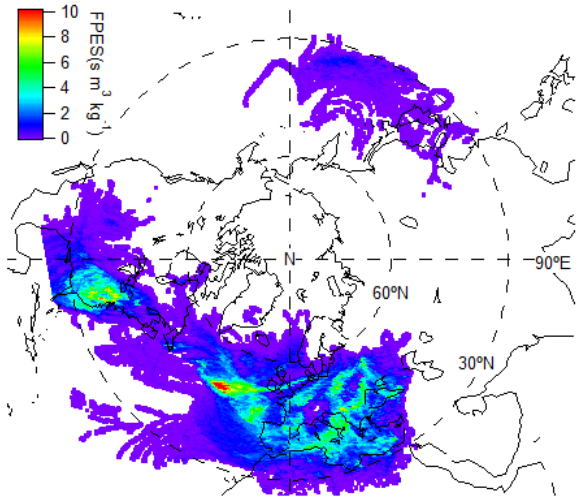


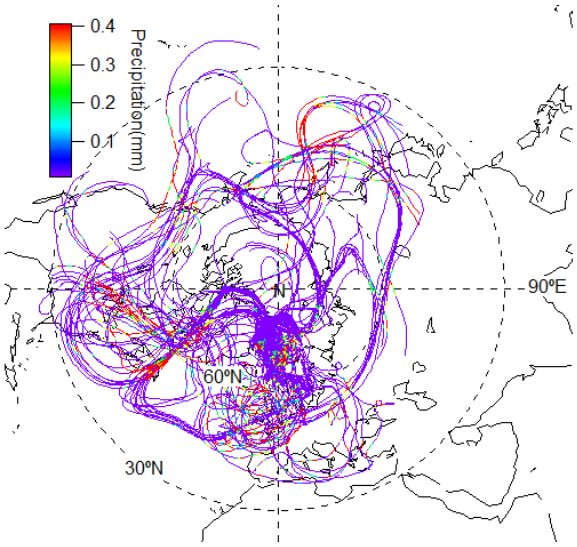


NA+EU

D1

D2

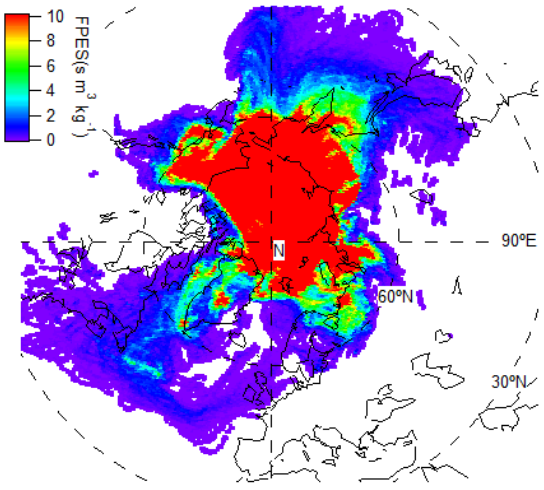




Arctic Background Air (BG)

E1

E2



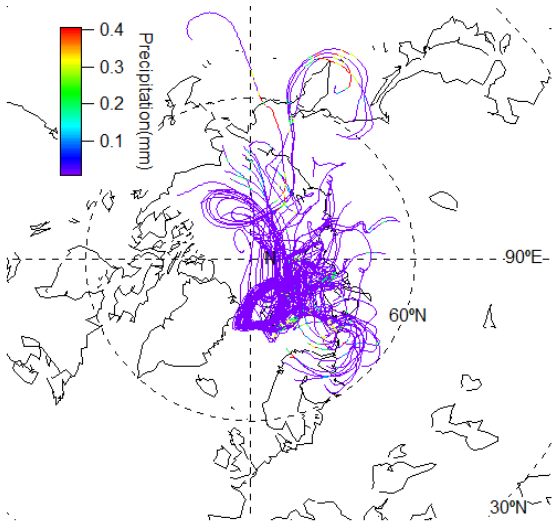


Fig. 5. The FLEXPART FPES (left panels from a1-e1) and the corresponding HYSPLIT backward trajectories (right panels from a2-e2) for each classified air mass origin, representing flight B759 10:50-11:15, B760 8:45-9:45, B761 12:10-12:25, B761 15:06-15:15 and B759 15:15-15:45 respectively. Each backward trajectory is coloured by precipitation along the pathway and the open circles with numbers in A2, B2 and C2 mark the time backwards along the trajectory in 1 day intervals.

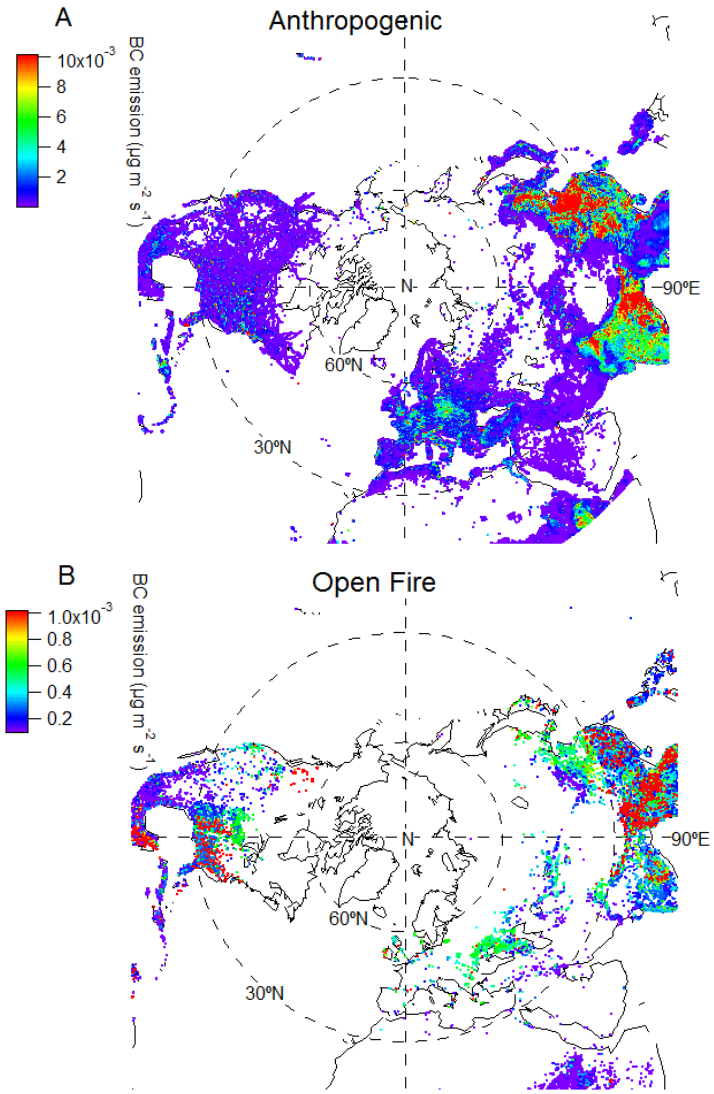
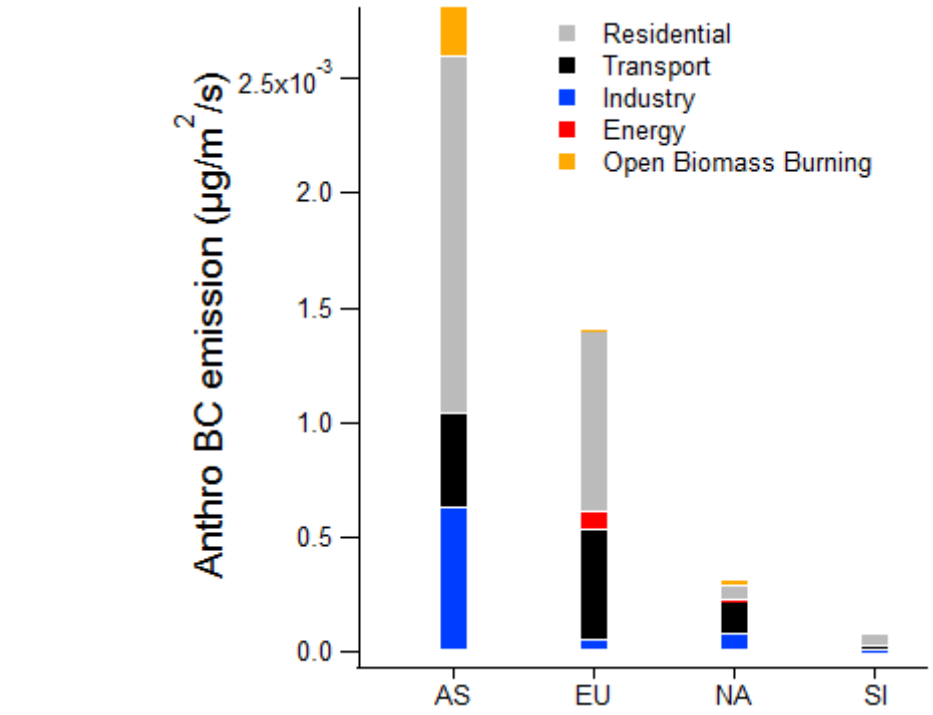


Fig. 6. The BC anthropogenic (A) in March 2010 and open fire (B) emission inventories in March 2013, from HTAPv2 and the FINN open fire biomass burning inventory respectively. The specified sectors for anthropogenic sources, i.e. residential activity, transport, industry, energy and flaring are shown in Fig. S3.

A



B

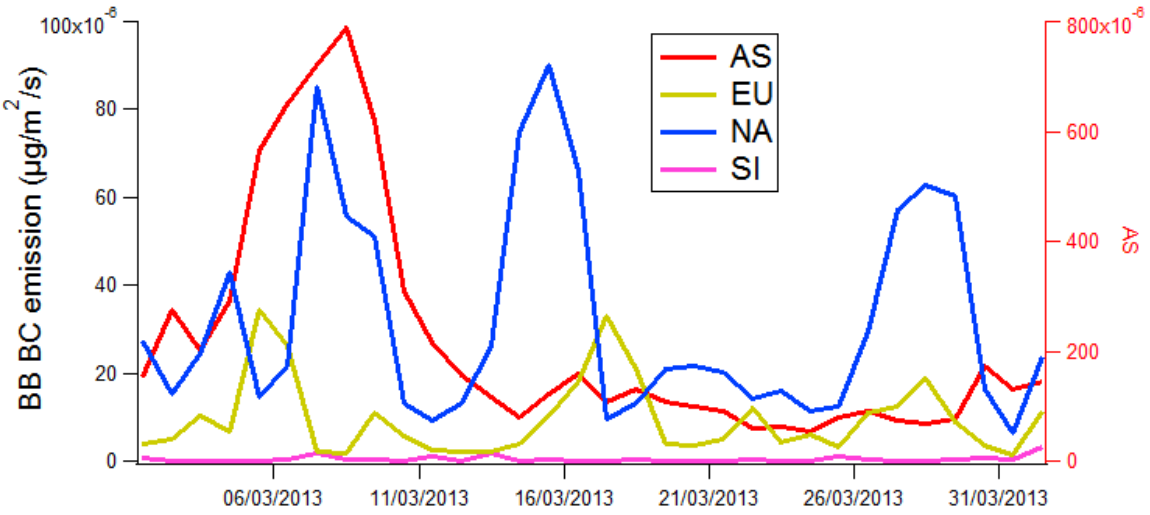


Fig. 7. BC emissions characterised by source region (as defined in Fig. 2). (a) comparison between different source sectors; (b) the temporal evolution of OBB sources throughout the experimental period. Note for AS, the right axis is used.

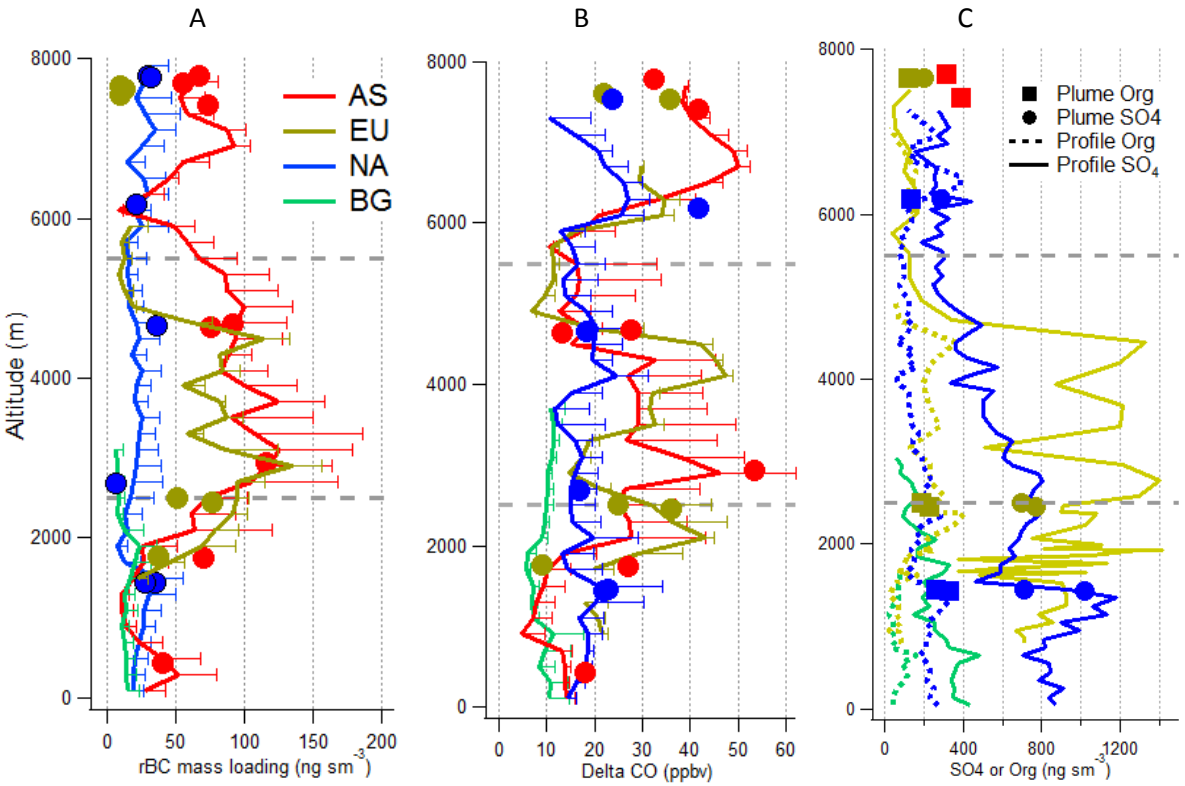


Fig. 8. Vertical profiles (binned in 200m altitude) and plumes classified by air mass origin: the lines show the mean values and +σ for data obtained during vertical profiles; the filled markers denote values derived during plume intercepts along straight and level runs for (a) rBC mass loading; (b) ΔCO; (c) sulphate and organic matter. Note there was no sulphate or organic data available for AS vertical profiles therefore only AS plume information is shown in (c). The horizontal dash lines on each graph show the bounds of LT/MT/UT.

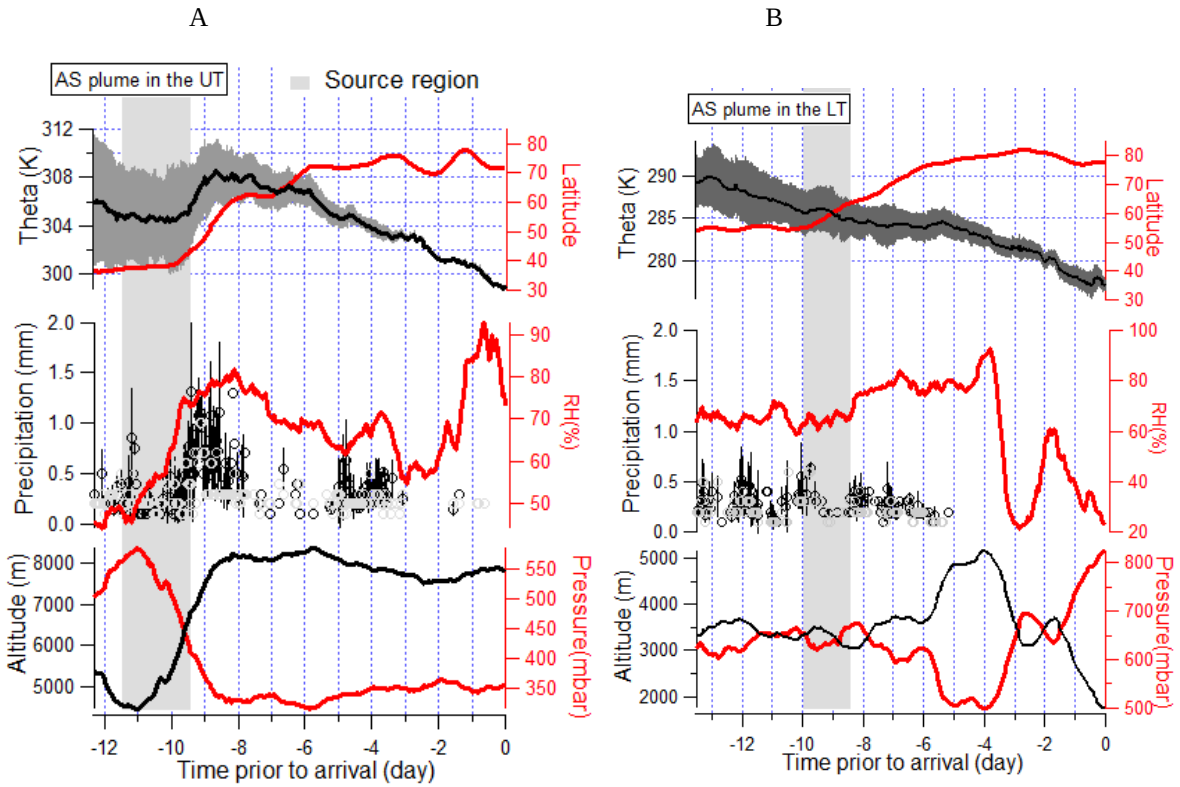


Fig. 9. Average characteristics of sampled air parcel plumes arriving at receptor locations along 12 day backward trajectory pathways. (A) and (B) show the cases for AS plume in the UT (corresponding to B759 i plume in Table 2) and LT (B760 ii in Table 2) respectively. All parameters shown are mean values averaged over all back trajectories sampled throughout the specified plume. The bars on the precipitation and potential temperature (θ) data denote the stand deviation ±σ of the ensemble of all back trajectories, and the grey markers on precipitation show the median value. The grey bars mark the time when the air masses passed over the source regions according to FPES. The identical plots for the other air mass regions are shown in Fig. S5.

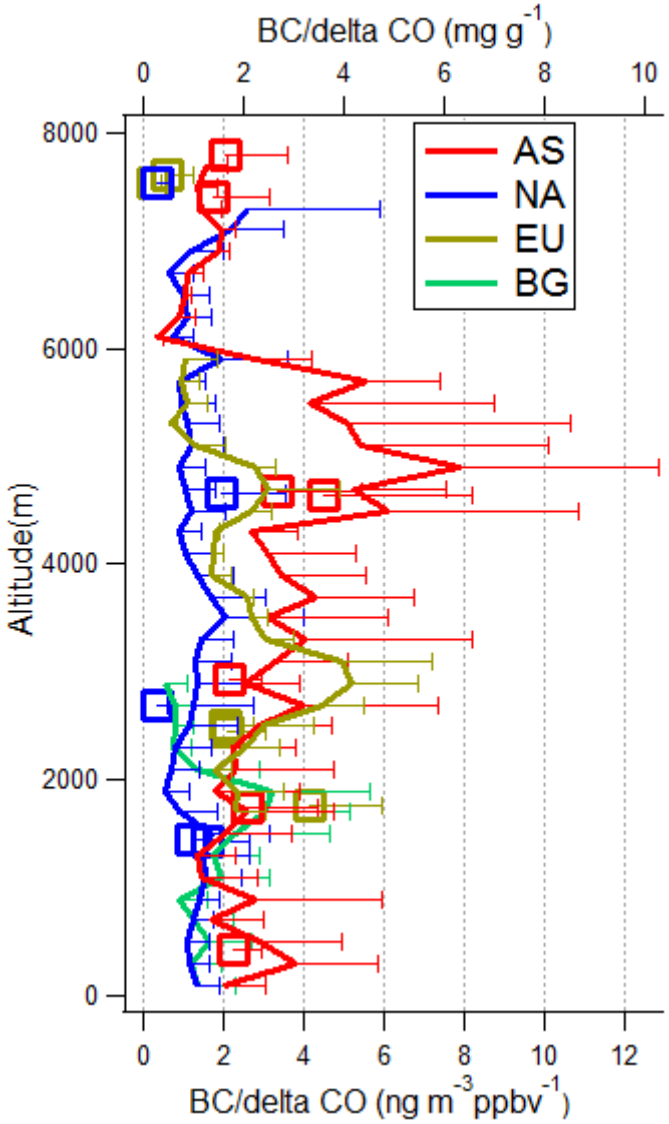


Fig. 10. BC/ΔCO measured during ACCACIA for different air mass origins, with the lines and square markers showing the ratio for vertical profiles and plumes respectively. The bars show the +σ.

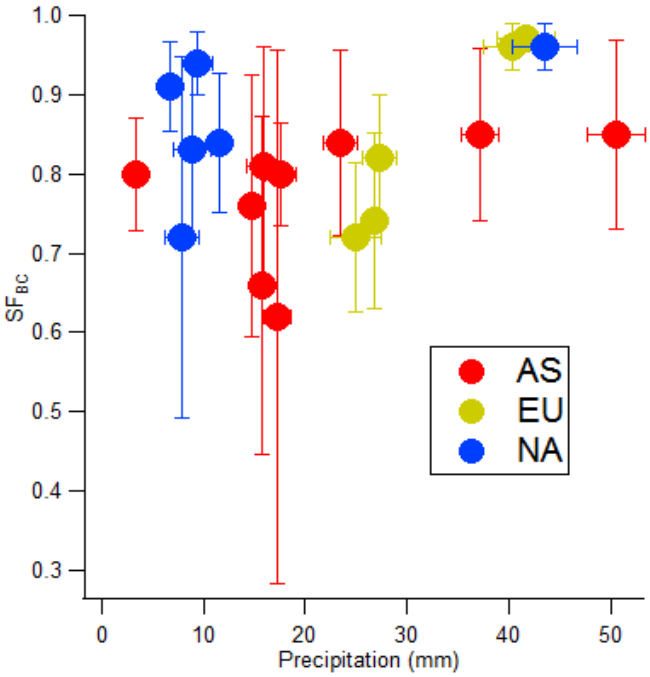
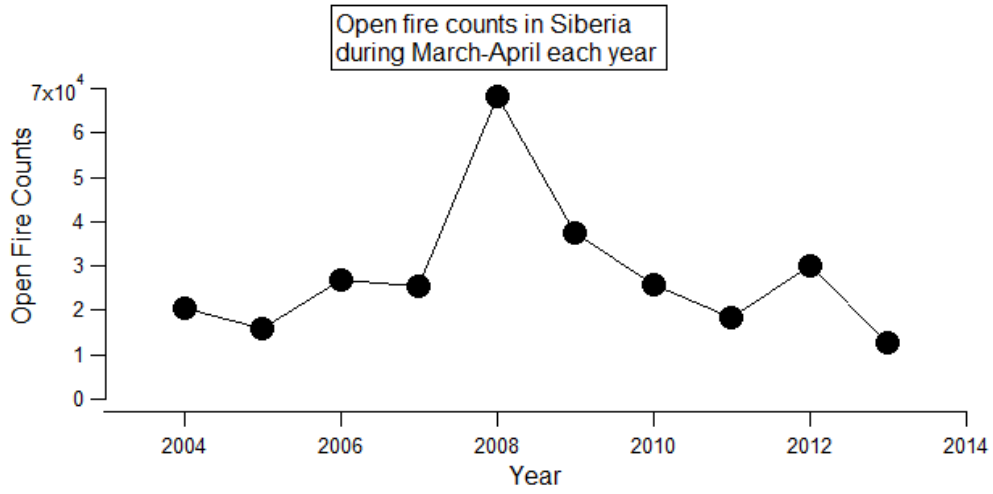


Fig. 11. The calculated scavenged fraction of BC (SF_{BC}) versus accumulated precipitation, with the bars showing the ±propagated uncertainties for both precipitation and SF_{BC}.

A



B

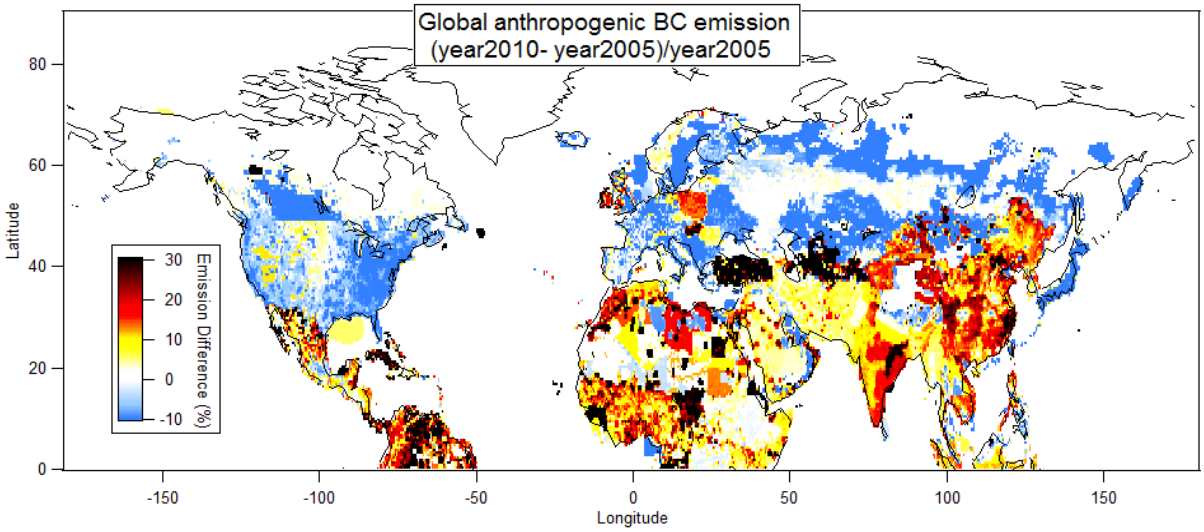


Fig. 12. A) Fire counts from FINN in Siberia region (50-70°N, 30-180°E) during March and April from 2004 to 2013. B) Percentage difference between total anthropogenic BC emissions from the ECLIPSE global BC inventory in 2010 and 2005.