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.

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 m^-3.

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.

p. 14848, line 4 - 7: This sentence is confusing. Please edit for clarity. This has been clarified.

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".

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". *p. 14849, line 22: Corrected for ambient what? Aerosol?* This is corrected.

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.

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, lines 16 - 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."

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 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.

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 x10⁴ 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.

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:

"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. 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.

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.

"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.

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.

The paragraph 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.

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.

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.

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.

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.