Author's reply to peer-review comments on

"An 800 year high-resolution black carbon ice-core record from Lomonosovfonna, Svalbard", by Dimitri Osmont et al., submitted to ACP.

We would like to thank the referees for the time they spent on our manuscript and for their constructive comments which helped us to improve the quality of this paper. Please find below our responses to your comments (in blue) and our changes to the manuscript (in red).

Reviewer 1 (RC1, Mauro Rubino):

Osmont and colleagues present a new record of black carbon concentration from ice cores from Svalbard. The authors measure the concentration of black carbon in two ice cores sampled from the Lomonosovfonna ice field in 2009 and 2011 covering the periods 1222-2004 and 2004-2011 respectively, and show that the results from the two ice cores are compatible for the overlapping year (2004). Then they discuss the long term trends of the black carbon concentration record by converting it into a black carbon flux record. After realising that the black carbon concentration completely changes after the industrial revolution, they focus on the industrial period and interpret the industrial black carbon record by comparing it with proxies of anthropogenic aerosol emissions (such as nitrate, sulphate, and ammonium). In doing so, they attribute an anthropogenic origin to part of the measured black carbon. Then, the authors compare their record to records of black carbon from Greenland, and use the HYSPLIT model to identify the geographical source of anthropogenic black carbon in the Lomonosovfonna ice core. They conclude that Siberia and Northern Europe are the main sources of black carbon for Svalbard. However, since they are unable to provide an explanation for some of the features found in the industrial black carbon record, they discuss the possibility that post-depositional processes induced by summer melting could have been the cause for them. Their discussion is qualitative, as they state, but it is realistic and leads to the conclusion that melting and refreezing has affected part of the industrial record of black carbon. Finally, they interpret the pre-industrial record as a reconstruction of biomass burning events by comparing it with records of other proxies of biomass burning such as ammonium, formate, vanillic acid and para-hydroxybenzoic acid, and performing a Principal Component Analysis. In the end, they also correlate the frequency of past biomass burning events recorded with the main indexes of climate variability (e.g.: the summer temperature anomalies and the Palmer drought severity index). The study is of interest for ACP. Though not extremely innovative, the investigation provides a new record of black carbon from a site which had not been explored before. The methods used are appropriate and rigorous. The interpretation of the results has no flaw, as far as I can see. Therefore, the manuscript can be published in the present form. However, I have provided several comments on points that I did not find very clear, or that I think could be improved (e.g.: some figures could be merged). I have reported my comments on the manuscript in the attachment. I hope the authors will take my comments into consideration before the manuscript is accepted.

Please also note the supplement to this comment:

https://www.atmos-chem-phys-discuss.net/acp-2018-244/acp-2018-244-RC1-supplement.pdf

We acknowledge your overall positive evaluation. We considered all your comments and revised the manuscript accordingly. We reported your comments from the supplement below and give detailed responses.

Page 5, line 2: Could you provide a number here? From figure 2a it is not possible to see what type of reproducibility you have got.

We calculated the relative errors between each sample and its respective replicate and averaged the values for the 120 samples, leading to a total relative error of 23 %. We added this information in the manuscript p.5 lines 2–3.

Page 6, line 20: I would not say that the concentration of rBC in LF-11 is lower than in LF-09. Without reading the following sentence, this sentence made me think that the concentration of rBC measured in LF-11 is lower than that measured in LF-09 in the overlapping year (2004). To prevent confusion, I would rather say that the concentration of rBC measured in LF-11 after 2004 is comparable to preindustrial values measured in LF-09.

We clarified this sentence to avoid misunderstandings. It reads now: "rBC concentrations in the LF-11 ice core are comparable to preindustrial values (before 1800) measured in the LF-09 ice core, with an average of 0.5 ± 0.4 ng g^{-1}, a median of 0.3 ng g^{-1} and a range from LOD to 2.4 ng g^{-1}."

Page 7, line 22: Either you say 'almost the same' or 'similar'. Also the flux and concentration records cannot be similar, rather their trends are similar.

The reviewer is right to mention that trends are similar but not concentrations and fluxes. We corrected the sentence and specified the small discrepancies between the two records as follows: "Trends in the rBC flux and concentration records are almost the same (except that the highest fluxes were recorded in the 1870s)…"

Page 8, line 6: It would be much easier to see the correlation if you merge figure 5 and 6. Could you move the emission inventories from figure 6 to figure 5 below the Fiescherhorn EC record?

Figures 5 and 6 were merged according to your suggestion.

Page 8, line 27: declining

Replaced.

Page 8, line 29: I do not see these similarities. To me, you can only say that the LF rBC record and the HDF EC record are less dissimilar than the LF rBC records and the rBC records from Greenland ice cores. Try to quantify the degree of similarity, if you believe they are similar.

We agree with the reviewer's statement that the LF and HDF records are less dissimilar than the LF and Greenland records and that resemblances between the HDF and LF record are not obvious. We deleted the discussion about their similarities and rephrased this part accordingly: "Despite their common Svalbard origin, similarities between the HDF and LF records are not obvious. The LF and HDF records are less dissimilar than the LF and Greenland records as they do not show a uniform decline since the 1910s maximum but a second increase from the 1930s to 1960s in HDF and 1940s to 1970s in LF."

Page 8, line 40: use of coal and oil

Corrected.

Page 10, line 1: I suggest you name the different plots in figure 7 with letters, as you did in figure 3, and cite them here when you mention them. So, Svalbard airport temperature series would be figure 7d, for example.

We added letters to each figure containing several panels when they were missing (new figures 4-5-6-7) and included more references to the figures in the text. We also added in Fig. 7a (Fig. 6a in the new version) the time periods when algae were visible in the LF ice cores (red bars).

Page 11, line 37: I do not understand why you cite this reference (Owens et al., 2017) here. It does not look like they talk about biomass burning in their paper.

This reference was provided to justify the term "Little Ice Age". As it does not bring any useful material to the discussion, we deleted it.

Page 12, line 13: I would not provide the list of years when there is matching between rBC and ammonium peaks or peaks of other proxies. This is difficult and boring to read. I suggest you provide the total number of matching peaks only (as you did for ammonium and rBC ('In total, 21 peaks...').

We agree with your suggestion. This succession of years does not read well and is in addition not very relevant given the dating uncertainties. We decided to remove it and we added a table describing the total number of matching peaks between the different proxies with the percentages of peaks this represents for each proxy (Table 3):

Table 3: Number of peaks matching within ±1 year between the different fire proxies (in bold) for the time period 1222–1800. Italic numbers below give the percentage of matching peaks in relation to the total number of peaks for each proxy.

	rBC	Ammonium	Formate	VA	p-HBA
rBC	-				
Ammonium	**21** *42 % rBC* *60 % ammonium*	-			
Formate	**19** *38 % rBC* *31 % formate*	**19** *54 % ammonium* *31 % formate*	-		
VA	**17** *34 % rBC* *38 % VA*	**10** *29 % ammonium* *22 % VA*	**21** *34 % formate* *47 % VA*	-	
p-HBA	**16** *32 % rBC* *37 % p-HBA*	**12** *34 % ammonium* *28 % p-HBA*	**17** *27 % formate* *40 % p-HBA*	**16** *36 % VA* *37 % p-HBA*	-

In addition, the respective paragraph in section 3.4 reads now:

Table 3 shows the number of peaks matching (within ±1 year due to the different sampling) between the different proxies, associated with the percentage it represents in relation to the total number of peaks for each proxy. The best agreement is obtained between rBC and ammonium, with 21 peaks matching, which corresponds to 60 % (42 %) of the total number of ammonium (rBC) peaks, respectively.

Reviewer 2 (RC2, Michel Legrand):

The paper presents the first long-term continuous and high-resolution refractory black carbon (rBC) record extracted from ice cores drilled in Svalbard and covering the last 800 years. The record is discussed in terms of anthropogenic (fossil fuel) emissions as well as past biomass burning (boreal fires) occurrence. The contribution of biomass burning to the rBC variations is addressed with complementary information from other potential proxies of biomass burning including ammonium, formate, vanillic and p-hydroxybenzoic acids.

Data on black carbon in ice deposits in the Arctic basin are definitely of major interest when considering our future climate. Past frequency of boreal fires also are of importance since boreal forest is an important carbon reservoir and experiences natural fires of which the severity is expected to change with the on-going global warming. At the opposite to Canadian fires, Siberian fires are far less documented except for the very last decades (satellite data). The Svalbard experiencing air masses from Siberia and to a lesser extent from Europe and North America, this paper provides new information together with those recently extracted from the Akademii Nauk on Siberian fires over the past. The paper is therefore of great interest for scientific communities working on forest fire records in ice cores and lake sediments as well as for the general topic of climate/fire conditions/vegetation interactions.

Overall the manuscript is well organized and clearly written. The discussion of data is very well conducted, and I enjoyed reading it. I therefore recommend publication of the manuscript, after authors consider the following (minor) points rise below.

Evaluation:

Among others, I identify two very positive aspects in this paper:

Inherent to this region, present climatic conditions may, in some extent, disturb the ice record. This difficulty is well addressed in the manuscript.

I would like to congratulate the authors for reporting and discussing together several potential biomass-burning proxies (too many previous studies only focusing on one or two proxies).

We thank you for the very positive evaluation and the compliments which are a motivation to pursue our efforts. We carefully took into account your suggestions and provide a detailed response below.

Minor points:

Section 3.2.1:

Line 4: “Inflection point”: please reword.

We reworded this as follows: “The most striking feature is the increase in rBC concentrations and variability from 1800 on that we attribute to rising anthropogenic BC emissions.”

Line 9: You can also cite here the work from McConnell.

We included the citation.

Line 26: remove “aerosol” since nitrate is partly in the form of gaseous nitric acid.

Removed.

Equation 2: why this use of molar ratio: why not mass ratio (0.25) ??? (all your data are in mass concentrations)

This equation is obtained by subtracting the proportional amount of Na^+ from the SO_4^{2-} according to the sea water ratio of Na^+ to SO_4^{2-}. We directly reused the equation presented in Wendl (2014) in section 2.7.2, that is

why we converted our mass concentrations to charge concentrations (µeq L^{-1}). We totally agree that we could have kept mass concentrations and used the appropriate ratio (0.25). This would give a similar result.

Section 3.4

Lines 35-39: I think the reference is here is not adequate: the year 1994 is not exceptional because documented by Dibb et al. in terms of back-trajectories. May certainly be more useful here to say that "the year 1994 was marked by a high fire activity in Canada (6.08 MHa burnt, see Legrand et al. 2016 for the complete references of data that I copied below). By the way, that permits to compare with your following discussions on 1980 and 1981.

Data on area burned in Canada are available since 1920 (Van Wagner, 1988) and became more accurate after 1959 with the Canadian National Fire Database (Canadian Forest Service, 2015, National Fire Database – Agency Fire Data, Northern Forestry Centre, Edmonton, Alberta; http://cwfis.cfs.nrcan.gc.ca/ha/nfdb) providing precise fire location, start date, and final size (Stocks et al., 2003).

We thank the reviewer for the interesting references and took his comment into consideration. Indeed, fires in 1994 in Canada burned a larger area than in 1980 or 1981. However, the rBC peak is lower in 1994 (12 ppb) than in 1980 (39 ppb) or 1981 (32 ppb), thus highlighting the importance of transport and deposition processes. We reorganized this paragraph as follows: "For instance, the highest rBC concentrations of the record in 1980 and 1981 could be linked with strong biomass burning seasons in Canada (4.8 and 6.1 Mha, respectively, Stocks et al., 2003) potentially related to the ammonium spikes noted in Greenland ice cores (Legrand et al., 2016). The clear rBC peak visible in the LF record in summer 1994 could reflect the high fire activity in Canada for the year 1994, when 6.1 Mha burned (Stocks et al., 2003). Dibb et al. (1996) documented the advection of a biomass burning plume from the Hudson Bay lowlands, Canada, to Greenland on August 5th 1994, suggested to be responsible for an increase in NH_4^+, K^+ and light carboxylic acid concentrations in the snowpack."

Lines 15-26, second page: I like this discussion.

Line 24, second page of this section: "secondary production of formate (from formaldehyde) is possible: Please add « formaldehyde and numerous volatile organic compounds including alkenes for instance (see Figure 1 in the review from Legrand et al. (2016)" is possible.

We added this sentence.

End of the review

Reviewer 3 (RC3, Anonymous Referee #2):

This paper describes a rBC chronology from Svalbard that extends from 1222 to 2009. This chronology is supplemented with an impressive multi-proxy approach to determine rBC source attribution as well as a forest fire history. I'm impressed with the body of work presented here, but I am concerned that several weaknesses exist that merit attention before publication. For example, I find that the discussion of rBC and snowpack melting is weak and contains inconsistencies (details below). Further, and I think that this is not unique to this paper, when you're making conclusions based on labile and/or unstable organic proxies, there's been no consideration given to postdepositional processes affecting those proxies, nor possible changes to those proxies during analysis. For example, did you monitor changes in formate during analysis? Where duplicate samples run, etc… Changes in formate, VA, p-HBA, levoglucosan, either post-depositionally or during the analysis are going to have an effect on your interpretation and so should be considered, or at least controlled. More detailed comments follow:

We are grateful to the reviewer for his comments. We would like to point out that major ions (including formate) in the LF-09 ice core were measured by Wendl et al. (2015) while VA and p-HBA were analyzed by Grieman et al. (2018). We have full confidence in the quality of the analyses, and we kindly ask the reviewer to refer to the respective publications for further information.

Organic acids and specific molecules (such as VA or p-HBA) are known for experiencing post-deposition processes or atmospheric degradation (Legrand et al., 2016). We are aware of these limitations and reminded it throughout the paper (e.g. p.2 2nd paragraph or p.11 1st paragraph). For specific organic markers, little is still known about their preservation. Unfortunately, without detailed field studies (snowpits), it is impossible to quantitatively assess the impact of those processes on the glaciochemical records.

However, the analytical process should not affect the concentration of these species as we worked in clean conditions to prevent sample contamination as much as possible. We did not monitor changes in formate during analysis as it is normally not required. Instrumental and procedural blanks as well as duplicate samples were prepared in the same way as for rBC. Samples were cut from the inner part of the ice core to limit contamination from ambient air. Samples for ionic analyses were melted under a N_2 atmosphere (Wendl et al., 2015). They were analyzed rapidly after melting (in a few hours at the maximum) and covered with aluminium foil to prevent contamination by lab air. We agree with the reviewer on the fact that formate analysis is not trivial as contamination can easily occur, either from plastic materials or formic acid traces from ambient air (Legrand and Saigne, 1988).

Intro Line 35: Not that I think that you should get into a prolonged discussion about the causes of Arctic amplification, but Serreze and Barry (Global and Planetary Change, 2011) might be a more suitable citation as they explain that it's not simply an albedo issue.

We thank the reviewer for the interesting reference. We added it to the paper. The first sentence is now: "In the last decades, the Arctic region has experienced the strongest surface air temperature increases globally, referred to as the Arctic amplification (Serreze and Barry, 2011), leading to a range of severe consequences on glaciers, sea ice, wildlife and local human societies and partially explained by strong snow and sea ice feedbacks implying surface albedo changes."

Page 2, line 14: consider changing the word "nowadays".

We replaced "nowadays" by "currently".

Page 2, line 20…: I find this argument to be somewhat weak. I don't think that this study seeks to determine source attribution on a global scale, but rather is limited to the Arctic. "BC in the environment" implies a global scale, especially when it follows a summary of northern-hemisphere BC chronologies. Rather than "BC in the environment", maybe consider "BC in the Arctic"?

We agree with the reviewer and replaced "environment" by "Arctic".

Page 7, line 22: The rBC flux and concentration records are not similar, but the trends in each record are…

This point has already been mentioned by reviewer 1. We rephrased this sentence; please see the corresponding comment in RC1.

Page 7, line 25: Is this significant correlation between annually averaged rBC and anthropogenic aerosols, or the 11-year moving averages? How was this correlation calculated, because the data does not appear to be normally distributed.

It is with the 11-year moving averages. For the significance, we took into account the loss of degrees of freedom induced by the averaging process. Data were not log-transformed.

Page 8, line 29: Your qualitative treatment of the "similarities" between LF and HDF rBC records is weak. Arguably, they don't look similar, and the similarities that you propose are lagged, or seem to be… Invoking "local differences in transport, deposition and melting effects" to explain why they don't match is a cop out unless you can provide evidence that these mechanisms are in play.

We agree with the reviewer that resemblances between the two records are not evident. This point was also mentioned by reviewer 1 and we rephrased it accordingly (see comment above). Our interpretation is that, despite the discrepancies (particularly after 1970), the HDF and LF rBC records are less dissimilar than the Greenland and LF rBC records. Lines 33 to 36 give some elements to justify those site-induced differences: "Beaudon et al. (2013) showed that the HDF ice core was more affected by melting, with outstanding melting features after 1970, than the LF ice core during the 20th century. In addition, the LF site is more frequently located above the thermal inversion layer in winter, in contrast to the HDF site, thus being more exposed to long-range pollution from the free troposphere (Beaudon et al., 2013)". In addition, we have already discussed in section 3.1, lines 31–41, the analytical differences between rBC and EC measurements and what they could imply.

Page 8, line 40: Is it strange that you don't find the impact of World War II in the rBC record?

rBC values start to increase again in the 1940s. rBC annual averages show a clear rise from 1944 on but given the dating uncertainty (±3 years at that time), we cannot fully exclude or include any influence from World War II. Fires due to bombings and fighting or sustained industrial and energy production to support the war effort could have enhanced rBC emissions. The HDF EC record does not show a clear impact of World War II either. This is not very surprising as Svalbard stayed away from the main centres of conflict. Moreover, rBC local sources were reduced at that time since mining activities were stopped after 1941 and most of the coal mines were destroyed during World War II (Hisdal, 1998).

Page 9, line 18: Again, invoking melting without providing evidence that it's happened is a cop out.

This sentence was meant to introduce the next section. We added a reference to section 3.3: "As discussed in the next section, we suggest that post-depositional effects induced by summer melting are mostly responsible of these features."

Page 9, line 30 and onward: This section is difficult to follow with several inconsistencies.

1) evidence for surface melting as indicated by the presence of "local algae" between 1900 and 1940 doesn't match your dip in rBC from 1915-1935. The Svalbard airport temperature record really doesn't tell us anything about conditions that might produce melting at any point during the year, nor does the Arctic temperature anomaly. To me, the strongest indication of melt is the melt index, which spans between 1915-1935, as indicated by your shading, but doesn't cover the entirety of your low rBC period, that spans from ~1905-1935.

We kindly ask the reviewer to refer to the publication from Hicks and Isaksson (2006), figure 2, presenting the algae record for the LF-97 ice core (and not our LF-09 ice core). Algae are only observed between 1901 and 1940, with a maximum between 1909 and 1932. We do not understand your assertion that it does not match our

rBC minimum extending roughly between 1915 and 1935. Obviously, we cannot expect the dates to perfectly match within one year given the dating uncertainties, the disturbances induced by melting and the fact that we are comparing two different ice cores drilled 4.6 km apart.

Of course the Svalbard airport temperature record and the Arctic temperature anomaly cannot indicate local conditions that prevailed on the Lomonosovfonna glacier at that time but these latter are and will remain unknown. However, the Longyearbyen weather station is the only long-term and continuous record available for Svalbard and it has been carefully studied and corrected. It is evident that both records show a sustained warming trend from the 1910s to 1930s that has been identified everywhere in the Arctic (Bengtsson et al., 2004). It seems reasonable to think that this warming also affected the LF site which is not located at a high altitude (1200 m) and can experience positive temperatures in summer.

The melt index behaves like an instantaneous proxy for melting as it is based on the different elution of two ions that have similar sources. It shows that most of the melting took place between 1915 and 1935, which is supported by the temperature and algae records. On the contrary, the melt% based on melt features or ice lenses acts as a time-lagged proxy as meltwater first percolates through to snowpack and refreezes lower down, forming this characteristic transparent ice features. This is why the melt% peak is visible a few years before (or meters below), from 1900 to 1915.

Page 10, line19: Have you provided any evidence that these algae layers exist?

This information was taken from the logbook of the drilling expedition of the LF-09 ice core, mentioning that reddish layers were visible in tubes 171 and 173 spanning 1883–1885 and 1879–1881, respectively. The logbook also indicates the occurrence of ice lenses and dust layers. These red layers do not stem from volcanic eruption (ashes) as none of these layers (7 in total) match historic eruptions. We think that this information is reliable and is worth being presented.

Page 10, line 21: You use possible artifacts associated with sample treatment and/or analytical treatment to explain an anonymously low rBC decrease that doesn't correspond to melt layers. If we accept this, then how would this artifact affect any rBC reading that you've provided? If you're going to invoke the artifact argument to explain an anonymous result, doesn't that undermine the result for any sample that was treated in the same way?

It has been shown that rBC concentrations were lower for samples experiencing melting and refreezing during laboratory studies. We suggest that similar processes can naturally happen in the snowpack. We therefore wanted to warn that rBC measurements are probably biased (under-estimated) when dealing with ice-core sections having high levels of melt, like in LF for the time periods 1910–1940 and 1980–1990. Even if rBC is not the most labile compound in ice cores, it seems more affected during the 1910–1940 time period than ions, even compared to the most mobile ones such as nitrate or sulfate (Fig. 4). This artefact is specific to the SP2 method as other analytical methods (ion chromatography, thermal-optical analysis for EC) are not impacted by melting. However, when little melting occurs, the SP2 method remain reliable and rBC values not biased. This is the case for the pre-industrial record.

Page 11, line 24: "Formate can also undergo post-depositional effects", such as? Are you referring to, at least in part, biogeochemical processing in the snowpack? Did you test for changes in formate during sample analysis? Were the samples fixed to inhibit microbial alteration during sample preparation and analysis?

Formate, similarly to other light carboxylic acids, is a relatively weak acid and can be remobilized depending on pH. Once deposited in the snowpack, it can re-volatilize and diffuse in the snowpack (De Angelis and Legrand, 1995). Nevertheless, it is less affected than other carboxylic acids and was confidently used as a biomass burning proxy (Legrand et al., 2016). Here we did not refer to biogeochemical degradation of formate in the snowpack. To our knowledge, this process has not been documented. It happens in liquid phase but remains negligible in frozen samples, that is why samples are stored in frozen state and are melted just prior to analysis. Legrand and De Angelis (1995) stated: "Samples were kept frozen until the time of the analysis, and it was

therefore not necessary to treat them with a biocide to protect them against bacterial activity as it is required for storage of liquid samples."

We added some information in the manuscript: "Formate, like other light carboxylic acids, can also undergo post-depositional effects such as re-volatilization and diffusion in the snowpack (De Angelis and Legrand, 1995)."

Page 12, line 14 and onward: Can these dates be presented as a table? It's difficult to keep track of when presented in the text.

We agree with this suggestion. As already explained to reviewer 1, we created a new table (Table 3) listing the total number of matching peaks between the different proxies with the percentages of peaks this represents, similar to what we did for ammonium and rBC. We therefore deleted the list of years, which is not of great relevance given the dating uncertainty.

Page 12, line 34: Are you suggesting that fires in Tibet deposited rBC in Svalbard? Do you have any evidence to suggest that rBC would stay aloft long enough to travel that distance? Earlier, you suggested that rBC could be transported from Canada if the right atmospheric conditions prevailed. I would think that transport from Tibet would be more of a stretch.

We never said that fires in Tibet were contributing to the rBC deposited in Svalbard. We reported ice-core evidence from Tibet that the 1790s decade experienced severe drought conditions at a continental scale due to South Asian Monsoon failure and a strong El Niño event (Thompson et al., 2000). Other drought evidence is presented in the same paper. Moreover, fires in Tibet are rare owing to the scarce vegetation cover and the cold climate due to the high altitude. However, the Altai region, located in Southern Siberia, 2000 km north of Tibet, and extensively covered with forests and meadows, is a potential source area for the rBC deposited in Svalbard as fires in this region are part of the ecosystem (Eichler et al., 2011). Atmospheric back-trajectories studies performed for the LF site showed that Siberia was the major source region (Grieman et al., 2018). We therefore used in Fig. 9a (Fig. 8a now) temperature and precipitation records from one Siberian region (the Altai) for which available records exist to discuss the potential influence of regional climatic conditions on the LF rBC record.

We clarified this paragraph as follows: "In Fig. 8 an exhaustive comparison is made between enhanced background concentration periods and peak years versus summer temperature anomalies and drought reconstructions from two regions of Northern Eurasia for which datasets are available, namely Northern Europe (Cook et al., 2015; Esper et al., 2014) and the Altai (Büntgen et al., 2016; Cook et al., 2010)."

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