Author's response to referee comments on

"Tracing devastating fires in Portugal to a snow archive in the Swiss Alps: a case study", by Dimitri Osmont et al., submitted to TC

We would like to thank the referee for the time spent on our manuscript and for the detailed and 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 grey and italic).

Anonymous Referee #2

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This manuscript attempts to link emissions from a known fire event and their deposition on snow close to JFJ in the Swiss Alps through extensively investigating a severe fire event occurred on 17-24 June 2017 in Portugal. This study is attempting to provide an interesting approach, connecting a set of valuable record of charcoal, black carbon, and ions in the snow pit and a combination of atmospheric in-situ measurement, remote sensing, air mass trajectory calculation, and transport simulation, to better understand the information of particle deposition in snow in European high-altitude sites. This approach is very useful to understand atmospheric processes of aerosol particles how to transport over long distances, be scavenged by snow fall, and be deposited to snow by providing clues. However, there is an issue that should be considered seriously to publish the results. Authors measured charcoal, refractory black carbon (rBC), and ions in snow collected near JFJ and compared their profiles with equivalent black carbon (eBC) measured in the atmosphere at JFJ. The elevations of charcoal in the upper layer of C in snow (sample "2" in figure 2e) and eBC in atmosphere on 22 June (figure 2a) were obvious with an increase relative to background level by _6 times and _10 times, respectively, indicating that the fire plume reached JFJ. In contrast to them, rBC in sample "4-6" (figure 2d) increased a little and changes in ion concentrations are even indiscernible. Readers reasonably expect that potassium, ammonium, and nitrate can be elevated in their concentrations if the big fire plume indeed reached and is detectable by charcoal and eBC. In the same snow layer, the rBC concentration was elevated just 1.5 times relative to the second peak with rBC concentration of _5 ng/g, that is probably from local fires as suggested by authors. Discuss this issue in dept and provide more evidences and/or assumption to support your argument.

This is a really helpful suggestion, and the point was raised also by referee 1. We were from the beginning puzzled by not finding elevated concentrations of the other fire tracers (ammonium, formate, and acetate) in the samples 4-6. Re-considering this in view of both referee's comments, we revised our interpretation. Charcoal originating from the Portugal fires was deposited by dry deposition during the period 22nd-24th June, when the fire plume was detected by atmospheric eBC measurements at JFJ. Since no snowfall occurred during that period, the majority of BC was not deposited, since dry deposition is not efficient for submicron particles. Dry deposition of charcoal most likely resulted in a confined layer, which was not resolved with the 10 cm sampling resolution. With the snowfall on 25th June, air mass transport changed, ending the advection of the fire plume to the JFJ as indicated by the backward trajectories arriving at JFJ at 18 UTC (will be included in Fig. 4 in the revised version). Instead more regional polluted air masses were scavenged. That the charcoal and

rBC peaks partly overlap is most likely due to the coarse resolution of the charcoal samples. The change of air masses does explain the lack of a peak in the other fire tracers, e.g. ammonium, and the time delay between the charcoal and the BC peak in the model output and the snow pit data. The manuscript was changed accordingly.

Abstract:

According to modelled emissions of the FINN v1.6 database, the fire emitted a total amount of 203.5 tons BC from a total burned area of 501 km^2 as observed on the basis of satellite fire products. Backward trajectories unambiguously linked a peak of atmospheric equivalent BC observed at the Jungfraujoch research station on 22nd June, with elevated levels until the 25th June, with the highly intensive fires in Portugal. The atmospheric signal is in correspondence with an outstanding peak in microscopic charcoal observed in the snow layer, depositing nearly as many charcoal particles as during an average year in other ice archives. In contrast to charcoal, the amount of atmospheric BC deposited during the fire episode was minor due to a lack of precipitation. Simulations with a global aerosol climate model supported that the observed microscopic charcoal particles originated from the fires in Portugal and that their contribution to the BC signal in snow was negligible. Our study revealed that microscopic charcoal can be transported over long distances (1500 km), and that snow and ice archives are much more sensitive to distant events than sedimentary archives, for which the signal is dominated by local fires. The findings are important for future ice-core studies, as they document that for BC as fire tracer the signal preservation depends on precipitation. Single events, like this example, might not be preserved due to unfavorable meteorological conditions.

Fire tracers:

We hypothesize that charcoal originating from the Portugal fires was deposited by dry deposition during the period 22nd to 24th June, when the fire plume arrived at the JFJ as detected by elevated atmospheric eBC concentrations. Since no snowfall occurred during that period, the majority of BC was not deposited. Dry deposition most likely resulted in a confined charcoal layer, which was not separated from the rBC peak in the snowpit due to the coarse sampling resolution of 10 cm. With the beginning of snowfall on 25th June, air mass transport changed, ending the advection of the fire plume to the JFJ as indicated by backward trajectories (see below and Fig. 4). Instead more regional polluted air masses were scavenged, which explains the absence of ionic fire tracers and of a shift in the rBC size distribution.

Atmospheric transport:

The back trajectories with arrival at JFJ on 25th June at 18 UTC indicates a major change in the synoptic situation to more north-westerly flow directions. We can only speculate that this happened concomitant with the onset of precipitation, since the timing of the latter is not precisely known.

Furthermore, other BC sources than fires seem to dominate in our simulations, which is in agreement with our hypothesis that during fire plume arrival at JFJ the majority of fire-related BC was not deposited due to the lack of snowfall. With the beginning of snowfall at JFJ, air mass transport changed and more regional polluted air masses with minor or without fire contribution were scavenged.

Conclusions:

Dry deposition of microscopic charcoal resulted in an outstanding peak in the snowpack. This event deposited nearly as many charcoal particles as during an average year in other ice archives.

For rBC, in contrast, concentrations in the snow were not exceptionally high. In combination with the absence of a peak in ionic fire tracers such as ammonium, this suggest that the majority of atmospheric BC was not deposited during the fire episode due to a lack of precipitation. Instead the observed rBC peak was mostly likely caused by scavenging of air masses containing regional pollution with the beginning of snowfall on 25th June, which ended the advection of the fire plume to the JFJ. rBC scavenging ratios were in line with previous studies, giving additional evidence that rBC was predominantly scavenged by wet deposition. Simulations with a global aerosol climate model supported that the observed microscopic charcoal particles originated from the fires in Portugal, whereas their contribution to the BC signal in snow was minor. The findings of our case study are important for future ice-core studies, as they document that for BC as fire tracer the signal preservation depends on precipitation and wet deposition. Single events, like this example, might not be preserved due to the unfavorable meteorological conditions.

L135: Since the fire event studied in this work was a well-documented recent extreme event, authors need to calculate forward air mass trajectories to see if the plume departing from the exact fire spot reach the JFJ site. Consider that a starting height of air mass trajectories is not necessarily just above ground because fire-emitted particles can be directly injected into the free troposphere up to > 3 km. Also, it would be great to see if the area-averaged time series of AOD match with forward air mass trajectories. For AOD, just simply check GIOVANNI platform (https://giovanni.sci.gsfc.nasa.gov/giovanni/#service=DiArAvTs&starttime=&endtime=&bbox=- 180,-90,180,90). Intense fire plumes are often easily captured by AOD, and thus AOD near JFJ should be elevated day-by day.

We calculated forward trajectories as suggested by the reviewer and they basically agree with the backward trajectories. We prefer to keep the backward trajectories in the manuscript, since they are more conclusive, but added the one arriving at JFJ on 25th June 18 UTC to show the change to north-westerly air flow. We show now trajectories calculated 5-days backward at 20 equidistant levels in pressure coordinates between 700 hPa and 500 hPa, in accordance with the detected smoke plume height.

Thanks for the suggestions of the GIOVANNI database to explore changes in AOD concentrations. We had a look into different daily AOD products (e.g. from OMAEROe v003 at 0.25° (smallest spatial resolution in the GIOVANNI database) to the MODIS-Terra MOD08_D3 v6.1. at 1° spatial resolution) and their area-averaged time series for 17th until 26th of June. However, this didn't help to address your question because 1) of their rather coarse spatial resolution, 2) cloud coverage which obscured the satellite's view. The data are inclusive because of their poor spatial resolution.

In addition, we looked at AOD data with a spatial resolution of 10 km at nadir from the Modis sensor flying onboard of the satellites Terra and Aqua, which support that between 20th and 22nd of June the JFJ site received air masses with elevated AOD concentrations from Portugal. The AOD concentration levels increased over Switzerland mainly North of the Alps

during that time, in agreement with the backward trajectories (Fig. 4). From 23rd to 25th of June the area was cloud covered, so AOD could not be retrieved.

Since AOD data are not fully conclusive, we decided not to include them in the manuscript.

L206: The upper size limit of BBHG is generally <300 nm, although which depends on the instrumental setting. Please check if authors mentioned "BBLG" instead of "BBHG".

Thank you for discovering this! The sentence was changed accordingly.

The mass size distribution for the broadband low gain (BBLG) channel of the SP2 remained similar throughout the profile with 306 nm and 291 nm for the two series of replicates, respectively (Fig. 2d).

L205-214: In figure 2d, the diameter of rBC particles in sample "4-6" (figure 2d) seem similar to that of other layers except for sample "13". The size of atmospheric rBC particles generated from biomass combustion is generally larger than that from urban fossil emissions. If the rBC size is not obviously large in sample "4-6", the possible reason should be suggested, regarding for example, cloud or wet-scavenging during transport and/or scavenging by snowfall, etc.

This finding fits now better with our revised interpretation, see discussion above, and changed the discussion about the size distribution.

Only sample 13 shows a bigger fraction of large rBC particles, in association with a small peak in rBC and charcoal concentration. On the contrary, no shift in the rBC size distribution is visible for samples 4 to 6 during the peak in rBC and charcoal concentrations.

L224: Authors mentioned that smoke particles can be lifted up to free troposphere and travel over long distance. It can be true not only for charcoal but also for rBC particles. As noted above, it should be seriously considered why rBC and ionic particles did not elevated unlike charcoal. rBC particles are small with diameters < 1 micrometer, which thus can travel longer distances, as found in previous studies so far. Also, authors may need to check if the number of particles in SP2 scattering channel ("SCLG" or "SCHG") is elevated in the snow layers. The profile of scattering particles, i.e., number concentration, might correspond to that of charcoal.

We agree. rBC particles should have been transported as well. We think that charcoal was deposited by dry deposition whereas rBC was not scavenged until 25th June due to the lack of precipitation, see comment above. We revised our interpretation accordingly, see comment above.

Charcoal particles detected microscopically had a major axis >10 µm. Such large particles are not aerosolized with the nebulizer set-up and, thus, do not enter the SP2. In addition, it is not possible to analyse insoluble scattering-only particles in aerosolized snow (or other liquid) extracts since extremely large numbers of scattering particles are produced by aerosolization and drying of the dissolved components in the solution. The numbers of such particles overwhelm any SP2 scattering signals from the insoluble components.

L230: Both ammonia (NH3) and NOx can be emitted from fires, and during the transport time in the atmosphere, ammonium nitrate (NH4NO3) and other forms of nitrate and ammonium can be formed particularly under high relative humidity via aqueous reaction. Authors should seriously consider why such inorganic ions are not sufficiently detected in the snow layer corresponding to smoke plume. Meteorological conditions, for example too

dry condition in FT and/or too warm in PBL, were not favorable for the aerosol formation? Moreover, potassium has been broadly used as an indicator of biomass combustion so far. Did authors observe a peak of potassium in the snow layer?

We agree and revised our interpretation accordingly, see above.