

# 1 Refractory black carbon mass concentrations in snow and 2 ice: method evaluation and inter-comparison with 3 elemental carbon measurement

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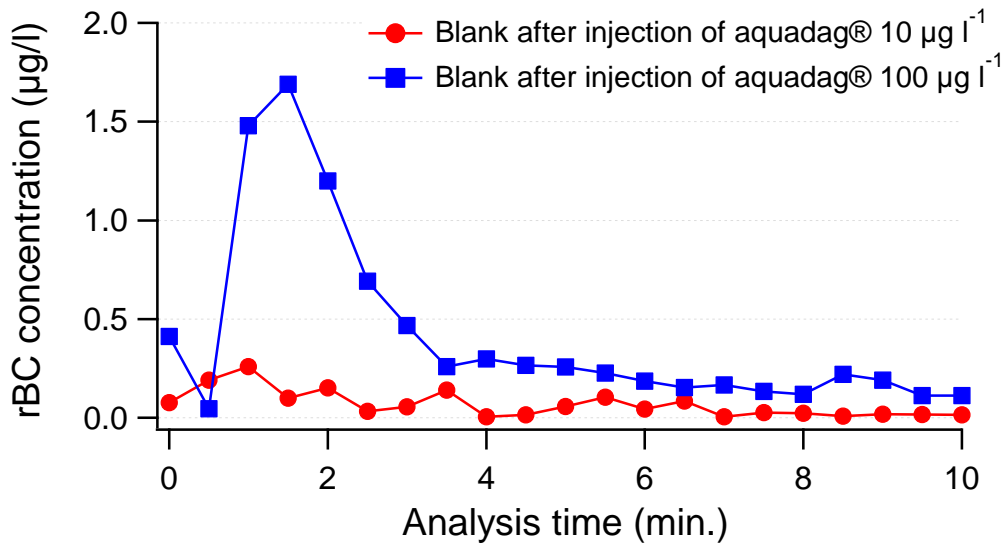
## 14 **Supplementary material**

### 15 **Fate of unaerosolized rBC particles in the APEX-Q**

16 We investigated potential memory effect of the APEX-Q nebulizer. To do so, we sequentially  
17 analyzed aquadag® solutions (10 or 100  $\mu\text{g l}^{-1}$ ) and ultrapure water. Figure S1 reveals a rBC  
18 residue of the aquadag® solution lasting for ~10 minutes when measuring the ultrapure water.  
19 The total amount of measured rBC residue was about 5-10 % of the injected rBC, suggesting  
20 that ~5-10 % of the injected rBC mass adheres to the system surfaces between the sample  
21 injection tube and the SP2 optics. Such rBC residue does not represent a significant memory  
22 effect and contamination for measurements of snow and firn samples reported in this study, as  
23 observed rBC concentrations range above a few  $\mu\text{g l}^{-1}$  (Table 2). However, this artifact may  
24 impact analysis of very clean, low rBC levels, samples (e.g., rBC concentrations  $< \sim 1.0 \mu\text{g l}^{-1}$ ),  
25 and cleaning the system with blank water between analyses would then be required. To fully  
26 understand the fate of rBC during aerosolization, we also analyzed the drain of an aquadag®  
27 solution of 10  $\mu\text{g l}^{-1}$  measured. Surprisingly, less than 5 % of injected rBC mass was found  
28 in the drain. Consequently, among rBC mass loss of 35 % (=100 % - 65 %), only ~15 % was  
29 identified both in the drain and on the surfaces of the system. The remaining rBC loss was not  
30 located probably due to the following reasons; (i) no specific aerosolization calibration was  
31 applied when measuring drains, (ii) a minor contribution of mass of hydrogen and oxygen

1 may coexist with carbon of aquadag®, and (iii) the flow rate of sample injection and drain  
2 might be too low to effectively clean out the surface area in the system.

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6 Fig. S1. Measured rBC residues of aquadag® of 10 µg l<sup>-1</sup> and 100 µg l<sup>-1</sup> as a function of time  
7 during 10 minutes when ultra-pure water (i.e., blank conditions) is injected right after  
8 aquadag®.

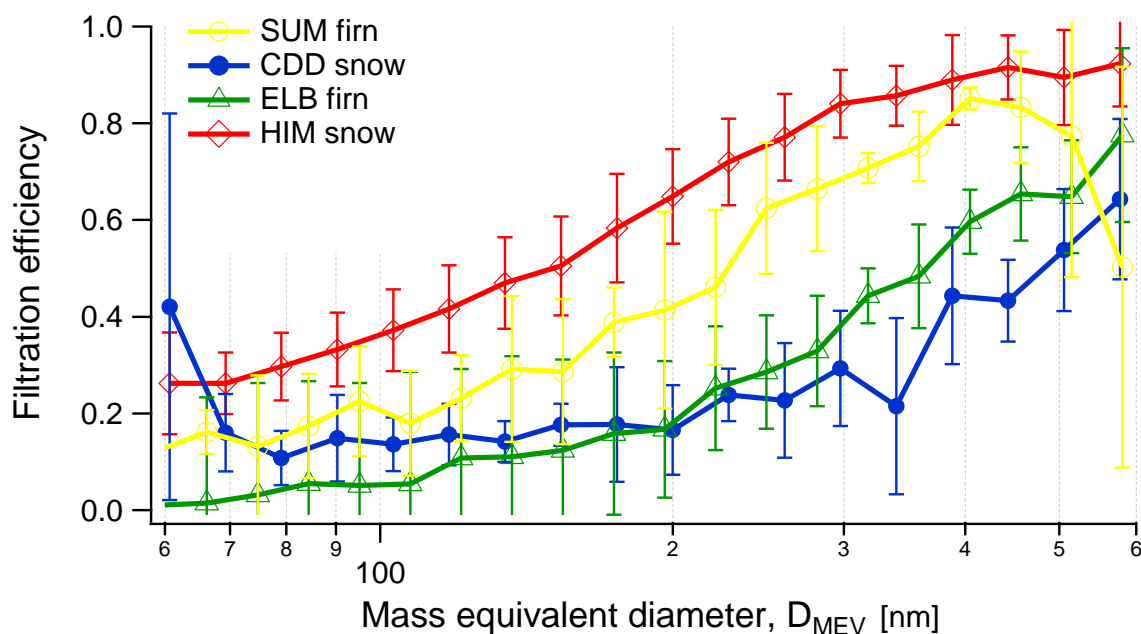
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## 12 **Filtration and chemical decarbonation for thermal-optical method**

13 The EUSAAR-2 protocol was applied to snow and firn by filtering melted samples on quartz  
14 filters (Pall Tissuquartz™ filter, Pall corp.) using sample volumes of 0.16-2.00 liters.  
15 Insoluble EC particles initially present in snow and firn were collected on the filters  
16 introduced into the OC-EC analyzer. Evaluation of the filtration procedure was performed  
17 using the CDD snow and the following optimized procedure was applied to sample filtration.  
18 Melted snow and firn samples were carefully filtrated on a pre-heated quartz fiber filter (pre-  
19 heating conducted at 500 °C during 8 hours) by using a hand pump (e.g., at ~169 mbar). The  
20 separation of water insoluble particles was achieved by using a filter unit equipped with a  
21 stainless steel mask allowing particles concentrated onto a circular spot of 10 mm diameter.  
22 We observed very negligible EC levels in back-up filters, thus a single filter was used.  
23 Filtration was performed twice on a filter. At the end of filtration, a glass unit was rinsed with  
24 ultrapure water (e.g., ~100 ml) to ensure complete transfer of EC particles to filter. Filters  
25 were dried in a laminar flow cabinet all the night through, and then stored in a fridge until

1 before analysis. Whole filters were analyzed, i.e. without punching a specific part of these  
2 filters since we observed different EC concentrations in punched areas of a filter, which  
3 differed up to around factor of two. Filters were decarbonated chemically prior to analysis to  
4 avoid potential positive EC artifact from carbonate carbon(Lavanchy et al., 1999). Briefly,  
5 filters were acidified with 20 % HCl over-night and then neutralized with NaOH pellets for  
6 two days to reach a pH of 7 in a pre-cleaned glass container without air exchange. We  
7 observed that positive EC artifact from calcium carbonate (~6  $\mu\text{g}$ ) was efficiently removed  
8 when this chemical decarbonation procedure was applied to a filter loaded with both  
9 aquadag® (10  $\mu\text{g}$ ) and calcium carbonate (200  $\mu\text{g}$ , a level in the range frequently observed in  
10 CDD ice cores (Legrand et al., 2007; Preunkert et al., 2001).

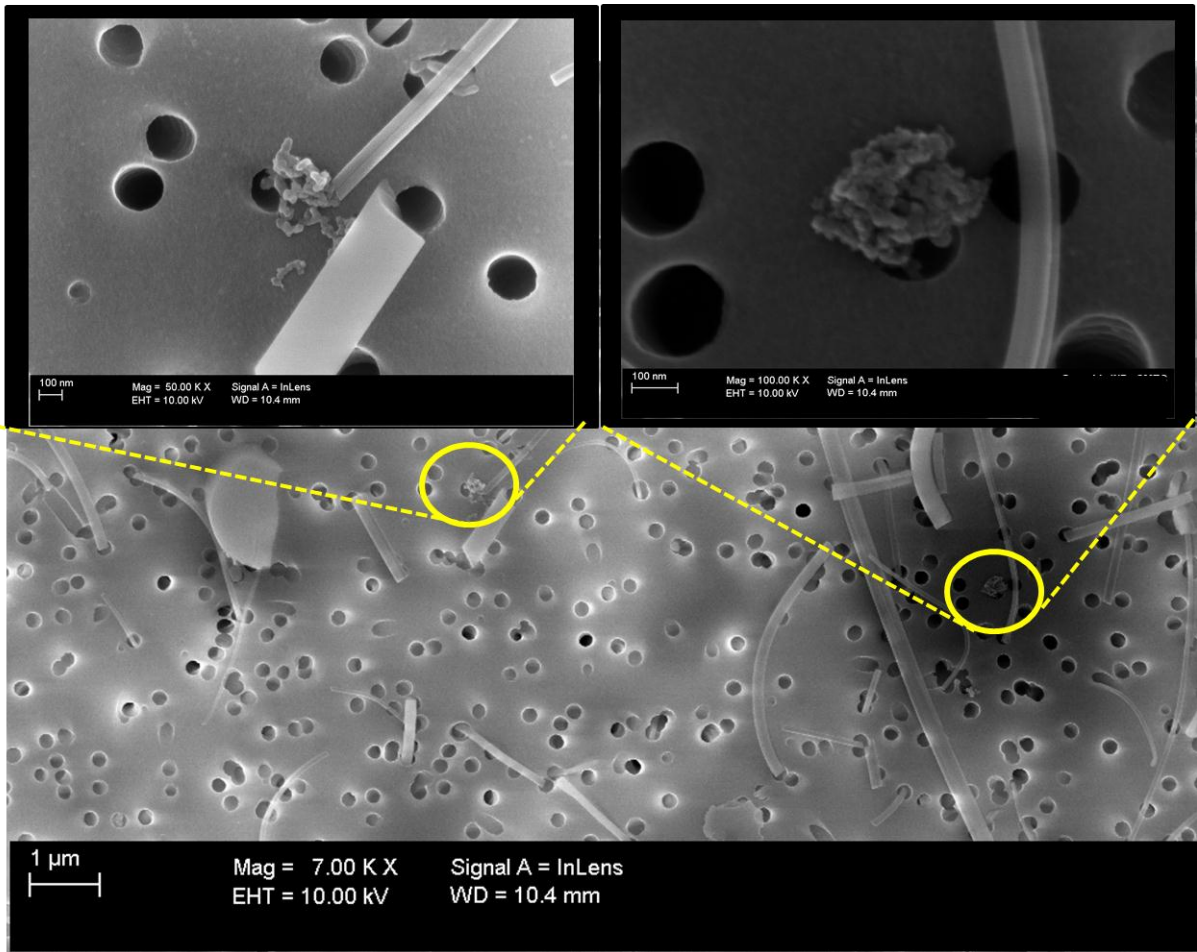
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15 Fig. S2. SP2-based filtration efficiency of rBC particles in snow and firn on the quartz fiber  
16 filter. The whiskers stand for one standard deviation.

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2 Fig. S3. An SEM image illustrating rBC particles retaining on a 0.2  $\mu$ m- nuclepore filter after  
3 first filtration on a quartz fiber filter. 10-mL volume CDD surface snow sample was used to  
4 avoid particle agglomeration on the filters.

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