1 SUPPLEMENTARY MATERIAL

- 2 Black Carbon Seasonal and Diurnal Variation in surface snow in Svalbard and its
- **3 Connections to Atmospheric Variables**
- 4 Michele Bertò^{1#}, David Cappelletti^{2,7}, Elena Barbaro^{1,3}, Cristiano Varin¹, Jean-Charles
- 5 Gallet⁴, Krzysztof Markowicz⁵, Anna Rozwadowska⁶, Mauro Mazzola⁷, Stefano
- 6 Crocchianti², Luisa Poto^{1,3}, Paolo Laj⁸, Carlo Barbante^{1,3} and Andrea Spolaor^{1,2}.

7

- 8 ¹ Ca' Foscari University of Venice, Dept. Environmental Sciences, Informatics and Statistics,
- 9 via Torino, 155 30172 Venice-Mestre, Italy;
- ² Università degli Studi di Perugia, Dipartimento di Chimica, Biologia e Biotecnologie,
- 11 Perugia, Italy;
- ³ CNR-ISP, Institute of Polar Science National Research Council –via Torino, 155 30172
- Venice-Mestre, Italy;
- ⁴ Norwegian Polar Institute, Tromsø, Norway.
- ⁵ University of Warsaw, Institute of Geophysics, Warsaw, Poland;
- ⁶ Institute of Oceanology, Polish Academy of Sciences, Sopot, Poland;
- ⁷ CNR-ISP, Institute of Polar Science National Research Council Via Gobetti 101,
- 18 Bologna;
- ⁸ Univ. Grenoble-Alpes, CNRS, IRD, Grenoble-INP, IGE, 38000 Grenoble, France

20

- 21 ** Now at Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, 5232 Villigen PSI,
- 22 Switzerland

23

24

25

26

27

28 29

30

31

32

33

1. Details of the statistical analyses

The two multiple regression models were fitted on the logarithm scale because the distribution of rBC concentrations in both the experiments is characterized by a significant skewness. Coarse mode particles number concentrations and conductivity were also log-transformed to linearize their relationships with log(rBC). Graphical inspection of residuals plots and normal probability plots confirmed that after the logarithm transformations, the regression models meet the assumptions of linearity, constant error variance (called homoschedasticity in the statistical literature) and normal errors. The regression model fitted on the 80-days (daily sampling resolution) experiments is:

$$\log(\text{rBC}) = \beta_0 + \beta_1 \log(\text{dust}) + \beta_2 \text{eBC} + \beta_3 \text{temp} + \beta_4 \text{snow} + \beta_5 \text{SWR} + \beta_6 \log(\text{cond}) + \epsilon$$

In the regression model 'dust' indicates coarse mode particles number concentrations, 'temp' is the snow temperature, 'snow' is an indicator for the solid precipitation, 'SWR' is solar incoming shortwave radiation, 'cond' is the conductivity and ε is a normal error. In the 3-days experiment (hourly resolution), the model includes also trigonometric components $\sin(2p \text{ hour}/24)$ and $\cos(2p \text{ hour}/24)$ to account for the hourly periodicity of the incoming solar energy. The statistical analyses were performed with the statistical language R (R Core Team, 2020).

2. Calibration of the SP2

The empirical calibration of the SP2, performed using size selected fullerene soot particles is linear up to 500 nm, and the assumption is that it is also linear beyond that size (and the corresponding mass). However, when a massive particle enters the laser beam the incandescence signal might saturate the detector; therefore, in this analysis only the particles below 700 nm were considered. The evaluation of the BC mass for the samples showing a BC geometric mean mass equivalent diameter above 300/400 nm, might therefore be more influenced resulting in an underestimation of the mass. However, the evaluation of the missing mass is beyond the scope of this manuscript and require further analyses. The losses of mass due to the presence of undetected small particles, below 80/70 nm of MED, is not significant given that the geometric mean of the MED of the mass size distributions is always above 150 nm.

3. Possible interferences during the SP2 rBC mass concentration in Arctic snow

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

88

89

90

91

92

93

94

95

96

97

98 99

The surface snow of the Svalbard archipelago is normally characterized by high content of sea salt particles, for example those containing Na, due to its geographical position surrounded by the ocean. The Na concentration in the samples analyzed in this work is, on average, about 500 ng g⁻¹. We cannot exclude that the rBC mass concentration variability for the 80-days experiment presented in this work might have been slightly influenced by the Na content (tracer of sea salt deposition), however any clear relation appeared from the comparison between the rBC and the Na mass concentrations during the "80 days" experiment (Fig. SI 3). As a precaution, all samples from the 3-days experiments were diluted five times with Milli-Q water prior to the SP2 analyses. The SP2 instrumental performances during the analyses, in terms of laser power and incandescence signal quality, were monitored and constant. At the best of our knowledge, no study focusing on the possible effects of the Na particles on the rBC mass concentration retrieved from the SP2 has been published in the literature. Further analyses of snow samples collected in areas characterized by a high influence of marine-like aerosols could shed light on the effects of salt particles on the rBC mass and number concentration via SP2 measurements, as well as establishing a shared procedure to avoid measuring artifacts. Moreover, mineral dust particles might have an influence on the SP2 measurements, depending on their chemical and physical properties. Currently, the literature lacks investigation on these specific SP2-mineral dust particles interactions. However, a few studies on snow and ice samples measured with an SP2 take the assumptions that mineral dust particles are not detected by the SP2 as incandescence signals, but only as scattering signals (Kaspari et al., 2011). Recent studies, based on atmospheric measurements suggest that the SP2 rBC mass concentration measurement can potentially be interfered by the presence of metals and metals-oxide (Moteki et al., 2017), of volcanic ashes or of dust (Kupiszewski et al., 2016).

4. Ancillary measurements for the 3-days samples: Na, Mn, EC, OC

Ancillary data were measured/gathered for the 3-days experiment in order to strengthening the interpretations even though they were not considered in the statistical exercise. The Mn concentrations are considered to be a good proxy for mineral dust aerosols (Baker et al., 2006). In **Errore. L'origine riferimento non è stata trovata.** the Mn concentration profile was compared with that of the coarse mode particles concentration

showing a very similar behavior and suggesting a common source. Despite some differences, a similar pattern is also clearly visible for all the various chemical species.

Every six hours a surface snow sample was collected in parallel to that of the hourly sampling. These samples water volume was of about 1618 ± 290 mL cm³ and they were used for TC, EC and OC measurement, and the results are shown in Fig. SI 4. A different profile is shown for the three compounds compared to that of the rBC mass concentration. As reported above the two measuring techniques are different and, in particular, the size range of particles detected by the two instruments is different, from 80 to 600 nm for the SP2 whereas a much broader dimensional spectrum for the Sunset, potentially explaining part of the observed difference. Interestingly, the EC daily values increased of one order of magnitude, from 1 to 10 µg 1⁻¹, during the sampling period, showing a maximum during the precipitation episode. For more info and results about the comparison between rBC and EC snow/ice measurement check Sigl et al. (2018). On the contrary, the OC atmospheric concentration showed a decreasing trend showing the highest values at the beginning of the sampling period and the lowest at the end, similarly to the atmospheric eBC behavior. Remarkably, the highest OC concentration was found in the same sample were the highest concentration of all the other measured compounds was found (at the very beginning of the snow episode) suggesting a common atmospheric scavenging process (although not above the average for the rBC mass concentration).

Table S1. Abakus Klotz selected dimensional bins.

Channel	Size (µm)
1	0.8
2	0.9
3	1
4	1.1
5	1.2
6	1.3
7	1.4
8	1.6
9	1.8
10	2

11	2.2
12	2.4
13	2.6
14	2.9
15	3.2
16	3.5
17	3.9
18	4.3
19	4.8
20	5.3
21	5.8

22	6.4
23	7.1
24	7.8
25	8.6
26	9.5
27	10.5
28	11.6
29	12.8
30	14.1
31	15.5
32	80

Figure S1. a) Potential source contribution function analysis (PSCF) of eBC recorded in the 80-days experiment (8 April-29 June). 10 days back-trajectories for two selected days: b) 22 April and c) 23 May. Four BTs were calculated for the two selected days, with a 6 hours interval. The red crosses represent the fires taking place during the last 10 days (data from the MODIS active fire products (https://firms. modaps.eosdis.nasa.gov/firemap).

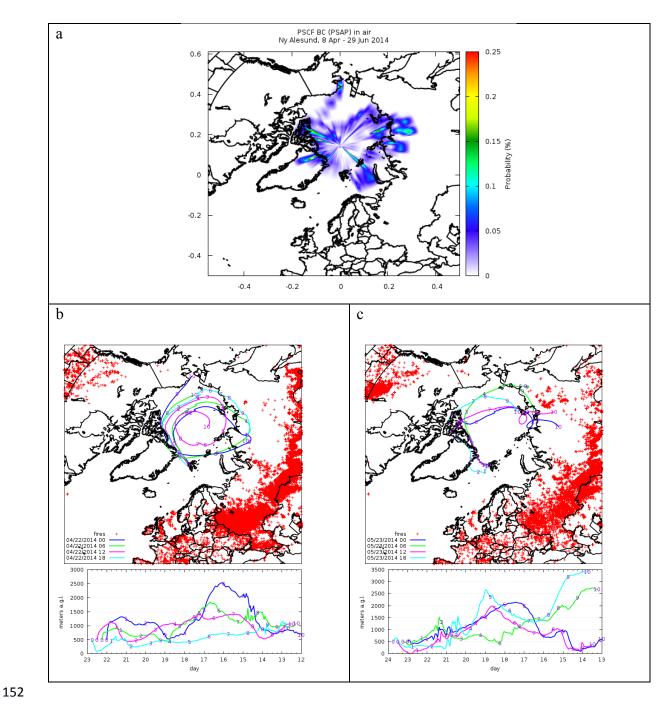


Figure S2. 10 days back-trajectory results for the 3-days experiment (three per day with a 8 hours interval). The red crosses represent the fires taking place during the last 10 days (data from the MODIS active fire products (https://firms. modaps.eosdis.nasa.gov/firemap/).

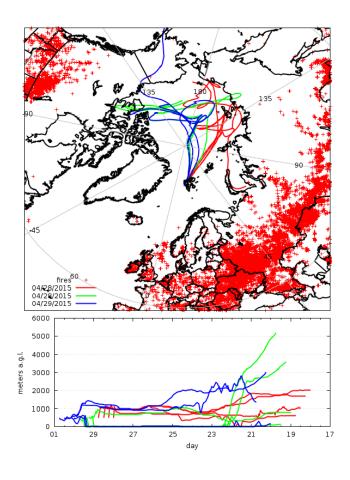


Figure S3. Results for the 80-days experiment from ancillary measurements. Upper panel: rBC mass concentration (gray line), Na concentration (red line) and conductivity (green line). Lower panel: atmospheric eBC mass concentration (black line) and ammonia as measured at the Zeppelin station, Svalbard (gray bars).

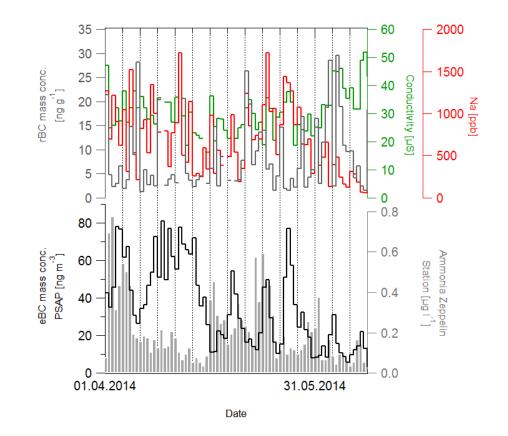
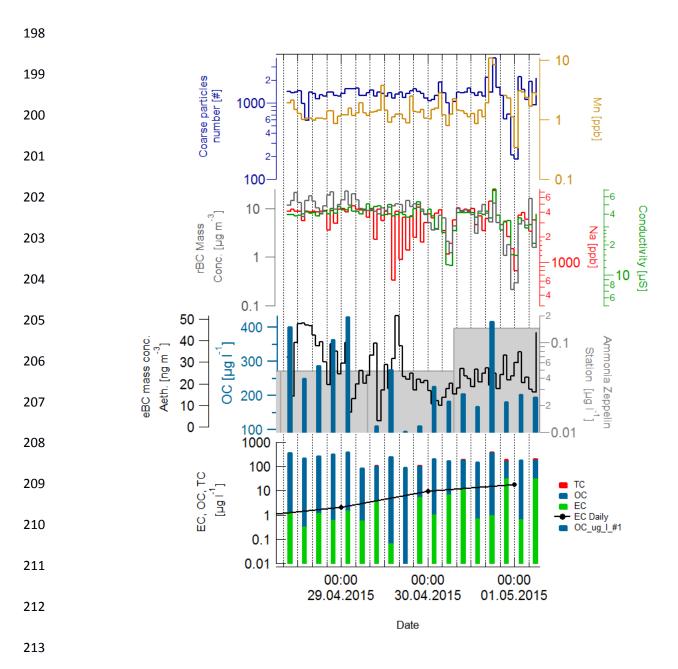


Figure S4. The 3-days experiments results from ancillary measurements. Uppermost panel: manganese (Mn, dark-yellow line) mass concentration and the coarse mode particles number (blu line). Second uppermost panel: rBC mass concentration (gray line) with sodium (Na) concentration (red line) and conductivity (green line). Second lowermost panel: atmospheric eBC mass concentration (black line), snow OC concentration (blue bars) and daily average of atmospheric ammonia as measured at the Zeppelin station (gray bars). Lowermost panel: TC (red bars), OC (blue bars) and EC (green bars) and EC daily average (black line).



216 References:

- Kaspari, S. D., Schwikowski, M., Gysel, M., Flanner, M. G., Kang, S., Hou, S. and
- Mayewski, P. A.: Recent increase in black carbon concentrations from a Mt. Everest ice
- core spanning 1860–2000 AD, Geophysical Research Letters, 38(4),
- doi:10.1029/2010GL046096, 2011.
- Kupiszewski, P., Zanatta, M., Mertes, S., Vochezer, P., Lloyd, G., Schneider, J., Schenk, L.,
- Schnaiter, M., Baltensperger, U., Weingartner, E. and Gysel, M.: Ice residual properties in
- 223 mixed-phase clouds at the high-alpine Jungfraujoch site, Journal of Geophysical Research:
- 224 Atmospheres, 121(20), 12,343-12,362, doi:10.1002/2016JD024894, 2016.
- Moteki, N., Adachi, K., Ohata, S., Yoshida, A., Harigaya, T., Koike, M. and Kondo, Y.:
- Anthropogenic iron oxide aerosols enhance atmospheric heating, Nature Communications,
- 8, 15329, doi:10.1038/ncomms15329, 2017.
- Sigl, M., Abram, N. J., Gabrieli, J., Jenk, T. M., Osmont, D. and Schwikowski, M.: 19th
- century glacier retreat in the Alps preceded the emergence of industrial black carbon
- deposition on high-alpine glaciers, The Cryosphere, 12(10), 3311–3331,
- doi:https://doi.org/10.5194/tc-12-3311-2018, 2018.
- R Core Team: R: A language and environment for statistical computing. Vienna, Austria: R
- Foundation for Statistical Computing; 2020. URL https://www.R-project.org