1    Supplement of

3    **Composition and sources of carbonaceous aerosol in**

4        **the European Arctic at Zeppelin Observatory,**

5                        **Svalbard**

Table S1: Equations used for source apportionment of carbonaceous aerosol, following the Latin Hypercube Sampling (LHS) approach. See sect. 2.8 and Yttri et al. (2011a) for more details.

| | |
|---|---|
| $EC = [EC] \times \Phi_{EC}$ | (eq.1) |
| $OC = [OC_p] + \Phi_{NA} \times ([OC_F] - [OC_B])$ | (eq.2) |
| $TC = OC + EC$ | (eq.3) |
| $F_{14C\text{-}TC} = [F_{14C\text{-}TC}] \times \Phi^{14}C\text{-}TC$ | (eq.4) |
| $F_{14C\text{-}EC} = [F_{14C\text{-}EC}] \times \Phi^{14}C\text{-}EC$ | (eq.5) |
| $F_{14C\text{-}OC} = (F_{14C\text{-}TC} - F_{14C\text{-}EC} \times EC/TC) / (1 - EC/TC)$ | (eq.6) |
| $F_{EC\text{-}NF} = F_{14C\text{-}EC} / F_{14C\text{-}BB}$ | (eq.7) |
| $EC_{BB} = F_{EC\text{-}NF} \times EC$ | (eq.8) |
| $OC_{BB} = EC_{BB} \times (OC/EC)_{BB}$ | (eq.9) |
| $EC_{FF} = [EC] - [EC_{BB}]$ | (eq.10) |
| $OC_{PBC} = [Cellulose] \times (OC / Cellulose)_{PBC}$ | (eq.11) |
| $OC_{PBS} = [Mannitol] \times (OC / Mannitol)_{PBS}$ | (eq.12) |
| $OC_{BSOA} = \underline{F_{14C\text{-}OC} \times OC - (F_{14C\text{-}BB} \times OC_{BB} + F_{14C\text{-}PBC} \times OC_{PBC} \times OC_{PBS})}$ | (eq.13) |
| $\qquad\qquad\qquad\qquad F^{14}C\text{-}BSOA$ | |

Notes:
Square brackets denote measured quantities.
Equation (6) is derived from:

$TC \times F_{14C\text{-}TC} \qquad = OC \times F_{14C\text{-}OC} \; 0 \; EC \times F_{14C\text{-}EC}$

$\quad F_{14C\text{-}OC} \qquad\qquad = (TC \times F_{14C\text{-}TC} \; 1 - EC \times F_{14C\text{-}EC})/OC = (TC \times F_{14C\text{-}TC} - EC \times F_{14C\text{-}EC})/(TC\text{-}EC) \; = F_{14C\text{-}TC} - EC/TC \times F_{14C\text{-}EC})/(1 - EC/TC)$

Table S2. Emission ratios used for the LHS source apportionment approach of carbonaceous aerosol. Low, central, and high values are listed. See sect. 2.8, Yttri et al. (2011a) for more details.

| Parameter | Low | Central | High | Eqn. | Accounts for |
|-----------|-----|---------|------|------|--------------|
| $\Phi_{EC}$ | 0.75 | 1.0 | 1.25 | 1 | Uncertainty in EC measurement |
| $\Phi_{NA}$ | 0.0 | 0.2 | 1.0 | 2 | Negative sampling artefact of OC |
| $\Phi^{14}C\text{-}TC$ | 0.95 | 1.0 | 1.05 | 4 | Uncertainty in $^{14}C\text{-}TC_{Measurement}$ |
| $\Phi^{14}C\text{-}EC$ | 0.85 | 1.0 | 1.15 | 5 | in $\Phi14C-EC_{Measurement}$ |
| $\Phi_{F^{14}C}$ | 1.055 | - | 1.25 | 7 | Variability in modern carbon age of BB; Notation includes RWC, WF  and AWB |
| $(OC/EC)_{BB}$ | 3.3 | 6.2 | 9.1 | 9 | Wildfires. Derived from Saarikoski et al., (2007); Yttri et al. (2014). |
| $(OC/Cellulose)_{PBC}$ | 0.8 | 1.6 | 3.2 | 11 | $OC_{PBC}$ calculation |
| $(OC/Mannitol)_{PBS}$ | 5.2 | - | 10.8 | 12 | $OC_{PBS}$ calculation |
| $F_{14CBSOA}$ | | 1.055 | | 13 | Modern carbon |
| $F_{14CPBS}$ | 1.055 | - | 1.25 | 13 | Fungal Spores |
| $F_{14CPBC}$ | 1.055 | - | 1.25 | 13 | Plant debris |

**Table S3:** Source apportionment of CA by LHS for samples collected at the Zeppelin Observatory (2017 to 2018). Mean (10th, 50 th, 90th percentiles) are provided. Unit: ng C m$^{-3}$.

| Start date | End date | OC$_{BSOA}$ | OC$_{FF}$ | OC$_{BB}$ | OC$_{PBC}$ | OC$_{PBS}$ | EC$_{FF}$ | EC$_{BB}$ |
|---|---|---|---|---|---|---|---|---|
| 23.02.2017 | 02.03.2017 | 33 (4.4-27-70) | 92 (67-93-115) | 137 (107-132-175) | 5.7 (3.0-5.1-9.0) | 0.1 (0.1-0.1-0.1) | 7.7 (3.3-8.0-12) | 32 (28-32-37) |
| 05.05.2017 | 15.05.2017 | 55 (9.3–49-113) | 5.7 (0.8-4.9-12) | 97 (63-98-128) | 0.7 (0.4-0.6-1.1) | 0.0 (0.0-0.0-0.0) | 9.0 (6.7-9.0-11) | 15 (13-15-17) |
| 31.05.2017 | 26.06.2017 | 64 (37–58-98) | 25 (18-24-34) | 24 (15-24-34) | 3.4 (1.8-3-5.4) | 0.41 (0.29-0.41-0.52) | 2.1 (1.4-2.1-2.7) | 3.9 (3.3-3.9-4.6) |
| 08.09.2017 | 28.09.2017 | 52 (27-47-83) | 10 (5.7-10-16) | 26 (16-25-36) | 2.9 (1.6-2.6-4.7) | 1.2 (0.84-1.2-1.5) | 1.9 (1.2-1.9-2.5) | 4.1 (3.5-4.1-4.8) |
| 28.09.2017 | 06.10.2017 | 422 (241-384-657) | 27 (4.9-24-51) | 194 (119-195-266) | 13 (7-12-21) | 8.9 (6.4-8.9-11) | 22 (17-22-26) | 30 (26-30-35) |
| 06.10.2017 | 24.10.2017 | 18 (3.6-16-37) | 20 (13-19-27) | 33 (22-31-46) | 1.4 (0.7-1.3-2.3) | 0.9 (0.6-0.9-1.1) | 2.0 (1.1-2.1-2.9) | 6 (5.1-5.9-6.9) |
| 05.12.2017 | 13.12.2017 | N/A | N/A | N/A | N/A | N/A | N/A | N/A |
| 23.01.2018 | 31.01.2018 | 73 (48-67-106) | 74 (56-67-102) | 21 (13-21-29) | 1.3 (0.7-1.2-2.1) | 0.1 (0.1-0.1-0.1) | 13 (12-13-13) | 3.4 (2.9-3.4-3.9) |
| 21.03.2018 | 29.03.2018 | 50 (12-46-94) | 14 (5.5-13-23) | 62 (39-62-87) | 0.7 (0.4-0.6-1.1) | 0.1 (0.1-0.1-0.1) | 7.6 (5.9-7.6-9.1) | 10 (8.9-10-12) |
| 06.04.2018 | 16.04.2018 | 66 (24-62-114) | 13- (5.1-13-22) | 58 (36-57-81) | 0.4 (0.2-0.3-0.6) | 0.1 (0.0-0.1-0.1) | 7.6 (6.1-7.6-9) | 9.4 (8-9.4-11) |
| 12.07.2018 | 30.07.2018 | 30 (17-27-47) | 18 (13-17-24) | 13 (7.7-12-18) | 1.4 (0.7-1.2-2.2) | 1.4 (1.0-1.4-1.7) | 1.0 (0.6-1.0-1.3) | 2 (1.7-2-2.4) |
| 30.07.2018 | 15.08.2018 | 192 (130-174-280) | 18 (6.8-17-30) | 42 (25-41-59) | 4.8 (2.5-4.3-7.7) | 2.1 (1.5-2.2-2.7) | 2.3 (1.2-2.2-3.3) | 6.7 (5.7-6.7-7.8) |
| 23.11.2018 | 03.12.2018 | 10 (1.5-7.9-21) | 23 (15-24-30) | 33 (25-31-42) | 1.9 (1-1.7-3.1) | 0.2 (0.2-0.2-0.3) | 5.6 (4.4-5.7-6.6) | 7.4 (6.4-7.3-8.6) |

**Table S4:** Source apportionment of CA by LHS for samples collected at the Zeppelin Observatory (2017 to 2018). Mean (10th, 50 th, 90th percentiles) percentiles are provided. Unit: %

| Start date | End date | $OC_{BSOA}$/TC | $OC_{FF}$/TC | $OC_{BB}$/TC | $OC_{PBC}$/TC | $OC_{PBS}$/TC | $EC_{FF}$/TC | $EC_{BB}$/TC | $EC_{FF}$/EC | $EC_{BB}$/EC |
|---|---|---|---|---|---|---|---|---|---|---|
| 23.02.2017 | 02.03.2017 | 11 (1.7-9.1-21) | 30 (25-30-34) | 45 (36-45-53) | 1.9 (1.0-1.8-2.9) | 0.0 (0.0-0.0-0.0) | 2.6 (1.0-2.6-4.2) | 11 (8.7-10-13) | 19 (8.3-20-29) | 81 (71-80-92) |
| 05.05.2017 | 15.05.2017 | 28 (6-28-52) | 3.1 (0.4-2.7-6.6) | 54 (34-55-73) | 0.4 (0.2-0.3-0.6) | 0.0 (0.0-0.0-0.0) | 5.1 (3.3-5.1-7.1) | 8.5 (6.2-8.5-11) | 38 (53-38-72) | 62 (28-62-47) |
| 31.05.2017 | 29.06.2017 | 51 (37-52-63) | 20 (16-20-24) | 21 (12-20-31) | 2.9 (1.4-2.6-4.8) | 0.3 (0.2-0.3-0.5) | 1.8 (1.1-1.7-2.5) | 3.3 (2.3-3.4-4.3) | 34 (24-35-44) | 66 (56-65-76) |
| 08.09.2017 | 28.09.2017 | 51 (34-53-67) | 11 (6.2-11-15) | 27 (15-26-41) | 3.1 (1.6-2.8-5.2) | 1.2 (0.8-1.2-1.7) | 2.0 (1.1-1.9-2.9) | 4.4 (3.1-4.4-5.7) | 31 (20-31-42) | 69 (58-69-80) |
| 28.09.2017 | 06.10.2017 | 57 (41-58-72) | 3.7 (0.7-3.5-7.0) | 28 (16-28-42) | 1.9 (1.0-1.7-3.7) | 1.3 (0.8-1.2-1.8) | 3.2 (2.1-3.1-4.3) | 4.4 (3.1-4.4-5.6) | 42 (33-42-51) | 58 (49-58-67) |
| 06.10.2017 | 24.10.2017 | 22 (5-21-39) | 24 (19-24-29) | 41 (27-41-55) | 1.8 (0.9-1.7-3.0) | 1.1 (0.71-1.1-1.6) | 2.6 (1.3-2.6-4.1) | 1.9 (1.0-1.7-3.7) | 25 (14-26-36) | 75 (64-74-86) |
| 05.12.2017 | 13.12.2017 | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A |
| 23.01.2018 | 31.01.2018 | 38 (32-39-45) | 40 (37-40-43) | 12 (7-11-18) | 0.7 (0.4-0.7-1.2) | 0.0 (0.0-0.0-0.1) | 7.1 (5.1-7.4-8.7) | 7.6 (5.6-7.5-10) | 79 (75-79-82) | 21 (18-21-25) |
| 21.03.2018 | 29.03.2018 | 33 (9-34-55) | 9.3 (4.1-9.3-14) | 44 (26-43-64) | 0.5 (0.2-0.4-0.8) | 0.1 (0.1-0.1-0.1) | 5.4 (3.7-5.4-7.3) | 7.4 (5.4-7.5-9.5) | 42 (33-42-51) | 58 (49-58-67) |
| 06.04.2018 | 16.04.2018 | 41 (18-43-61) | 8.5 (3.5-8.5-13) | 39 (22-38-58) | 0.2 (0.1-0.2-0.4) | 0.0 (0.0-0.0-0.1) | 5.1 (3.5-5.1-6.7) | 6.3 (4.5-6.38.1) | 45 (36-45-53) | 55 (47-55-64) |
| 12.07.2018 | 30.07.2018 | 44 (31-45-56) | 27 (23-27-30) | 20 (11-19-30) | 2.2 (1.1-1.9-3.6) | 2.1 (1.4-2.1-3.0) | 1.5 (0.9-1.5-2.2) | 3.2 (2.2-3.2-4.1) | 32 (21-3.2-42) | 68 (58-68-79) |
| 30.07.2018 | 15.08.2018 | 71 (60-72-81) | 6.8 (2.7-6.7-11) | 16 (9-15-24) | 1.9 (0.9-1.7-3.2) | 0.8 (0.5-0.8-1.2) | 0.9 (0.9-1.4-2.0) | 2.6 (1.8-2.6-3.4) | 25 (13-26-37) | 75 (63-74-87) |
| 23.11.2018 | 03.12.2018 | 11 (2.1-10-23) | 28 (23-28-33) | 41 (31-41-49) | 2.5 (1.3-2.3-3.8) | 0.3 (0.2-0.3-0.4) | 7.2 (5.0-7.0-9.9) | 9.4 (7.4-9.2-12) | 43 (34-44-51) | 57 (59-56-66) |

**Table S5:** Annual and seasonal mean concentrations of OC, EC, and organic tracers at Birkenes Observatory (Norway) and at Ispra (Italy), 2017 to 2020.

| | OC (µg C m⁻³) | EC (µg C m⁻³) | Levoglucosan (ng m⁻³) | Mannosan (ng m⁻³) | Galactosan (ng m⁻³) | Arabitol (ng m⁻³) | Mannitol (ng m⁻³) | Glucose (ng m⁻³) | Trehalose (ng m⁻³) | 2-methylerythritol (ng m⁻³) | 2-methylerythritol (ng m⁻³) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| *Birkenes* | | | | | | | | | | | |
| **2017** | **0.72** | **0.05** | **8.24** | **1.35** | **0.32** | **5.52** | **5.70** | **5.17** | **3.37** | **0.37** | **0.10** |
| DJF | 0.57 | 0.07 | 15.77 | 2.49 | 0.65 | 1.16 | 1.40 | 2.03 | 1.03 | 0.01 | 0.01 |
| MAM | 0.65 | 0.05 | 7.62 | 0.98 | 0.23 | 3.90 | 3.65 | 4.11 | 1.58 | 0.05 | 0.02 |
| JJA | 0.92 | 0.03 | 2.37 | 0.48 | 0.06 | 9.15 | 8.45 | 7.09 | 4.21 | 1.32 | 0.35 |
| SON | 0.77 | 0.06 | 7.92 | 1.31 | 0.29 | 8.18 | 9.49 | 7.58 | 6.61 | 0.17 | 0.06 |
| **2018** | **0.96** | **0.08** | **9.77** | **1.62** | **0.39** | **5.76** | **5.65** | **4.16** | **2.83** | **0.45** | **0.16** |
| DJF | 0.49 | 0.07 | 13.50 | 2.21 | 0.60 | 0.72 | 0.94 | 2.74 | 0.60 | 0.01 | 0.01 |
| MAM | 1.32 | 0.11 | 13.64 | 2.04 | 0.54 | 4.18 | 4.21 | 3.59 | 1.91 | 0.24 | 0.08 |
| JJA | 1.20 | 0.05 | 1.38 | 0.25 | 0.04 | 8.66 | 7.94 | 4.52 | 2.90 | 1.24 | 0.42 |
| SON | 0.81 | 0.08 | 10.64 | 2.01 | 0.41 | 9.38 | 9.47 | 5.79 | 5.91 | 0.25 | 0.12 |
| **2019** | **0.93** | **0.08** | **8.30** | **1.37** | **0.29** | | | | | | |
| DJF | 0.55 | 0.09 | 16.8 | 2.73 | 0.61 | | | | | | |
| MAM | 1.44 | 0.12 | 8.36 | 1.20 | 0.26 | | | | | | |
| JJA | 1.13 | 0.05 | 1.32 | 0.27 | 0.04 | | | | | | |
| SON | 0.57 | 0.05 | 6.91 | 1.29 | 0.23 | | | | | | |
| **2020** | **0.82** | **0.08** | **7.73** | **1.31** | **0.33** | | | | | | |
| DJF | 0.31 | 0.06 | 7.73 | 1.29 | 0.33 | | | | | | |
| MAM | 0.96 | 0.10 | 10.7 | 1.69 | 0.42 | | | | | | |
| JJA | 1.34 | 0.06 | 1.62 | 0.32 | 0.04 | | | | | | |
| SON | 0.70 | 0.09 | 10.61 | 1.88 | 0.50 | | | | | | |
| Mean ±SD | | | | | | | | | | | |
| **Annual** | **0.86±0.11** | **0.07±0.01** | **8.51±0.88** | **1.41±0.14** | **0.33±0.04** | **5.64±0.17** | **5.68±0.04** | **4.67±0.71** | **3.10±0.38** | **0.41±0.06** | **0.13±0.04** |
| DJF | 0.48±0.12 | 0.07±0.01 | 13.45±4.05 | 2.18±0.63 | 0.55±0.15 | 0.94±0.31 | 1.17±0.33 | 2.39±0.50 | 0.82±0.30 | 0.01±0.00 | 0.01±0.00 |
| MAM | 1.09±0.36 | 0.10±0.03 | 10.08±2.71 | 1.48±0.48 | 0.36±0.14 | 4.04±0.20 | 3.93±0.40 | 3.85±0.37 | 1.75±023 | 0.15±0.13 | 0.05±0.04 |
| JJA | 1.15±0.18 | 0.05±0.01 | 1.67±0.48 | 0.33±0.10 | 0.05±0.01 | 8.91±0.35 | 8.20±0.36 | 5.81±1.82 | 3.56±0.93 | 1.28±0.06 | 0.39±0.05 |
| SON | 0.71±0.11 | 0.07±0.02 | 9.02±1.90 | 1.62±0.38 | 0.36±0.12 | 8.78±0.85 | 9.48±0.01 | 6.69±1.27 | 6.26±0.49 | 0.21±0.06 | 0.09±0.04 |
| H-S | 0.75±0.18 | 0.08±0.02 | 11.90±1.79 | 1.91±0.30 | 0.48±0.08 | 2.50±0.11 | 2.55±0.14 | 3.94±0.75 | 1.39±0.10 | 0.09±0.04 | 0.03±0.02 |
| NH-S | 1.02±0.09 | 0.06±0.01 | 3.70±0.52 | 0.70±0.09 | 0.12±0.02 | 10.11±0.12 | 10.14±0.46 | 6.81±1.55 | 5.56±1.17 | 0.86±0.04 | 0.28±0.06 |
| *Ispra* | | | | | | | | | | | |
| **2017** | **5.53** | **1.11** | | | | | | | | | |
| DJF | 10.5 | 2.19 | | | | | | | | | |
| MAM | 3.37 | 0.66 | | | | | | | | | |

| | OC (µg C m⁻³) | EC (µg C m⁻³) | Levoglucosan (ng m⁻³) | Mannosan (ng m⁻³) | Galactosan (ng m⁻³) | Arabitol (ng m⁻³) | Mannitol (ng m⁻³) | Glucose (ng m⁻³) | Trehalose (ng m⁻³) | 2-methylerythritol (ng m⁻³) | 2-methylerythritol (ng m⁻³) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| JJA | 2.67 | 0.37 | | | | | | | | | |
| SON | 5.62 | 1.23 | | | | | | | | | |
| **2018** | **3.75** | **0.77** | | | | | | | | | |
| DJF | 8.30 | 1.59 | | | | | | | | | |
| MAM | 2.50 | 0.52 | | | | | | | | | |
| JJA | 1.65 | 0.31 | | | | | | | | | |
| SON | 2.86 | 0.73 | | | | | | | | | |
| **2019** | **3.82** | **0.80** | | | | | | | | | |
| DJF | 7.74 | 1.68 | | | | | | | | | |
| MAM | 2.58 | 0.57 | | | | | | | | | |
| JJA | 2.46 | 0.28 | | | | | | | | | |
| SON | 2.40 | 0.63 | | | | | | | | | |
| **2020** | **4.33** | **0.85** | | | | | | | | | |
| DJF | 7.93 | 1.74 | | | | | | | | | |
| MAM | 3.06 | 0.47 | | | | | | | | | |
| JJA | 1.96 | 0.25 | | | | | | | | | |
| SON | 4.48 | 0.97 | | | | | | | | | |
| Mean ±SD | | | | | | | | | | | |
| **Annual** | 4.36±0.82 | 0.88±0.16 | | | | | | | | | |
| DJF | 8.62±1.28 | 1.80±0.27 | | | | | | | | | |
| MAM | 2.88±0.41 | 0.56±0.08 | | | | | | | | | |
| JJA | 2.19±0.46 | 0.30±0.05 | | | | | | | | | |
| SON | 3.84±1.48 | 0.89±0.27 | | | | | | | | | |
| H-S | 5.69±0.97 | 1.19±0.20 | | | | | | | | | |
| NH-S | 2.51±0.64 | 0.45±0.09 | | | | | | | | | |

1. OC, EC, and organic tracers for Birkenes 2017 and 2018 is taken from Yttri et al., (2021), whereas for data for 2019 and 2020 is extracted from ebas.nilu.
2. OC and EC for Ispra is extracted from ebas.nilu

**Table S6:** Monthly mean ratios of levoglucosan/mannosan, mannitol/arabitol and 2-methylthreitol/2-methylerythritol at Zeppelin Observatory (2017 to 2020) and Birkenes Observatory (2017 to 2018).

| | Levoglucosan/ Mannosan | | Mannitol/ Arabitol | | 2-methylthreitol/ 2-methylerythritol | |
|---|---|---|---|---|---|---|
| Mean (±SD) | Zeppelin Obs. | Birkenes Obs. | Zeppelin Obs. | Birkenes Obs. | Zeppelin Obs. | Birkenes Obs. |
| January | 7.9 ± 1.4 | 6.5 ± 0.1 | 0.7 ± 0.2 | 1.4 ± 0.8 | 0.72 ± 0.28 | 0.69 ± 0.00 |
| February | 9.0 ± 2.7 | 6.8 ± 0.6 | 1.0 ± 0.5 | 2.3 ± 1.0 | 0.51 ± 0.06 | 0.76 ± 0.10 |
| March | 7.9 ± 1.2 | 6.2 ± 0.3 | 1.2 ± 0.2 | 1.2 ± 0.2 | 0.59 ± 0.13 | 0.76 ± 0.07 |
| April | 7.4 ± 3.0 | 6.6 ± 0.0 | 1.9 ± 1.2 | 1.0 ± 0.1 | 0.61 ± 0.10 | 0.60 ± 0.00 |
| May | 7.2 ± 1.6 | 7.5 ± 1.0 | 1.6 ± 0.7 | 0.9 ± 0.1 | 0.65 ± 0.09 | 0.29 ± 0.05 |
| June | 5.3 ± 1.2 | 5.5 ± 0.2 | 1.3 ± 0.4 | 1.0 ± 0.2 | 0.64 ± 0.07 | 0.29 ± 0.03 |
| July | 4.1 ± 0.8 | 5.7 ± 0.6 | 1.0 ± 0.3 | 0.8 ± 0.2 | 0.54 ± 0.20 | 0.29 ± 0.28 |
| August | 4.2 ± 0.9 | 4.4 ± 0.2 | 1.1 ± 0.3 | 0.9 ± 0.1 | 0.52 ± 0.13 | 0.34 ± 0.00 |
| September | 5.0 ± 1.7 | 6.1 ± 1.4 | 1.1 ± 0.1 | 1.1 ± 0.2 | 0.48 ± 0.07 | 0.42 ± 0.01 |
| October | 5.3 ± 1.1 | 5.4 ± 0.2 | 1.2 ± 0.3 | 1.1 ± 0.0 | 0.54 ± 0.11 | 0.55 ± 0.10 |
| November | 5.7 ± 0.8 | 5.6 ± 0.5 | 1.2 ± 0.1 | 1.0 ± 0.1 | 0.65 ± 0.17 | 0.74 |
| December | 6.5 ± 1.8 | 5.4 ± 0.4 | 1.0 ± 0.5 | 0.8 ± 0.0 | 0.74 ± 0.16 | 0.77 ± 0.09 |
| **Annual** | **6.0 ± 1.8** | **6.1 ± 0.1** | **1.1 ± 0.5** | **1.0 ± 0.0** | **0.55 ± 0.16** | **0.32 ± 0.05** |
| DJF | 7.8 ± 1.9 | 6.3 ± 0.7 | 0.9 ± 0.4 | 1.5 ± 0.9 | 0.65 ± 0.13 | 0.74 ± 0.07 |
| MAM | 7.5 ± 2.1 | 6.7 ± 0.8 | 1.5 ± 0.8 | 1.1 ± 0.2 | 0.62 ± 0.03 | 0.55 ± 0.22 |
| JJA | 4.5 ± 1.2 | 5.2 ± 0.7 | 1.1 ±0.3 | 0.9 ± 0.1 | 0.56 ± 0.06 | 0.31± 0.05 |
| SON | 5.9 ± 1.1 | 5.7 ± 0.7 | 1.1 ± 0.2 | 1.1 ± 0.1 | 0.56 ± 0.09 | 0.54 ± 0.14 |
| NH-S | 4.8 ± 1.2 | 5.4 ± 0.8 | 1.1 ± 0.3 | 1.0 ± 0.1 | 0.54 ± 0.06 | 0.38 ± 0.11 |
| H-S | 7.5 ± 1.9 | 6.4 ± 0.8 | 1.2 ± 0.6 | 1.2 ± 0.6 | 0.64 ± 0.08 | 0.65 ± 0.18 |

Tale S7: Annual mean concentrations of Antarctic baseline aerosol (ABA)[1] OC, EC, and positive artifact corrected OC ($OC_P$) at Trollhaugen Observatory (Antarctica), 2016 to 2018 and 2021 to 2022. Unit ng C m$^{-3}$.

| | EC | OC | $OC_P$ |
|---|---|---|---|
| **2016** | 2.0 | 12.0 | 7.4 |
| **2017** | 2.3 | 14.1 | 10.9 |
| **2018** | 1.4 | 10.0 | 7.5 |
| **2021** | 2.0 | 14.2 | 9.3 |
| **2022** | 1.7 | 10.8 | 7.1 |
| **Mean ± SD** | **1.9 ± 0.3** | **12.2 ± 1.9** | **8.4 ± 1.6** |

Antarctic baseline aerosol (ABA): To differentiate between Antarctic baseline aerosol (ABA) and non-ABA, we relied on input from in-situ on-line nephelometer (TSI 3563) measurements, as defined by Fiebig et al. (2014). We considered values below the 4 weeks running 5th percentile x 2.5 of the aerosol scattering coefficient ($\sigma$sp, 550 nm) as ABA, while values above this threshold were defined as non-ABA. We made sure not to sample at wind speeds > 10 m s$^{-1}$ to avoid filter damage. We collected ABA and non-ABA using two high-volume samplers (Digitel DHA-80 with a PM$_{10}$ inlet) both with double quartz fibre filters (Pallflex 2500) (McDow and Huntzicker, 1990) to estimate of the positive sampling artifact caused by semi volatile organic compounds (SVOC). The inlet of the nephelometer and high-volume samplers were positioned 5.5 meters above the ground.

Table S8. Mean PBAP and BSOA tracer concentrations for those of the top ten concentration samples collected in the non-heating season and with a levoglucosan concentration higher than the long-term annual mean at Zeppelin Observatory (2017 to 2020).

| | Mean conc. top ten samples (ng m$^{-3}$) | Mean conc. excl. H-Season and with levo > levo long-term mean (ng m$^{-3}$) | Mean conc. and percentile levoglucosan (ng m$^{-3}$) (percentile) |
|---|---|---|---|
| Arabitol | 1.2 | 1.3 n=8) | 2.7 (96) |
| Mannitol | 1.0 | 1.1 (n=7) | 2.4 (95) |
| Trehalose | 1.0 | 1.0 (n=7) | 2.4 (95) |
| Fructose | 0.7 | 1.1 (n=5) | 3.6 (98) |
| Glucose | 3.5 | 3.5 (n=9) | 2.5 (95) |
| Cellulose | 5.7 | 6.4 (n=3) | 4.3 (98) |
| 2-Methyltetrols | 6.1 | 7.8 (n=7) | 2.2 (94) |
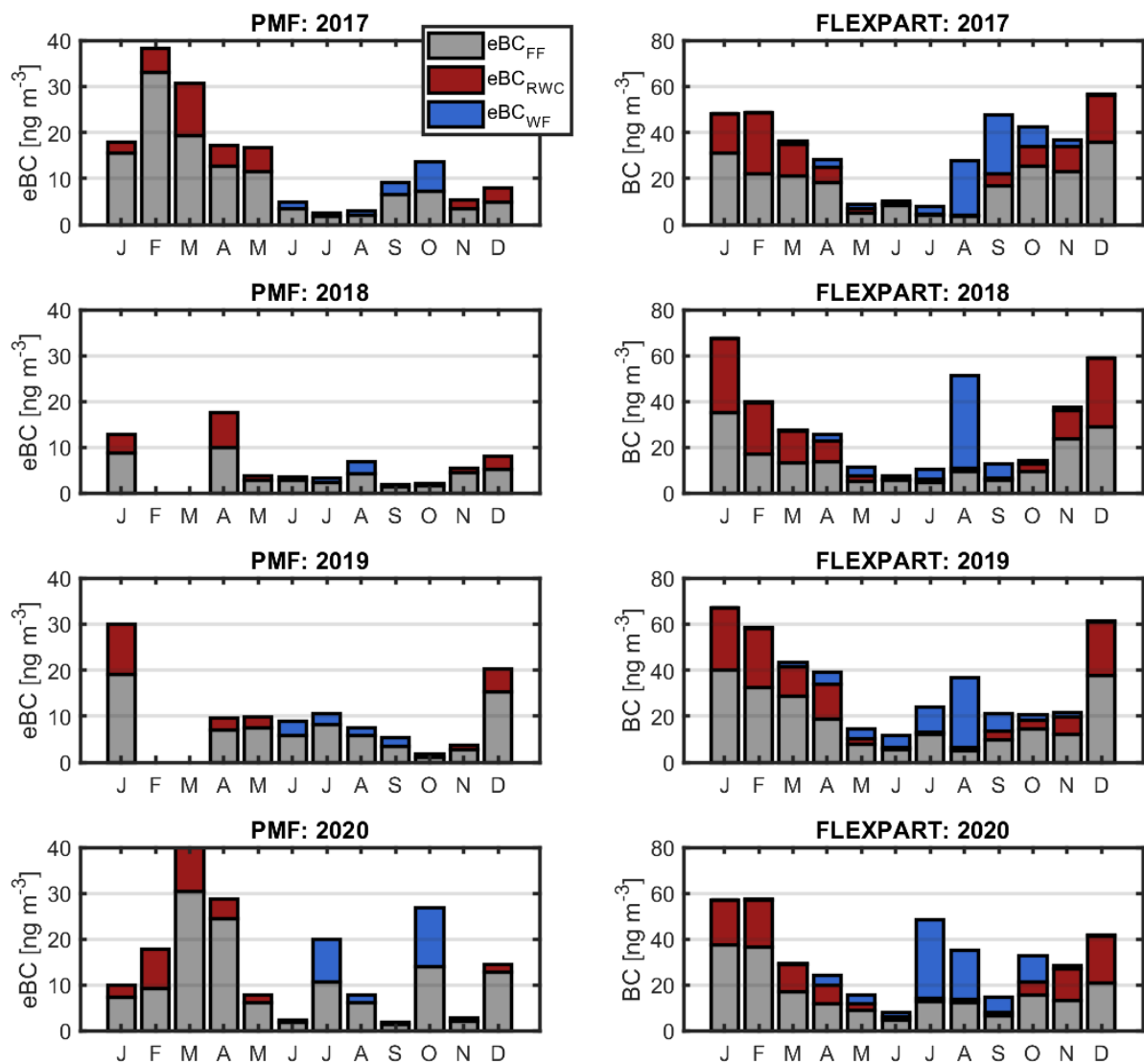
Figure S1. Monthly mean eBC concentrations (PMF) (left) and BC (FLEXPART) (right) apportioned to FF combustion and BB. BB is categorized as RWC in H-Season (red color) and WF in NH-season (blue color). Zeppelin Observatory 2017 to 2020.
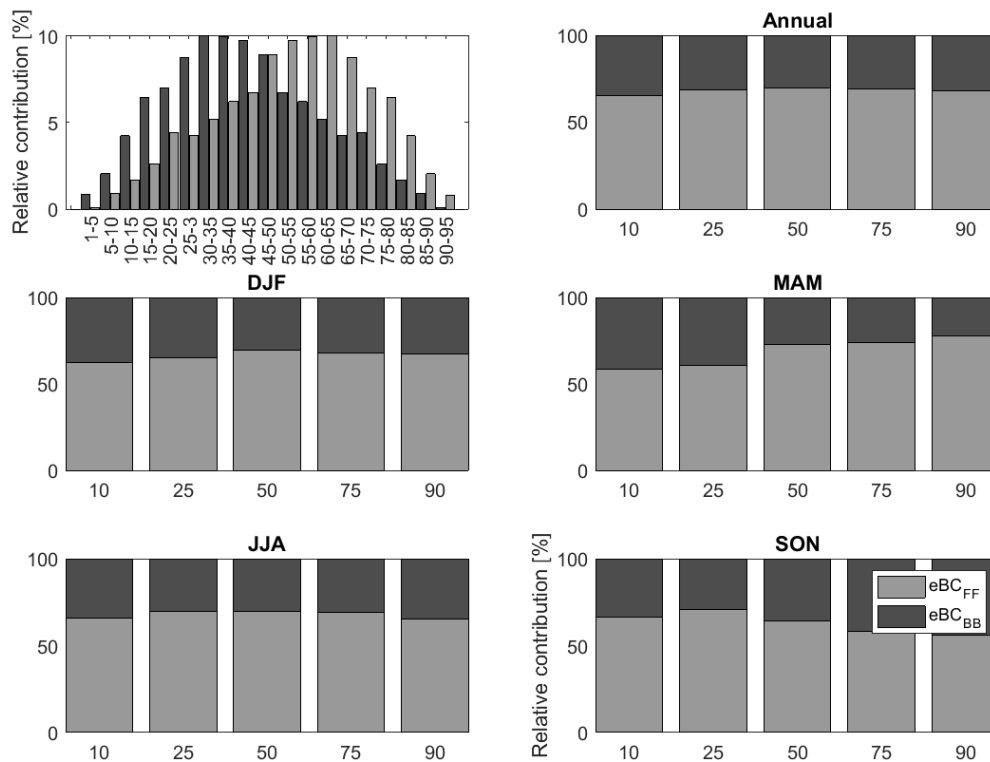
Figure S2. Frequency distribution of $eBC_{BB}/eBC$ and $eBC_{FF}/eBC$ fractions (left top panel), and average $eBC_{BB}/eBC$ and $eBC_{FF}/eBC$ fractions for eBC concentrations below the 10th and the 25th percentile, all observations (50th percentile), and above the 75th and 90th percentiles, for the Zeppelin Observatory, 2017 to 2020. Results obtained by PMF.
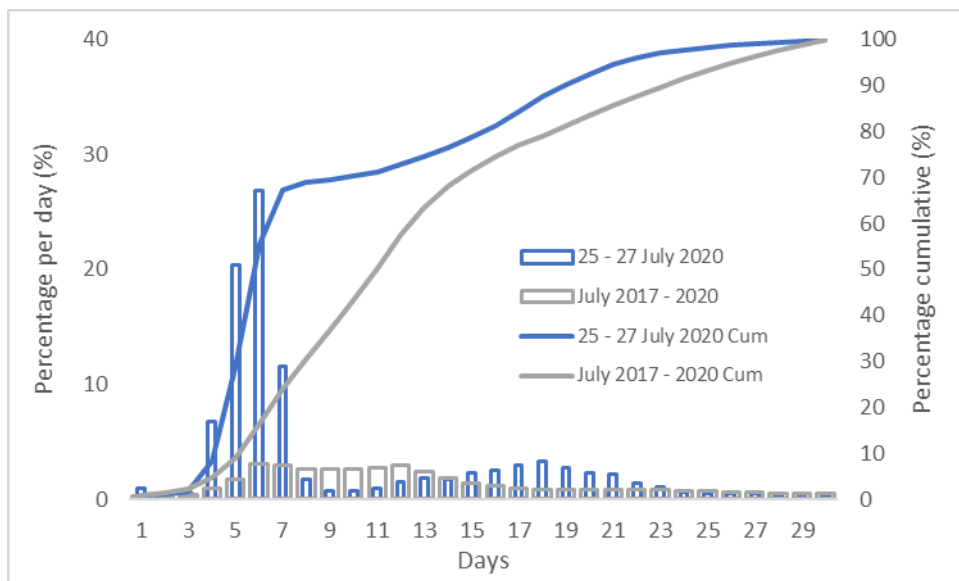
Figure S3. Transport time of modelled BC concentrations from the respective emission sources to Zeppelin Observatory. The blue line are the results for the part of the pollution episode spanning from 25 to 27 July 2020 compared to the reference period (July 2017 to 2020, grey line).