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Origin of elemental carbon in snow from western Siberia and northwestern European Russia during winter–spring 2014, 2015 and 2016

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1 Quality assurance of the EC measurements

Minerals, such as calcite ($CaCO_3$) and dolomite [$CaMg(CO_3)$], contain CO_3^{2-}- carbon, which can be detected by the thermal-optical measurement principle. Depending on the chemical speciation, CO_3^{2-} - carbon will evolve over a broad range of temperatures (Cavalli et al., 2010; Gunasekaran and Anbalagan, 2007), which coincides with the temperature range used in thermal-optical analysis (TOA). Whether CO_3^{2-} - carbon evolves as part of the organic carbon (OC) or the elemental carbon (EC) fraction, or both, depends on the protocol applied, but this question is still not fully explored even for the most commonly used protocols (NIOSH; IMPROVE; EUSAAR-2). Typically, acid fumigation has been used prior to analysis to eliminate and/or quantify the CO_3^{2-}- carbon contribution to the OC and/or EC signal (Chow et al., 1993). However, this approach can cause loss of volatile organic acids (Chow et al., 1993), induce severe charring (Jankowski et al., 2008), and importantly, the time of CO_3^{2-}- carbon evolving is defined prior to the analysis. Experience based on the thermal-oxidative pretreatment approach (Jankowski et al., 2008), and subsequent analysis by the EUSAAR-2 protocol (as used in the current study) of ambient aerosol filter samples from Eastern Europe, Caucasus and Central Asia (the EECAA countries) and from the Arabian Peninsula, suggests that CO_3^{2-} - carbon evolves both as OC and EC. A similar finding is made for the filtered snow samples in the current study. The results presented in Table S 2 indicate a minor influence of CO_3^{2-} - carbon evolving as EC for samples collected in 2015 and 2016, whereas it was a factor of 2 – 4 times higher for the samples collected in 2014. For a few samples though, CO_3^{2-} - carbon made a substantial (>50%) contribution to the EC signal.

The thermal-oxidative pre-treatment approach coupled with the EUSAAR-2 protocol applied in the current study, has been demonstrated to provide similar results as that of the acid fumigation approach, with respect to concentrations of CO_3^{2-} - carbon from calcite (Fagerli et al., 2015). This is successful in removing OC and EC, but not carbonate carbon, during the thermal-oxidative pre-treatment step.

All filter samples in the current data set had an EC level of <15 µg C cm^{-2}, which is considered the upper limit when analysing filter samples by TOA (Subramanian † et al., 2006; Wallén et al., 2010). The mean $EC_{CO_3^{2-}}^{corr} - TC_{CO_3^{2-}}^{corr}$ ratio of the filtered snow samples ranged from 0.031 – 0.091, which is in the lowest range of what has been observed for atmospheric aerosol particles in the Northern European rural background environment (Yttri et al., 2007). Albeit crude, EC is considered as a tracer of anthropogenic activity; however,

the very low $EC_{CO_3^{2-}}^{corr} - TC_{CO_3^{2-}}^{corr}$ ratio cannot be used as an argument of low anthropogenic influence, as there can be non-atmospheric sources contributing to $TC_{CO_3^{2-}}^{corr}$.

Table S 1. Information about the samples collected in springtime of 2014, 2015 and 2016 in Western Russia.

Latitude	Longitude	Day	Month	Year	Snow depth (m)	Surface sampled (m^2)	Snow Volume (L)	Water equivalent volume (L)
56.52	84.15	19	02	2014	0.05	0.56	28.13	30.00
57.10	83.90	19	02	2014	0.05	0.56	28.13	30.00
57.33	83.93	19	02	2014	0.05	0.56	28.13	30.00
58.07	82.82	19	02	2014	0.05	0.56	28.13	30.00
67.90	74.80	25	02	2014	0.05	0.56	28.13	30.00
67.93	75.08	25	02	2014	0.05	0.56	28.13	30.00
67.77	75.50	25	02	2014	0.05	0.56	28.13	30.00
67.62	75.90	25	02	2014	0.05	0.56	28.13	30.00
67.53	76.17	25	02	2014	0.05	0.56	28.13	30.00
67.40	76.35	25	02	2014	0.05	0.56	28.13	30.00
67.25	76.43	25	02	2014	0.05	0.56	28.13	30.00
66.80	76.40	25	02	2014	0.05	0.56	28.13	30.00
65.98	77.67	25	02	2014	0.05	0.56	28.13	30.00
65.78	78.17	25	02	2014	0.05	0.56	28.13	30.00
65.70	78.02	25	02	2014	0.05	0.56	28.13	30.00
63.80	75.55	26	02	2014	0.05	0.56	28.13	30.00
63.80	75.57	28	02	2014	0.05	0.56	28.13	30.00
65.38	77.75	26	02	2014	0.05	0.56	28.13	30.00
64.28	75.73	26	02	2014	0.05	0.56	28.13	30.00
63.80	75.57	28	02	2014	0.05	0.56	28.13	30.00
63.82	75.57	01	03	2014	0.05	0.56	28.13	30.00
61.48	74.25	03	03	2014	0.05	0.56	28.13	30.00
60.50	76.97	04	03	2014	0.05	0.56	28.13	30.00
65.08	41.11	16	03	2015	0.03	0.09	2.70	1.00
66.53	33.13	19	03	2015	0.14	0.09	12.60	3.00
66.53	33.13	19	03	2015	0.56	0.09	50.40	3.10

66.55	33.03	20	03	2015	0.14	0.09	12.60	1.00
66.53	33.03	21	03	2015	0.06	0.09	5.40	0.75
66.53	33.12	22	03	2015	0.10	0.09	9.00	0.50
66.53	33.08	23	03	2015	0.09	0.09	8.10	0.75
66.53	33.13	23	03	2015	0.04	0.09	3.15	0.95
66.52	33.08	24	03	2015	0.08	0.09	7.20	0.75
66.53	33.05	24	03	2015	0.07	0.09	6.30	1.00
66.55	33.08	25	03	2015	0.04	0.09	3.60	1.25
64.53	38.85	29	02	2016	0.05	0.09	4.50	0.25
65.08	41.11	29	02	2016	0.15	0.09	13.50	0.50
66.55	33.14	01	03	2016	0.05	0.09	4.50	0.25
66.54	33.05	02	03	2016	0.05	0.09	4.50	0.25
64.32	40.79	03	03	2016	0.28	0.09	25.20	0.25
64.32	40.78	03	03	2016	0.30	0.09	27.00	0.25
64.32	40.79	03	03	2016	0.17	0.09	15.30	0.50
64.32	40.79	03	03	2016	0.19	0.09	17.10	0.50
64.32	40.79	03	03	2016	0.15	0.09	13.50	0.50
66.55	33.10	04	03	2016	0.05	0.09	4.50	0.25
66.54	33.15	06	03	2016	0.05	0.09	4.50	0.25
72.26	68.82	09	04	2016	0.45	0.01	3.53	1.05
72.94	65.36	22	04	2016	0.80	0.01	6.28	2.35
78.45	70.89	23	04	2016	0.25	0.01	1.96	0.86
78.70	67.47	23	04	2016	0.45	0.01	3.53	1.57
69.93	71.78	28	04	2016	0.19	0.01	1.49	0.43
71.76	71.51	29	04	2016	0.09	0.01	0.71	0.19
71.44	71.09	29	04	2016	0.19	0.01	1.49	0.52
71.28	69.75	30	04	2016	0.09	0.01	0.71	0.41
69.81	69.84	01	05	2016	0.07	0.01	0.55	0.47

Table S 2. $EC_{CO_3^{2-}}^{corr}$ to EC ratio (Mean ± SD; Min - Max), showing overestimation of EC due to $EC_{CO_3^{2-}}$ in the filtered snow samples.

	2014	**2015**	**2016**
EC	66±107	87±36	63±45
(µg L^{-1})	4–476	49–153	8–178
$EC_{CO_3^{2-}}^{corr}$	40±50	83±37	56±39
(µg L^{-1})	3–219	46–152	7–161
TC	949±1258	955±428	1469±1278
(µg L^{-1})	50–5927	550–1827	226–4504
$TC_{CO_3^{2-}}^{corr}$	865±1146	949±428	1449±1263
(µg L^{-1})	48–5580	546–1864	211–4493
$EC_{CO_3^{2-}}^{corr}/TC_{CO_3^{2-}}^{corr}$	5.3±2.4	9.1±2.5	5.7±3.8
(%)	2–12	6–14	1–15
$EC_{CO_3^{2-}}^{corr}/EC$	77±16	94±6	90±6
(%)	31–100	77–99	76–100
CO_3^{2-}	84±191	6±6	20±32
(µg L^{-1})	0–753	2–21	1–119

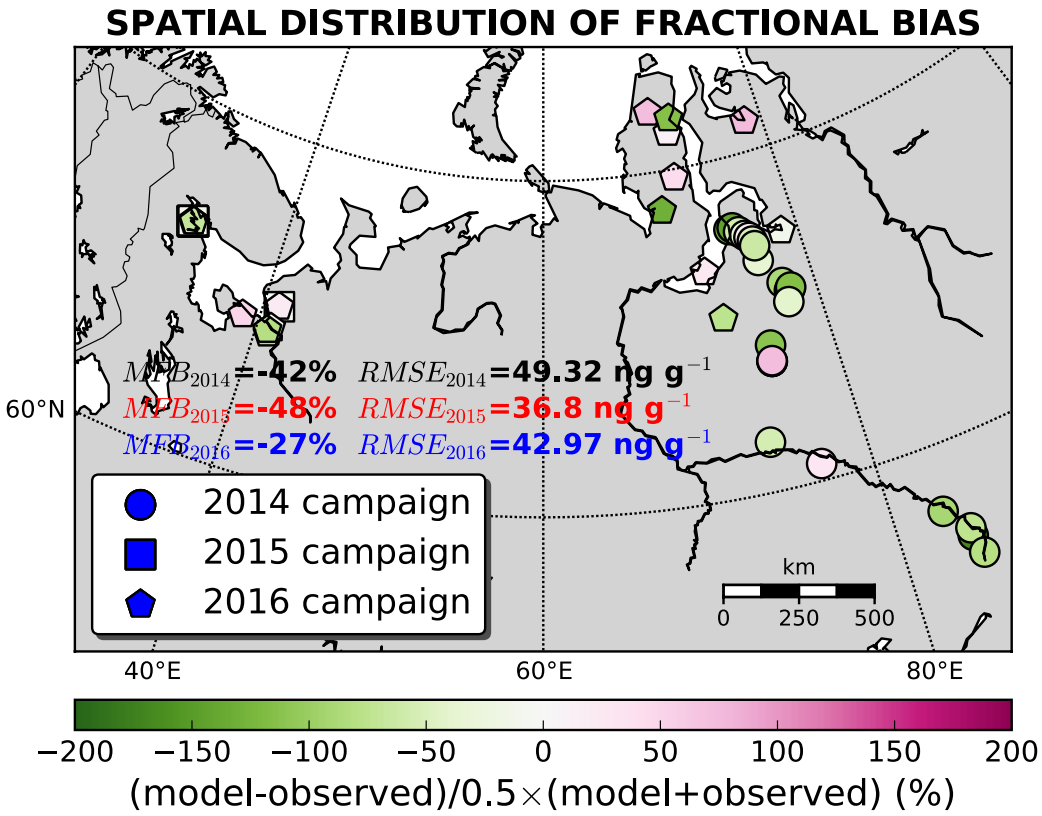

Figure S 1. Fractional bias ($FB = [(C_m - C_o)/(C_m + C_o) \times 0.5] \times 100\%$) for all samples collected from the three campaigns in Western Siberia and northwestern European Russia in 2014, 2015 and 2016. MFB (mean fractional bias) is the fractional bias averaged for all snow samples from 2014, 2015 and 2016, whereas RMSE is the root mean square error in ng g^{-1}).

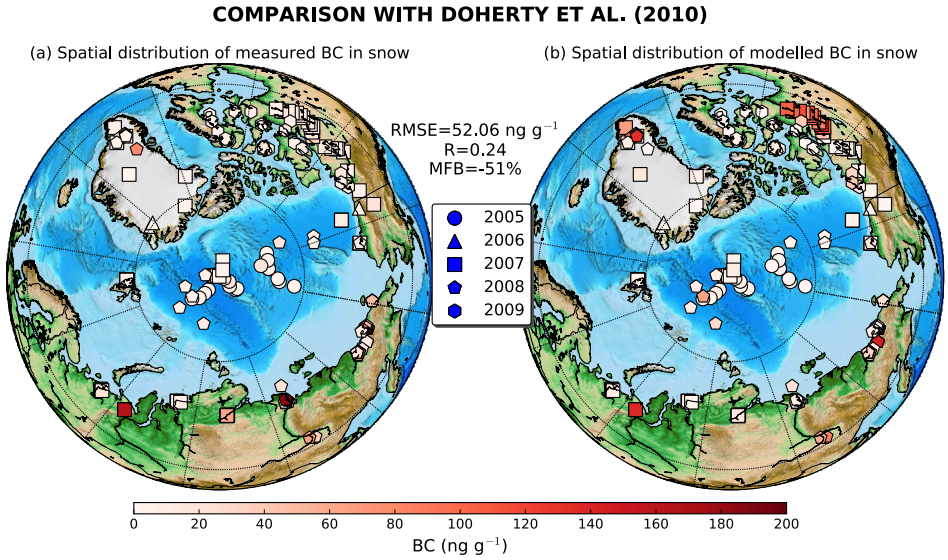

Figure S 2. (a) Distribution of snow measurements of BC adopted from Doherty et al. (2010) in the Arctic from 2005 to 2009. (b) Simulated (FLEXPART) BC concentrations in snow for the same period (right). MFB, RMSE and correlation coefficient (R) values are further given.

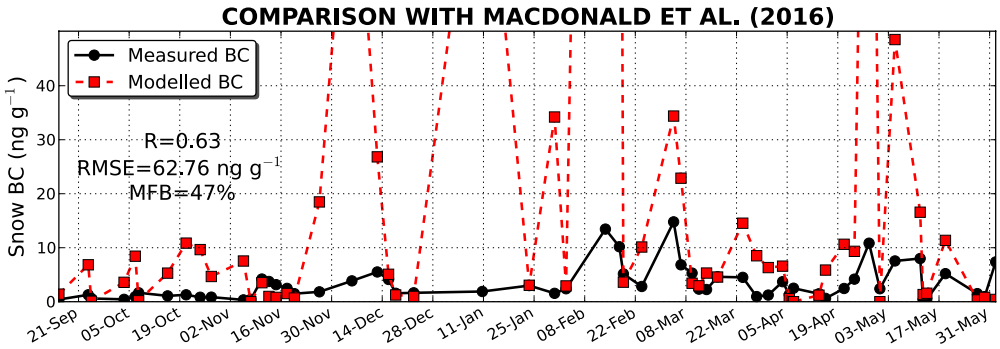

Figure S 3. Timeseries of simulated and measured BC concentrations in snow collected in Alert (Macdonald et al., 2017). Correlation coefficient (R) between modelled and measured BC, RMSE and MFB values are also shown.

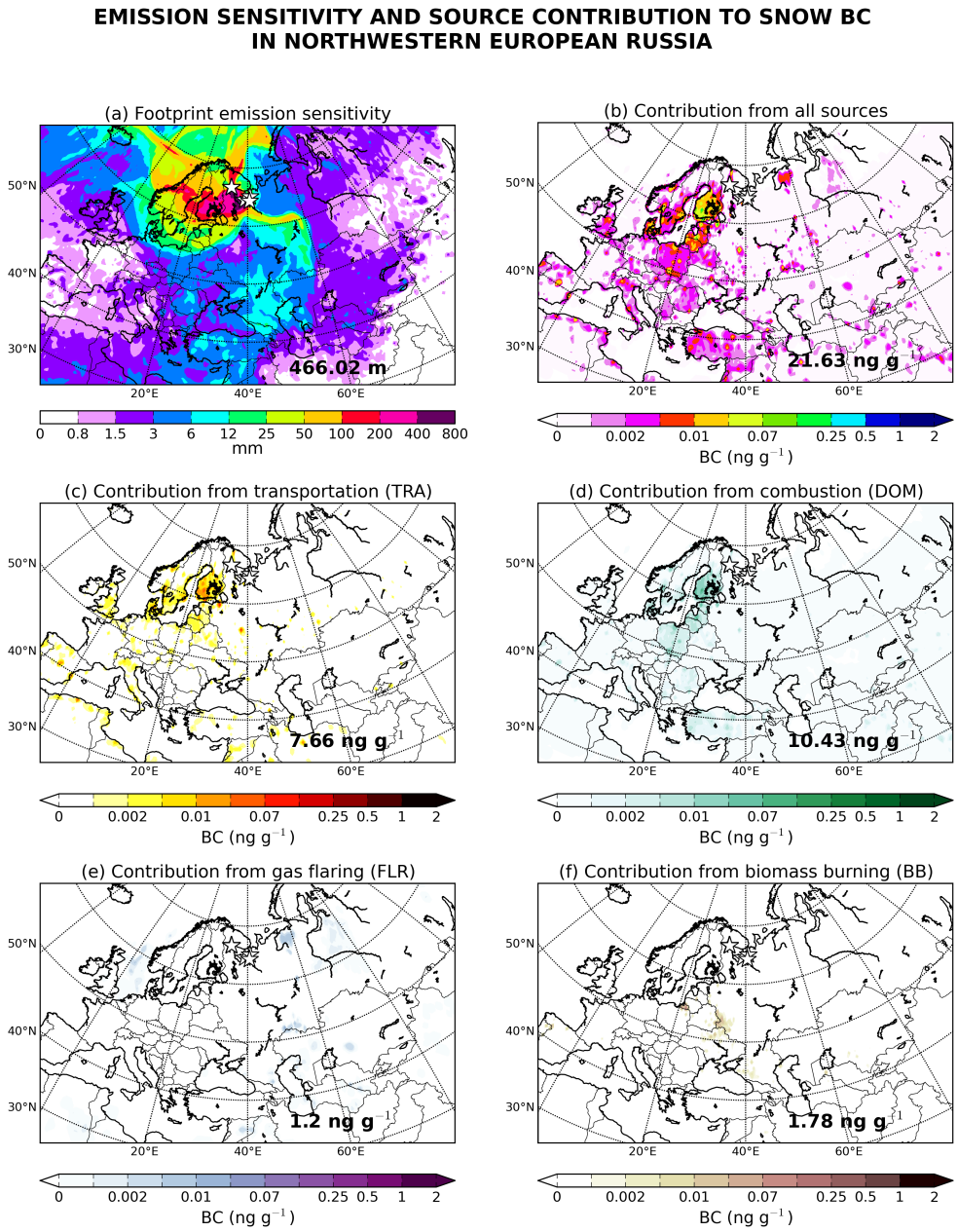

Figure S 4. (a) Average footprint emission sensitivity and (b–f) source contribution (from all sources, TRA, DOM, FLR and BB) for all the samples located in Northwestern European Russia.

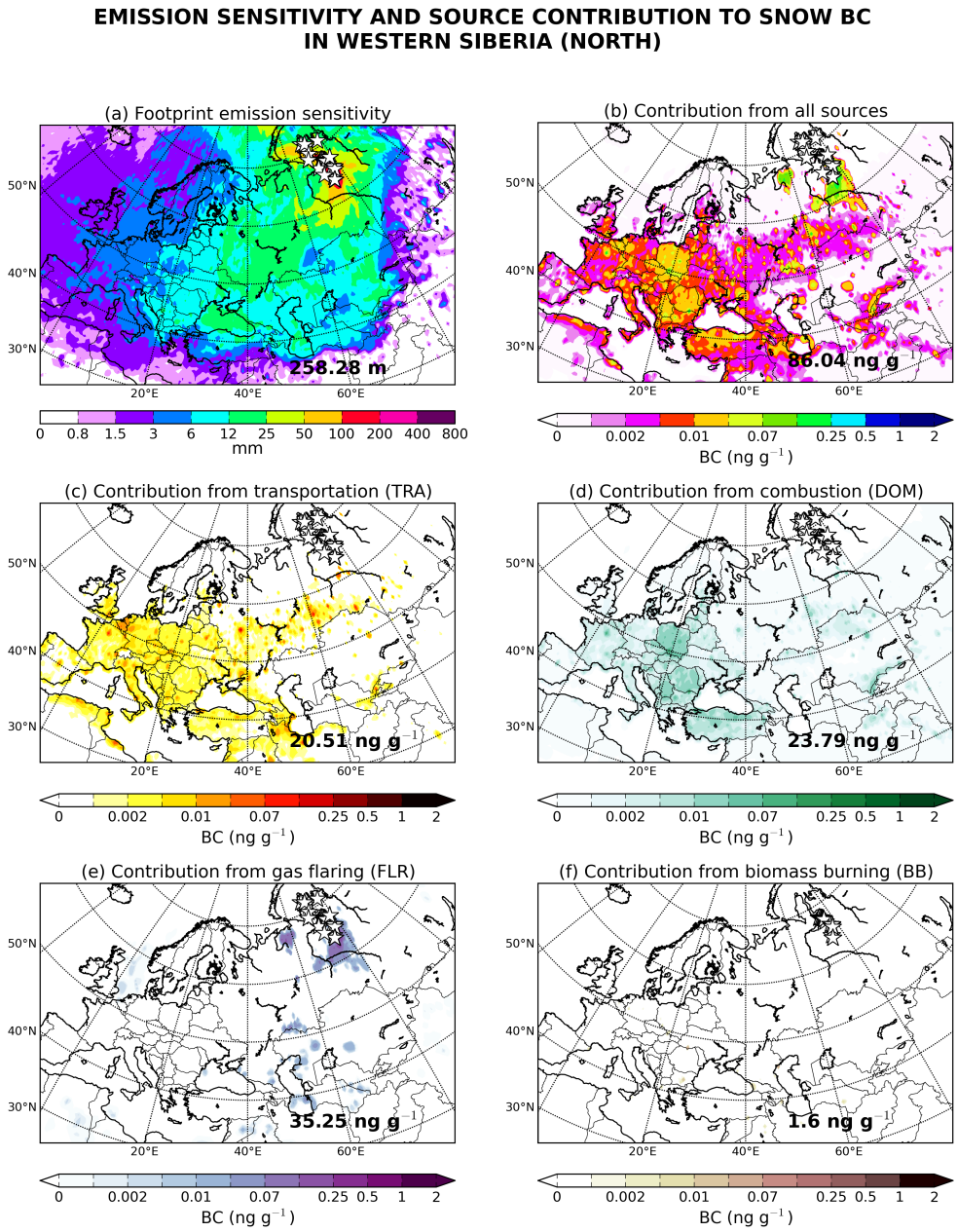

Figure S 5. (a) Average footprint emission sensitivity and (b–f) source contribution (from all sources, TRA, DOM, FLR and BB) for all the samples located in Western Siberia (north of 62 °N).

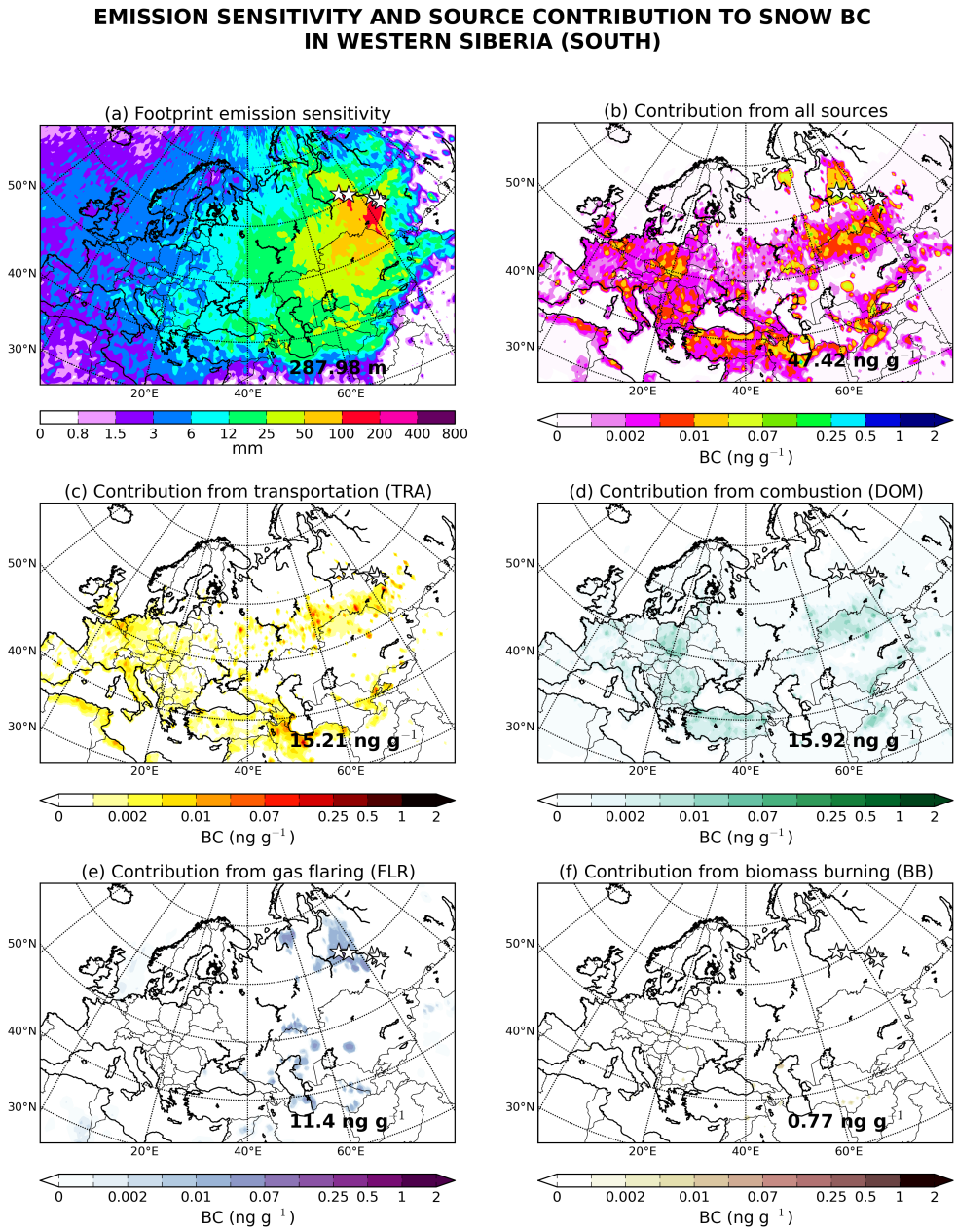

Figure S 6. (a) Average footprint emission sensitivity and (b–f) source contribution (from all sources, TRA, DOM, FLR and BB) for all the samples located in Western Siberia (south of 62 °N).

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