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Fossil fuel combustion and biomass burning sources of global black carbon from GEOS-Chem simulation and carbon isotope measurements

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Table S1. Carbon isotope analysis of BC sources (fossil fuel versus biomass burning) in the atmosphere

Pt	Region	Site	Lat	Lon	Alt (m)	Year	Mon	Season	f_{bb} (%)	BC isolation method	References
1	Arctic	Zeppelin	78.9	11.9	478	2009	Jan-Mar	winter	52±15[5]	NIOSH 5040[6]	Winiger et al., 2015
2	Arctic	Abisko	68.4	19.1	359	2011-13	Jan-Mar	winter	35±10	NIOSH 5040	Winiger et al., 2016
3	Arctic	Abisko	68.4	19.1	359	2011-13	Apr-Aug	summer	58±15	NIOSH 5040	Winiger et al., 2016
4	Arctic	Barrow	71.2	-156.6	11	2012-13	Dec-Feb	winter	32±9	NIOSH 5040	Barrett et al., 2015
5	Arctic	Barrow	71.2	-156.6	11	2012-13	Feb_Mar	winter	51±6	NIOSH 5040	Barrett et al., 2015
6	Arctic	Barrow	71.2	-156.6	11	2013	Mar-May	spring	18±7	NIOSH 5040	Winiger et al., 2019
7	Arctic	Barrow	71.2	-156.6	11	2013	Jul-Aug	summer	34±5	NIOSH 5040	Winiger et al., 2019
8	Arctic	Barrow	71.2	-156.6	11	2013	Sep-Nov	fall	9±5	NIOSH 5040	Winiger et al., 2019
9	Arctic	Barrow	71.2	-156.6	11	2013	Dec-Feb	winter	34±9	NIOSH 5040	Winiger et al., 2019
10	Arctic	Alert	82.3	-62.3	210	2014-15	Feb	winter	39±5	NIOSH 5040	Winiger et al., 2019
12	Arctic	Alert	82.3	-62.3	210	2014-15	Mar	spring	39±5	NIOSH 5040	Winiger et al., 2019
13	Arctic	Alert	82.3	-62.3	210	2014	May	spring	39±5	NIOSH 5040	Winiger et al., 2019
11	Arctic	Alert	82.3	-62.3	210	2014	Jul	summer	37±5	NIOSH 5040	Winiger et al., 2019
14	Arctic	Alert	82.3	-62.3	210	2014	Nov	fall	40±5	NIOSH 5040	Winiger et al., 2019
15	Arctic	Alert	82.3	-62.3	210	2014	Dec	winter	44±5	NIOSH 5040	Winiger et al., 2019
16	Arctic	Tiksi	71.4	128.5	11	2012-14	Mar-May	spring	25±0.2	NIOSH 5040	Winiger et al., 2017
17	Arctic	Tiksi	71.4	128.5	11	2012-14	Jun-Aug	summer	45±0.1	NIOSH 5040	Winiger et al., 2017
18	Arctic	Tiksi	71.4	128.5	11	2012-14	Sep-Nov	fall	48±0.1	NIOSH 5040	Winiger et al., 2017
19	Arctic	Tiksi	71.4	128.5	11	2012-14	Dec-Feb	winter	28±0.2	NIOSH 5040	Winiger et al., 2017
20	South Asia	MCOH[1]	6.8	73.3	15	2006	Jan_Mar	winter	68±6	NIOSH 5040	Gustafsson et al., 2009
21	South Asia	MCOH	6.8	73.3	15	2008-09	Dec_Mar	winter	53±5	NIOSH 5040	Budhavant et al., 2015
22	South Asia	MCOH	6.8	73.3	15	2008-09	Mar_Nov	summer	53±11	NIOSH 5040	Budhavant et al., 2015
23	South Asia	SINH[2]	18.3	73.7	1450	2006	Mar_Apr	spring	46±8	NIOSH 5040	Gustafsson et al., 2009
24	South Asia	SINH	18.3	73.7	1450	2008-09	Dec_Mar	winter	56±3	NIOSH 5040	Budhavant et al., 2015
25	South Asia	SINH	18.3	73.7	1450	2008-09	Mar_Nov	summer	48±8	NIOSH 5040	Budhavant et al., 2015
26	South Asia	Delhi	28.5	77.2	300	2011	Dec-Feb	witner	39	NIOSH 5040	Bikkina et al., 2019
27	South Asia	Delhi	28.5	77.2	300	2011	Mar-May	spring	24	NIOSH 5040	Bikkina et al., 2019
28	South Asia	Delhi	28.5	77.2	300	2011	Jun-Aug	summer	17	NIOSH 5040	Bikkina et al., 2019
29	South Asia	Delhi	28.5	77.2	300	2011	Sep-Nov	fall	31	NIOSH 5040	Bikkina et al., 2019
30	Europe	Göteborg	57.7	11.9	20	2005	Feb	winter	12±4	THEODORE[7]	Szidat et al., 2009
31	Europe	Göteborg	57.7	11.9	20	2006	Jun	summer	12±3	THEODORE	Szidat et al., 2009
32	Europe	Råö	57.3	11.9	10	2005	Feb	winter	38±5	THEODORE	Szidat et al., 2009
33	Europe	Zurich	47.3	8.5	410	2002	Aug	summer	8±1	THEODORE	Szidat et al., 2004
34	Europe	Zurich	47.3	8.5	410	2003	Feb	winter	29±5	THEODORE	Szidat et al., 2006

35	Europe	Zurich	47.3	8.5	410	2003	Mar	spring	15±5	THEODORE	Szidat et al., 2006
36	Europe	Zurich	47.3	8.5	410	2006	Jan	winter	29±4	THEODORE	Sandradewi et al., 2008a
37	Europe	Dübendorf	47.4	8.6	440	2007	Oct	fall	36±3	Swiss_4S[8]	Zhang et al., 2012
38	Europe	Roveredo	46.2	9.1	298	2005	Jan	winter	60±6	THEODORE	Szidat et al., 2007
39	Europe	Roveredo	46.2	9.1	298	2005	Mar	spring	58±6	THEODORE	Szidat et al., 2007
40	Europe	Roveredo	46.2	9.1	298	2005	Dec	winter	74±10	THEODORE	Sandradewi et al., 2008b
41	Europe	Roveredo	46.2	9.1	370	07/08-	Dec-Feb	winter	46	Swiss_4S	Zotter et al., 2014
42	Europe	Moleno	46.3	8.9	254	2005	Feb	winter	17±7	THEODORE	Szidat et al., 2007
43	Europe	Moleno	46.3	8.99	305	07/08-	Dec-Feb	winter	28	Swiss_4S	Zotter et al., 2014
44	Europe	Reiden	47.2	7.9	457	2006	Feb	winter	30±4	THEODORE	Sandradewi et al., 2008a
45	Europe	Reiden	47.2	7.9	510	07/08-	Dec-Feb	winter	34	Swiss_4S	Zotter et al., 2014
46	Europe	Massongex	46.2	6.1	400	2006	Nov	fall	36±4	THEODORE	Perron et al., 2010
47	Europe	Massongex	46.2	6.1	400	2006	Dec	winter	36±4	THEODORE	Perron et al., 2010
48	Europe	Massongex	46.2	6.1	452	08/09- 11/12	Dec-Feb	winter	54	Swiss_4S	Zotter et al., 2014
49	Europe	Saxon	46.1	7.1	460	2006	Dec	winter	32±4	THEODORE	Perron et al., 2010
50	Europe	Sion	46.2	7.3	505	2006	Dec	winter	20±3	THEODORE	Perron et al., 2010
51	Europe	Brigerbad	46.3	7.9	650	2006	Dec	winter	31±4	THEODORE	Perron et al., 2010
52	Europe	Payerne	46.8	6.9	456	2006	Jan	winter	60±4	Swiss_4S	Zhang et al., 2012
53	Europe	Payerne	46.8	6.9	456	2006	Jun	summer	44±3	Swiss_4S	Zhang et al., 2012
54	Europe	Payerne	46.8	6.9	539	07/08- 11/12	Dec-Feb	winter	51	Swiss_4S	Zotter et al., 2014
55	Europe	Barcelona	41.3	2.1	80	2009	Mar	spring	15±3	adapted THEODORE	Minguillón et al., 2011
56	Europe	Barcelona	41.3	2.1	80	2009	Jul	summer	9±4	adapted THEODORE	Minguillón et al., 2011
57	Europe	Montseny	41.8	2.3	720	2009	Mar	spring	37±4	adapted THEODORE	Minguillón et al., 2011
58	Europe	Montseny	41.8	2.3	720	2009	Jul	summer	23±5	adapted THEODORE	Minguillón et al., 2011
59	Europe	Bern-Bollwerk	46.9	7.6	506	08/09- 12/13	Dec-Feb	winter	22	Swiss_4S	Zotter et al., 2014
60	Europe	Sissach-West	47.5	7.8	410	07/08- 11/12	Dec-Feb	winter	43	Swiss_4S	Zotter et al., 2014
61	Europe	St.Gallen-Rorschacherstr asse	47.4	9.4	457	07/08- 11/12	Dec-Feb	winter	38	Swiss_4S	Zotter et al., 2014
62	Europe	Vaduz-Austrasse	47.1	9.5	706	07/08- 11/12	Dec-Feb	winter	45	Swiss_4S	Zotter et al., 2014
63	Europe	Zôrich-Kaserne	47.3	8.5	457	07/08-	Dec-Feb	winter	41	Swiss_4S	Zotter et al., 2014

						11/12						
64	Europe	Basel-St.Johann	47.6	7.6	308	07/08-08/09	Dec-Feb	winter	41	Swiss_4S	Zotter et al., 2014
65	Europe	Solothurn-Altwyberhôsli	47.1	7.6	502	07/08-11/12	Dec-Feb	winter	46	Swiss_4S	Zotter et al., 2014
66	Europe	Sch_chental	46.8	8.8	995	10/11-	Dec-Feb	winter	67	Swiss_4S	Zotter et al., 2014
67	Europe	Chiasso	45.8	9	291	07/08-11/12	Dec-Feb	winter	41	Swiss_4S	Zotter et al., 2014
68	Europe	Magadino-Cadenazzo	46.8	6.9	254	07/08-11/12	Dec-Feb	winter	48	Swiss_4S	Zotter et al., 2014
69	Europe	San-Vittore	46.2	9.1	330	07/08-11/12	Dec-Feb	winter	66	Swiss_4S	Zotter et al., 2014
70	North America	Salt Lake City	40.7	-111.8	1426	2012-14	annual	summer	11±1.1	adapted Swiss_4S	Mouteva et al., 2017
71	North America	Mexico City	19.5	-99.1	2240	2006	Mar	spring	16±4	THEODORE	Aiken et al., 2010
72	East Asia	Tokyo	35.6	139.6	40	2004	Oct	fall	36.4	adapted IMPROVE[9]	Yamamoto et al., 2007
73	East Asia	Tokyo	35.6	139.6	40	2004	Dec	winter	33.8	adapted IMPROVE	Yamamoto et al., 2007
74	East Asia	Tokyo	35.6	139.6	40	2004	Feb	winter	32.6	adapted IMPROVE	Yamamoto et al., 2007
75	East Asia	Tokyo	35.6	139.6	40	2004	Apr	spring	41.3	adapted IMPROVE	Yamamoto et al., 2007
76	East Asia	Tokyo	35.6	139.6	40	2004	Jun	summer	37.7	adapted IMPROVE	Yamamoto et al., 2007
77	East Asia	Tokyo	35.6	139.6	40	2004	Aug	summer	35.8	adapted IMPROVE	Yamamoto et al., 2007
79	East Asia	Beijing	39.9	116.4	55	2013	Jan	winter	30±2	Swiss_4S	Zhang et al., 2015
80	East Asia	Beijing	39.9	116.4	55	2013	Jan	winter	26±2	NIOSH 5040	Andersson et al., 2015
81	East Asia	Beijing	39.9	116.4	55	2010	Feb	winter	17±4	NIOSH 5040	Chen et al., 2013
82	East Asia	Shanghai	31.3	121.5	4	2013	Jan	winter	21±2	Swiss_4S	Zhang et al., 2015
83	East Asia	Shanghai	31.3	121.5	4	2013	Jan	winter	32±2	NIOSH 5040	Andersson et al., 2015
84	East Asia	Shanghai	31.3	121.5	4	2010	Jan	winter	17±4	NIOSH 5040	Chen et al., 2013
85	East Asia	Guangzhou	23.1	113.4	15	2013	Jan	winter	48±5	Swiss_4S	Zhang et al., 2015
86	East Asia	Guangzhou	23.1	113.4	15	2013	Jan	winter	32±2	NIOSH 5040	Andersson et al., 2015
89	East Asia	Xi'an	34.2	108.9	416	2013	Jan	winter	25±3		Zhang et al., 2015
90	East Asia	Xiamen	24.5	118	2	2009	Dec	winter	13±3	NIOSH 5040	Chen et al., 2013
93	East Asia	KCOG[3]	33.3	126.2	72	2011	Mar	winter	25±6	NIOSH 5040	Chen et al., 2013
94	East Asia	SCCO[4]	24.6	118.1	3	2009	Jan	winter	22±3	NIOSH 5040	Chen et al., 2013
95	Tibet	Jilong	28.2	86	4166	2013	Apr	spring	45	NIOSH 5040	Li et al., 2016
96	Tibet	Jilong	28.2	86	4166	2013	Jun	winter	41	NIOSH 5040	Li et al., 2016
97	Tibet	Nielamu	28.2	86	4166	2013	Nov	fall	40	NIOSH 5040	Li et al., 2016
98	Tibet	Dhunche	28.1	85.3	2051	2014	Jan	winter	49	NIOSH 5040	Li et al., 2016
99	Tibet	Dhunche	28.1	85.3	2051	2013	Aug	summer	16	NIOSH 5040	Li et al., 2016

100	Tibet	Dhunche	28.1	85.3	2051	2013	Sep	fall	41	NIOSH 5040	Li et al., 2016
101	Tibet	Bode	27.7	85.4	1386	2014	Jan	winter	42	NIOSH 5040	Li et al., 2016
102	Tibet	Bode	27.7	85.4	1386	2013	Apr	spring	33	NIOSH 5040	Li et al., 2016
103	Tibet	Bode	27.7	85.4	1386	2013	Aug	summer	16	NIOSH 5040	Li et al., 2016
104	Tibet	Bode	27.7	85.4	1386	2013	Nov	fall	28	NIOSH 5040	Li et al., 2016
105	Tibet	Zhongba	29.7	84	4704	2013	Apr	spring	70	NIOSH 5040	Li et al., 2016
106	Tibet	Jomsom	28.8	83.7	3048	2013	Apr	spring	57	NIOSH 5040	Li et al., 2016
107	Tibet	Pokhara	28.2	84	813	2013	Jul	summer	26	NIOSH 5040	Li et al., 2016
108	Tibet	Pokhara	28.2	84	813	2013	Apr	spring	65	NIOSH 5040	Li et al., 2016
109	Tibet	Lumbini	27.5	83.3	100	2013	Apr	spring	58	NIOSH 5040	Li et al., 2016
110	Tibet	Lumbini	27.5	83.3	100	2013	Jul	summer	42	NIOSH 5040	Li et al., 2016
111	Tibet	Lumbini	27.5	83.3	100	2013	Oct	fall	53	NIOSH 5040	Li et al., 2016
112	Tibet	Lumbini	27.5	83.3	100	2013	Dec	winter	49	NIOSH 5040	Li et al., 2016
113	Tibet	Namco	30.8	91	4730	2013	Apr	spring	54	NIOSH 5040	Li et al., 2016
114	Tibet	Namco	30.8	91	4730	2014	Jun	summer	63	NIOSH 5040	Li et al., 2016
115	Tibet	Namco	30.8	91	4730	2014	Jul	summer	49	NIOSH 5040	Li et al., 2016
116	Tibet	Namco	30.8	91	4730	2013	Nov	fall	58	NIOSH 5040	Li et al., 2016
117	Tibet	Lulang	29.8	94.7	3326	2014	Jun	summer	20	NIOSH 5040	Li et al., 2016
118	Tibet	Lulang	29.8	94.7	3326	2014	Jul	summer	23	NIOSH 5040	Li et al., 2016
119	Tibet	Lhasa	29.6	91	3640	2014	Jan	winter	18	NIOSH 5040	Li et al., 2016
120	Tibet	Lhasa	29.6	91	3640	2013	Apr	spring	24	NIOSH 5040	Li et al., 2016
121	Tibet	Lhasa	29.6	91	3640	2013	Jun	summer	7	NIOSH 5040	Li et al., 2016

[1] Maldives Climate Observatory in Hanimaadhoo

[2] Indian Institute of Tropical Meteorology in Sinhagad, India

[3] Korea Climate Observatory-Gosan

[4] South China Climate Observatory

[5] Standard deviation of observations

[6] National Institute for Occupational Safety and Health 5040

[7] Two-step Heating system for the EC/OC Determination of Radiocarbon in the Environment

[8] four-step (S1, S2, S3 and S4) thermal-optical protocol

[9] Integragency Monitoring of Protected Visual Environments

Table S2 Observed and GEOS-Chem simulated atmospheric f_{bb} in various regions (%)

Region	Observations	Simulation
The Himalayan-Tibetan plateau	39±17*	62±7
South Asia	37±16	55±5
East Asia	29±9	31±5
The Arctic	33±14	32±23
North America	14±4	29±2
Europe	43±16	14±3

*Standard deviation, reflecting variations of atmospheric f_{bb} among different sites during different seasons in each region.

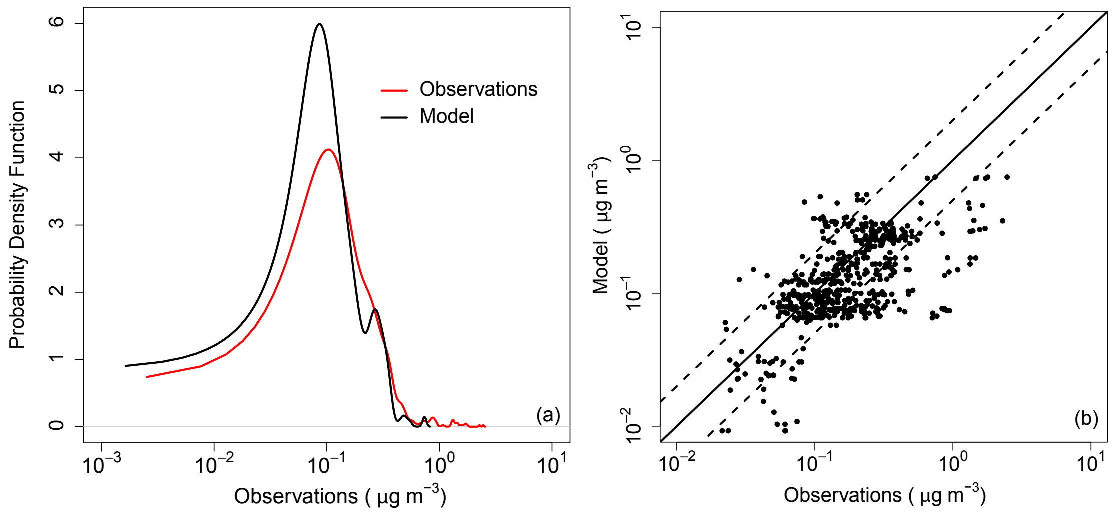

Figure S1. (a) Probability density function of observed (red line) and GEOS-Chem simulated (black) BC concentrations in surface air (μg m^{-3}) and (b) Observed and GEOS-Chem simulated annual BC concentrations in surface air. Data are for 2007–2013. Solid line is 1:1 ratio line and dashed lines are 1:2 (or 2:1).

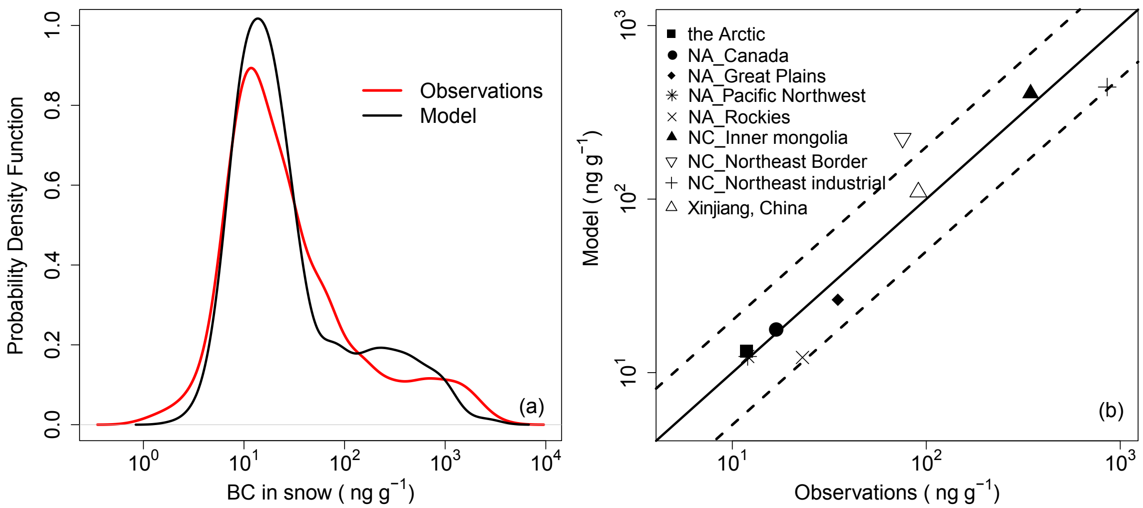

Figure S2. (a) Probability density function of observed (red line) and GEOS-Chem simulated (black) BC concentration in snow (ng g^{-1}) and (b) medians of observed and simulated BC in snow (ng g^{-1}) in the Arctic, North America (Canada, the Great Plains, the Pacific Northwest, and the Rockies, as defined in Doherty et al., 2014)), Northern China (Inner Mongolia, Northeast Border and Northeast Industrial, as defined by Wang et al., 2013), and Xinjiang, China. The regions are symbol-coded. Solid line – 1:1 ratio line; dashed lines – 1:2 (or 2:1) ratio lines.

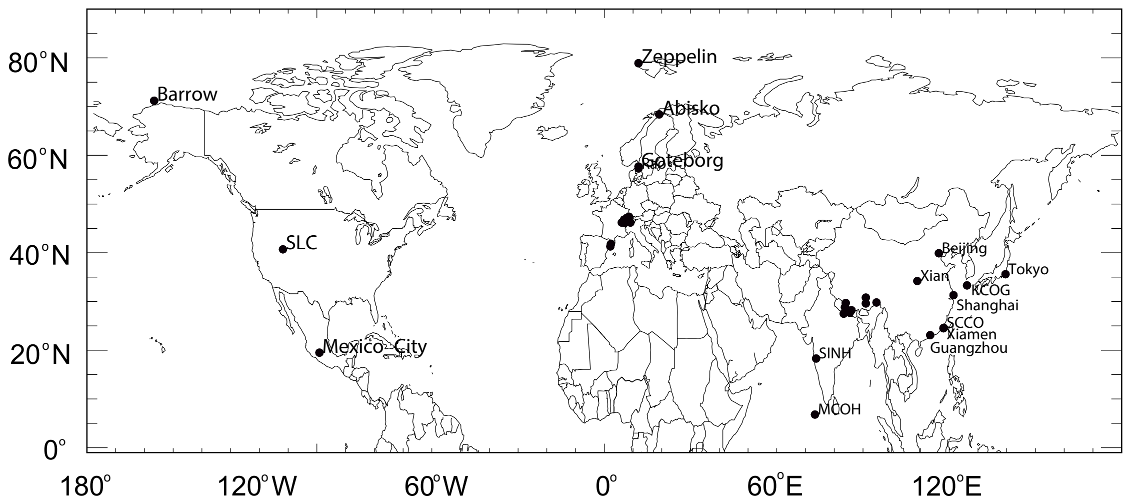

Figure S3. Carbon isotope measurement stations of BC as listed in Table S1.

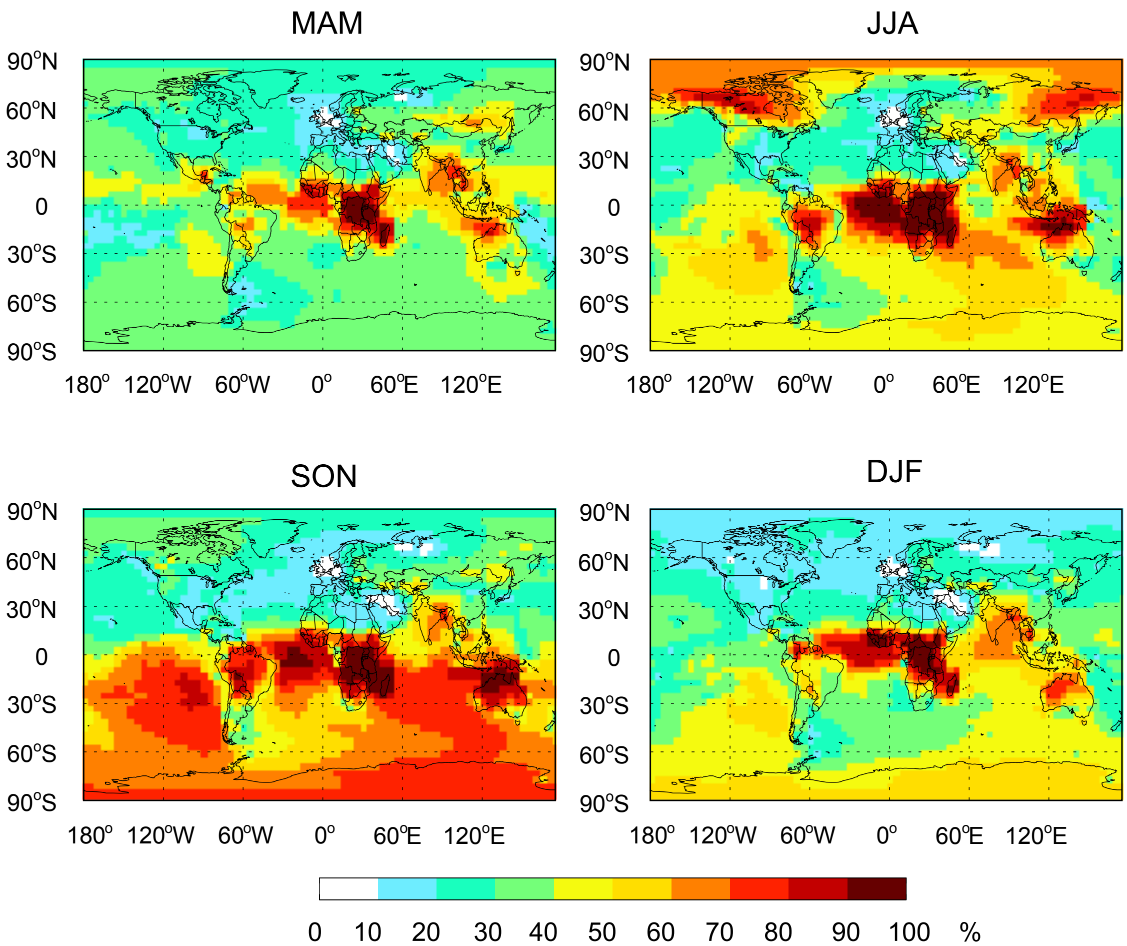

Figure S4. Average f_{bb} of BC in surface atmosphere during March–May (MAM), June–August (JJA), September–November (SON) and December–February (DJF) for 2007–2013.

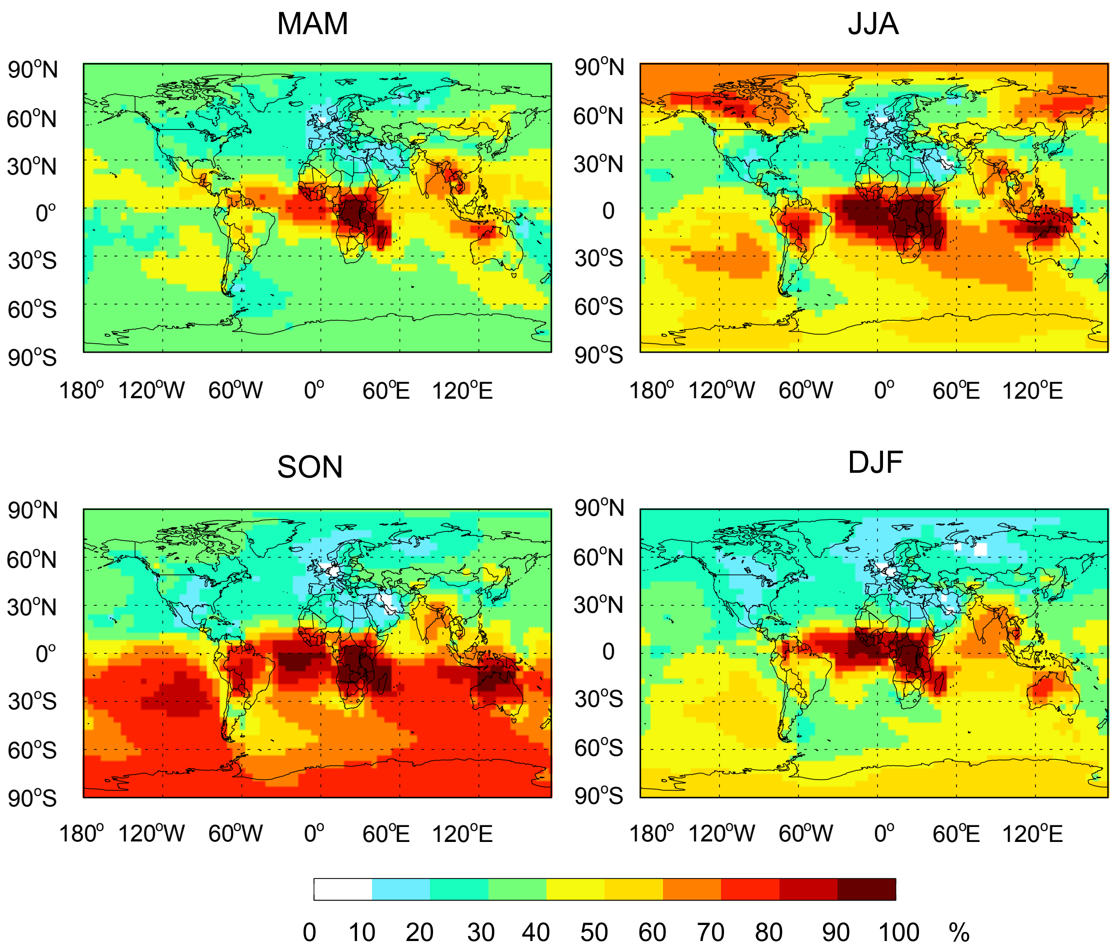

Figure S5. Same as Figure S4, but for BC deposition.

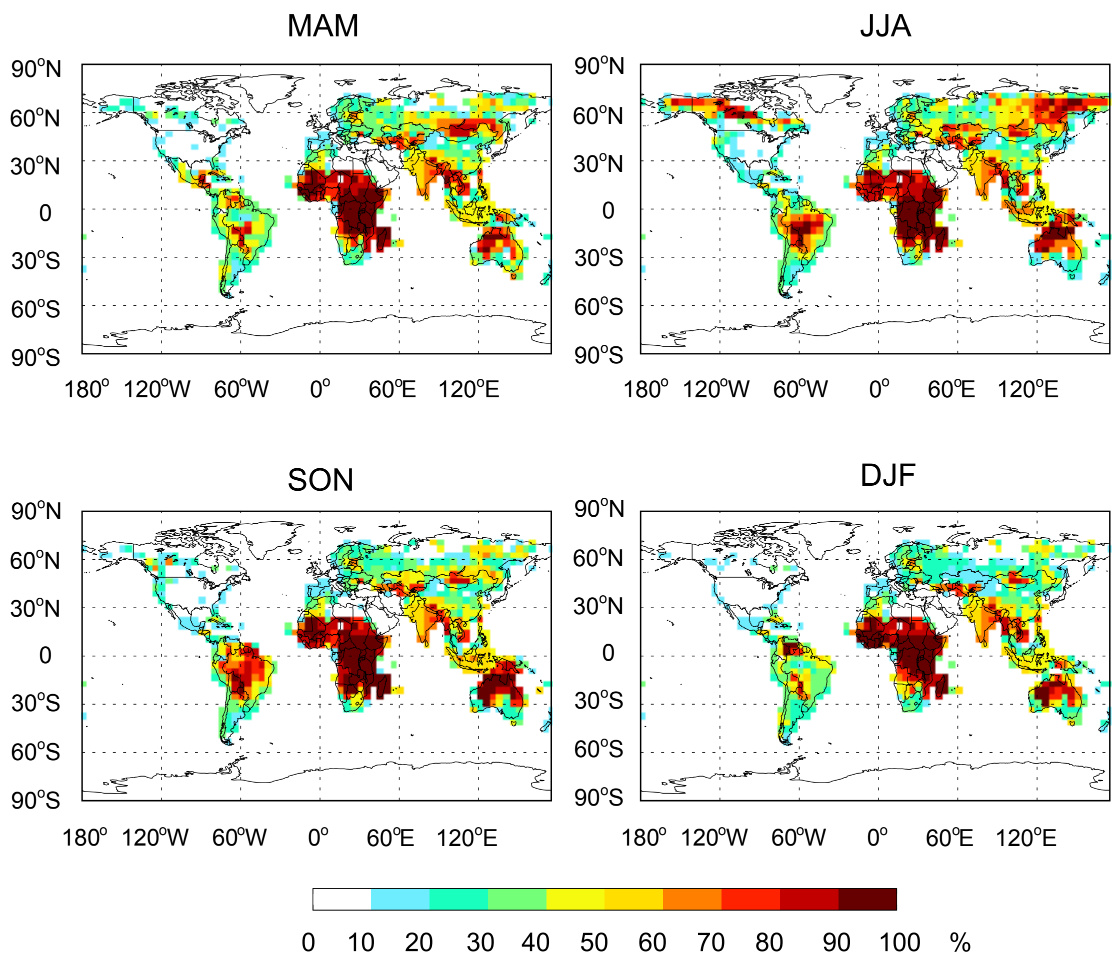

Figure S6. Same as Figure S4, but for BC emissions.

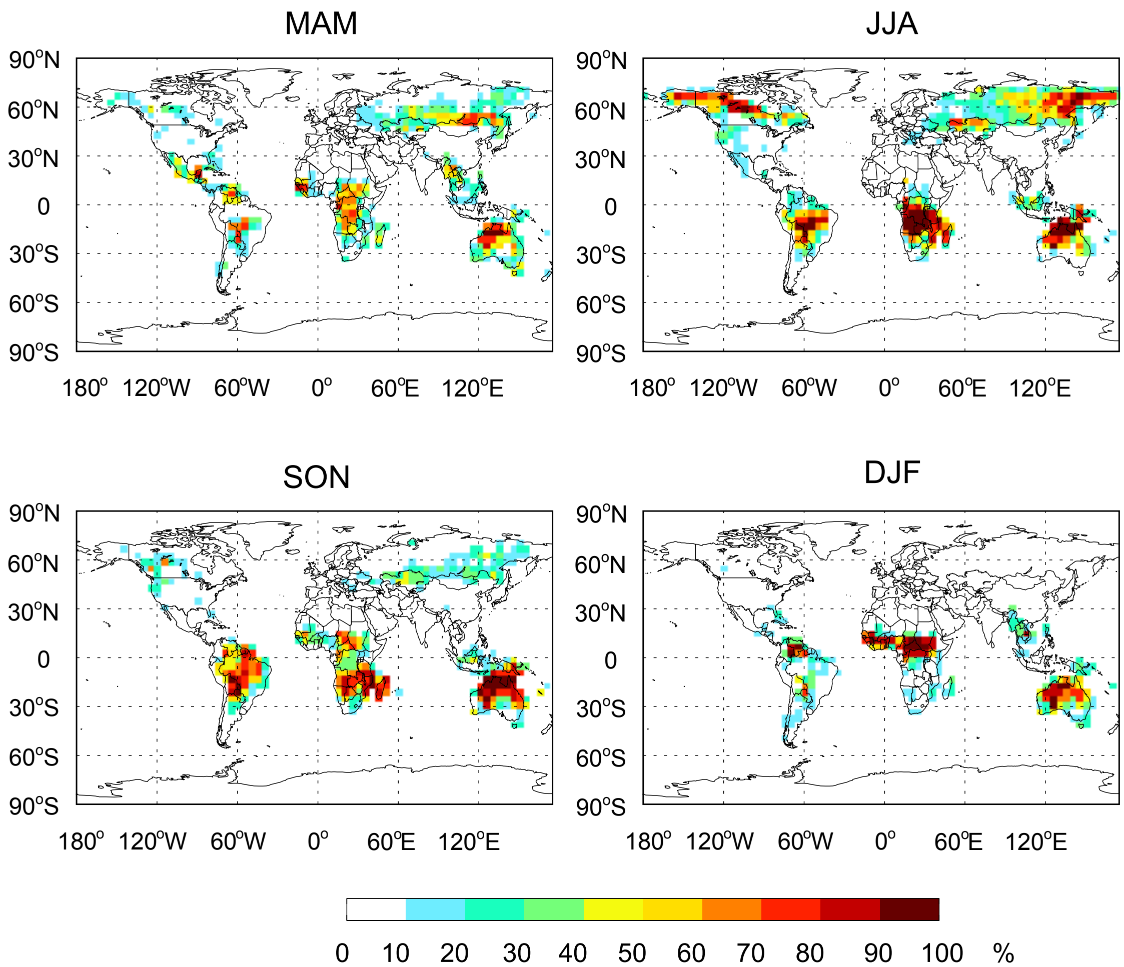

Figure S7. Average contribution of open burning to BC emissions (%) during March–May (MAM), June–August (JJA), September–November (SON) and December–February (DJF) for 2007–2013.

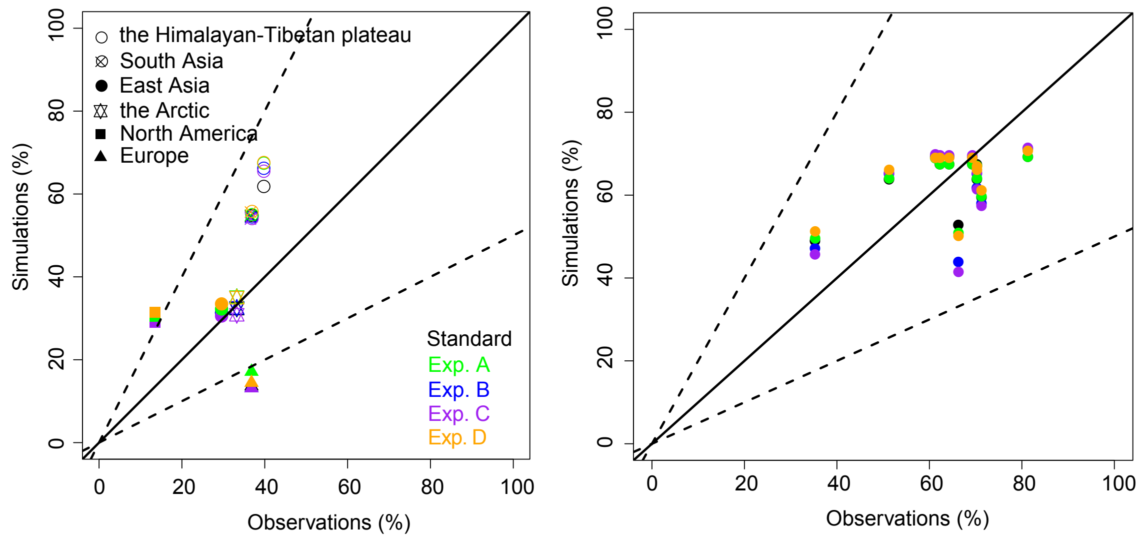

Figure S8. Observed and GEOS-Chem simulated mean f_{bb} (%) (a) of BC in the atmosphere in the six regions in Northern Hemisphere and (b) of BC deposited in snow over the Tibetan plateau. The regions are symbol-coded and the simulations are color-coded (see text for details). Solid lines are 1:1 and dashed lines are 1:2 (or 2:1).

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