



*Supplement of*

## **Impact of air pollution control measures and regional transport on carbonaceous aerosols in fine particulate matter in urban Beijing, China: insights gained from long-term measurement**

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Table S1. Annual variation in atmospheric  $T$ , RH, WS and ASC.

Year	$T$ (°C)	RH (%)	WS (m/s)	ASC (ton/day/km <sup>2</sup> )*
2013	13.4	54	1.6	2.87
2014	14.3	50	1.5	2.77
2015	13.5	54	1.5	2.99
2016	14.3	53	1.4	3.07
2017	14.0	48	1.4	3.01

\*ASC, a synthetical parameter connecting dilution by ventilation with wet scavenging, was obtained from 2018 *China Climate Bulletin* ([http://www.cma.gov.cn/root7/auto13139/201801/t20180117\\_460484.html](http://www.cma.gov.cn/root7/auto13139/201801/t20180117_460484.html), last access: 26 June 2019).

Table S2. Review of main emissions control measures for power plants, coal-fired boilers, residential heating and motor vehicles in Beijing from 2002-2017\*.

			Elimination of coal-fired boilers (ECFB)		
			Elimination of ECFB in suburbs of Beijing		
				Multiple stage ECFB	
Residential heating	<b>Electricity usage for selected old single-storey houses</b>				
		<b>Replacement with electricity for selected old single-storey houses in core areas</b>	<b>Usage of clean energy and no residential coal use in the core areas</b>	<b>No residential coal use in urban, urban-rural fringe and rural areas</b>	
Light-duty gasoline vehicles	<b>China I</b>	<b>China II</b>	<b>China III</b>	<b>China IV</b>	<b>China V</b>
High-duty diesel vehicles	<b>China I</b>	<b>China II</b>	<b>China III</b>	<b>China IV<sup>1</sup></b>	<b>China IV<sup>2</sup></b>
					<b>China V<sup>1</sup></b>
Traffic restriction			<b>Elimination of yellow-labeled vehicles</b>		

			<b>Elimination of light-duty gasoline vehicles with China I and II</b>
Clean energy and new energy vehicles	<b>Compressed Natural Gas public buses</b>	<b>New energy vehicles promoted</b>	<b>Electric public buses and taxies</b>
Temporary vehicular measures		<b>Alternate driving days for cars with even and odd numbered license plates</b>	<b>Banning private cars operating in Beijing's urban areas one work-day a week based on number plates</b>

\* Source: <https://www.unenvironment.org/resources/report/review-20-years-air-pollution-control-beijing>, last access: 26 June 2019

<sup>1</sup> Only implemented for public fleets; <sup>2</sup> for freight trucks and long-distance coaches

Table S3. Annual variations of gross domestic product (GDP), population, total energy consumption, population of vehicles, consumption of gasoline, diesel oil, coal, and natural gas in Beijing.

Category	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
GDP ( $10^8$ yuan)	4396	5104	6164	7141	8312	10071.9	11392	12419	14441.6	16627.9	18350	20330.1	21944.1	23685.7	25669.1	28000.4
Population ( $10^4$ )	1423.2	1456.4	1492.7	1538	1601	1676	1771	1860	1961.9	2018.6	2069.3	2144.8	2151.6	2170.5	2172.9	2170.7
Energy consumption ( $10^4$ ton)	4436.1	4648.2	5139.6	5049.8	5399.3	5747.7	5786.2	6008.6	6359.5	6397.3	6564.1	6723.9	6831.2	6852.6	6961.7	7130.6
Vehicular population ( $10^4$ )	176.5	212.4	229.6	258.3	287.6	312.8	350.4	398.1	480.9	498.3	520	543.7	559.1	561.9	571.7	590.9
Gasoline ( $\times 10^4$ tons)	152	165.22	198.39	235.3	278.16	324.72	340.92	363.61	371.53	389.79	415.9	423.61	440.61	462.76	470.37	489.85
Diesel oil ( $\times 10^4$ tons)	109	110.41	131.93	140.86	177.49	192.02	227.22	240.18	237.42	241.12	215.82	193.9	196.46	182.35	172.69	175.11
Coal ( $\times 10^4$ tons)	2531	2674	2939	3069	3055.7	2984.7	2747.7	2664.7	2634.6	2365.5	2269.9	2019.23	1736.54	1165.18	847.62	490.46
Natural gas ( $\times 10^8 \text{ m}^3$ )	21	21.19	27.02	32.04	40.65	46.64	60.65	69.4	74.79	73.56	92.07	98.81	113.70	145.37	160.30	162.24

Table S4. Ratio of TC to PM<sub>2.5</sub> in this and previous studies.

Cities	Method	Period	TC/PM <sub>2.5</sub> (%)	References
Budapest, Hungary	EA	June 2010- May 2011	40.0%	Szigeti et al., 2013
Istanbul, Turkey	EA	June 2010- May 2011	30.0%	Szigeti et al., 2013
Beijing, China	TOR	March 2005-February 2006	27.6%	Yang et al., 2011c
Chongqing, China	TOR	March 2005-February 2007	28.3%	Yang et al., 2011c
Shanghai, China	TOR	March 2005-February 2008	34.5%	Yang et al., 2011c
Guangzhou, China	TOR	March 2005-February 2009 the autumn–early winter period	26.4% 60–80%	Yang et al., 2011c Keywood et al., 2011
Melbourne, Australia	-	the summer months December 2006-January 2007 18/09/2001-29/10/2001&19/12/2002-	20–40% 30–50%	Keywood et al., 2011
Antwerp, Belgium	TA	23/02/2003 01/02/2002-26/03/2002&27/09/2002-	32.0%	Bencs et al., 2010
Hasselt, Belgium	TA	04/11/2002 16/05/2002-26/06/2002&05/11/2002-	23.0%	Bencs et al., 2010
Mechelen, Belgium	TA	03/01/2003	24.0%	Bencs et al., 2010
Ghent, Belgium	TOT	Jun. 10-Jul. 7, 2004	31.7%	Viana et al., 2007
Ghent, Belgium	TOT	Jan. 10-Feb.14, 2005	22.3%	Viana et al., 2007
Barcelona, Spain	TOT	Jul. 27–Aug. 31, 2004	32.6%	Viana et al., 2007
Barcelona, Spain	TOT	Nov. 11–Dec. 16, 2004	28.8%	Viana et al., 2007
Amsterdam, Holland	TOT	Jul. 4-Aug. 2, 2005	24.4%	Viana et al., 2007

Amsterdam, Holland	TOT	Jan. 9-Feb. 16, 2006	32.6%	Viana et al., 2007
Diamond Bar, USA	-	Oct. 31-Nov. 2, 1997	29.5%	Allen et al., 2000
Mira Loma, USA	-	Oct. 31-Nov. 2, 1997	36.6%	Allen et al., 2000
Los Angeles (Riverside), USA	-	Oct. 31-Nov. 2, 1997	30.5%	Allen et al., 2000
Mira Loma, USA	TOT	Sep. 2001-Feb. 2002	30.5%	Na et al., 2004
Rubidoux, USA	TOR	Jan.-Feb. 1999	33.7%	Tolocka et al., 2001
Phoenix, USA	TOR	Jan.-Feb. 1999	73.2%	Tolocka et al., 2001
Philadelphia, USA	TOR	Jan.-Feb. 1999	33.3%	Tolocka et al., 2001
Los Angeles, USA	TMO	Jan. 1995-Feb. 1996	36.9%	Kim et al., 2000
San Nicolas Island, USA	TMO	Jan. 1995-Feb. 1996	24.6%	Kim et al., 2000
Beijing, China	TOR	Jul. 1999-Sep. 2000	30.2%	He et al., 2001
Abbotsford, Canada	TOR	May 1994-Feb. 1995	48.0%	Brook and Dann, 1999
Kaohsiung, Taiwan, CN	EA	Nov. 1998-Apr. 1999	21.3%	Lin and Tai, 2001
Sao Paulo, Brazil	TA	Jul.11-Sep. 10, 1997	77.5%	Castanho and Artaxo, 2001
Hongkong (roadside site), China	TOT/TOR	2013	30.3%	a*
Hongkong (urban site), China	TOT/TOR	2013	44.5%	a*
Hongkong (suburban site), China	TOT/TOR	2013	27.0%	a*
Hongkong (new town site), China	TOT/TOR	2013	20.4%	a*
Hongkong (urban site), China	TOT/TOR	2013	26.7%	a*

Hongkong (new town site), China	TOT/TOR	2013	30.0%	a*
South Phoenix (traffic site), USA	TOT	Dec. 20, 2008 to Feb. 18, 2009	45.0%	Upadhyay et al., 2011
South Phoenix (airport site), USA	TOT	Dec. 20, 2008 to Feb. 18, 2009	49.2%	Upadhyay et al., 2011
South Phoenix (mixed site), USA	TOT	Dec. 20, 2008 to Feb. 18, 2009	48.9%	Upadhyay et al., 2011
Tongyu, China	TOR	Spring 2006	6.2%	Zhang et al., 2012
Zloty, Potok, Poland	TOT	2013	40.4%	Błaszcza et al., 2016
Raciborz, Poland	TOT	2011 and 2012	44.5%	Błaszcza et al., 2016
Shangdianzi, China	TOR	Four seasons (2009-2010)	20.4%	Zhao et al., 2013
Beijing, China	TOR	Four seasons (2009-2011)	19.8%	Zhao et al., 2013
Tianjin, China	TOR	Four seasons (2009-2012)	18.1%	Zhao et al., 2013
Shijiazhuang, China	TOR	Four seasons (2009-2013)	19.0%	Zhao et al., 2013
Chengde, China	TOR	Four seasons (2009-2014)	28.6%	Zhao et al., 2013
Fresno, China	TOR	Jan. 1-Dec. 26, 2000	43.5%	Watson and Chow, 2002
Xi'an (rural site), China	TOR	2010	16.4%	Wang et al., 2015
Xi'an (urban), China	TOR	2010	17.3%	Wang et al., 2015
Xi'an (downtown site), China	TOR	2010	15.4%	Wang et al., 2015
Xi'an (roadside site), China	TOR	2010	17.4%	Wang et al., 2015
Xi'an (urban site), China	TOR	2010	19.7%	Wang et al., 2015
Xi'an (reference site), China	TOR	2010	14.8%	Wang et al., 2015

Beijing (Tinghua site), China	TOT	2008	32.0%	Yang et al., 2011b
Beijing (Miyun site), China	TOT	2008	33.0%	Yang et al., 2011b
Beijing (Tinghua site), China	TOR	Jul 1999-Jun 2000	40.0%	He et al., 2001
Egbert, Canada	TOT	Jan. 2006-Nov 2007	38.0%	Yang et al., 2011a
Europe (average)	TOT	2008-2011	25.0%	Cavalli et al., 2016
Europe (average)	-	1996-2007	19.0%	Putaud et al., 2010
USA (average)	TOR	2005-2008	25.0%	Hand et al., 2011
China (average)	TOT/TOR/TMO	-	22.0%	Wang et al., 2016
India (average)	-	-	20.0%	Ram and Sarin, 2010, 2012; Bisht et al., 2015

a\* [http://www.epd.gov.hk/epd/sites/default/files/epd/english/environmentinhk/air/studyrpts/files/final\\_report\\_mvtmpms\\_2013.pdf](http://www.epd.gov.hk/epd/sites/default/files/epd/english/environmentinhk/air/studyrpts/files/final_report_mvtmpms_2013.pdf), last access: 26 June 2019

TOR: thermal-optical reflectance; TOR: thermal-optical transmittance; TMO: thermal manganese oxidation; EA: elemental analysis; TA: thermal analysis

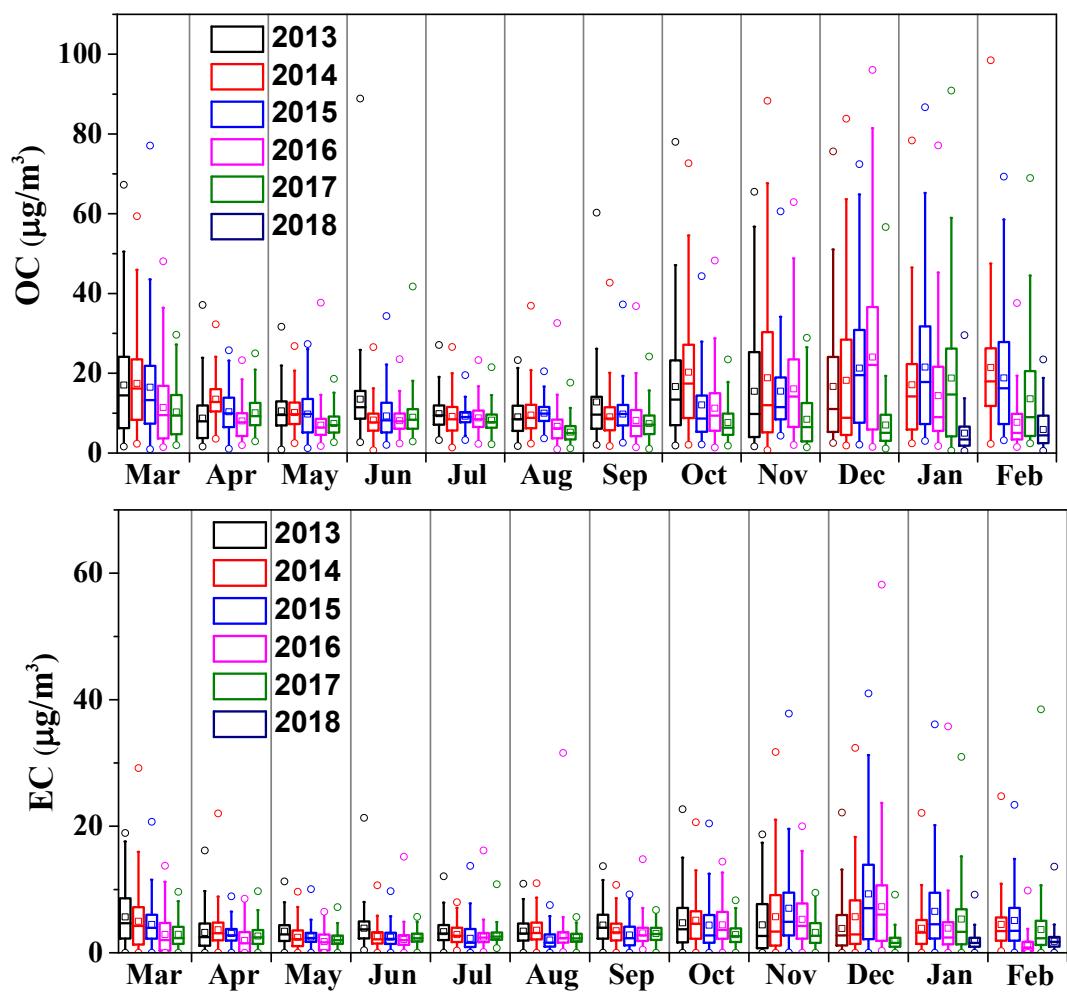


Fig. S1. Monthly variation of OC and EC concentrations from March 2013 to February 2018.

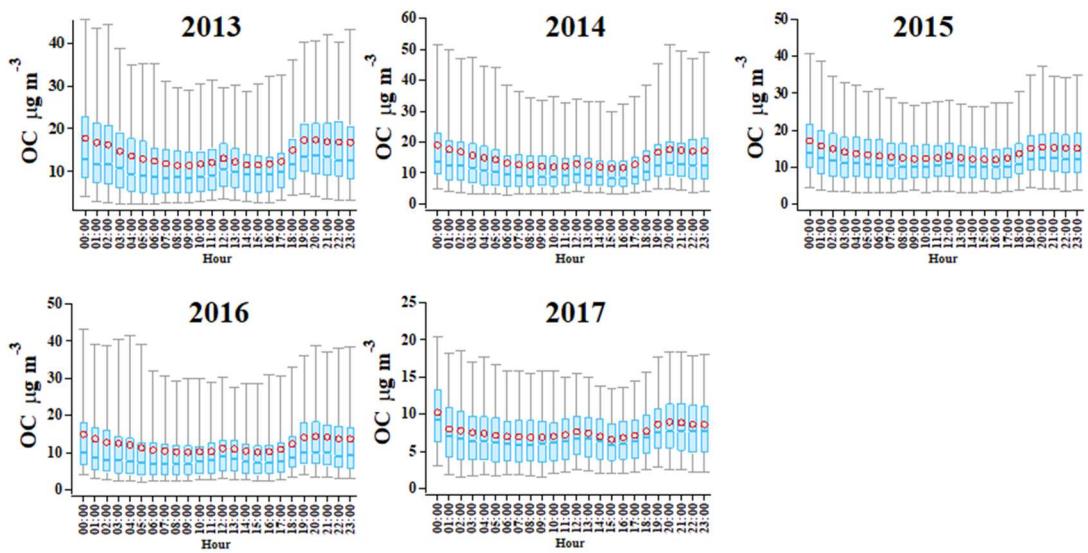


Fig. S2. Diurnal variation of OC concentrations from March 2013 to February 2018.

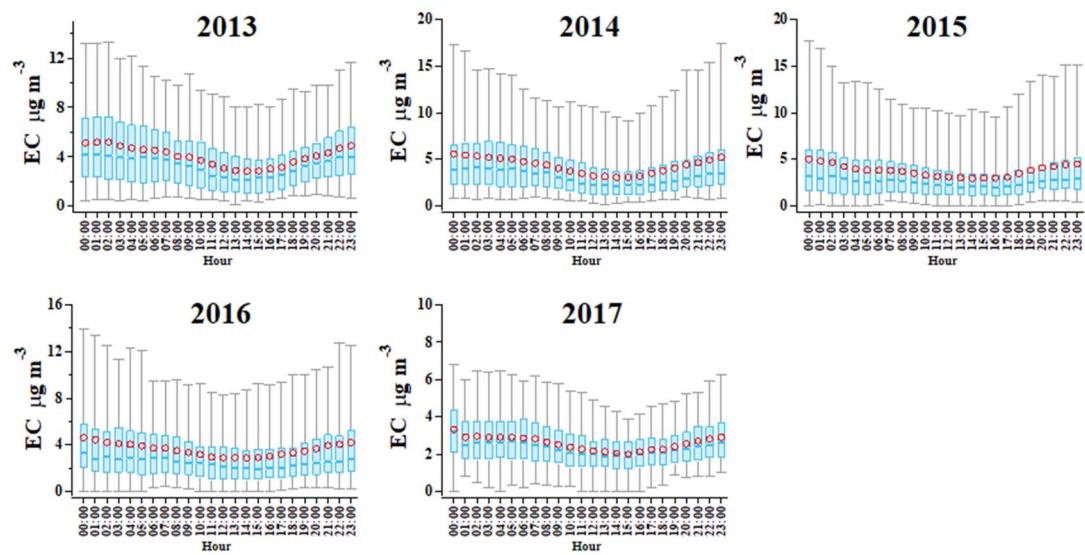


Fig. S3. Diurnal variation of EC concentrations from March 2013 to February 2018.

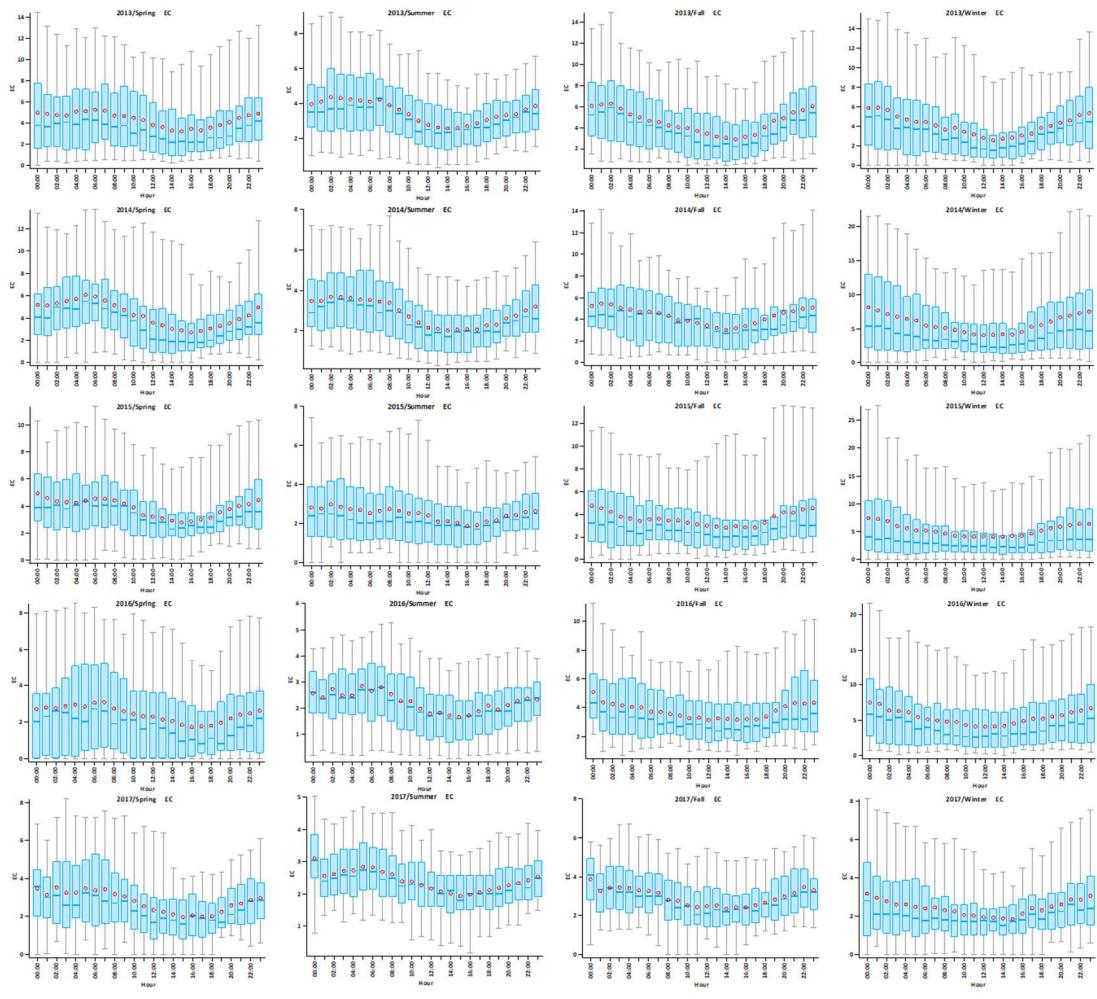


Fig. S4. Diurnal variation of OC concentrations (in  $\mu\text{g}/\text{m}^3$ ) in different seasons during the whole study period.

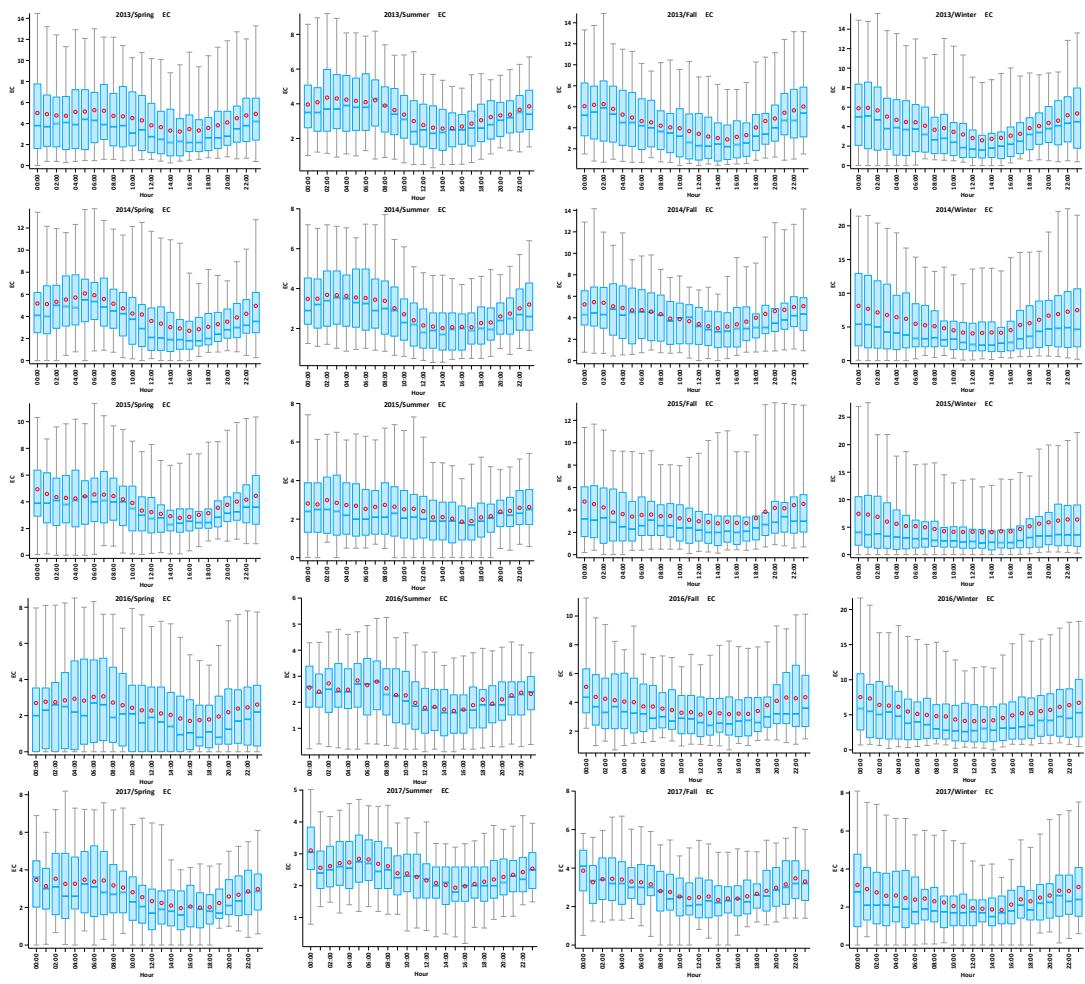


Fig. S5. Diurnal variation of EC concentrations (in  $\mu\text{g}/\text{m}^3$ ) in different seasons during the whole study period.

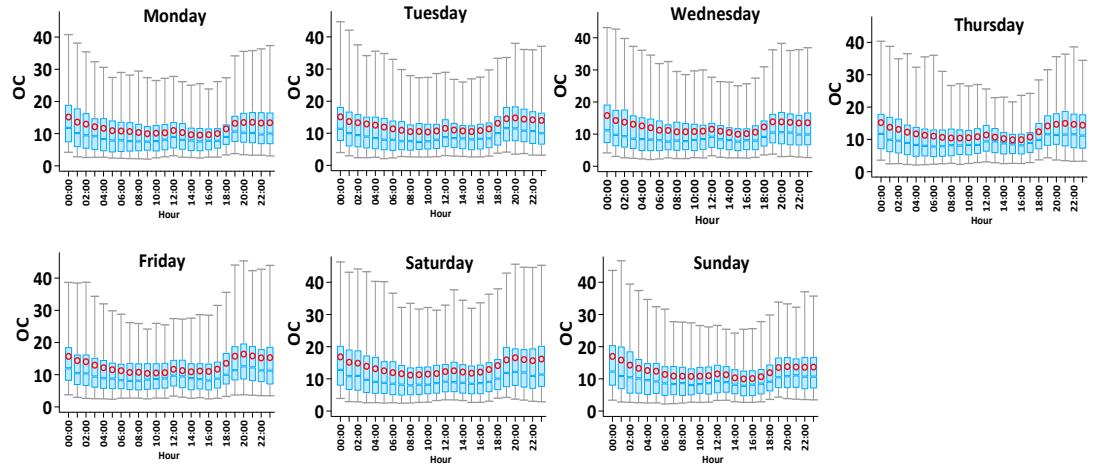


Fig. S6. Diurnal variation of OC concentrations (in  $\mu\text{g}/\text{m}^3$ ) on weekdays and weekend days during the whole study period.

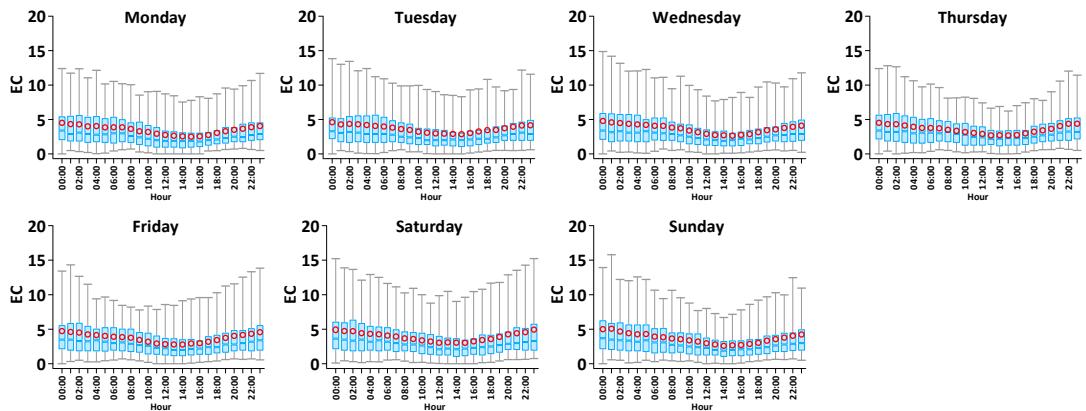


Fig. S7. Diurnal variation of EC concentrations (in  $\mu\text{g}/\text{m}^3$ ) on weekdays and weekend days during the whole study period.

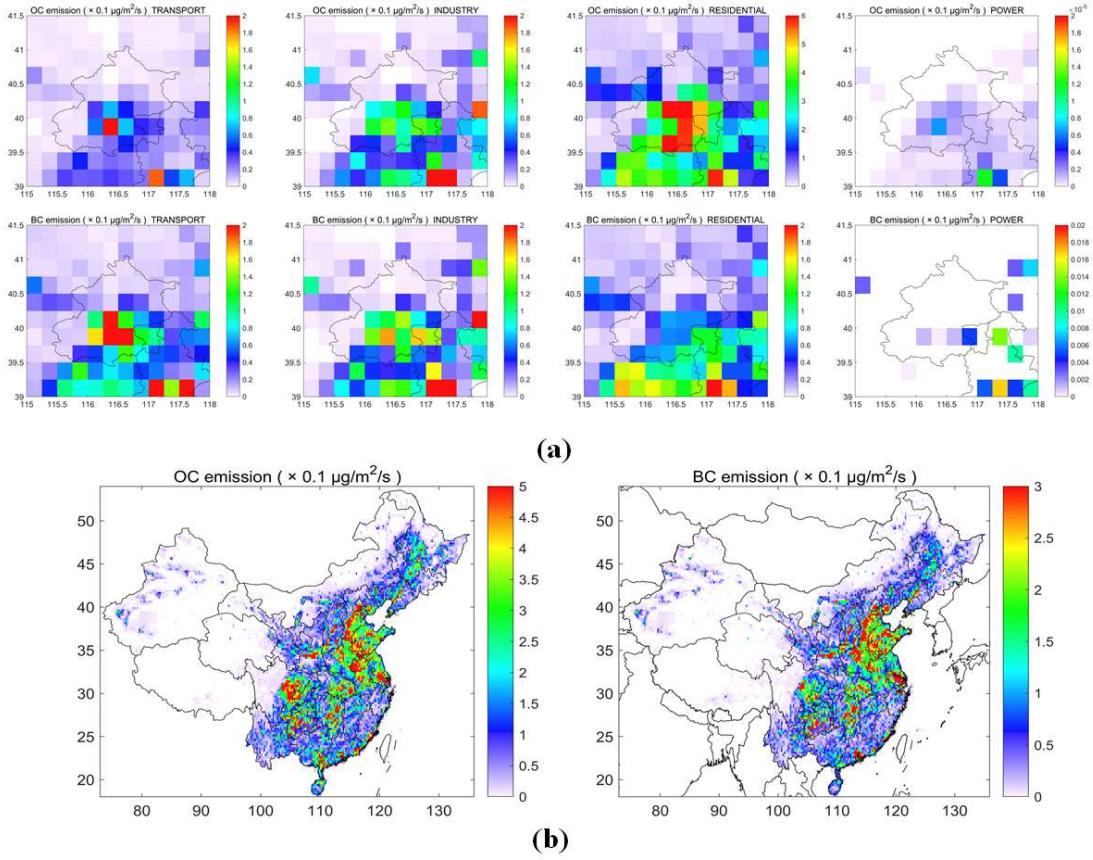


Fig. S8. Gridded emissions of OC and BC (proxy for EC) in Beijing (a) and China (b) from the MEIC (Multi-resolution Emission Inventory for China, <http://www.meicmodel.org>, unit:  $\mu\text{g}/\text{m}^3/\text{s}$ , last access: 26 June 2019).

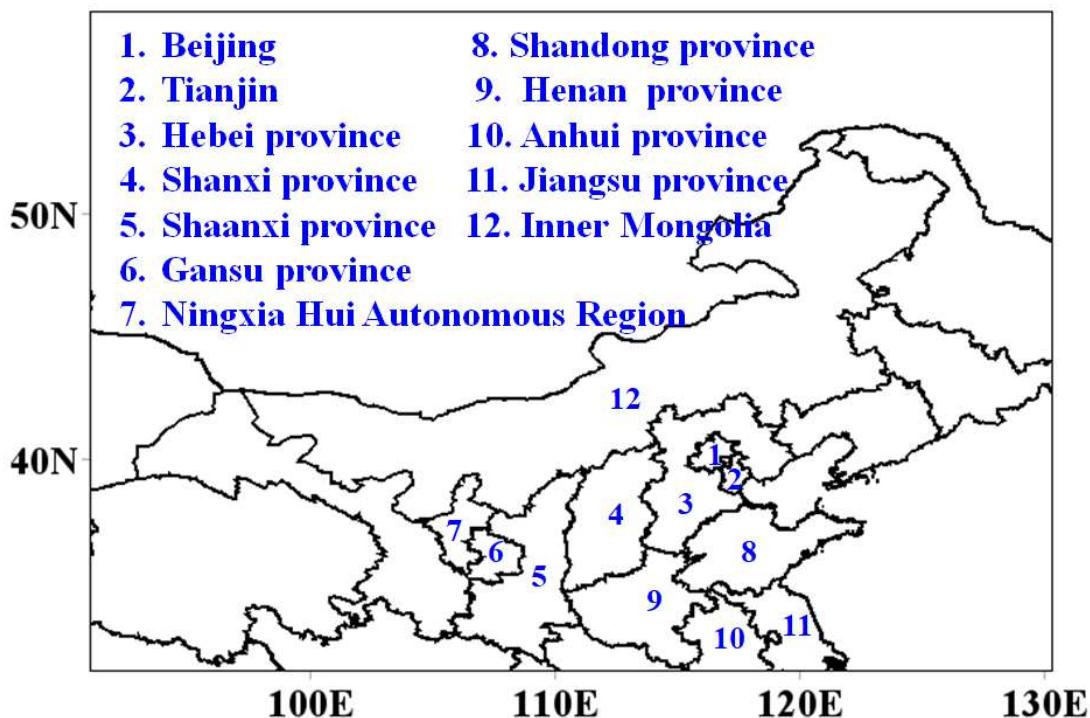


Fig. S9. Map with identification of the Chinese provinces, which are potential source areas of OC and EC in this study.

## References

- Allen, J. O., Hughes, L. S., Salmon, L. G., Mayo, P. R., Johnson, R. J., and Cass, G. R.: Characterization and evolution of primary and secondary aerosols during PM<sub>2.5</sub> and PM<sub>10</sub> episodes in the South Coast Air Basin. Environmental Engineering and Science Department California Institute of Technology Pasadena, CA. Final report, CRC Project No, A-22, 2000.
- Bencs, L., Ravindra, K., Hoog, J. D., Spolnik, Z., Bleux, N., Berghmans, P., Deutsch, F., Roekens, E., and Van Grieken, R.: Appraisal of measurement methods, chemical composition and sources of fine atmospheric particles over six different areas of northern Belgium. Environ. Pollut., 158(11), 3421-3430, 2010.
- Bisht, D. S., Srivastava, A. K., Pipal, A. S., Srivastava, M. K., Pandey, A. K., Tiwari, S., and Pandithurai, G.: Aerosol characteristics at a rural station in southern peninsular India during CAIPEEX-IGOC: physical and chemical properties. Environ. Sci. Pollut. Res., 22, 5293-5304, 2015.
- Błaszczał, B., Rogula-Kozłowska, W., Mathews, B., Juda-Rezler, K., Klejnowski, K., and Rogula-Kopiec, P.: Chemical compositions of PM<sub>2.5</sub> at two non-urban sites from the polluted region in Europe. Aerosol Air Qual. Res., 16(10), 2333–2348. doi:10.4209/aaqr.2015.09.0538, 2016.
- Brook, J. R. and Dann, T. F.: Contribution of nitrate and carbonaceous species to PM<sub>2.5</sub> observed in Canadian cities. J. Air and Waste Manage. Assoc., 49, 193–199, 1999.
- Castanho, A. D. A. and Artaxo, P.: Wintertime and summertime São Paulo aerosol source apportionment study. Atmos. Environ., 35(29), 4889–4902. doi:10.1016/s1352-2310(01)00357-0, 2001.
- Cavalli, F., Alastuey, A., Areskoug, H., Ceburnis, D., Čech, J., Genberg, J., Harrison, R. M., Jaffrezo, J. L., Kiss, G., Laj, P., Mihalopoulos, N., Perez, N., Quincey, P., Schwarz, J., Sellegri, K., Spindler, G., Swietlicki, E., Theodosi, C., Yttri, K. E., Aas, W., and Putaud, J. P.: A European aerosol phenomenology-4: Harmonized concentrations of carbonaceous aerosol at 10 regional background sites across Europe. Atmos. Environ., 144, 133-145, 2016.
- Hand, J. L., Copeland, S. A., Day, D. E., Dillner, A. M., Idresand, H., Malm, W. C., McDade, C. E., Moore, Jr., C. T., Pitchford, M. L., Schichtel, B. A., and Watson, J. G.: IMPROVE (Interagency Monitoring of Protected Visual Environments): Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States: Report V, CIRA Report ISSN: 0737-5352-87,

- <http://vista.cira.colostate.edu/improve/Publications/Reports/2011/2011.htm>, 2011.
- He, K., Yang, F., Ma, Y., Zhang, Q., Yao, X., Chan, C. K., Cadle, S., Chan, T., and Mulawa, P.: The characteristics of PM<sub>2.5</sub> in Beijing, China, *Atmos. Environ.*, 35, 4959-4970, [https://doi.org/10.1016/S1352-2310\(01\)00301-6](https://doi.org/10.1016/S1352-2310(01)00301-6), 2001.
- Keywood, M., Guyes, H., Selleck, P., and Gillett, R.: Quantification of secondary organic aerosol in an Australian urban location. *Environ. Chem.*, 8(2), 115-126, 2011.
- Kim, B. M., Teffera, S., and Zeldin, M. D.: Characterization of PM<sub>2.5</sub> and PM<sub>10</sub> in the south coast air basin of southern California: Part 1—Spatial variations, *J. Air Waste Manage. Assoc.*, 50:12, 2034-2044, DOI: 10.1080/10473289.2000.10464242, 2000.
- Lin, J. J. and Tai, H. S.: Concentrations and distributions of carbonaceous species in ambient particles in Kaohsiung City, Taiwan. *Atmos. Environ.*, 35, 2627–2636, 2001.
- Na, K., Sawant, A. A., Song, C., and Cocker, D. R.: Primary and secondary carbonaceous species in the atmosphere of Western Riverside County, California. *Atmos. Environ.*, 38(9), 1345–1355. doi:10.1016/j.atmosenv.2003.11.023, 2004.
- Putaud, J. P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Flentje, H., Fuzzi, S., Gehrig, R., Hansson, H. C., Harrison, R. M., Hermann, H., Hitzenberger, R., Hüglin, C., Jones, A. M., Kasper-Giebl, A., Kiss, G., Kousa, A., Kuhlbusch, T. A. J., Löschen, G., Maenhaut, W., Molnar, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz, M., Puxbaum, H., Querol, X., Rodriguez, S., Salma, I., Schwarz, J., Smolik, J., Schneider, J., Spindler, G., Ten Brink, H., Tursic, J., Viana, M., Wiedensohler, A., and Raes F.: A European aerosol phenomenology-3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe, *Atmos. Environ.*, 44(10), 1308-1320, 2010.
- Ram, K. and Sarin, M. M.: Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over northern India, *J. Aerosol Sci.*, 41(1), 88-98, 2010.
- Ram, K. and Sarin, M. M.: Carbonaceous aerosols over northern India: sources and spatio-temporal variability, *Proc. Indian Natl. Sci. Acad.*, 78, 523-533, 2012.
- Szigeti, T., Mihucz, V. G., Óvári, M., Baysal, A., Atilgan, S., Akman, S., and Záray, G.: Chemical characterization of PM<sub>2.5</sub> fractions of urban aerosol collected in Budapest and Istanbul, *Microchemical J.*, 107(3), 86-94, 2013.

- Tolocka, M. P., Solomon, P. A., Mitchell, W., Norris, G. A., Gemmill, D. B., Wiener, R. W., Vanderpool, R. W., Homolya, J. B., and Rice, J.: East versus west in the US: chemical characteristics of PM<sub>2.5</sub> during the winter of 1999. *Aerosol Sci. Tech.* 34, 88–96, 2001.
- Upadhyay, N., Clements, A., Fraser, M., and Herckes, P.: Chemical speciation of PM<sub>2.5</sub> and PM<sub>10</sub> in South Phoenix, AZ. *J. Air Waste Manage. Assoc.*, 61(3), 302–310. doi:10.3155/1047-3289.61.3.302, 2011.
- Viana, M., Maenhaut, W., Brink, H. M., Chi, X., Weijers, E., Querol, X., Alastuey, A., Mikuska, P., and Vecera, Z.: Comparative analysis of organic and elemental carbon concentrations in carbonaceous aerosols in three European cities. *Atmos. Environ.*, 41(28), 5972–5983. doi:10.1016/j.atmosenv.2007.03.035, 2007.
- Wang, L. P., Zhou, X. H., Ma, Y. J., Cao, Z. Y., Wu, R. D., and Wang W. X.: Carbonaceous aerosols over China-review of observations, emissions and climate forcing, *Environ. Sci. Pollut. Res.*, 23, 1671-1680, 2016.
- Wang, P., Cao, J. J., Shen, Z. X., Han, Y. M., Lee, S. C., Huang, Y., Zhu, C. S., Wang, Q. Y., Xu, H. M., and Huang, R. J.: Spatial and seasonal variations of PM<sub>2.5</sub> mass and species during 2010 in Xi'an, China, *Sci. Total Environ.*, 508, 477-487, <https://doi.org/10.1016/j.scitotenv.2014.11.007>, 2015.
- Watson, J. G. and Chow, J. C.: Comparison and evaluation of in situ and filter carbon measurements at the Fresno Supersite. *J. Geophys. Res.* 107, D21. doi:10.1029/2001JD000573, 2002.
- Yang, F., Huang, L., Sharma, S., Brook, J. R., Zhang, W., Li, S. -M., and Tan, J.: Two-year observations of fine carbonaceous particles in variable sampling intervals, *Atmos. Environ.*, 45, 2418–2426, 2011a.
- Yang, F., Huang, L., Duan, F., Zhang, W., He, K., Ma, Y., Brook, J. R., Tan, J., Zhao, Q., and Cheng, Y.: Carbonaceous species in PM<sub>2.5</sub> at a pair of rural/urban sites in Beijing, 2005-2008, *Atmos. Chem. Phys.*, 11, 7893–7903, 2011b.
- Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., Chen, G., and Zhao, Q.: Characteristics of PM<sub>2.5</sub> speciation in representative megacities and across China. *Atmos. Chem. Phys.*, 11(11), 1025-1051, 2011c.
- Zhang, R. J., Tao, J., Ho, K. F., Shen, Z. X., Wang, G. H., Cao, J. J., Liu, S. X., Zhang, L. M., and Lee, S. C.: Characterization of atmospheric organic and elemental carbon of PM<sub>2.5</sub> in a typical semi-

arid area of northeastern China, *Aerosol Air Qual. Res.*, 12: 792–802, 2012.  
Zhao, P. S., Dong, F., He, D., Zhao, X. J., Zhang, X. L., Zhang, W. Z., and Liu, H. Y.: Characteristics  
of concentrations and chemical compositions for PM<sub>2.5</sub> in the region of Beijing, Tianjin, and Hebei,  
China. *Atmos. Chem. Phys.*, 13(9), 4631–4644. doi:10.5194/acp-13-4631-2013, 2013.