Supplement of

Measurement report: Source characteristics of water-soluble organic carbon in $PM_{2.5}$ at two sites in Japan, as assessed by long-term observation and stable carbon isotope ratio

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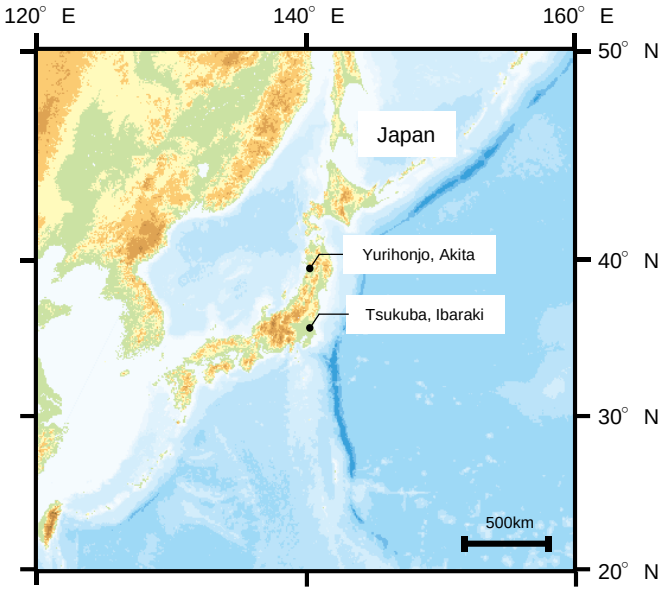

Figure S1. Sampling locations at the Japan Automobile Research Institute (Tsukuba, Ibaraki) and Akita Prefectural University (Yurihonjo, Akita). This map plots the sampling locations on a standard map provided by the Geospatial Information Authority of Japan.

Table S1. $\delta^{13}C_{WSOC}$ in $PM_{2.5}$ determined in the present and other published studies.

Sampling site	Site type	Particle size	Sampling period	*n*	Analytical method	$\delta^{13}C_{WSOC}$ (‰)		Reference
						Average ± SD	Range	
Tsukuba, Japan	Suburban	$PM_{2.5}$	Jul 2017–Jul 2019	62	Wet oxidation/IRMS	−25.2 ± 1.1	−26.7 to −21.8	This study
Yurihonjo, Japan	Rural	$PM_{2.5}$	Aug 2017–Jul 2019	45	Wet oxidation/IRMS	−24.6 ± 2.4	−28.4 to −19.8	This study
Delhi, India	Urban	$PM_{2.5}$	Jan–Mar 2016	7	Combustion-EA/IRMS	−25.4 ± 1.0	−	Dasari et al. (2019)
Bhola, Bangladesh	Rural	$PM_{2.5}$	Jan–Mar 2016	12	Combustion-EA/IRMS	−24.2 ± 0.6	−	Dasari et al. (2019)
Hanimaadhoo, Maldives	Rural	$PM_{1.0}$	Jan–Mar 2016	15	Combustion-EA/IRMS	−20.9 ± 0.6	−	Dasari et al. (2019)
Seoul, Korea	Urban	TSP	Mar 2015–Jan 2016	78	TOC analyzer/IRMS	−24.0 ± 1.5	−27.5 to −21.0	Han et al. (2020)
Nanjing, China	Suburban	$PM_{2.5}$	Jan 2015	−	GasBench/IRMS	−	−26.24 to −23.35	Zhang et al. (2019)
Beijing, China	Urban	$PM_{2.5}$	Jan, Jun, 2013	10	Combustion-EA/IRMS	−22.51 ± 0.49	−	Yan et al. (2017)
						−25.40 ± 0.46	−	Yan et al. (2017)
Hanimaadhoo, Maldives	Rural	$PM_{2.5}$	Feb–Mar 2012	14	Combustion-EA/IRMS	−20.8 ± 0.7	−22.13 to −19.64	Bosch et al. (2014)
Jeju, Korea	Rural	TSP/$PM_{2.5}$	Mar 2011	10	Combustion-EA/IRMS	−	−	Kirillova et al. (2014a)
New Delhi, India	Urban	$PM_{2.5}$	Oct 2010–Mar 2011	20	Combustion-EA/IRMS	−24.1 ± 0.9	−26.3 to −22.4	Kirillova et al. (2014b)
Sapporo, Japan	Rural	TSP	Sep 2009–Oct 2010	21	Combustion-EA/IRMS	−24.2 ± 1.59	−26.7 to −21.2	Pavuluri and Kawamura (2017)
Sapporo, Japan	Forest	TSP	Jun 2009–Dec 2010	−	Combustion-EA/IRMS	−	−	Miyazaki et al. (2012)
Stockholm, Sweden	Forest	TSP	Aug–Oct 2009	3	Combustion-EA/IRMS	−	−25.6 to −25.1	Kirillova et al. (2010)
Hanimaadhoo, Maldives	Rural	TSP	Jan 2008–Apr 2009	12	Combustion-EA/IRMS	−18.4 ± 0.5	−20.8 to −17.5	Kirillova et al. (2013)
Sinhagad, India	Rural	TSP	Jan 2008–Apr 2009	12	Combustion-EA/IRMS	−20.4 ± 0.5	−23.7 to −19.8	Kirillova et al. (2013)
Millbrook, USA	Rural	TSP	Mar, May, Aug 2007	3	Combustion-EA/IRMS	−24.7	−25.1 to −24.4	Wozniak et al. (2012b)
Harcum, USA	Rural	TSP	Feb, Apr, Aug 2007	3	Combustion-EA/IRMS	−25.4	−26.1 to −24.8	Wozniak et al. (2012b)
Millbrook, USA	Rural	TSP	May 2006–May 2007	9	Combustion-EA/IRMS	−25.2 ± 0.2	−26.0 to −23.9	Wozniak et al. (2012a)
Harcum, USA	Rural	TSP	Jun 2006–Jun 2007	10	Combustion-EA/IRMS	−25.3 ± 0.6	−27.4 to −21.1	Wozniak et al. (2012a)

Supplement S1 Water-soluble ion analysis

A portion of each quartz fiber filter (1.44 cm^2) was extracted in 3 mL of Milli-Q water under ultrasonic agitation for 15 min. The extract was filtered through a syringe filter (Chromatodisc Type A 0.45 μm, GL Sciences, Japan) to remove insoluble materials. Anion concentrations were determined in the filtrate using an IonPac AS17-C column and IonPac AG17-C guard column (Thermo Fisher Scientific Inc.), with a 1–40 mM gradient of potassium hydroxide as the eluent. Cation concentrations were determined using a CS12A column and CG12A guard column (Thermo Fisher Scientific Inc.), with 20 mM methanesulfonic acid as the eluent. Calibration curves were prepared using cation mixed standard solution 2 and anion mixed standard solution 4 (Kanto Chemical Co., Inc., Tokyo, Japan). The coefficient of determination was >0.999 for all compounds, and the detection limits were 4 ppb (Cl^-), 5 ppb (NO_2^-), 12 ppb (NO_3^-), 8 ppb (SO_4^{2-}), 8 ppb (Na^+), 5 ppb (NH_4^+), 15 ppb (K^+), 15 ppb (Mg^{2+}), and 20 ppb (Ca^{2+}). These values were comparable to those used in other research (Shen et al., 2009).

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