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Supplement of

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

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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. (2014)
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. (2014)
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)
Zurich, Switzerland	Urban	−	Aug 2002–Mar 2003	−	GasBench/IRMS	−	−25 to −23	Fisseha et al. (2009)
Zurich, Switzerland	Urban	−	−	−	GasBench/IRMS	−24.0 ± 0.7	−	Fisseha et al. (2006)

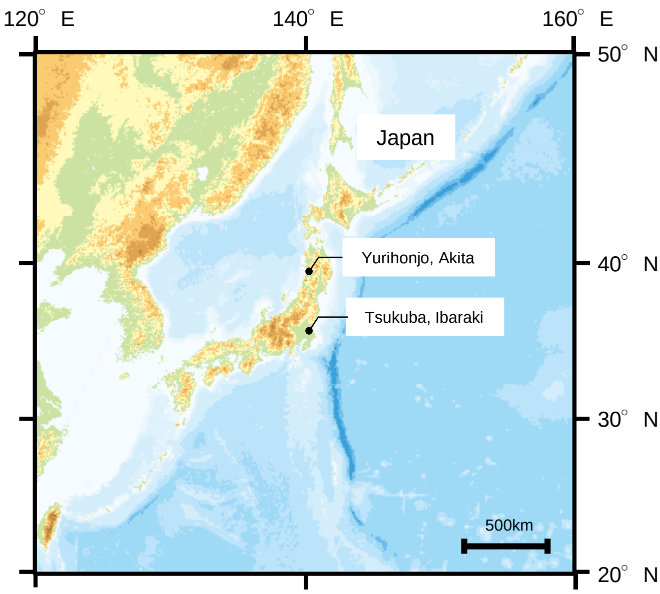

Figure S1. Sampling locations at the Japan Automobile Research Institute (Tsukuba, Ibaraki) and Akita Prefectural University (Yurihonjo, Akita). This map plots the sampling locations on a standard map provided by the Geospatial Information Authority of Japan.

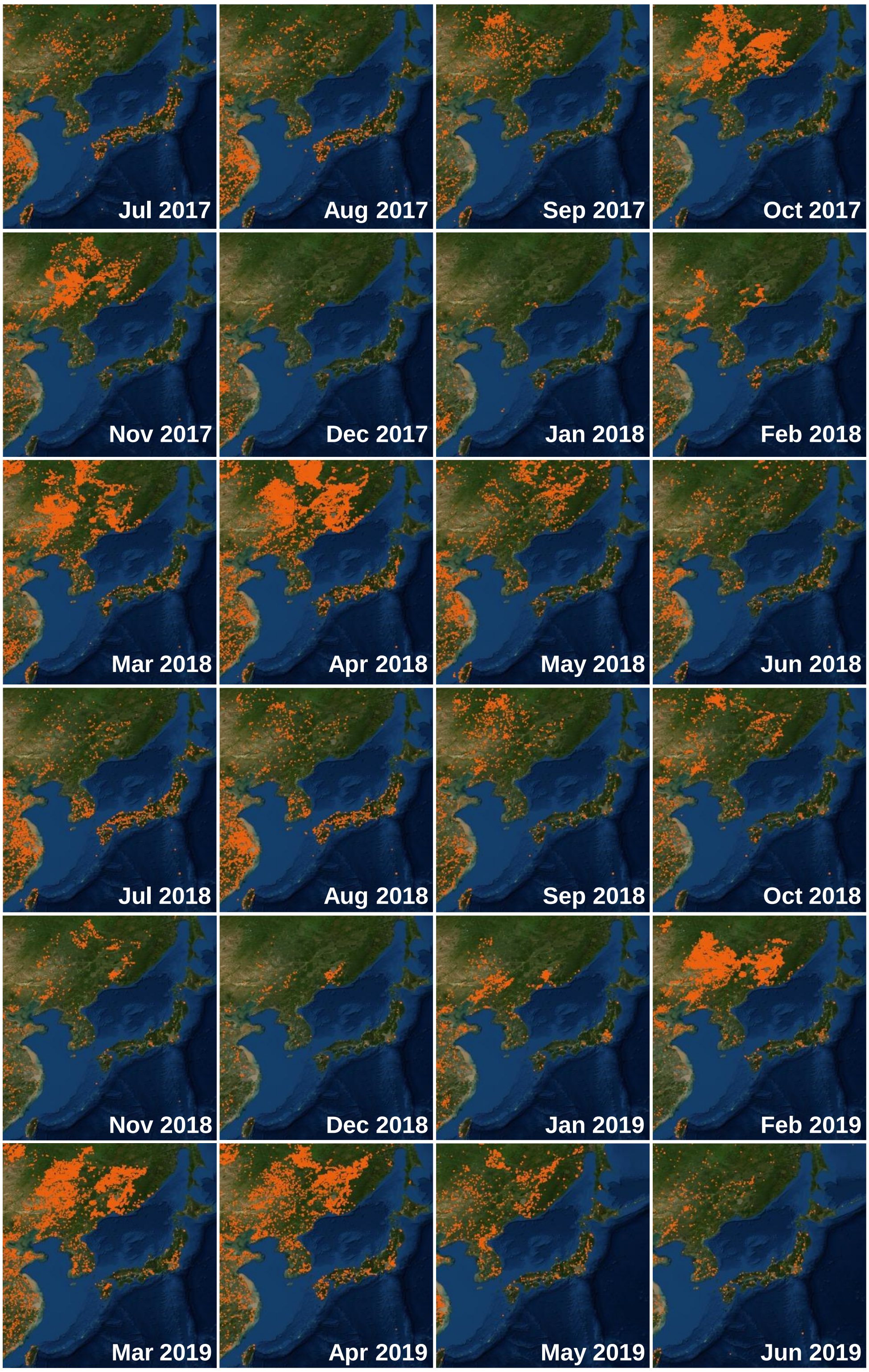

Figure S2. Monthly fire spots from July 2017 to June 2019 as determined by MODIS (moderate resolution imaging spectroradiometer) in Fire Information for Resource Management System (FIRMS) (NASA, 2017). In order to identify burning events, fire spot data were obtained from the National Aeronautics and Space Administration's (NASA) Fire Information for Resource Management System (FIRMS). This data was obtained from the MODIS (moderate resolution imaging spectroradiometer) sensor on NASA's Earth Observing System satellites (Terra and Aqua).

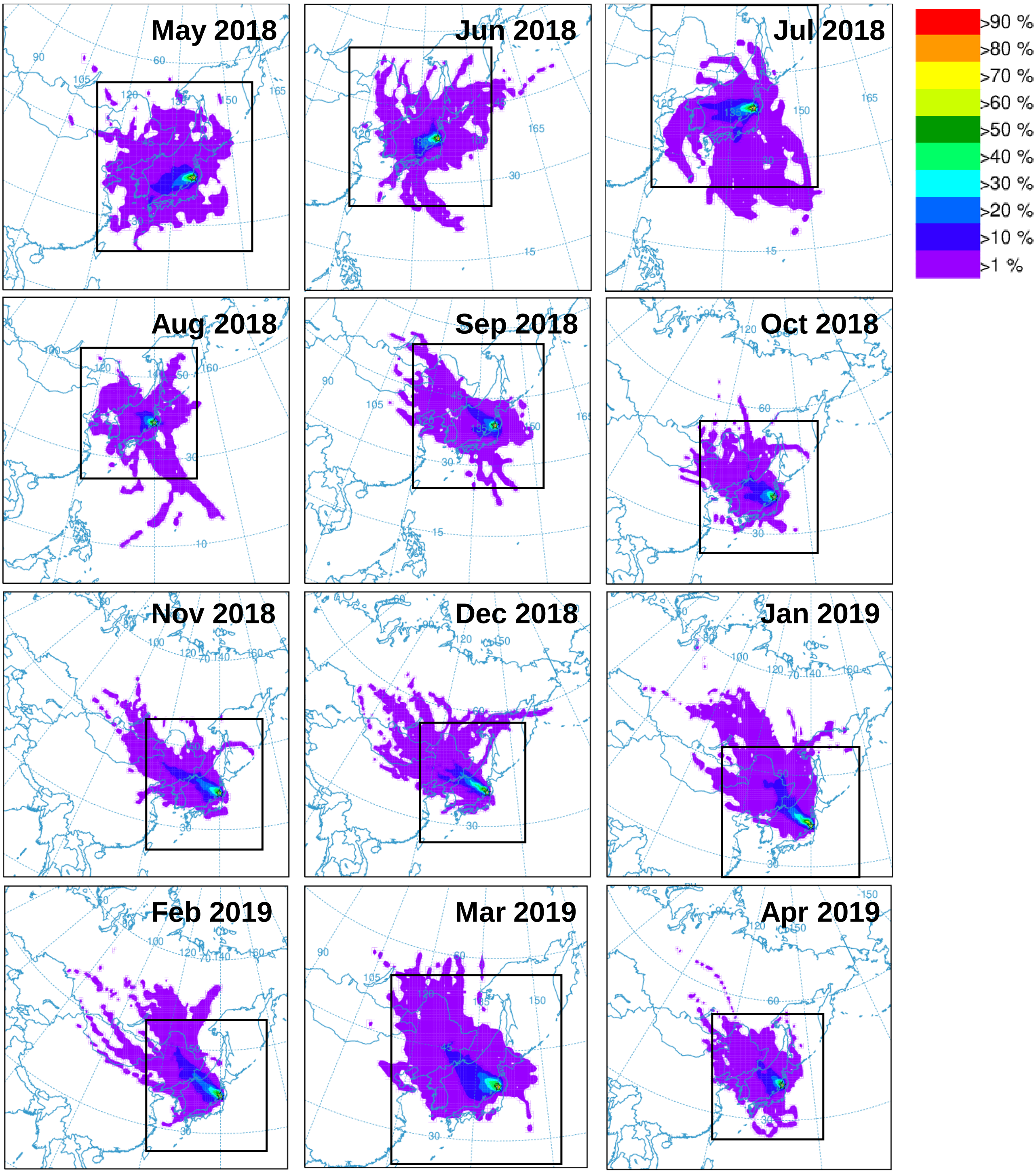

Figure S3. Monthly air masses backward trajectories frequency at Yurihonjo from May 2018 to April 2019. Air mass backward trajectories were calculated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model to understand the source regions and transport pathways (Draxler and Rolph, 2013). We used 72 hours back trajectory with a daily resolution arriving at the Yurihonjo sampling site at 1000 m above ground level. The trajectory frequency option will start a trajectory from a single location and height every 6 hours and then sum the frequency that the trajectory passed over a grid cell and then normalize by either the total number of trajectories or endpoints. A trajectory may intersect a grid cell once or multiple times. The grid resolution was selected as 1.0 deg.

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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