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Mercury isotopic compositions in fine particles and offshore surface seawater in a coastal area of East China: implications for Hg sources and atmospheric transformations

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Table S1 Spearman correlation of $Hg_{PM2.5}$ content with chemical species in $PM_{2.5}$ at the CX and DMS.

Chemical species	CX		DMS	
	r	*p-value*	*r*	*p-value*
Cl^-	**0.411**[a]	0.003	-0.064	0.729
NO_3^-	**0.398**	0.004	-0.004	0.984
SO_4^{2-}	**-0.514**	0.000	-0.063	0.732
Na^+	**-0.319**	0.023	-0.203	0.265
NH_4^+	0.045	0.753	-0.014	0.941
K^+	**0.572**	0.000	0.110	0.550
Ca^{2+}	-0.040	0.782	0.139	0.449
Mg^{2+}	0.063	0.659	-0.293	0.104
OC	**0.436**	0.001	**0.380**	0.032
EC	-0.075	0.600	**0.521**	0.002

[a] *r* values higher than 0.30 are marked in bold

Table S2 Mass concentrations and isotopes of $Hg_{PM2.5}$ at the CX and DMS, as well as concentrations and isotopes of total Hg in the offshore surface seawater.

Date (Y/M/D)	Hg content	$\delta^{202}Hg$ (‰)	2σ	$\Delta^{199}Hg$ (‰)	2σ	$\Delta^{201}Hg$ (‰)	2σ	$\Delta^{200}Hg$ (‰)	2σ
	$Hg_{PM2.5}$ (μg g^{-1})					$Hg_{PM2.5}$ at the CX			
2017.11.8	0.76	-0.48	0.25	-0.10	0.03	-0.07	0.13	0.13	0.07
2017.11.17	0.69	-1.11	0.25	0.03	0.03	0.09	0.13	0.17	0.07
2017.12.19	1.10	0.08	0.25	0.22	0.03	0.34	0.13	0.12	0.07
2017.12.21	0.51	-1.01	0.25	0.35	0.03	0.36	0.13	0.23	0.07
2018.3.21	0.76	-0.72	0.25	-0.17	0.03	-0.06	0.13	0.08	0.07
2018.4.4	0.61	-0.80	0.25	0.52	0.03	0.48	0.13	0.06	0.07
2018.4.25	0.87	-0.62	0.25	0.41	0.03	0.31	0.13	0.09	0.07
2018.4.26	0.54	-0.26	0.25	0.20	0.03	0.26	0.13	-0.01	0.07
2018.5.25	0.90	-0.50	0.25	0.09	0.03	0.20	0.13	0.14	0.07
2018.6.6	0.78	-0.68	0.25	0.15	0.03	0.19	0.13	0.06	0.07
	$Hg_{PM2.5}$ (μg g^{-1})					$Hg_{PM2.5}$ at the DMS			
2017.7.26	0.40	0.34	0.35	0.22	0.08	0.08	0.10	0.27	0.04
2017.8.9	1.19	-0.68	0.35	0.20	0.08	0.46	0.10	0.06	0.04
2017.11.1.	1.81	-0.40	0.35	-0.15	0.08	-0.07	0.10	0.14	0.04
2017.11.8	1.55	0.54	0.35	0.05	0.08	0.02	0.10	0.09	0.04
2017.12.20	0.34	0.49	0.35	0.32	0.08	0.62	0.10	0.17	0.04
2018.1.10	1.96	0.54	0.35	0.40	0.08	0.41	0.10	0.17	0.04
2018.1.17	0.69	-0.44	0.35	-0.07	0.08	-0.20	0.10	0.08	0.04
2018.2.13	0.95	0.43	0.35	0.47	0.08	0.65	0.10	0.22	0.04

2018.2.7	2.80	1.10	0.35	0.38	0.08	0.59	0.10	0.15	0.04
2018.3.21	0.59	-0.78	0.35	-0.22	0.08	-0.29	0.10	0.07	0.04
	$Hg_{seawater}$ (ng L^{-1})			Total Hg in the surface seawater					
2017.7.7	10.94	-0.32	0.15	-0.03	0.08	-0.01	0.10	0.11	0.04
2017.7.19	4.66	-0.32	0.15	0.12	0.08	0.09	0.10	0.04	0.04
2017.8.3	6.19	-0.67	0.15	-0.32	0.08	-0.25	0.10	-0.08	0.04
2017.8.31	6.41	-0.32	0.15	-0.03	0.08	0.02	0.10	0.02	0.04
2017.9.19	16.06	-0.31	0.15	0.19	0.08	0.10	0.10	0.10	0.04
2017.9.28	10.12	-0.40	0.15	0.02	0.08	0.05	0.10	-0.01	0.04
2017.10.25	23.55	-0.33	0.15	-0.01	0.08	-0.01	0.10	0.01	0.04
2017.11.14	15.84	-0.48	0.15	-0.04	0.08	0.13	0.10	-0.01	0.04
2017.11.28	12.51	-0.22	0.15	0.05	0.08	-0.04	0.10	0.02	0.04
2017.12.18	10.02	-0.21	0.15	0.07	0.08	0.08	0.10	0.06	0.04
2017.12.29	8.80	-0.54	0.15	-0.21	0.08	-0.08	0.10	0.04	0.04
2018.1.16	11.06	0.16	0.15	-0.01	0.08	0.02	0.10	0.05	0.04
2018.1.30	10.97	-0.53	0.15	-0.03	0.08	-0.01	0.10	0.08	0.04
2018.3.20	10.25	-0.49	0.15	-0.03	0.08	0.00	0.10	0.08	0.04
2018.3.30	14.60	-0.40	0.15	-0.05	0.08	0.02	0.10	0.07	0.04
2018.4.16	6.81	-0.19	0.15	-0.09	0.08	-0.10	0.10	0.04	0.04
2018.4.27	6.80	-0.20	0.15	0.13	0.08	0.06	0.10	0.03	0.04
2018.5.14	6.24	-0.43	0.15	-0.07	0.08	-0.04	0.10	0.04	0.04
2018.6.15	1.88	-0.40	0.15	-0.04	0.08	-0.04	0.10	0.07	0.04
2018.6.29	15.93	-0.37	0.15	-0.01	0.08	0.03	0.10	0.06	0.04

Table S3 Summary of isotopic compositions of Hg_{PM} in recent studies

Locations	$\delta^{202}Hg$ (‰)	$\Delta^{199}Hg$ (‰)	$\Delta^{201}Hg$ (‰)	$\Delta^{200}Hg$ (‰)	References
Industrial (CX), Ningbo, China	-0.61 ± 0.35 (-1.11~0.08)	0.17 ± 0.22 (-0.17~0.52)	0.21 ± 0.18 (-0.07~0.48)	0.11 ± 0.07 (-0.01~0.23)	This study
Mountain (DMS), Ningbo, China	0.12 ± 0.63 (-0.78~1.10)	0.16 ± 0.24 (-0.22~0.47)	0.23 ± 0.36 (-0.29~0.66)	0.14 ± 0.07 (0.06~0.28)	
Coast of Grand Bay, Mississippi, USA	-0.87 (-1.61~-0.12)	0.83 (0.36~1.36)	0.73 (0.30~1.20)	0.14 (0.06~0.26)	Rolison et al., 2013
Mt. Dameishan, Ningbo, China	-0.26	-0.07		0.03	Yu et al., 2016
Mt. Waliguan, high altitude site	-0.83 ± 0.39 (-1.41~-0.19)	0.27 ± 0.22 (-0.02~1.05)	0.26 ± 0.18 (-0.06~0.80)	0.07 ± 0.03 (0.01~0.14)	Fu et al., 2019
Mt. Ailao, high altitude site	-0.84 ± 0.29 (-1.38~-0.30)	0.66 ± 0.32 (0.06~1.27)	0.56 ± 0.30 (-0.02~1.19)	0.09 ± 0.05 (-0.05~0.20)	
Mt. Changbai, Forest side	-1.45 ± 0.45 (-2.48~-0.75)	0.36 ± 0.34 (-0.15~1.17)	0.23 ± 0.31 (-0.23~0.99)	0.10 ± 0.04 (0.02~0.19)	
Huaniao island, marine boundary layer site	-0.88 ± 0.31 (-1.37~-0.30)	0.35 ± 0.34 (-0.20~1.08)	0.17 ± 0.29 (-0.34~0.79)	0.08 ± 0.04 (0.02~0.18)	
Beijing, China	-0.71 (-2.18~0.51)	0.05 (-0.53~0.57)	0.02	0.09 (0.02~0.17)	Huang et al., 2016
Central Beijing, China	-1.00 (-1.39~-0.51)	-0.04 (-1.39~0.46)	-0.09 (-1.41~0.41)		Huang et al., 2020
Beijing, China	-0.53 (-1.49~0.55)	0.14 (-0.53~1.04)		0.07 (-0.02~0.21)	Huang et al., 2019
Beijing, China	-1.11 (-1.56~-0.71)	-0.31 (-1.12~0.37)	-0.35 (-1.17~0.19)	0.02 (-0.03~0.07)	Xu et al., 2019

Changchun, China	-1.60 (-2.46~-0.65)	-0.12 (-0.54~0.21)	-0.17 (-0.54~0.09)	0.03	(-0.06~0.14)	
Chengdu, China	-0.74 (-1.75~1.07)	-0.02 (-0.45~0.15)	-0.11 (-0.47~0.10)	0.03	(-0.03~0.11)	
Guiyang, China	-1.42	0.19	0.10		0.06	Huang et al., 2015
Xi'an, China	-0.80 (-1.45~0.08)	-0.02 (-0.34~0.25)				Xu et al., 2017
Downtown, Guiyang, China	-1.26	0.05			0.01	
Downtown, Guiyang, China	-0.42	0.04			0.03	Yu et al., 2016
Suburban/industrial, Guiyang, China	-0.64	0.02			0.02	
Landfill site, Urban Kolkata, India	-0.58	0.03	0.04		0.03	
Industrial site, Urban Kolkata, India	-2.41	-0.15	-0.13		0.03	Das et al., 2016
Traffic site, Urban Kolkata, India	-1.65	0.02	-0.01		0.04	
Near CFPP, Xiamen, China	-1.01 (-3.05~0.01)	-0.27 (-0.77~0.20)	-0.26 (-0.60~0.20)	0.10	(-0.06~0.27)	Huang et al., 2018

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Table S4 Spearman correlation of $\delta^{202}Hg$ and $\Delta^{199}Hg$ with chemical components contents in $PM_{2.5}$ at the CX and DMS.

Components	CX				DMS			
	$\delta^{202}Hg$		$\Delta^{199}Hg$		$\delta^{202}Hg$		$\Delta^{199}Hg$	
	r	*p-value*	*r*	*p-value*	*r*	*p-value*	*r*	*p-value*
Levoglucosan	**-0.820**[a]	0.007	0.109	0.781	0.480	0.160	0.430	0.214
Cl^-	**-0.778**	0.014	0.201	0.604	0.128	0.725	-0.152	0.676
NO_3^-	0.418	0.229	-0.042	0.907	0.407	0.243	0.139	0.701
SO_4^{2-}	-0.564	0.090	-0.188	0.603	-0.614	0.059	-0.309	0.385
Na^+	-0.055	0.881	0.370	0.293	-0.571	0.084	-0.406	0.244
NH_4^+	0.030	0.934	-0.612	0.060	-0.128	0.725	-0.176	0.627
K^+	-0.055	0.881	0.042	0.907	0.396	0.257	**0.784**	0.007
Ca^{2+}	0.164	0.651	**0.758**	0.011	0.061	0.867	0.213	0.555
Mg^{2+}	0.188	0.603	**0.818**	0.004	0.000	1.000	-0.188	0.603
OC	0.036	0.920	0.462	0.179	0.036	0.920	0.309	0.385
EC	-0.200	0.580	-0.103	0.777	-0.231	0.521	0.176	0.627

[a] *r* values higher than 0.30 with *p-value* < 0.05 are marked in bold

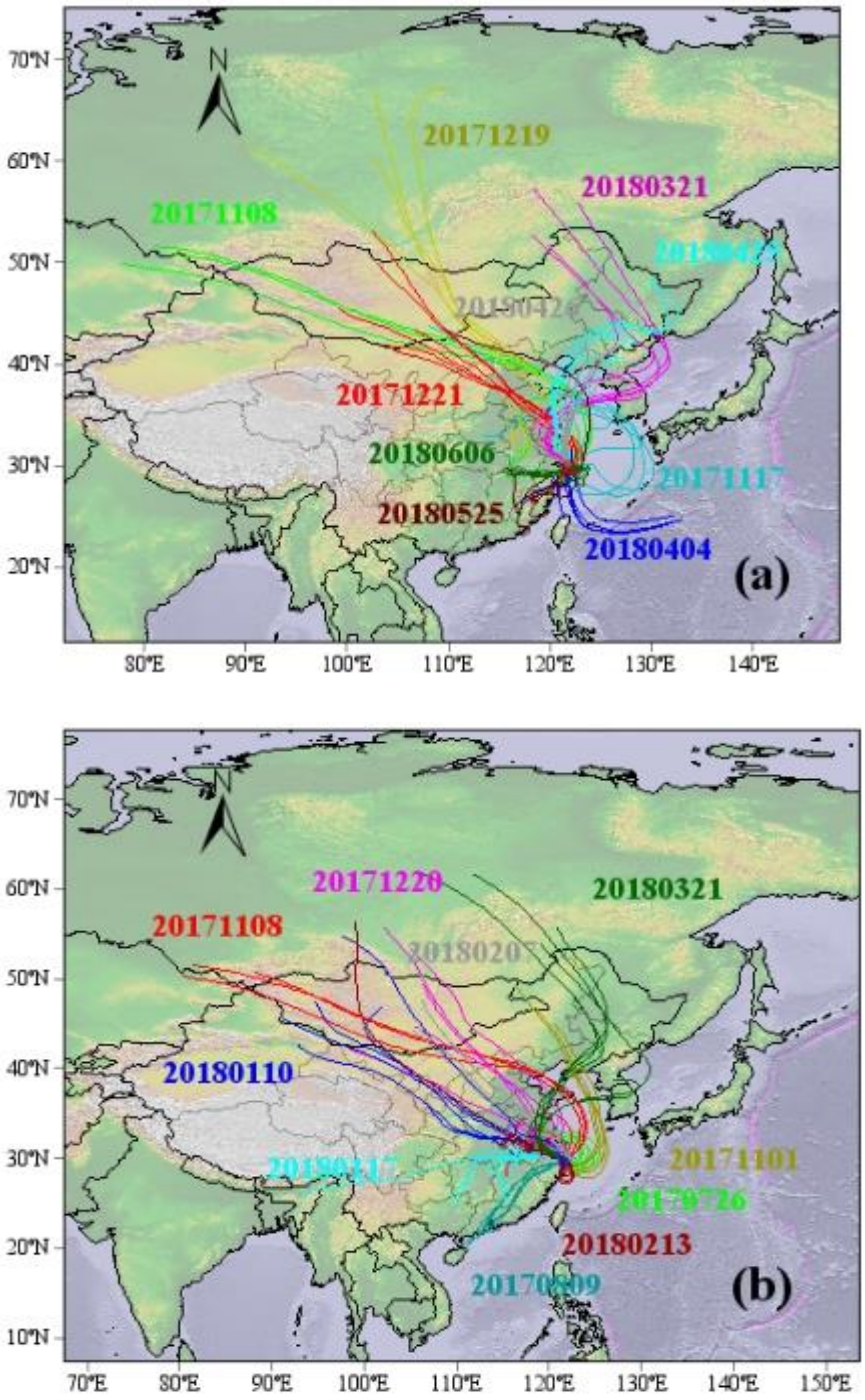

Fig. S1 72-h backward air trajectories associating with $PM_{2.5}$ samples for Hg isotopes analysis starting every 4 h arriving at the (a) CX and (b) DMS (6 trajectories for one sample).

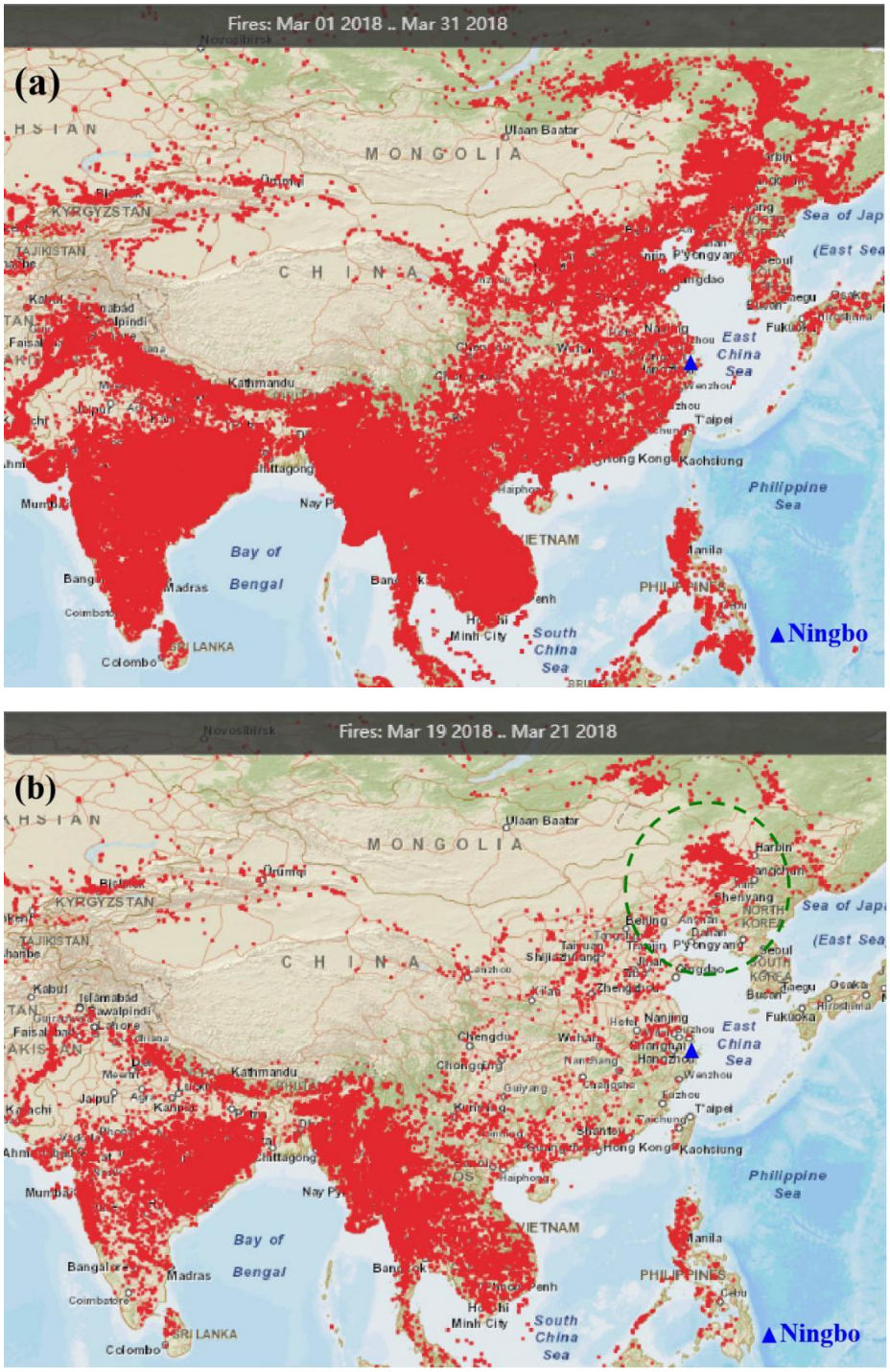

Fig. S2 Fire map in (a) March 2018 and (b) March 19~21, 2018. The study region is marked by blue triangle. *https://firms.modaps.eosdis.nasa.gov/*

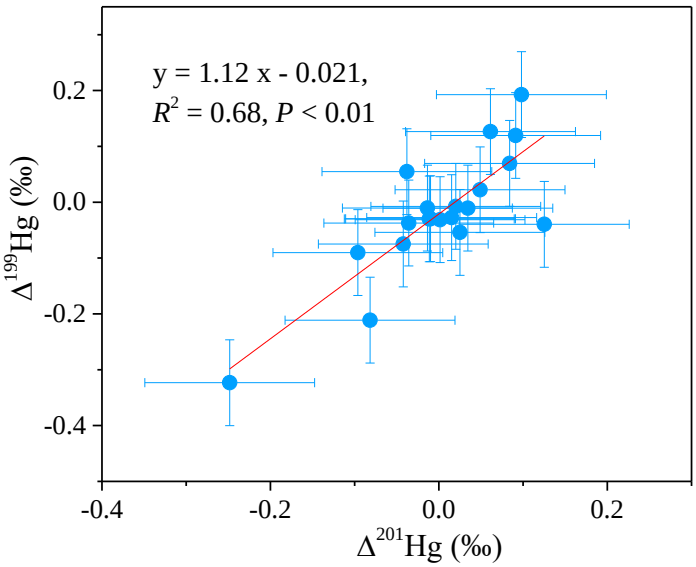

Fig. S3 Linear relationships of Δ^{199}Hg vs. Δ^{201}Hg in surface seawater adjacent to the industrial area.