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Article

Comparative Analysis of $PM_{2.5}$-Bound Polycyclic Aromatic Hydrocarbons (PAHs), Nitro-PAHs (NPAHs), and Water-Soluble Inorganic Ions (WSIIs) at Two Background Sites in Japan

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Text S1. Quartz fiber Filter treatment

To ensure the accuracy of the calculation of $PM_{2.5}$ concentration, quartz fiber filters were put into the desiccator with constant room temperature (21.5 ± 1.5 °C) and humidity (50 ± 5%) for 48 h. Before sampling, each filter was weighed and take the average of twice, then packaged by aluminum foil. After sampling, the filters were put into the desiccator under the same condition as before sampling, also weighed and take the average of twice. After weighing, the filters were packaged by aluminum foil and kept sealed in plastic bags and stored at -25 °C until analyzed. The concentration of each $PM_{2.5}$ sample was calculated as: $PM_{2.5}$ concentration = (weigh $_{after}$ – weigh $_{before}$)/ total volume.

Text S2. Sample pretreatment

For PAHs and NPAHs, one-third of each $PM_{2.5}$ filter (about 130 cm^2) was cut into small pieces and placed in flasks. PAH and NPAH internal standards (pyrene-d_{10} (Pyr-d_{10}), benzo[*a*]pyrene-d_{12}, (BaP-d_{12}, and 2-fluoro-7-nitrofluorene (FNF)) were added to the filter. Then, benzene: ethanol (3: 1, v/v) was added for extracting. After ultrasonic extraction twice, the solution was filtered, and the extracts were washed successively with 5% sodium hydroxide solution, 20% sulfuric acid solution, and distilled water. The solution was concentrated with a rotary evaporator to 100 μL after adding dimethyl sulfoxide. Then, adding ethanol to make the residue up to 1 mL. Finally, the solution was filtered into a vial by a 0.45 μm membrane filter (HLC-DISK13, Kanto Chemical CO., Inc., Tokyo, Japan). After pretreatment, the analysis solution was injected into an HPLC and the target PAHs and NPAHs were determined by the fluorescence detection system.

For WSIIs, one-third of each $PM_{2.5}$ filter (about 130 cm^2) was cut into small pieces and placed in polypropylene tubes. Ultrapure water was added to each tube and put at room temperature for 10 min. After ultrasonic extraction for 30 min, the solution was transferred through a 0.45 μm filter (SLHPX13NK, Merck Millipore Ltd. Tullagreen, Carrigtwohill, Co. Cork IRELAND) filtered into two portions. After pretreatment, the analysis solution was injected into an ion chromatography system to separately detect anions and cations.

Text S3. Quality control and quality assurance

To ensure the accuracy of chemicals detection, quartz fiber filters were roasted at 600 °C for 4 h. To checking the background contamination during transportation, blank filters were also analyzed, and no target chemicals were determined, indicating there was no contamination during the transport. To check the HPLC and IC analysis methods, PAHs, NPAHs, and WSIIs standard solution had injected into the systems before actual samples detecting, respectively. The calibration curves of all PAHs, NPAHs, and WSIIs had good linearity ($r > 0.998$). Pyr-d_{10} was used to quantify the 4-ring (FR, Pyr, BaA, and Chr) PAHs, BaP-d_{12} was used to quantify the 5- (BbF, BkF, and BaP) and 6-ring (BgPe and IDP) PAHs and FNF was used to quantify the three NPAHs (1-, 2-NPs and 2-NFR). The recoveries of internal standards of PAHs and NPAHs ranged from 80% to 95% in this study. The limit of determination (LOD) of each PAH, NPAH, and WSII are shown in **Table S1**.

Text S4. Calculated methods of cation equivalent (CE) and anion equivalent (AE)

The value of AE and CE were calculated as followed:

$$AE = [SO_4^{2-}]/48 + [NO_3^-]/62 + [Cl^-]/35.5 + [Br^-]/79.9 \quad \text{(Equation 1)}$$

$$CE = [NH_4^+]/18 + [Mg^{2+}]/12.2 + [Ca^{2+}]/20 + [K^+]/39 + [Na^+]/23 \quad \text{(Equation 2)}$$

$[SO_4^{2-}]$, $[NO_3^-]$, $[Cl^-]$, $[Br^-]$, $[NH_4^+]$, $[Mg^{2+}]$, $[Ca^{2+}]$, $[K^+]$, and $[Na^+]$ are the concentrations.

Text S5. Calculated methods of non-sea-salt WSIIs.

90 The value of non-sea-salt (nss-) WSIIs were calculated as followed:

$$[nss\text{-}SO_4^{2-}] = [SO_4^{2-}] - [Na^+] \times 0.2516 \quad \text{(Equation 3)}$$

$$[nss\text{-}Ca^{2+}] = [Ca^{2+}] - [Na^+] \times 0.038 \quad \text{(Equation 4)}$$

$$[nss\text{-}K^+] = [K^+] - [Na^+] \times 0.037 \quad \text{(Equation 5)}$$

91 $[SO_4^{2-}]$, $[Ca^{2+}]$, $[K^+]$, and $[Na^+]$ are the concentrations.

92 **Table S1.** Abbreviation and limit of detection (LOD) of PAHs, NPAHs, and WSIIs.

Species	**Abbreviation**	**LOD** [a]
Fluoranthene	FR	16.2
Pyrene	Pyr	30.3
Benz[*a*]anthracene	BaA	9.1
Chrysene	Chr	34.3
Benzo[*b*]fluoranthene	BbF	55.5
Benzo[*k*]fluoranthene	BkF	8.8
Benzo[*a*]pyrene	BaP	8.8
Benzo[*ghi*]perylene	BgPe	55.3
Indeno[1,2,3-*cd*]pyrene	IDP	82.9
Total PAHs	ΣPAHs	
1-Nitropyrene	1-NP	12.4
2-Nitropyrene	2-NP	6.2
2-Nitrofluoranthene	2-NFR	5.0
Total NPAHs	ΣNPAHs	
Sodium	Na^+	0.28
Ammonium	NH_4^+	0.14
Potassium	K^+	0.43
Calcium	Ca^{2+}	0.13
Magnesium	Mg^{2+}	0.30
Chloride	Cl^-	0.23
Sulfate	SO_4^{2-}	0.02
Nitrate	NO_3^-	0.08
Bromine	Br^-	0.03
Total WSIIs	ΣWSIIs	

93 [a]: Unit of pg/mL for PAHs and NPAHs, μg/mL for WSIIs.

Table S2. Daily concentrations of $PM_{2.5}$, each individual PAH, NPAH, and WSII at WAMS in two sampling periods.

	East Asian winter monsoon period (Period 1)											East Asian summer monsoon period (Period 2)				
	4/10	4/11	4/12	4/13	4/14	4/15	4/16	4/17	4/18	4/19	4/20	6/25	6/26	6/27	6/28	6/29
$PM_{2.5}$ (μg/m^3)	6.90	2.33	4.14	17.2	21.2	6.60	10.9	7.99	4.50	5.82	7.26	9.43	12.4	6.30	5.84	5.47
PAHs (pg/m^3)																
FR	111	57.3	120	143	238	111	115	44.3	38.0	78.6	95.0	6.45	38.4	13.5	6.29	9.23
Pyr	62.9	35.2	70.5	81.0	147	70.8	73.0	26.7	22.3	52.4	57.6	20.3	42.1	23.9	19.0	21.5
BaA	11.4	6.06	11.4	13.0	26.1	18.0	14.9	5.05	4.74	11.6	12.2	1.49	9.29	2.42	0.89	2.04
Chr	31.9	17.6	33.8	45.8	88.2	45.2	55.1	17.3	13.7	24.5	28.0	2.13	20.9	6.08	0.81	1.57
BbF	43.5	22.8	37.6	51.3	103	51.1	56.7	22.9	18.0	28.5	32.5	2.78	32.4	8.84	2.12	5.41
BkF	15.4	8.02	13.1	16.4	35.5	19.5	20.4	7.62	6.24	10.4	12.4	0.68	11.3	2.86	0.39	1.37
BaP	23.2	12.1	18.5	23.1	53.0	30.8	29.5	11.9	9.21	17.2	22.1	1.53	19.2	4.40	0.89	2.61
BgPe	48.8	26.6	42.4	53.7	116	62.4	61.7	25.5	22.3	37.7	44.5	3.35	39.4	7.87	2.23	4.69
IDP	22.9	12.6	18.2	25.4	49.8	26.4	27.7	12.9	9.74	16.3	19.3	2.12	28.3	5.95	1.56	3.36
NPAHs (pg/m^3)																
2-NFR	3.89	1.41	1.12	3.47	10.1	4.29	6.24	2.28	1.16	2.65	1.33	0.71	1.04	0.14	0.07	0.15
2-NP	0.21	0.07	0.11	0.25	0.67	0.25	0.33	0.16	0.08	0.33	0.12	0.04	0.15	0.04	0.02	0.03
1-NP	0.48	0.26	0.54	1.00	2.13	0.87	1.20	0.46	0.23	0.84	0.32	0.12	0.19	**0.02**	**0.02**	**0.02**
WSIIs (μg/m^3)																
Na^+	0.083	**0.008**	0.091	0.116	0.132	0.160	0.106	0.156	0.301	0.233	0.079	**0.006**	**0.006**	**0.006**	**0.006**	0.065
NH_4^+	0.554	0.429	0.435	0.744	1.862	1.294	1.826	0.612	0.407	0.293	0.535	0.639	1.043	0.157	0.270	0.111
K^+	0.131	**0.012**	0.150	0.158	0.180	0.089	0.095	0.068	0.087	0.082	0.113	**0.009**	**0.009**	**0.009**	**0.009**	**0.009**
Ca^{2+}	0.054	**0.003**	0.063	0.219	0.141	0.054	0.046	0.086	0.047	0.026	**0.003**	**0.003**	**0.003**	**0.003**	**0.003**	**0.003**
Mg^{2+}	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	**0.006**	**0.006**	**0.006**	**0.006**	**0.006**
Cl^-	0.051	**0.006**	0.059	0.068	0.085	0.006	0.006	0.094	0.141	0.087	0.039	**0.005**	**0.005**	**0.005**	**0.005**	**0.005**
Br^-	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	0.009	0.013	0.004	0.004	**0.001**
NO_3^-	0.206	0.056	0.214	0.454	0.827	0.120	0.221	0.069	0.095	0.172	0.104	0.042	0.029	**0.002**	**0.002**	0.015
SO_4^{2-}	1.418	1.195	1.278	2.193	4.825	3.898	5.151	2.040	1.689	1.074	1.510	2.054	2.992	0.512	0.772	0.476

Bold font means concentrations calculated by half of LOD to represent species which below LOD.

Table S3. Daily concentrations of $PM_{2.5}$, each individual PAH, NPAH, and WSII at FAMS in two sampling periods.

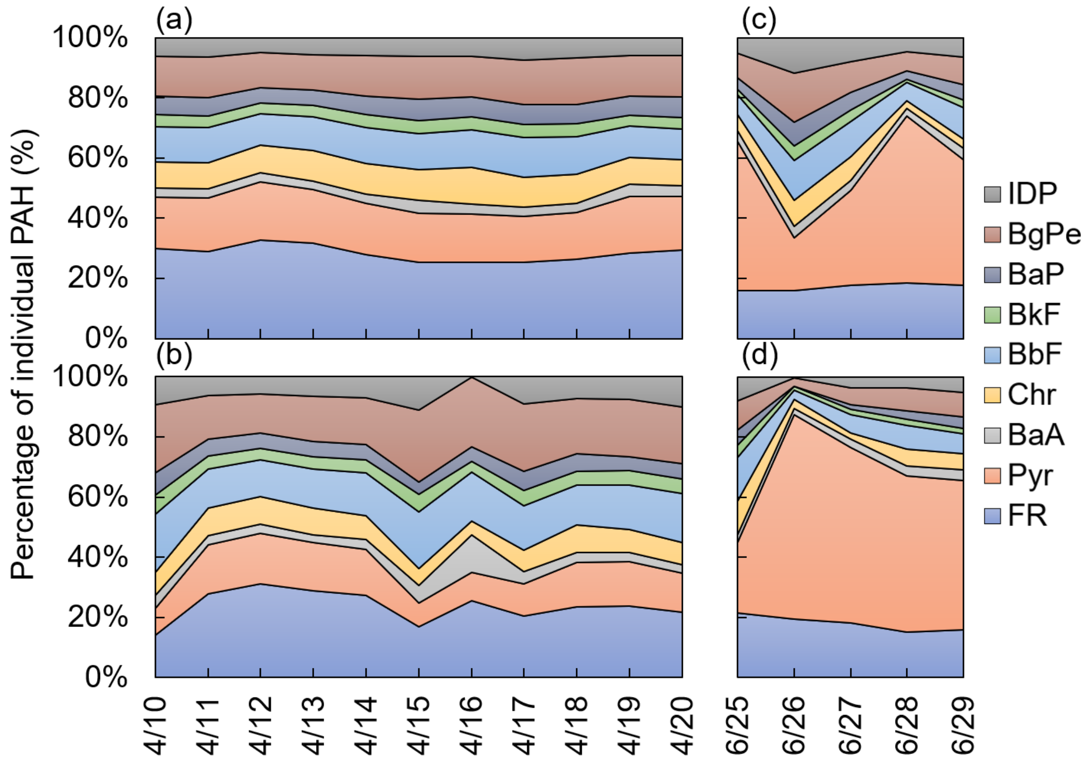
0	East Asian winter monsoon period (Period 1)											East Asian summer monsoon period (Period 2)				
	4/10	4/11	4/12	4/13	4/14	4/15	4/16	4/17	4/18	4/19	4/20	6/25	6/26	6/27	6/28	6/29
$PM_{2.5}$ (μg/m^3)	20.1	16.7	23.6	19.1	14.9	15.8	12.0	8.90	27.9	78.5	17.5	7.51	2.79	5.39	5.49	3.61
PAHs (pg/m^3)																
FR	68.5	226	216	158	44.8	9.74	7.01	40.5	143	98.8	63.9	35.6	5.55	6.24	5.99	6.42
Pyr	42.7	133	119	87.4	25.0	4.66	2.63	21.5	89.8	60.7	38.8	39.2	19.5	19.9	20.5	20.2
BaA	21.7	24.9	20.1	13.7	5.07	3.37	3.44	8.08	20.8	12.5	8.01	4.04	0.55	1.01	1.23	1.48
Chr	37.2	73.7	65.4	49.3	13.2	3.33	1.24	14.3	55.1	31.9	21.8	18.9	0.86	0.67	2.22	2.12
BbF	94.1	105	83.9	70.3	23.0	10.8	4.50	29.6	80.5	61.7	48.3	24.0	0.86	2.09	3.09	2.77
BkF	31.2	34.3	27.2	22.6	7.35	3.48	0.98	10.1	28.5	19.9	14.7	6.87	0.37	0.58	0.89	0.68
BaP	35.3	45.8	34.9	27.1	8.19	2.33	1.38	12.6	35.2	18.5	15.2	8.44	**0.05**	0.53	1.09	1.52
BgPe	111	118	92.1	82.4	25.5	13.9	6.33	44.3	111	79.6	55.6	16.3	0.79	1.97	3.03	3.33
IDP	44.7	51.2	39.3	35.7	11.3	6.49	**0.07**	18.2	44.7	31.1	29.7	13.1	**0.08**	1.23	1.40	2.11
NPAHs (pg/m^3)																
2-NFR	9.92	8.61	6.62	6.00	1.49	0.34	0.14	5.94	9.07	0.89	1.00	0.63	0.08	0.10	0.21	0.23
2-NP	0.70	0.84	10.6	0.96	0.11	0.03	0.02	0.36	0.64	1.25	0.17	0.11	0.06	0.06	0.06	0.09
1-NP	2.08	1.51	1.50	1.51	1.43	0.12	**0.02**	0.54	1.74	0.55	0.40	0.13	**0.02**	**0.02**	**0.02**	**0.02**
WSIIs (μg/m^3)																
Na^+	0.159	0.156	0.234	0.087	0.114	0.091	0.061	0.105	0.199	0.178	0.061	**0.007**	0.044	**0.006**	0.054	0.118
NH_4^+	0.510	1.705	0.870	1.433	1.209	1.557	0.604	1.082	1.877	0.521	0.651	1.013	0.223	0.746	1.102	0.635
K^+	0.012	0.164	0.140	0.148	0.109	0.091	0.081	**0.012**	0.134	0.171	0.096	**0.010**	**0.009**	**0.009**	**0.009**	**0.009**
Ca^{2+}	**0.004**	0.080	0.254	0.211	0.141	**0.004**	**0.004**	**0.004**	0.335	1.378	0.215	**0.003**	**0.003**	**0.003**	**0.003**	**0.003**
Mg^{2+}	**0.008**	**0.008**	0.056	**0.008**	**0.008**	**0.008**	**0.008**	**0.008**	0.056	0.093	**0.008**	**0.007**	**0.006**	**0.006**	**0.006**	**0.006**
Cl^-	0.206	0.042	0.069	**0.006**	**0.006**	**0.006**	**0.006**	**0.006**	0.079	0.283	**0.006**	**0.005**	**0.005**	**0.005**	**0.005**	**0.005**
Br^-	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	**0.001**	0.011	0.004	0.009	0.011	0.007
NO_3^-	0.089	1.563	0.663	0.931	0.055	0.020	0.020	0.280	0.900	1.201	0.165	**0.002**	**0.002**	**0.002**	**0.002**	0.026
SO_4^{2-}	3.008	3.505	2.492	3.442	3.508	4.483	1.811	3.019	4.755	1.981	2.222	2.930	0.783	2.467	3.616	2.174

Bold font means concentrations calculated by half of LOD to represent species which below LOD.

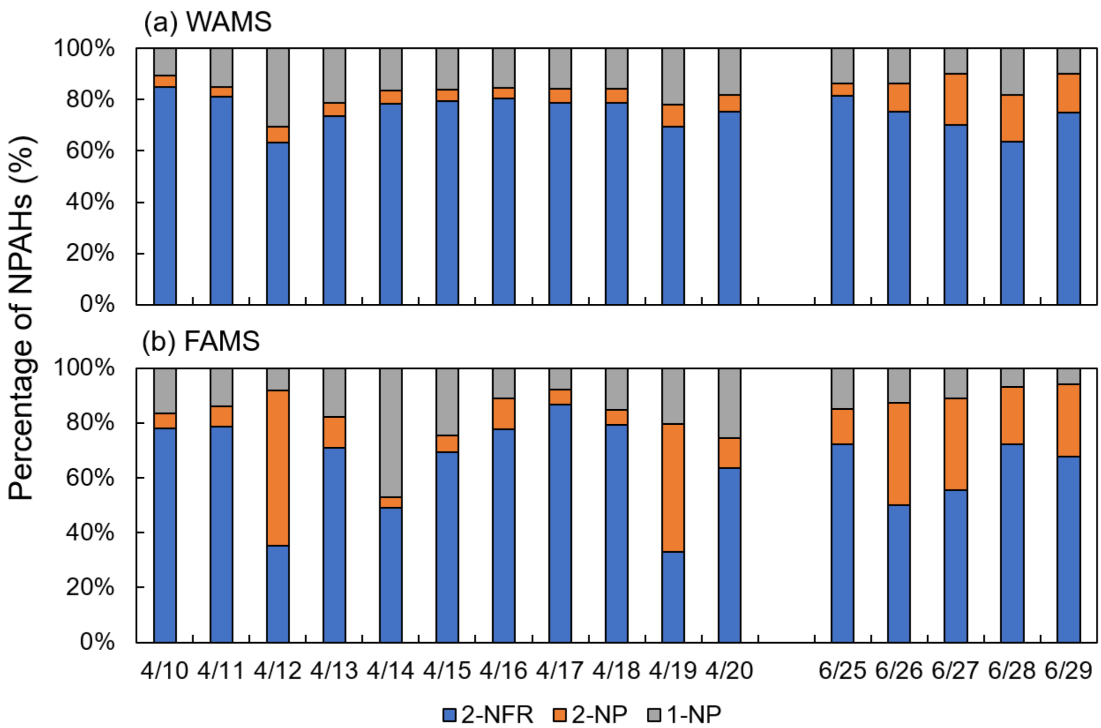
Table S4. Ratios of non-sea salt WSIIs factions at WAMS and FAMS during the East Asian winter monsoon period (Period 1).

	WAMS			FAMS		
	[nss-SO_4^{2-}]/[SO_4^{2-}]	[nss-Ca^{2+}]/[Ca^{2+}]	[nss-K^+]/[K^+]	[nss-SO_4^{2-}]/[SO_4^{2-}]	[nss-Ca^{2+}]/[Ca^{2+}]	[nss-K^+]/[K^+]
4/10	0.99	0.94	0.98	0.99	- [b]	0.50
4/11	- [a]	- [a]	- [a]	0.99	0.92	0.96
4/12	0.98	0.94	0.98	0.98	0.96	0.94
4/13	0.99	0.98	0.97	0.99	0.98	0.98
4/14	0.99	0.96	0.97	0.99	0.97	0.96
4/15	0.99	0.88	0.93	0.99	- [b]	0.96
4/16	0.99	0.91	0.96	0.99	- [b]	0.97
4/17	0.98	0.93	0.92	0.99	- [b]	0.67
4/18	0.96	0.75	0.87	0.99	0.98	0.95
4/19	0.95	0.66	0.89	0.98	1.00	0.96
4/20	0.99	0.12	0.97	0.99	0.99	0.98

[a]: Na^+ below the LOD; [b]: Ca^{2+} below the LOD.



102 **Figure S1.** Percentage of each individual PAH at WAMS ((a) and (c)) and FAMS ((b) and (d)) in the
103 East Asian winter monsoon period (Period 1, 4/10 – 4/20, 2017) and summer monsoon period (Period
104 2, 6/25 – 6/29, 2019).



105 **Figure S2.** Percentage of individual NPAHs at WAMS (a) and FAMS (b) in the East Asian winter
106 monsoon period (Period 1, 4/10 – 4/20, 2017) and summer monsoon period (Period 2, 6/25 – 6/29, 2019).

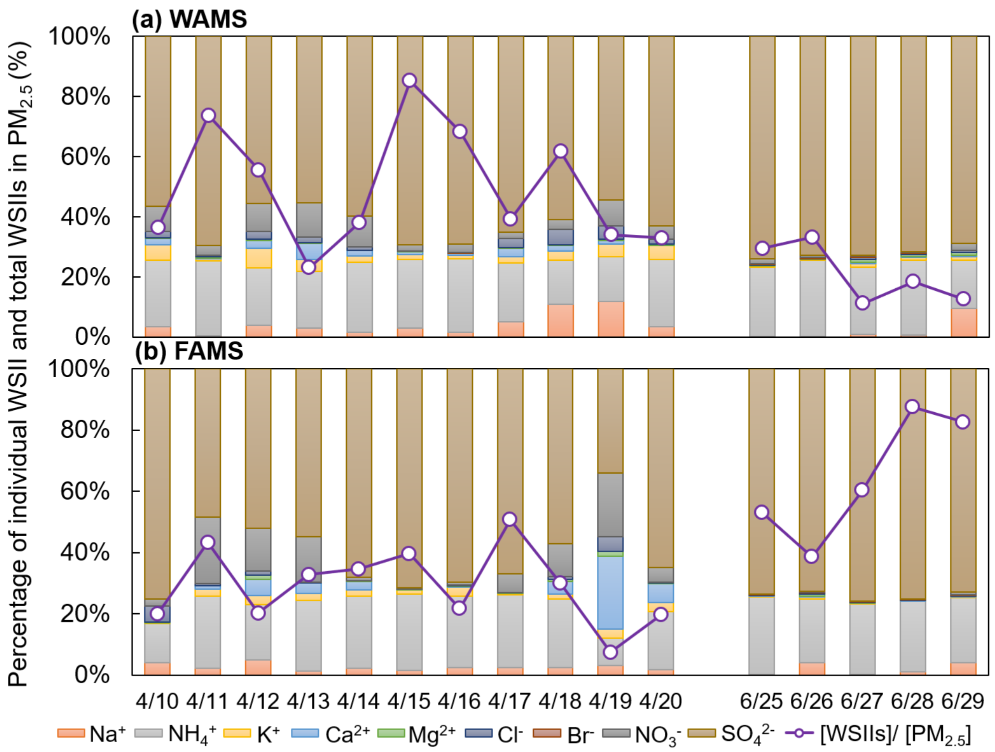


Figure S3. Percentage of individual WSII and [WSIIs]/[$PM_{2.5}$] at WAMS (a) and FAMS (b) in the East Asian winter monsoon period (Period 1, 4/10 – 4/20, 2017) and summer monsoon period (Period 2, 6/25 – 6/29, 2019).

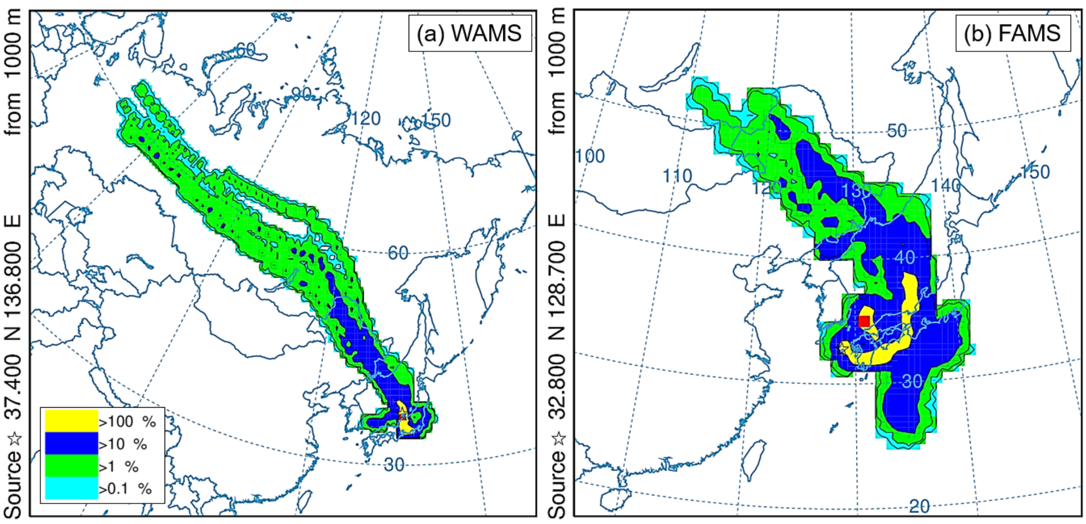


Figure S4. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 10, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

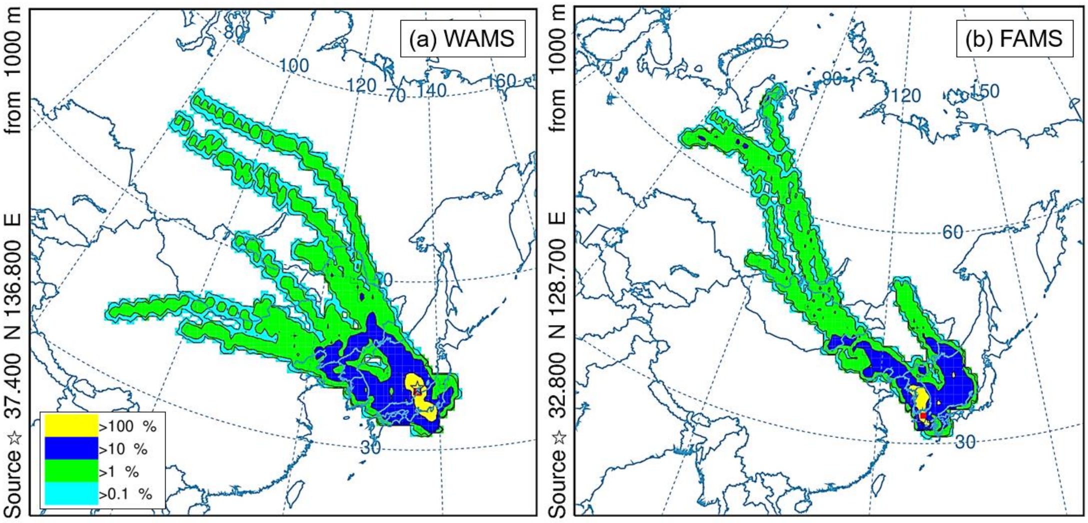


Figure S5. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 11, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

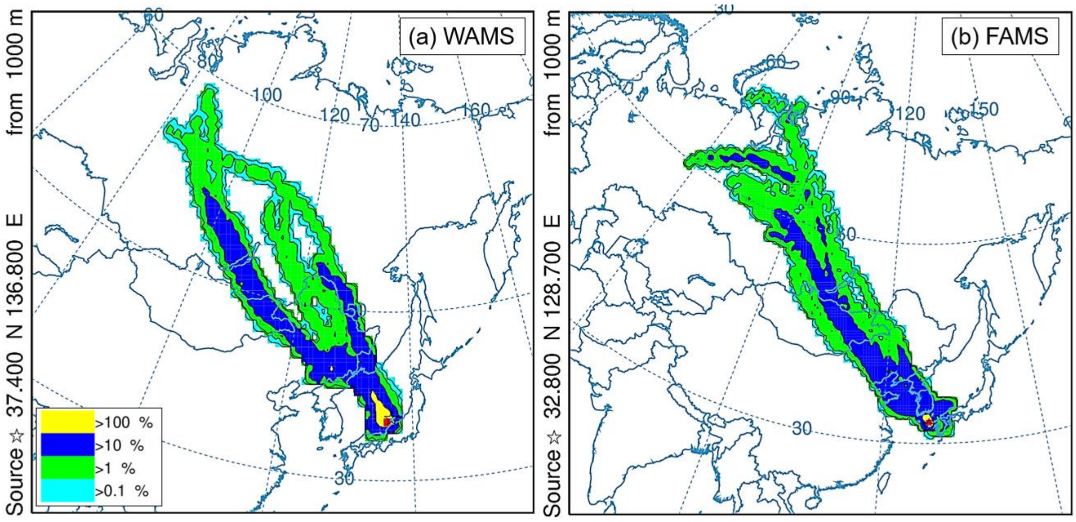


Figure S6. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 12, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

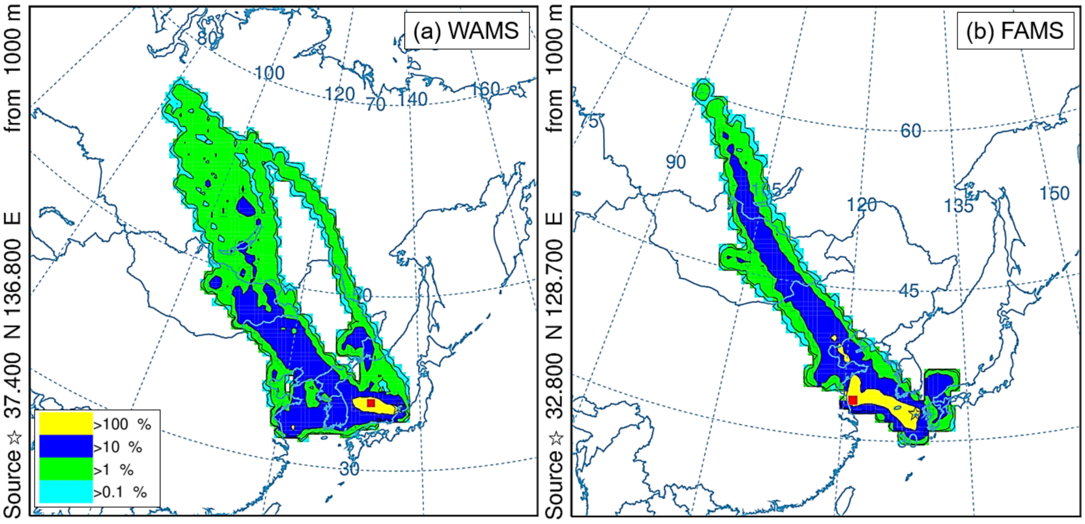


Figure S7. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 13, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

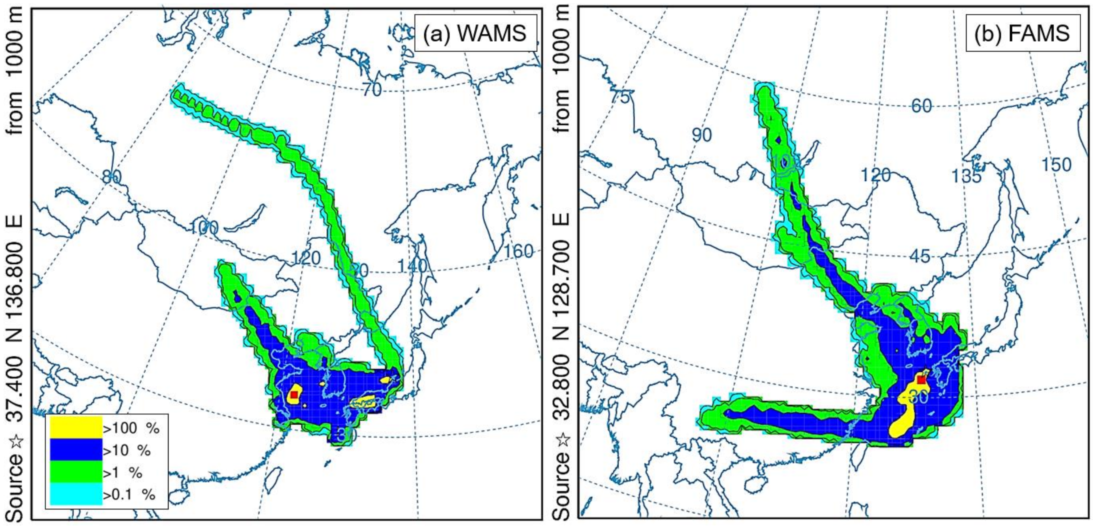


Figure S8. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 14, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

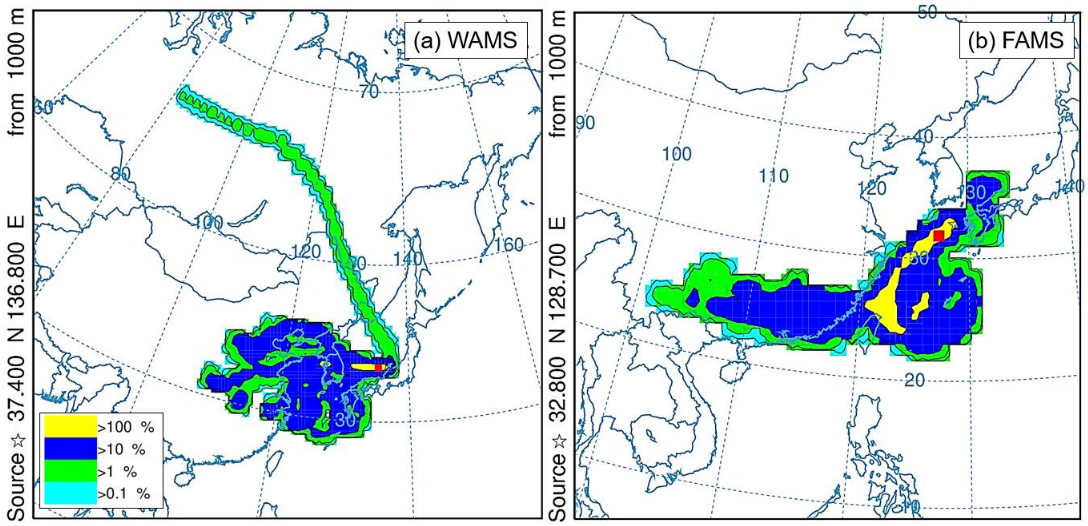


Figure S9. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 15, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

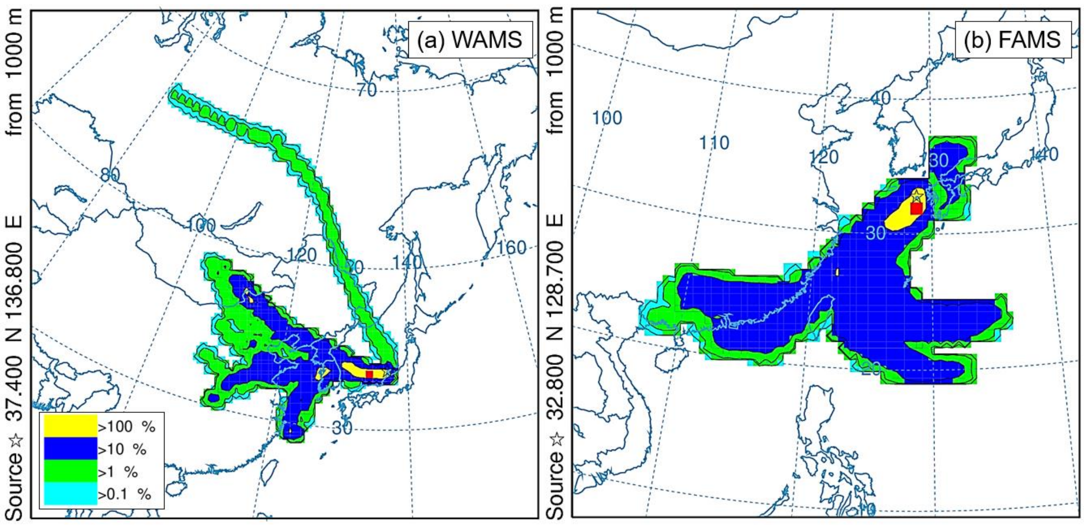


Figure S10. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 16, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

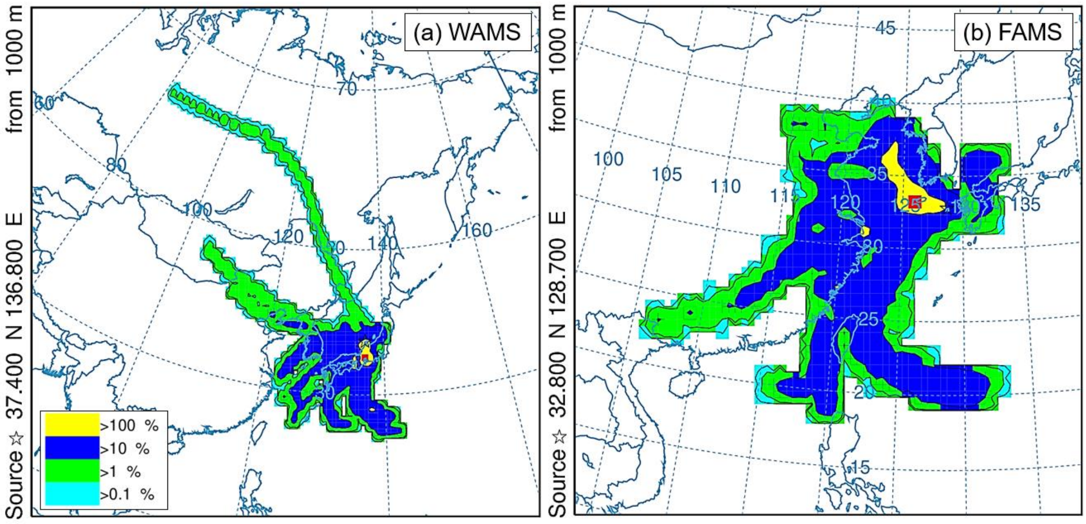


Figure S11. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 17, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

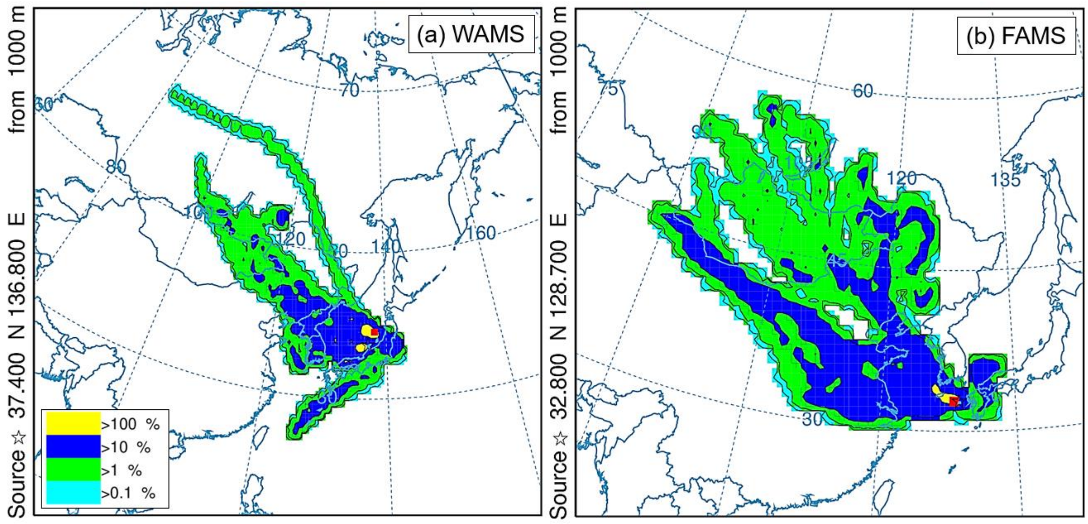


Figure S12. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 18, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

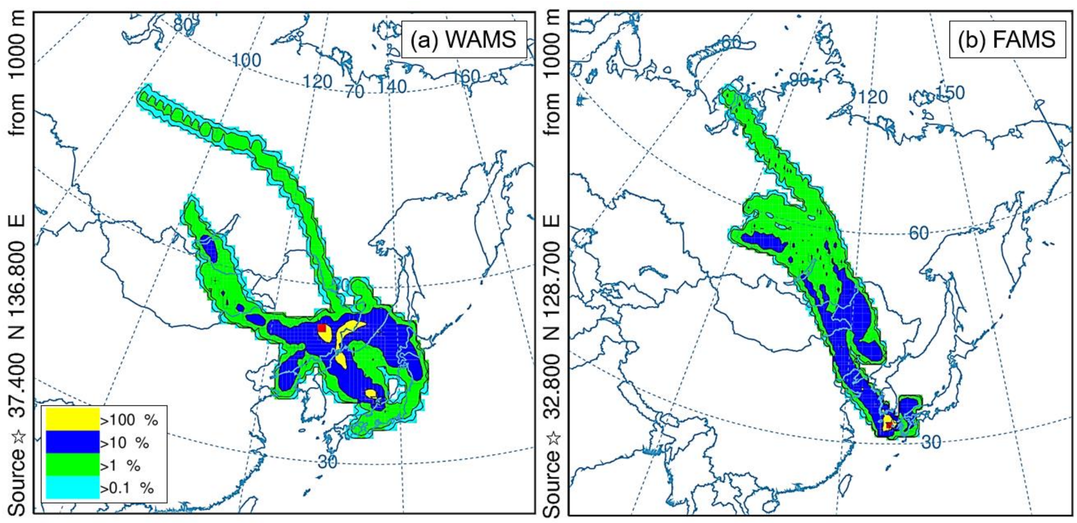


Figure S13. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 19, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

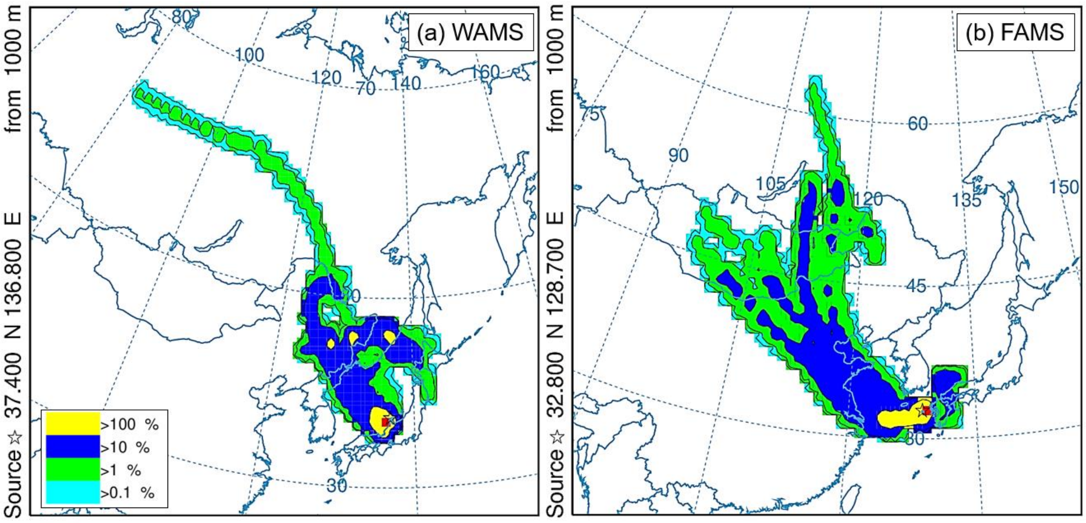


Figure S14. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on April 20, 2017. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

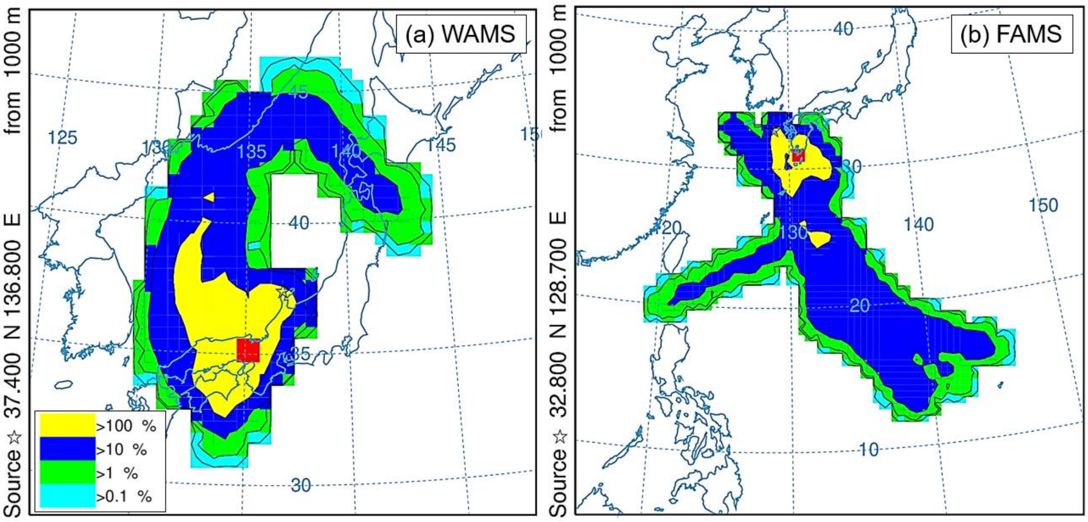


Figure S15. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on June 25, 2019. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

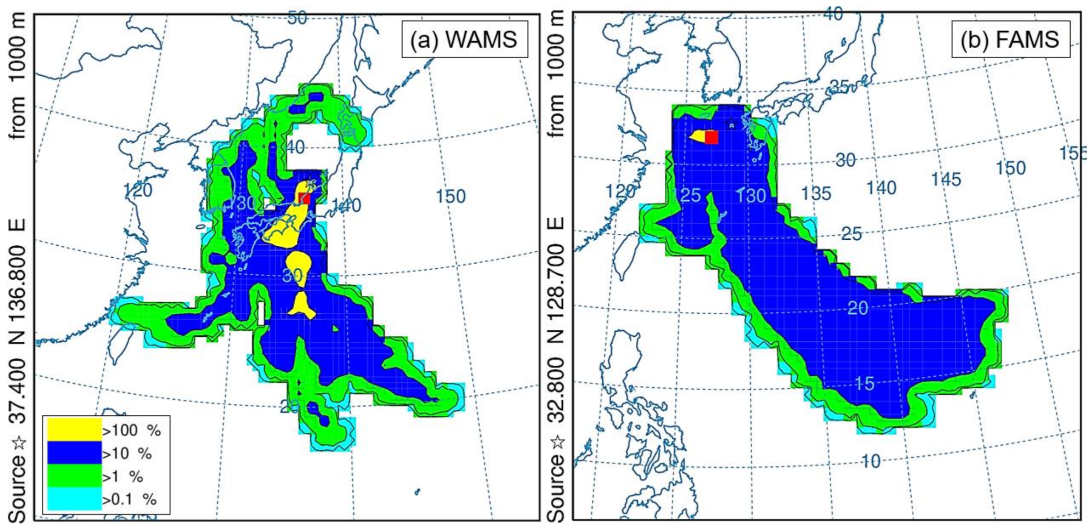


Figure S16. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on June 26, 2019. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

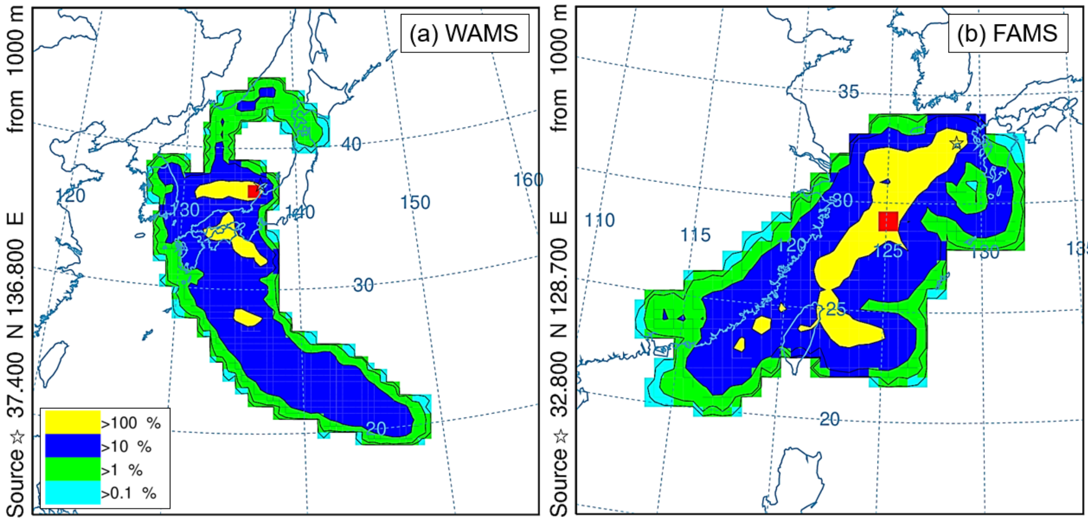


Figure S17. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on June 27, 2019. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

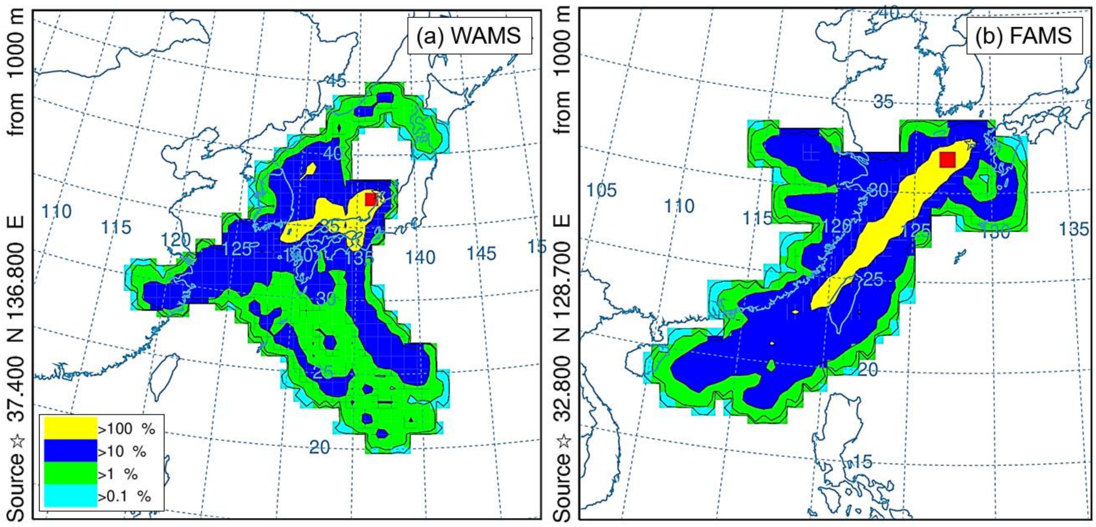


Figure S18. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on June 28, 2019. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

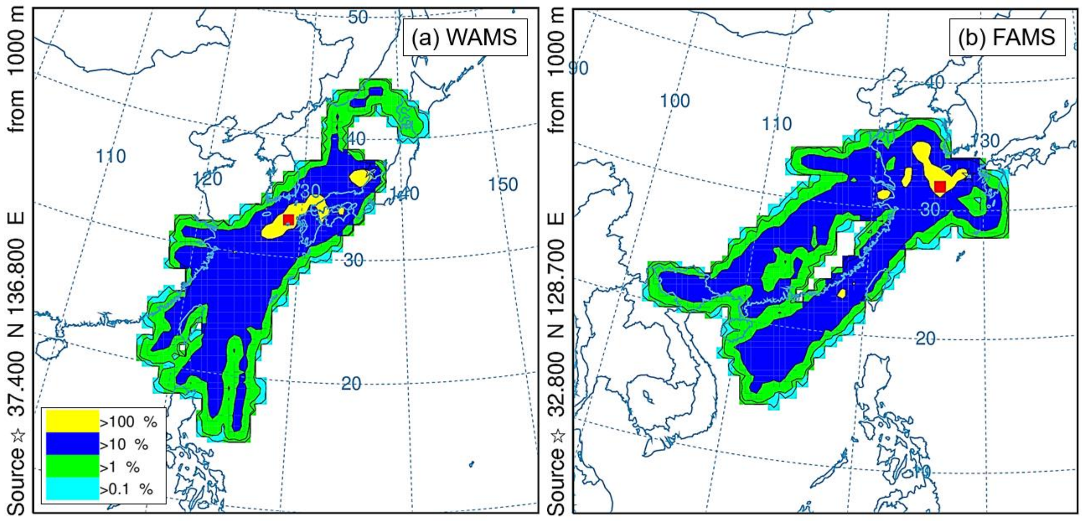


Figure S19. Frequency analysis of backward trajectories at WAMS (a) and FAMS (b) on June 29, 2019. (☆) means sampling sites at WAMS and FAMS, (■) means the longest of air mass residence area.

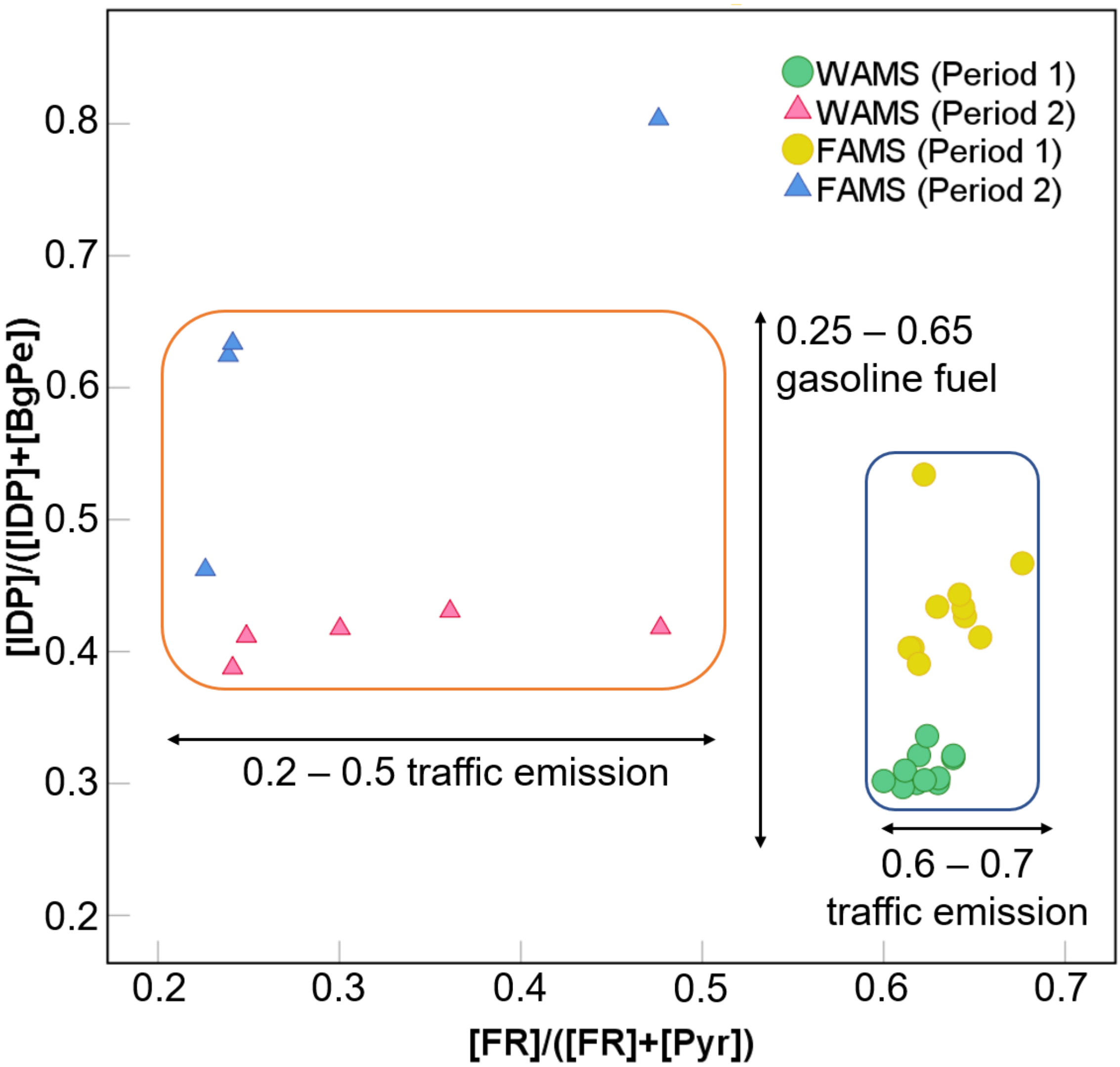


Figure S20. Ratios of [FR]/([FR]+[Pyr]) and [IDP]/([IDP]+[BgPe]) at WAMS and FAMS in the East Asian winter monsoon period (Period 1, 4/10 – 4/20, 2017) and summer monsoon period (Period 2, 6/25 – 6/29, 2019).