



Supplement of

Atmospheric chemical processing dictates aerosol aluminum solubility: insights from field measurement at two locations in Northern China

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S1. Sampling sites

At Xi'an, sampling in spring, summer and autumn was conducted on a building roof (~40 m above the ground level) at Shaanxi University of Science and Technology (34°22'N, 108°58'E). This site is surrounded by residential areas, with no major industrial sources nearby. Sampling in winter was carried out on a building roof (~10 m above the ground level) at the Institute of Earth Environment, Chinese Academy of Sciences (34°13'N, 108°53'E). This site is surrounded by commercial and residential areas. Both sampling sites are urban sites (Zhang et al., 2023).

At Qingdao, sampling was conducted on the rooftop of a 5-floor building in northeastern Qingdao (36°20'N, 120°39'E). This rural/suburban site, which is ~28 m above the ground level and ~1.3 km from the coastline, is surrounded by some residence and office buildings, villages and agricultural fields (Zhang et al., 2022).

15 **Table S1.** Brief summary of atmospheric sampling at Xi'an and Qingdao.

Location	Season	Sample period	Number of samples
Xi'an	spring	01 to 30 April 2021	28
	summer	12 July to 14 August 2021	32
	autumn	07 October to 06 November	30
	winter	01 November to 30 December 2020	36
Qingdao	spring	20 March to 22 April 2023	28
	summer	20 June to 26 July 2022	26
	autumn	04 to 31 October 2022	27
	winter	23 November to 24 December 2022	25

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19 **Table S2.** Summary of total aerosol Al concentrations ($\mu\text{g}/\text{m}^3$) measured over East Asian and the Pacific Ocean.

Location	Geographic coordinates	Sampling period	Size range	Average	Range	reference
Aksu, China	40°16'N, 80°28'E	May to June 2001	TSP	24	/	Zhang et al. (2003)
Urumqi, China	43°49'N, 87°37'E	January 2004 to March 2007	TSP	8.05	/	Li et al. (2008)
Wuhai, China	39°39'N, 106°49'E	March 2021	PM _{2.5}	7.77±19.15	0.00-195.79	Meng et al. (2023)
Xi'an, China	34°22'N, 108°58'E	November 2020 to November 2021	$d_p > 1 \mu\text{m}$	1.42±0.86	0.16-3.70	This study
			$d_p < 1 \mu\text{m}$	2.28±2.35	0.14-14.88	
Yulin, China	38°17'N, 109°28'E	Spring 2006-2008	TSP	36.38 (dust)	/	Wang et al. (2011)
				6.91 (non-dust)	/	
Beijing, China	39°97' N, 116°21'E	March to May 2012	PM _{2.5}	3.97±1.26 (dust)	/	Liu and Bei (2016)
				0.76±0.61 (non-dust)	/	
Jinan, China	36°40'N, 117°09'E	March 2021	PM _{2.5}	1.10±2.56	0.00-20.68	Meng et al. (2023)
Qingdao, China	36°20'N, 120°39'E	November 2022 to March 2023	$d_p > 1 \mu\text{m}$	0.56±0.75	0.08-4.01	This study
			$d_p < 1 \mu\text{m}$	1.08±1.67	0.05-9.63	
Huaniao Island, China	30°51'N, 122°40'E	April 2010 to March 2011	TSP	/	0.075-16	Guo et al. (2014)
Higashi-Hiroshima, Japan	34°24'N, 132°42'E	December 2012 to December 2013	TSP	/	0.17-1.716	Sakata et al. (2023)
The Pacific Ocean (Hawaii)	21°15'N, 157°45'W	January and June 2002	TSP	/	0.001-0.056	Measures et al. (2010)
The Pacific Ocean	along 30°N	June to August 2004	TSP	0.078	0.012-0.475	Buck et al. (2013)

Table S3. Overview of total aerosol Al concentrations ($\mu\text{g}/\text{m}^3$) for submicron and supermicron particles at Xi'an and Qingdao.

Location	Season	submicron particles			supermicron particles		
		range	mean	median	range	mean	median
Xi'an	spring	0.14-14.88	4.29 $\pm$ 3.70	3.32	0.25-3.70	1.54 $\pm$ 0.89	1.59
	summer	0.17-2.01	0.95 $\pm$ 0.44	0.90	0.16-1.96	0.96 $\pm$ 0.54	0.97
	autumn	0.20-2.26	1.06 $\pm$ 0.48	1.03	0.22-2.75	1.22 $\pm$ 0.72	1.07
	winter	0.32-6.11	2.92 $\pm$ 1.47	3.02	0.31-3.53	1.91 $\pm$ 0.93	1.91
Qingdao	spring	0.18-9.63	1.88 $\pm$ 2.51	0.93	0.29-3.69	1.04 $\pm$ 1.12	0.48
	summer	0.05-0.85	0.35 $\pm$ 0.22	0.35	0.08-0.63	0.33 $\pm$ 0.18	0.37
	autumn	0.27-1.32	0.65 $\pm$ 0.22	0.63	0.13-0.57	0.31 $\pm$ 0.12	0.30
	winter	0.40-9.17	1.40 $\pm$ 1.82	0.81	0.12-4.01	0.51 $\pm$ 0.77	0.29

Table S4. Overview of dissolved aerosol Al concentrations (ng/m³) for submicron and supermicron particles at Xi'an and Qingdao.

Location	Season	submicron particles			supermicron particles		
		range	mean	median	range	mean	median
Xi'an	spring	0.3-359.4	65.4±79.2	40.1	1.2-47.2	23.1±10.9	24.8
	summer	3.3-100.9	23.2±23.4	17.3	3.6-37.2	15.0±8.7	13.1
	autumn	6.0-120.2	22.6±20.1	19.3	1.2-37.0	13.2±8.6	12.5
	winter	3.1-97.3	25.6±19.8	22.1	9.2-43.7	19.6±8.2	17.5
Qingdao	spring	2.5-20.9	8.7±5.8	6.2	1.1-41.3	8.8±10.8	5.3
	summer	0.4-30.6	10.2±8.2	8.9	1.3-40.2	12.8±11.1	9.4
	autumn	1.3-23.0	6.0±4.8	5.2	0.3-49.0	7.9±10.5	3.5
	winter	1.8-63.3	14.5±15.2	9.4	1.4-54.9	12.8±12.9	8.9

Table S5. Overview of aerosol Al solubility (%) for submicron and supermicron particles at Xi'an and Qingdao.

Location	Season	submicron particles			supermicron particles		
		range	mean	median	range	mean	median
Xi'an	spring	0.11-7.74	1.46±1.48	1.01	0.45-4.84	1.75±1.04	1.38
	summer	0.74-9.10	2.40±2.03	1.69	0.76-4.71	1.83±0.95	1.59
	autumn	1.21-7.36	2.23±1.40	1.82	0.25-2.64	1.17±0.54	1.04
	winter	0.31-3.55	0.99±0.75	0.74	0.43-7.70	1.42±1.47	1.01
Qingdao	spring	0.14-4.23	0.93±1.02	0.61	0.06-8.91	1.58±2.29	0.54
	summer	0.11-11.11	3.27±2.55	2.33	0.69-12.79	4.55±3.61	3.56
	autumn	0.18-3.44	1.01±0.86	0.69	0.18-14.77	2.74±3.56	1.17
	winter	0.32-5.03	1.26±1.05	0.93	0.30-23.35	4.21±5.35	2.39

Table S6. Overview of total Al ($\mu\text{g}/\text{m}^3$), dissolved Al (ng/m^3) and Al solubility (%) under different weather conditions for submicron and supermicron particles at Xi'an.

submicron particles				supermicron particles		
total Al	range	average	median	range	average	median
entire	0.14-14.88	2.28 $\pm$ 2.35	1.40	0.16-3.70	1.42 $\pm$ 0.86	1.31
clean	0.28-6.73	1.67 $\pm$ 1.39	1.30	0.16-2.49	1.07 $\pm$ 0.58	1.13
dust	2.75-14.88	8.54 $\pm$ 4.24	8.40	0.92-3.70	2.25 $\pm$ 1.06	2.22
haze	0.32-5.94	2.59 $\pm$ 1.32	2.21	0.31-3.53	2.08 $\pm$ 0.91	2.23
dissolved Al	range	average	median	range	average	median
entire	0.3-359.4	33.1 $\pm$ 44.7	20.4	1.15-47.3	17.7 $\pm$ 9.7	16.3
clean	0.3-125.8	28.1 $\pm$ 25.3	21.7	1.21-37.2	14.6 $\pm$ 8.3	13.4
dust	54.1-359.4	146.1 $\pm$ 109.6	139.1	24.8-47.2	34.5 $\pm$ 8.7	34.3
haze	7.6-80.8	20.0 $\pm$ 21.1	15.0	11.3-43.7	22.2 $\pm$ 9.3	21.2
Al solubility	range	average	median	range	average	median
entire	0.11-9.10	1.75 $\pm$ 1.57	1.32	0.25-7.70	1.54 $\pm$ 1.10	1.22
clean	0.11-9.10	1.96 $\pm$ 1.62	1.64	0.25-4.71	1.51 $\pm$ 0.75	1.29
dust	0.42-7.74	2.31 $\pm$ 2.49	1.86	0.77-4.84	2.08 $\pm$ 1.55	1.47
haze	0.31-3.22	0.96 $\pm$ 0.71	0.72	0.43-7.70	1.49 $\pm$ 1.71	1.01

Table S7. Overview of total Al ($\mu\text{g}/\text{m}^3$), dissolved Al (ng/m^3) and Al solubility (%) under different weather conditions for submicron and supermicron particles at Qingdao.

submicron particles				supermicron particles		
total Al	range	average	median	range	average	median
entire	0.05-9.63	1.08±1.67	0.68	0.08-4.01	0.56±0.75	0.36
clean	0.08-1.34	0.66±0.28	0.68	0.08-1.15	0.31±0.17	0.30
dust	1.05-9.63	6.42±3.08	7.14	0.66-4.01	2.90±1.10	2.96
haze	0.77-1.90	1.40±0.58	1.55	0.47-1.09	0.69±0.35	0.50
fog	0.18-1.09	0.53±0.35	0.42	0.17-0.52	0.38±0.14	0.37
dissolved Al	range	average	median	range	average	median
entire	0.4-63.3	9.8±9.6	6.1	0.3-54.9	10.5±11.4	6.3
clean	0.4-40.6	6.2±5.7	5.4	0.9-26.6	5.1±5.0	3.0
dust	2.5-63.3	25.9±20.9	20.6	1.9-23.6	11.5±7.5	10.4
haze	24.5-26.6	25.4±1.1	25.2	14.2-39.7	23.7±14.0	17.3
fog	4.0-19.5	7.4±5.5	5.2	8.7-54.6	29.4±17.2	34.8
Al solubility	range	average	median	range	average	median
entire	0.11-11.11	1.60±1.78	0.91	0.06-23.35	3.23±3.95	1.70
clean	0.11-8.30	1.15±1.26	0.77	0.18-15.58	2.10±2.72	0.99
dust	0.20-1.50	0.45±0.47	0.24	0.06-2.87	0.67±0.98	0.31
haze	1.40-3.27	2.09±1.03	1.58	2.85-3.70	3.40±0.47	3.64
fog	0.43-4.23	1.93 ±1.52	1.38	2.37-23.35	9.09±7.26	6.90

Table S8. Pearson correlations coefficient (r): aerosol Al solubility versus [sulfate]/[Al] and [nitrate]/[Al] for submicron and supermicron particles at Xi'an and Qingdao. In this table, r values which are >0.5 are highlighted in bold.

Species	Season	Xi'an		Qingdao	
		submicron particles	supermicron particles	submicron particles	supermicron particles
[sulfate]/[Al]	spring	-0.33	0.33	0.87	0.75
	summer	-0.17	0.36	0.68	0.66
	autumn	0.15	0.58	0.59	0.84
	winter	0.57	0.73 (0.41*)	0.17	0.96
[nitrate]/[Al]	spring	-0.37	0.25	0.89	0.71
	summer	-0.20	0.29	0.14	0.59
	autumn	-0.12	0.70	0.70	0.90
	winter	0.32	0.93 (0.44*)	0.30	0.88

* Pearson correlations coefficients (r) after three outliers for which aerosol Al solubility was very high were excluded.

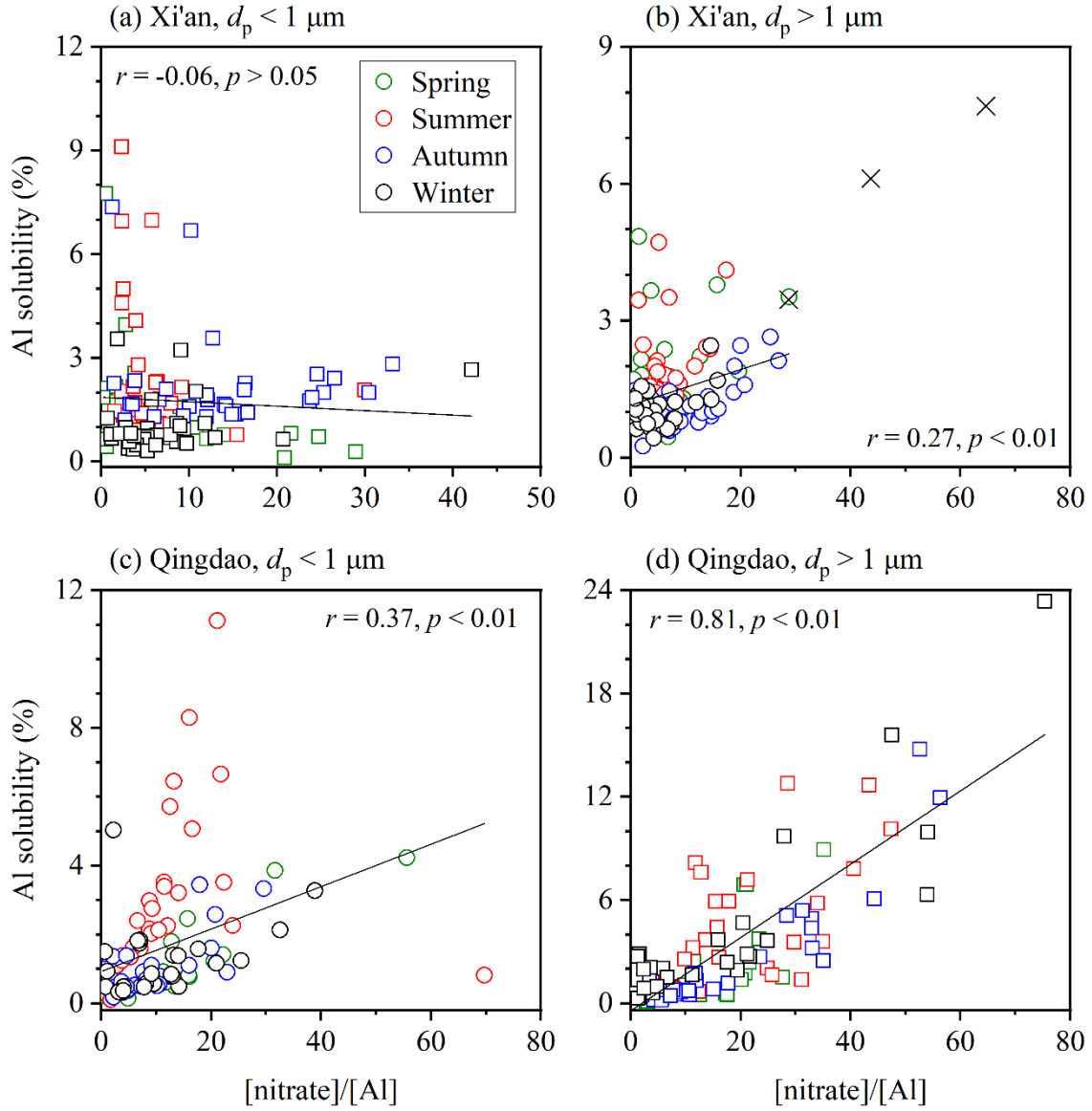


Figure S1. Aerosol Al solubility versus [nitrate]/[Al]: (a) submicron particles at Xi'an; (b) supermicron particles at Xi'an; (c) submicron particles at Qingdao; (d) supermicron particles at Qingdao (the r value changed from 0.81 to 0.77 if the data point with the highest Al solubility was excluded). Data represented by crosses are not included in fittings.

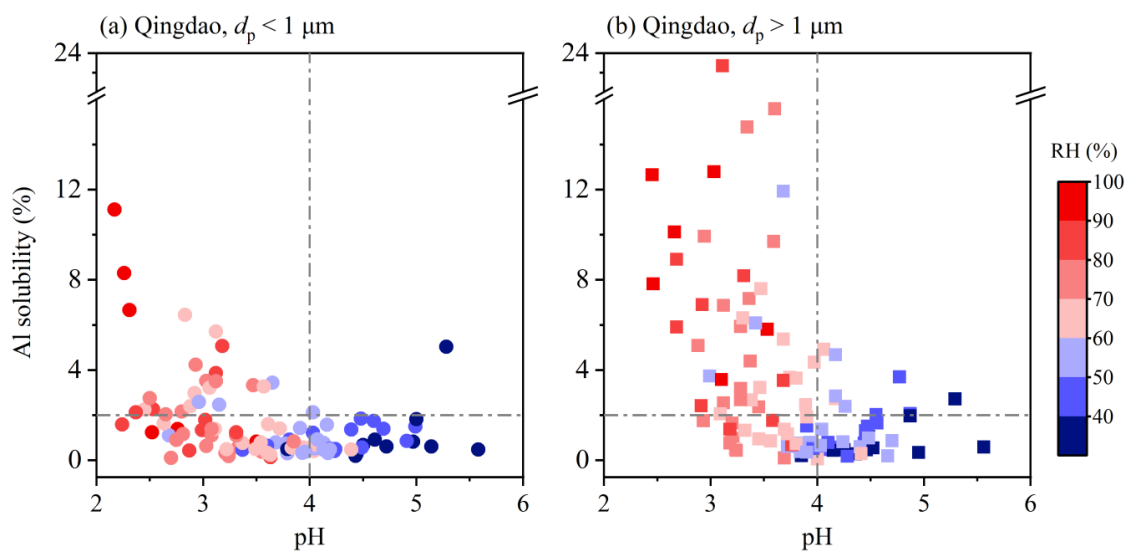


Figure S2. Dependence of aerosol Al solubility on relative humidity (RH) and aerosol pH at Qingdao: (a) submicron particles; (b) supermicron particles.

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