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Concentrations and solubility of trace elements in fine particles at a mountain site, southern China: regional sources and cloud processing

T. Li et al.

Correspondence to: Y. Wang (wy@sdu.edu.cn)

Comparison of various experimental conditions

No significant relationships were observed among various studies except the commonly low solubility for crustal elements such as Fe, Al and Cr, as shown in Fig. 7. Accordingly, environment type, extraction method and analysis instrument are compared in Table S5. The main differences are the type of environment and extraction method for total content of trace elements. The higher solubility for Mn, Cu and Fe at Mt. Lushan should be attributed the frequent cloud events at high altitude. Larger aerosol size (TSP) and stronger acid mixture (HNO_3-HF) were responsible for the various gap of solubility for Ba, Mo, Fe and Al in TSP over East China Sea and in $PM_{2.5}$ at Mt. Lushan. However, the differences of extraction method were not so much remarkable and the overall solubility trend for most elements were similar (decreasing from Zn to Cr) in each study, which indicated the much less significance of extraction method than the type of environment to the solubility of aerosol trace elements.

Table S1. Detection limits (μg L^{-1}) and recoveries (%) for trace elements analyzed by ICP-MS.

	Detection limit (μg L^{-1})	Recovery (%)
Al	1.15	75.7
Cr	0.10	79.9
Mn	0.05	87.2
Fe	0.80	78.3
Cu	0.08	90.6
Zn	0.70	88.4
As	0.12	78.7
Se	0.41	72.7
Mo	0.06	78.0
Cd	0.05	93.0
Ba	0.20	92.6
Pb	0.05	86.0

Table S2. Key meteorology parameters (Mean ± SD) for the summer of 2011 and the spring of 2012, as well as for the two selected cloud events on 11 September 2011 and 18 April 2012. Note that the statistical meteorology parameters for summer of 2011 and spring of 2012 were computed including fine-days, rainy-days and cloudy-days.

Periods	T (℃)	RH (%)	WS (m s^{-1})	Visibility (km)
Summer, 2011	20.0 ± 4.5	88 ± 12	12.7 ± 9.4	11.6 ± 7.7
Spring, 2012	13.6 ± 4.3	76 ± 26	3.6 ± 2.8	10.7 ± 8.1
11 September 2011	20.7 ± 0.4	99	1.7 ± 3.8	0
18 April 2012	13.3 ± 0.9	99	0.5 ± 0.5	0

Table S3. Comparison of $PM_{2.5}$ (μg m^{-3}) and trace elements (ng m^{-3}) at Mt. Lushan with typical mountains and megacities in China.

	Mt. Lushan (this study)	Mt. Tai (Deng et al., 2011)		Mt. Gongga (Yang et al., 2009a)	Mt. Dinghu (Yang et al., 2009b)	Beijing (Yang et al., 2011)	Guangzhou (Yang et al., 2011)	Shanghai [a] (Chen et al., 2008)
		Spring, 2006/2007	Summer, 2006	2006	2006	2005.3 – 2006.2	2008.12 – 2009.2	2004.4 – 2005.4
$PM_{2.5}$	55.2 ± 20.1	46.6/ 70.1	123.1	-	-	118.5	81.7	65
Al	449.1 ± 441.1	1200	1960	295.8 ± 248.9	914.1 ± 663.6	790 ± 320	-	-
Fe	331.1 ± 236.2	810	710	224.0 ± 167.2	569.8 ± 327.8	1130 ± 410	1850 ± 1130	950 ± 520
Zn	258.3 ± 162.8	400	450	154.6 ± 100.6	432.1 ± 241.3	530 ± 220	1360 ± 500	349 ± 154
Ba	68.2 ± 49.3	-	-	6.0 ± 5.7	14.2 ± 9.3	210 ± 160	70 ± 20	12 ± 11
Pb	63.8 ± 54.2	15.2	72	39.4 ± 26.0	216.2 ± 180.8	240 ± 120	450 ± 210	143 ± 117
Mn	22.2 ± 12.2	39.1	71.9	-	33.1 ± 20.6	90 ± 30	150 ± 70	51 ± 28
As	21.5 ± 19.6	2.3	3.6	4.3 ± 3.0	31.8 ± 26.5	20 ± 10	40 ± 30	28 ± 19
Cr	13.7 ± 17.2	22.9	85.4	-	-	50 ± 30	70 ± 20	15 ± 10
Cu	12.4 ± 9.6	24	21.8	2.2 ± 1.2	60.6 ± 47.4	70 ± 30	190 ± 80	29 ± 19
Se	7.0 ± 3.3	-	-	-	8.1 ± 6.2	20 ± 10	-	3.1 ± 1.9
Cd	2.5 ± 1.8	1	3.4	-	7.0 ± 5.0	50 ± 30	20 ± 10	3.7 ± 1.9
Mo	2.0 ± 2.0	-	-	-	-	-	-	-

-: Not reported.

[a]: An urban-residential site, Putuo.

Table S4. Correlation matrix of individual trace elements (water-soluble and total). Correlations higher than 0.60 are highlighted in bold.

Water soluble

Total

	Al	Cr	Mn	Fe	Cu	Zn	As	Se	Mo	Cd	Ba	Pb
Al		0.08	0.25*	**0.68****	-0.06	0.20	0.07	-0.11	0.16	0.06	0.24*	-0.18
Cr	0.12		0.05	0.40**	0.14	0.38**	0.55**	0.19	**0.67****	0.20	0.28*	-0.14
Mn	0.08	0.3		0.47**	0.25*	0.23	0.26*	0.35**	0.29*	0.29*	0.15	0.31**
Fe	**0.64****	**0.61****	0.40*		-0.06	0.26*	0.36**	0.03	0.46**	0.12	0.43**	-0.17
Cu	0.01	0.15	0.35**	0.32		0.52**	0.23	0.49**	0.19	0.33**	0.04	0.48**
Zn	-0.07	0.44*	0.59**	0.14	**0.77****		0.40**	0.25*	0.45**	0.29*	0.33**	0.31**
As	0.24*	-0.27	0.14	-0.17	0.02	0.12		0.46**	0.59**	0.31**	0.35**	0.28*
Se	-0.18	0.23	0.45**	0.02	0.54**	**0.72****	0.22		0.41**	0.40**	0.07	0.46**
Mo	0.04	0.41*	0.37**	0.28	0.37**	0.55**	0.22	**0.63****		**0.71****	0.47**	0.02
Cd	0.07	0.04	0.45**	0.17	**0.69****	**0.64****	0.14	**0.63****	0.28		0.12	0.19
Ba	0.19	0.15	-0.17	0.00	0.04	-0.08	0.13	0.00	0.06	-0.07		-0.23
Pb	-0.21	0.48**	**0.62****	0.17	**0.69****	**0.82****	0.01	**0.82****	**0.61****	**0.66****	-0.03	

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

Table S5. Differences of environment type, extraction method and instrument in determining aerosol element solubility in various studies.

Location	Type of environment	Extraction method		Instrument	Reference
		Soluble	Total		
Mt. Lushan ($PM_{2.5}$)	Rural mountain	Ultrapure water	HNO_3-H_2O_2, microwave digestion	ICP-MS	This study
East China Sea (TSP)	Sea surface	Milli-Q water	HNO_3-HF, microwave digestion	ICP-MS	(Hsu et al., 2010)
Edinburgh,UK ($PM_{2.5}$)	Urban background	Ultrapure water	HNO_3-HCl, heating	ICP-MS	(Heal et al., 2005)
Nanjing, China ($PM_{2.5}$)	Urban city	Glycine	HNO_3-H_2O_2, microwave digestion	ICP-OES & ICP-MS	(Hu et al., 2012)

Table S6. Solubility (%) of individual elements in $PM_{2.5}$ before and after two cloud events at Mt. Lushan. Sulfate concentration in $PM_{2.5}$ ($\mu g\ m^{-3}$) and cloud water ($mg\ L^{-1}$) are listed.

Species	11 September 2011 (cluster E)			18 April 2012 (cluster L)		
	Pre-cloud (4h)	In-cloud (3h)	Post-cloud (4h)	Pre-cloud (9h)	In-cloud (9h)	Post-cloud (9h)
Al	1.67	pH=3.36	28.68	12	pH=3.98	32.35
Cr			2.02	7.79		3.37
Mn	90.35		87.18	65.17		90.8
Fe	0.27		8.8	20.98		78.59
Cu	49.54		84.43	59.09		80.15
Zn	29.17		93.39	68.02		64.21
As	68.33		89.12	74.87		83.33
Se	34.6		85.89	65.02		66.6
Mo	8.83		21.74	54.08		60.18
Cd	18.92		56.95	65.33		60.99
Ba	10.63		33.63	28.52		70.57
Pb	47.11		57.8	33.38		36.28
SO_4^{2-}	14.77	41.70	24.72	15.96	26.78	17.34

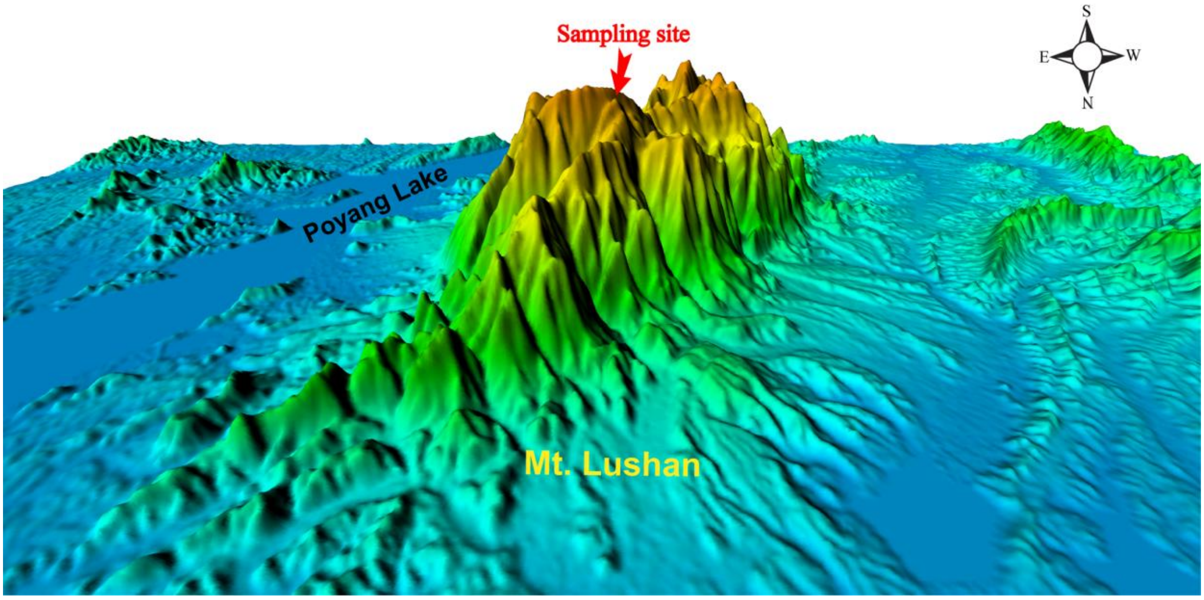

Figure S1. Topographical map of Mt. Lushan and the sampling site (Guniubei). The geographic data are available from the CGIAR-CSI SRTM 90m Database v4.1 (http://srtm.csi.cgiar.org; Jarvis et al., 2008).

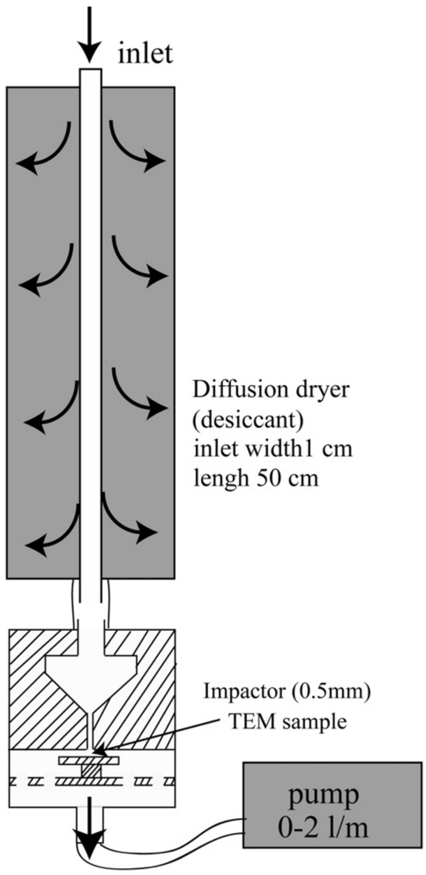

Figure S2. Schematic of the cloud droplet residues sampling system (Li et al., 2011).

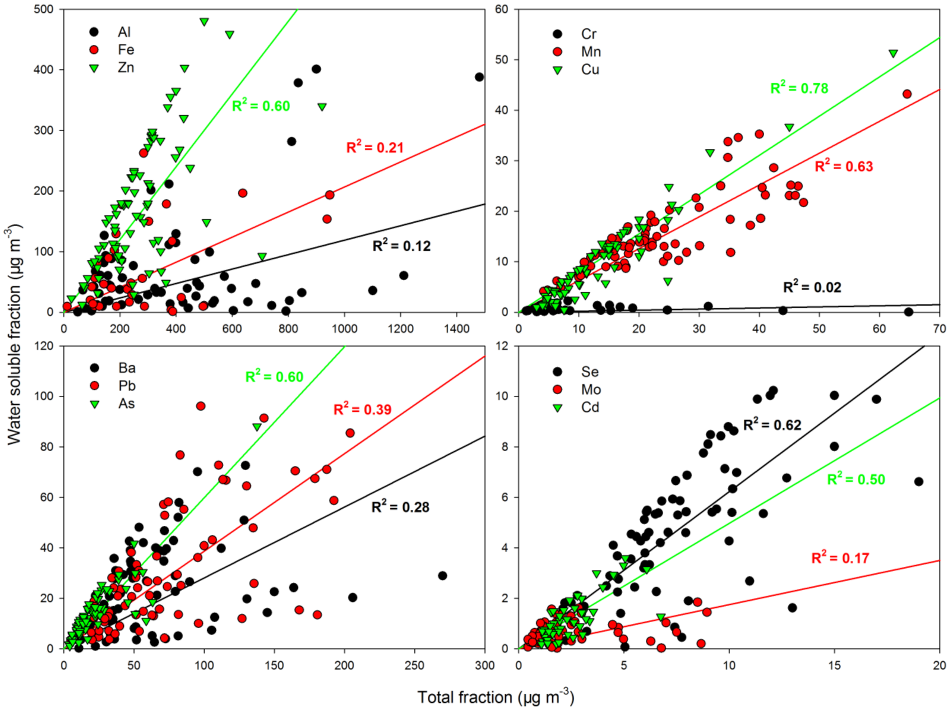

Figure S3. Linear regression of concentration for individual elements between water soluble fraction and total fraction.

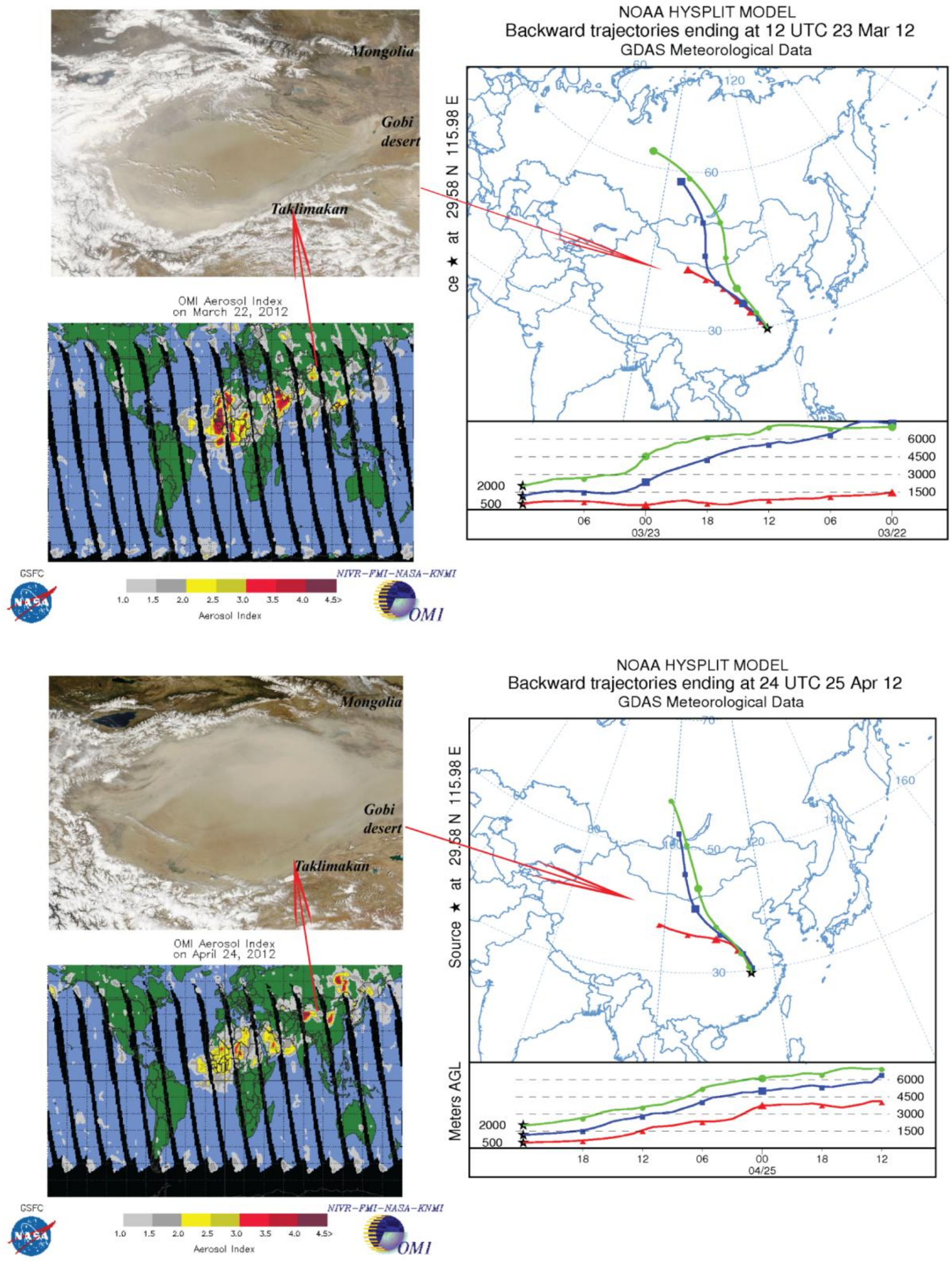

Figure S4. Satellite image of Taklimakan desert captured by MODIS on NASA's Aqua satellite (http://earthobservatory.nasa.gov/?eocn=topnav&eoci=home), OMI Aerosol Index (http://macuv.gsfc.nasa.gov/OMIAerosol.md) and 36-h backward trajectory arriving at Mt. Lushan for dust storm events during 23–25 March 2012 (top) and 25–26 April 2012 (bottom).

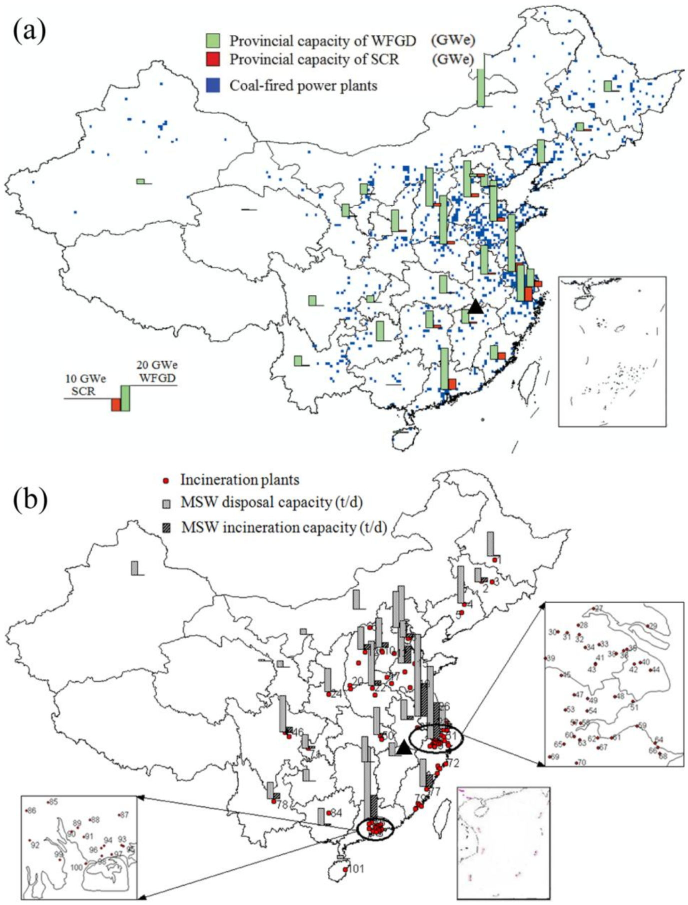

Figure S5. Geographical distribution of (a) coal-fired power plants (Tian et al., 2014) and (b) municipal solid waste (MSW) incineration plants (Tian et al., 2012) in China, 2010. The triangle indicates Mt. Lushan.

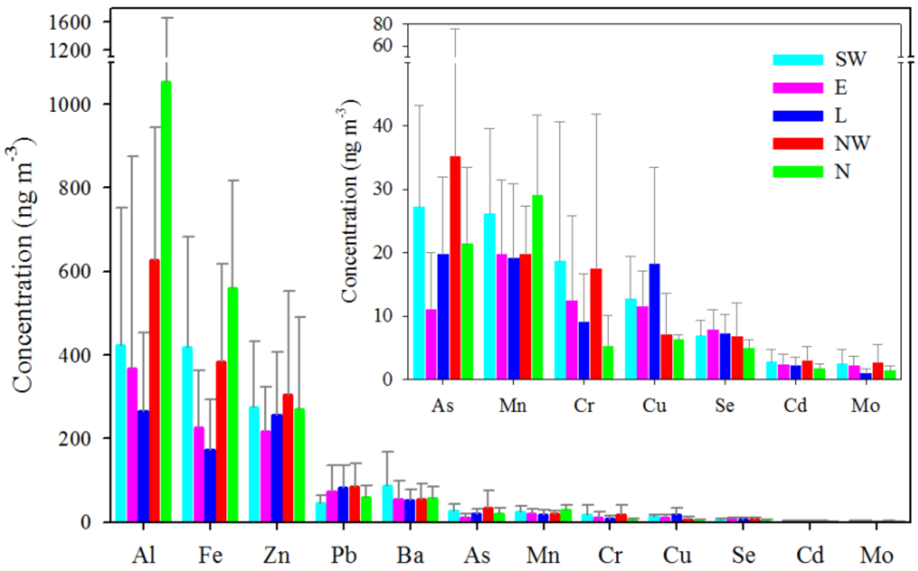

Figure S6. Trace elements concentrations in $PM_{2.5}$ at Mt. Lushan for five clusters.

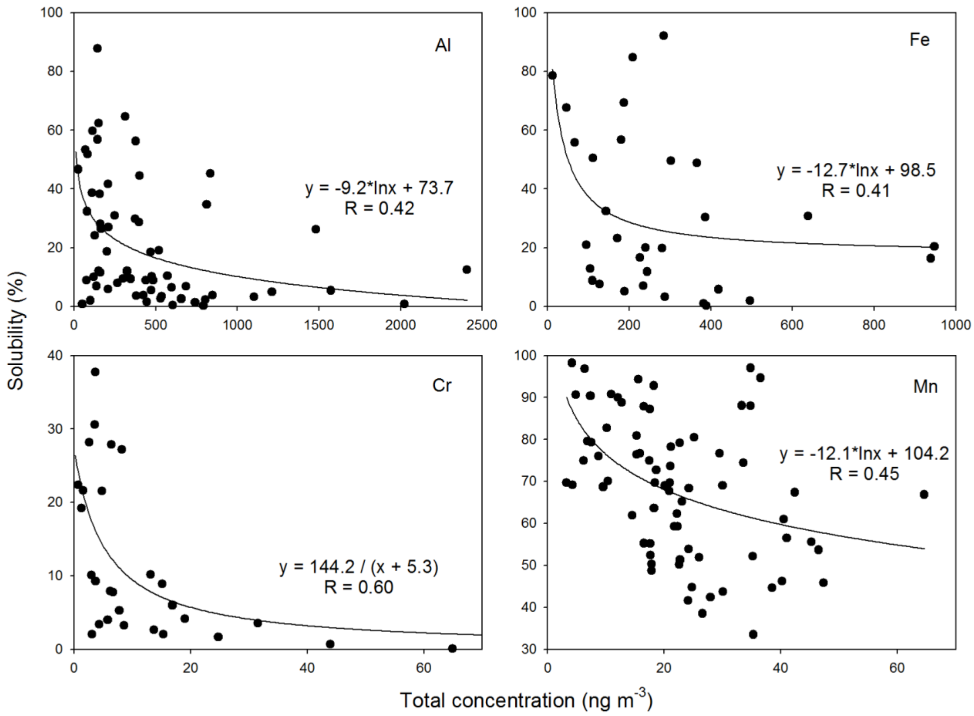

Figure S7. Inverse relationships between total concentration and element solubility for Al, Fe, Cr and Mn.

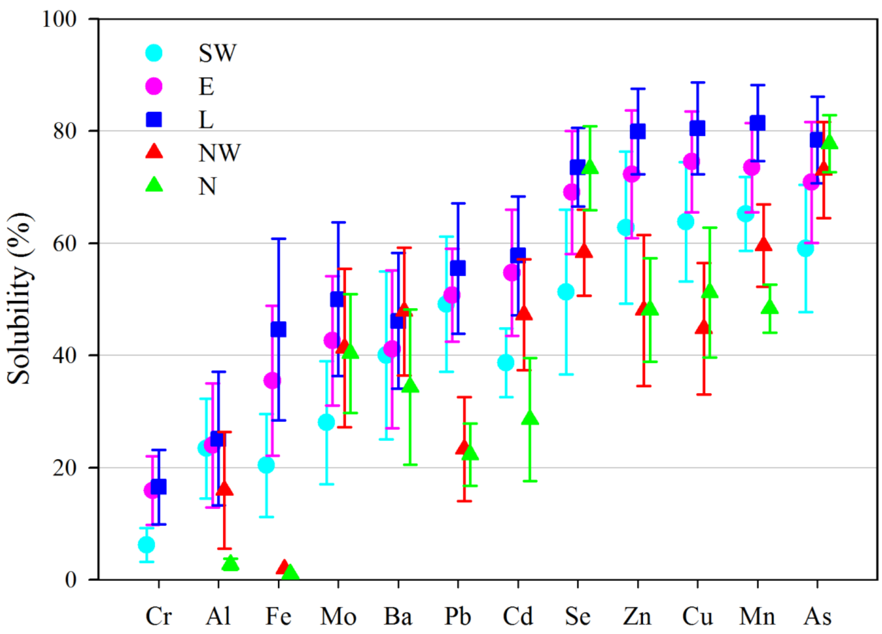

Figure S8. Solubility of trace elements in $PM_{2.5}$ at Mt. Lushan for five air mass clusters. Solid points and whiskers represent mean values and half of standard deviations, respectively.

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