



*Supplement of*

## **Atmospheric wet and dry deposition of trace elements at ten sites in Northern China**

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1    **Supplementary material includes Tables S1, S2, S3 and S4, and**  
2    **Figures S1, S2, S3 and S4.**

3

4    Figure S1. Locations of the study area (a) and sampling sites (b) in Northern China  
5    with lead deposition and SO<sub>2</sub> emission distributions. The total lead deposition data are  
6    means of 3-yr observations from December 2007 to November 2010. The emission  
7    data for SO<sub>2</sub> are from 2006 ([Zhang et al., 2009](#)) and have a resolution of 0.5 °×0.5 °. In  
8    Northern China, the annual SO<sub>2</sub> emission unit of kt grid<sup>-1</sup> is approximately 400 mg  
9    m<sup>-2</sup>.

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11    Figure S2. The automated wet and dry deposition sampler used in this study. The  
12    agricultural station of Yucheng in Shandong Province (left) and a schematic diagram  
13    of the sampler with the main apparatus (right): rainfall sampler (a), dry deposition  
14    bucket (b), and PUF filter for sampling dry-deposited particles (c).

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16    Figure S3. Temperature-controlled microwave digestion of a PUF filter with  
17    dry-deposited particles using 5 ml HNO<sub>3</sub>, 2 ml H<sub>2</sub>O<sub>2</sub> and 0.2 ml HF ([Pan et al., 2010](#)).

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19    Figure S4. Annual wet deposition fluxes of trace elements vs. precipitation in  
20    Northern China.

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Table S1 Atmospheric dry deposition fluxes of metals within and outside China ( $\text{mg m}^{-2} \text{ yr}^{-1}$ , but  $\text{kg ha}^{-1} \text{ yr}^{-1}$  for Na, Ca, Mg, Fe, Mn and Al)

Site	Period	Na	Ca	Mg	Al	Mn	Fe	Cu	Pb	Zn	Cd	Co	Ni	Cr	V	Reference
Xinghua Bay, China	2004-2005	6.69	/	/	7.83	/	6.83	2.71	3.83	14.39	0.08	0.28	7.82	11.63	/	(Wu et al., 2006)
Yellow sea, China	1995-1996	12.42	6.32	2.89	4.49	0.08	2.55	0.23	1.92	2.91	0.04	0.2	0.63	1.11	1	(Liu et al., 1998)
East Sea China	2005-2007	/	/	/	0.15	0.02	0.14	4.38	0.91	6.94	0.07	/	0.09	0.08	0.55	(Hsu et al., 2010)
Taiwan China	2009-2010	/	/	/	/	0.08	1.52	20.8	20.03	18.03	/	/	/	9.20	/	(Zhang et al., 2012)
Hong Kong ,China	1998-1999	6.67	/	1.15	0.83	0.07	0.78	5.25	28.98	27.95	/	/	/	/	0.19	(Zheng et al., 2005)
Matsuura, Japan	2004-2006	/	/	/	1.32	0.09	/	3.01	1.55	/	0.06	/	3.72	4.28	0.54	(Sakata and Asakura, 2011)
Seoul, S. Korea	Spring,1998	/	23.0	/	19.7	/	/	/	73.0	73.0	/	/	40.2	/	/	(Yi et al., 2001a)
Maagan Michale, Israel	1994-1997	/	/	/	4.97	0.11	4.32	0.23	1.54	1.98	0.07	/	/	1.27	/	(Herut et al., 2001)
Chicago, U.S.	1990s	/	25.51	9.12	/	0.4	/	69.4	46.4	266.5	4.4	/	/	19.3	/	(Fang, 1992)
Chicago, U.S.	1993-1995	/	/	8.29	3.80	0.21	/	23.0	13.87	43.80	/	/	/	2.08	1.21	(Yi et al., 2001b)
Sleeping Bear Dunes, U.S.	1993-1995	/	/	0.21	0.27	0.01	/	29.0	12.78	24.82	/	/	/	0.58	0.06	(Yi et al., 2001b)
South Heaven, U.S.	1993-1995	/	/	1.90	1.24	0.08	/	11.3	8.40	18.62	/	/	/	0.27	0.44	(Yi et al., 2001b)
Tor Paterno, Italy	1999	/	/	/	/	/	/	11.03	11.41	43.64	0.36	/	12.23	17.04	/	(Morselli et al., 2004)
Cap Ferat, France	1988-1989	/	/	/	1.20	0.02	0.88	1.19	1.85	3.20	/	0.02	0.33	0.38	/	(Chester et al., 1999)
Amman, Jordan	1995	/	/	/	/	/	/	5.55	4.20	29.69	0.15	/	/	/	/	(Momani et al., 2000)
Izmir, Turkey	2000-2001	/	157.32	11.32	/	0.49	44.13	45.3	80.3	697.2	8.8	/	47.1	5.8	/	(Odabasi et al., 2002)
Bursa, Turkey	2002-2003	/	88.44	12.88	/	0.62	29.23	71.17	55.84	366.46	1.10	2.92	46.36	22.26	/	(Tasdemir and Kural, 2005)

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27 Table S2 Atmospheric wet deposition fluxes of metals within and outside China ( $\text{mg m}^{-2} \text{ yr}^{-1}$ , but  $\text{kg ha}^{-1} \text{ yr}^{-1}$  for Fe, Mn and Al)

Site	Period	Fe	Mn	Al	Cu	Pb	Zn	Cd	Cr	Co	Ni	V	Reference
Nam Co, central Tibetan Plateau, China	2007-2008	0.05	0.003	0.055	0.23	0.06	0.27	0.002	/	0.007	0.09	0.03	(Cong et al., 2010)
Hong Kong, China	1998-1999	0.78	0.04	0.62	4.67	86.94	33.15	/	/	/	/	2.65	(Zheng et al., 2005)
Yellow Sea, China	2000-2002	/	/	/	1.99	0.37	0.12	37.4	/	/	/	/	(Liu et al., 2003)
Kathmandu, Nepal	2011-2012	2.47	0.08	2.10	1.95	1.42	24.44	0.10	1.60	1.00	0.71	/	(Tripathee et al., 2014)
Dhunche, Nepal	2011-2012	1.00	0.04	0.98	0.02	0.02	0.18	0.00	0.00	0.01	0.02	/	(Tripathee et al., 2014)
Chuncheon, Korea	2006-2009	/	0.03	0.10	1.21	1.06	6.93	0.05	/	/	0.37	0.10	(Kim et al., 2012)
Nakanoto, Japan	2003-2005	/	0.07	/	1.8	10	27	0.31	0.40	/	1.40	0.78	(Sakata and Asakura, 2009)
Higashi-Hiroshima, Japan	1995-1997	/	0.02	<0.01	0.89	1.78	6.84	0.09	/	/	0.42	0.33	(Takeda et al., 2000)
Singapore	2000	0.62	0.07	0.48	14.56	8.84	18.72	0.78	4.16	1.56	10.14	9.10	(Hu and Balasubramanian, 2003)
Fiordland, New Zealand	1993-1995	3.70	0.001	/	0.02	0.04	0.07	0.00	/	/	/	/	(Halstead et al., 2000)
New Castle, U.S.	1996-1997	0.14	/	0.17	0.67	0.78	8.33	0.12	0.08	0.12	0.42	0.72	(Pike and Moran, 2001)
Chesapeake and Delaware Bay, U.S.	1995-1996	0.14	0.01	0.20	0.97	0.35	3.60	0.04	0.04	/	0.82	/	(Kim et al., 2000)
Massachusetts, U.S.	1992-1993	0.36	0.01	0.29	0.70	0.86	2.70	0.21	1.50	0.01	3.00	/	(Golomb et al., 1997)
Bermuda, U.S.	1982-1983	0.016	0.001	/	0.07	0.31	0.66	0.02	/	/	0.08	0.07	(Church et al., 1984)
Ankara, Turkey	1993-1994	0.007	/	0.003	0.45	0.87	2.84	1.32	0.16	/	0.20	0.21	(Kaya and Tuncel, 1997)
North Sea	1992-1994	/	/	/	10.50	11.00	31.00	/	1.40	/	/	/	(Injuk et al., 1998)
Dutch Delta, The Netherlands.	1990	/	/	/	0.23	4.23	12.67	0.07	/	/	0.88	/	(Nguyen et al., 1990)

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30 Table S3 Atmospheric total deposition fluxes of metals within and outside China ( $\text{mg m}^{-2} \text{ yr}^{-1}$ )

Site	Period	Cd	Cu	Ni	Pb	Zn	As	Mn	V	Reference
Pearl River Delta, China	2001–2002	/	18.6	/	12.7	104	/	/	2.1	(Wong et al., 2003)
Hong Kong ,China	1998–1999	/	9.92	/	115.92	61.1	/	10.29	2.84	(Zheng et al., 2005)
Kushiro, Japan	2008	0.02	0.56	0.72	0.98	4.02	/	4.74	/	(Okubo et al., 2013)
Tokyo Bay, Japan	2004–2005	0.39	16	6.8	9.9		2.9	87	6.9	(Sakata et al., 2008)
Virolahti, Finland	2007	0.04	1.00	0.14	1.1	3.8	0.09	2.3	0.36	(Kyll önen et al., 2009)
Paris, France	2001–2002	0.24	6	0.62	4.2	30	/	/	/	(Motelay-Massei et al., 2005)
Massachusetts Bay, US	1992–1993	0.27	2.5	1.5	1.8	7.8	0.02	3.4		(Golomb et al., 1997)
Chesapeake Bay, US	1992–1993	0.05	0.26	0.26	0.56	1.34	/	/	/	(Motelay-Massei et al., 2005)
Lake Superior, US	1993–1994	0.46	3.1	0.8	1.5	8.8	0.17	4.2	0.34	(Sweet et al., 1998)
Lake Michigan, US	1993–1994	0.45	1.9	0.61	1.6	6	0.14	2.8	0.14	(Sweet et al., 1998)
Lake Erie, US	1993–1994	0.49	4.2	0.74	1.8	17	/	/	/	(Sweet et al., 1998)
Fiorland, New Zealand	1993–1995	0.004	0.023	0.035	0.025	/	/	/	/	(Halstead et al., 2000)
Varna, Bulgaria, Black Sea	2008–2009	0.02	17.8	0.41	0.73	15.18	/	2.01	1.1	(Theodosi et al., 2013)
North Sea	1993–1994	/	1.24	/	3.52	6.5	0.25	2.6	1.1	(Injuk et al., 1998)
Irish Sea	1993–1994	/	2.6	/	1.62	/	/	5.07	/	(Williams et al., 1998)
Mediterranean Coast	1988–1993	0.31	2.6	0.57	3.8	/	/	/	/	(Guieu et al., 1997)
Ligurian Sea	1997–1998	0.06	1.28	1.1	1.2	41.2	/	/	/	(Sandroni and Migon, 2002)

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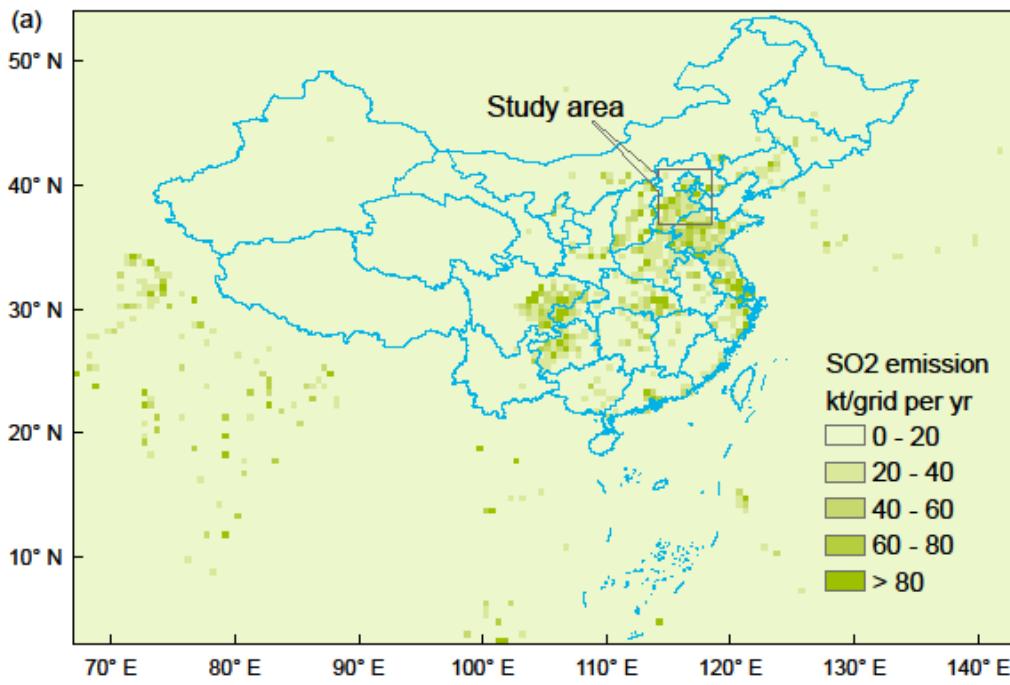
33 Table S4 Average enrichment (increment) of elemental content in top soil (0–10 cm) relative to deep soil (60–100 cm) vs. atmospheric total  
 34 deposition flux of metals at agricultural sites on an annual basis ( $\text{mg m}^{-2}$ )

Site	Item	Mo	Mn	Zn	Cu	Fe	Se	Cd	Pb	Cr	Ni	As
LC	Increment	22.3	-4684.6	662.1	749.4	-231178.5	43.6	0.6	420.8	-838.8	-349.7	-217.7
	Deposition	0.5	49.7	135.0	8.4	2453.0	1.5	0.6	21.8	4.2	5.0	3.0
	Ratio	<b>0.02</b>	<b>-0.01</b>	<b>0.20</b>	<b>0.01</b>	<b>-0.01</b>	<b>0.03</b>	<b>1.03</b>	<b>0.05</b>	<b>0.00</b>	<b>-0.01</b>	<b>-0.01</b>
YC	Increment	-12.8	3536.9	1709.3	10.1	-86889.7	13.0	2.1	347.4	232.5	-190.4	35.5
	Deposition	0.6	43.0	57.0	7.9	2388.8	1.4	0.4	16.2	3.4	11.2	3.6
	Ratio	<b>-0.05</b>	<b>0.01</b>	<b>0.03</b>	<b>0.78</b>	<b>-0.03</b>	<b>0.11</b>	<b>0.18</b>	<b>0.05</b>	<b>0.01</b>	<b>-0.06</b>	<b>0.10</b>

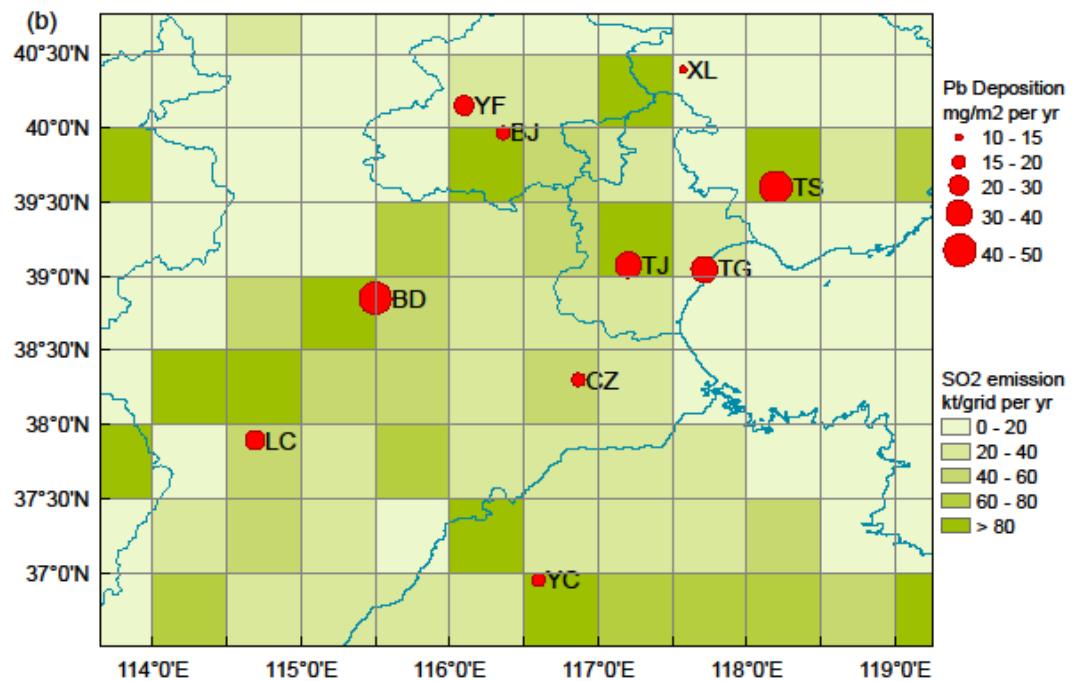
35 Data shown in bold was the ratio of total deposition to increment of metals in soil.

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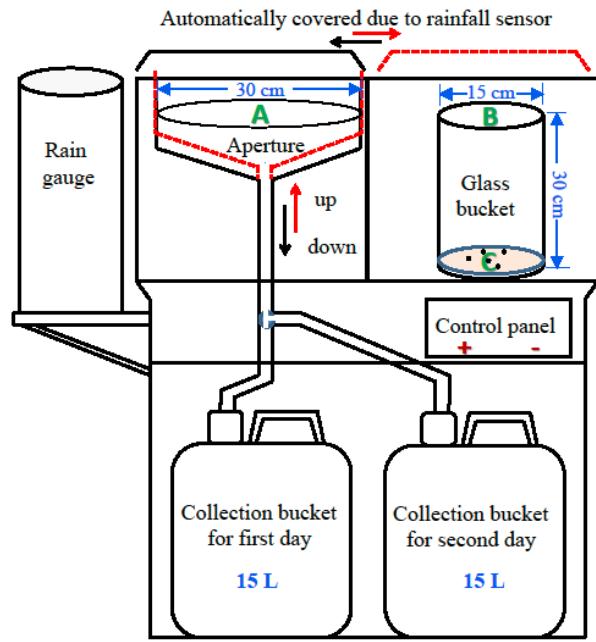


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39 Figure S1. Locations of the study area (a) and sampling sites (b) in Northern China  
 40 with lead deposition and SO<sub>2</sub> emission distributions. The total lead deposition data are  
 41 means of 3-yr observations from December 2007 to November 2010. The emission  
 42 data for SO<sub>2</sub> are from 2006 ([Zhang et al., 2009](#)) and have a resolution of 0.5 °×0.5 °. In  
 43 Northern China, the annual SO<sub>2</sub> emission unit of kt grid<sup>-1</sup> is approximately 400 mg  
 44 m<sup>-2</sup>.

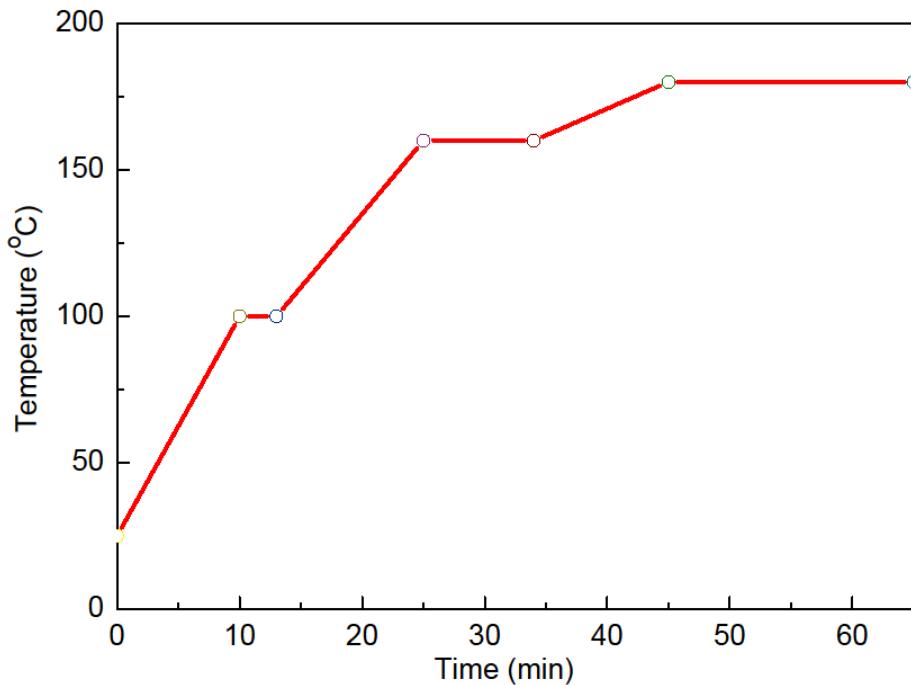
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47 Figure S2. The automated wet and dry deposition sampler used in this study. The  
 48 agricultural station of Yucheng in Shandong Province (left) and a schematic diagram  
 49 of the sampler with the main apparatus (right): rainfall sampler (a), dry deposition  
 50 bucket (b), and PUF filter for sampling dry deposited particles (c).

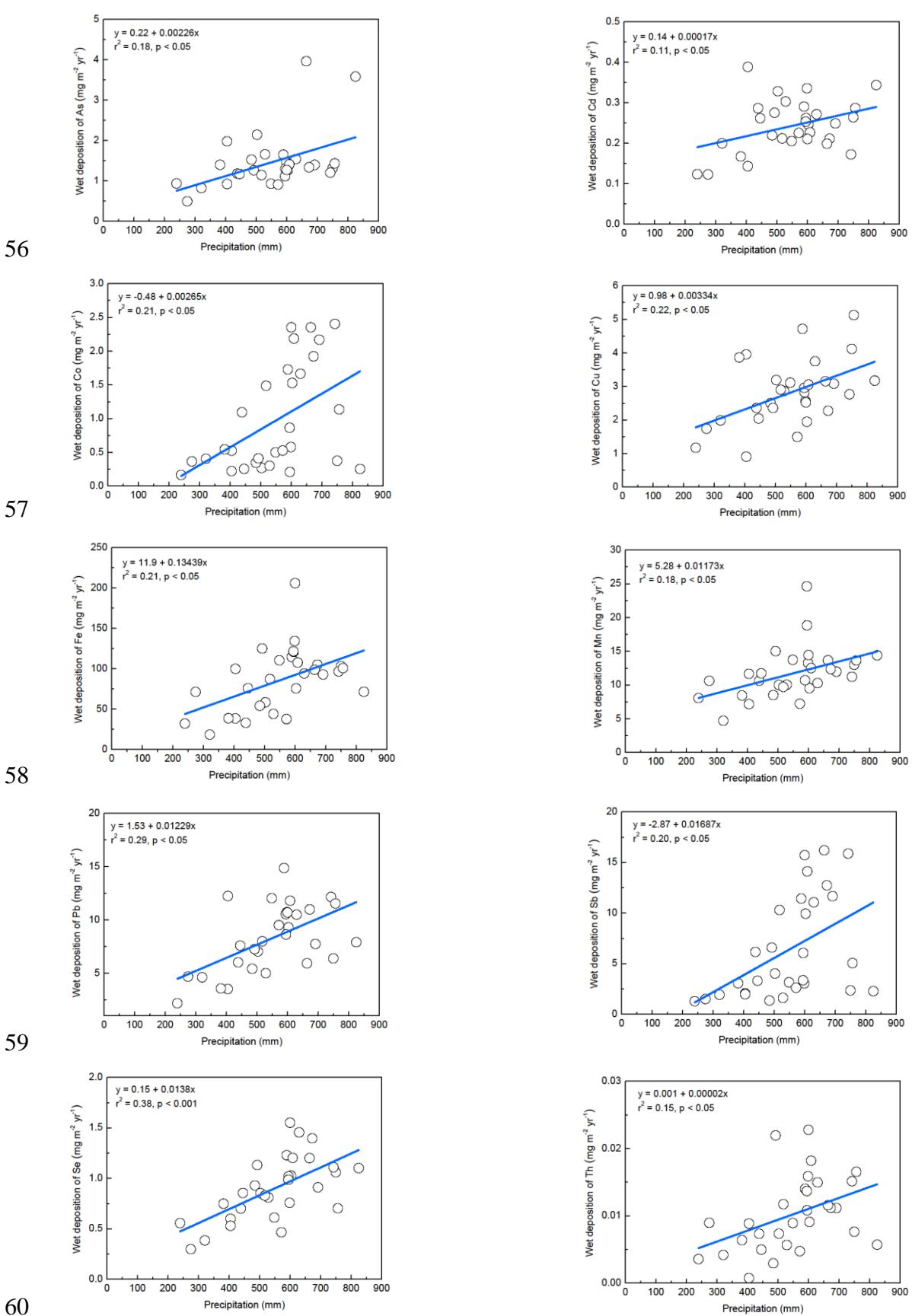
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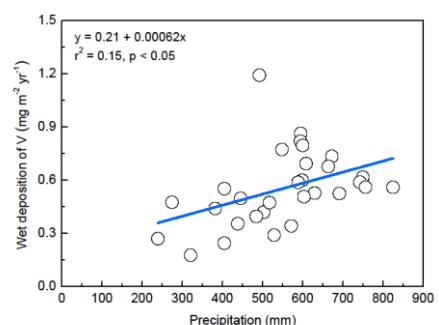
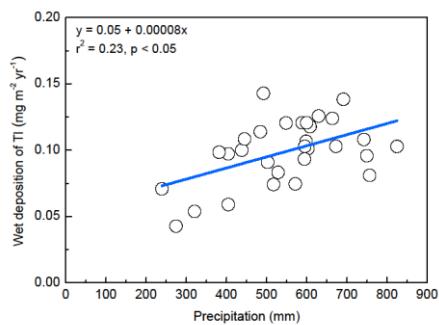
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53 Figure S3. Temperature-controlled microwave digestion of a PUF filter with  
 54 dry-deposited particles using 5 ml HNO<sub>3</sub>, 2 ml H<sub>2</sub>O<sub>2</sub> and 0.2 ml HF ([Pan et al., 2010](#)).

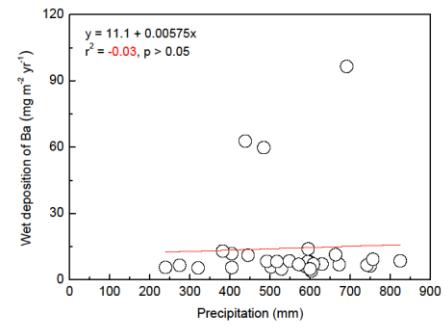
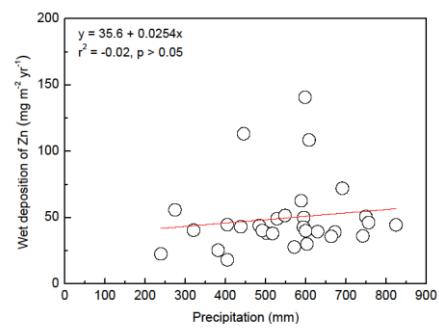
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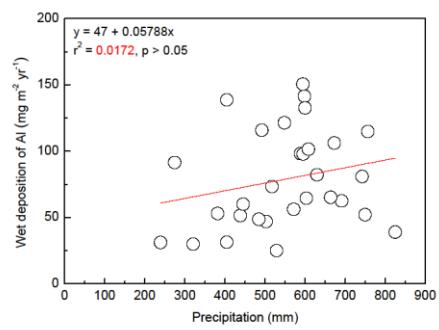
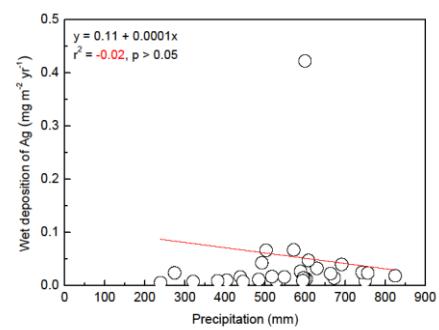
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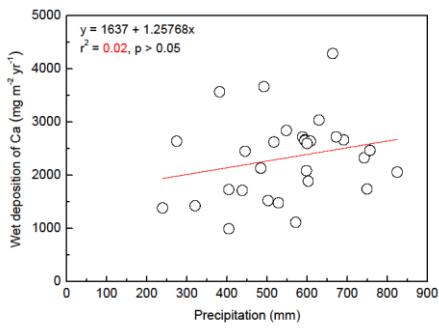
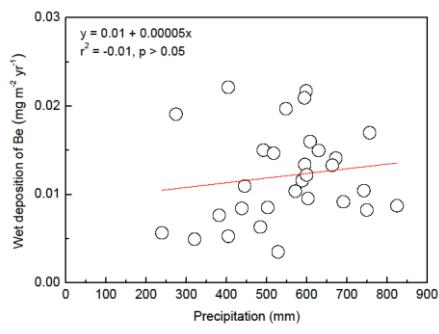
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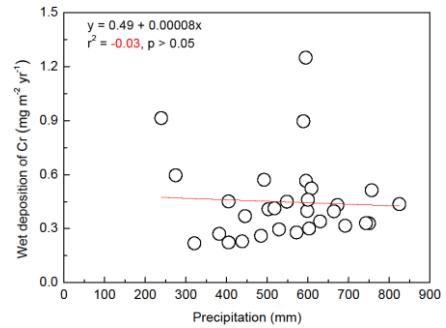
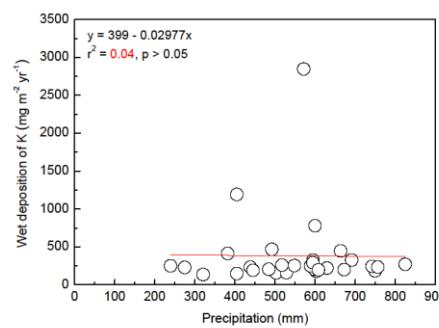
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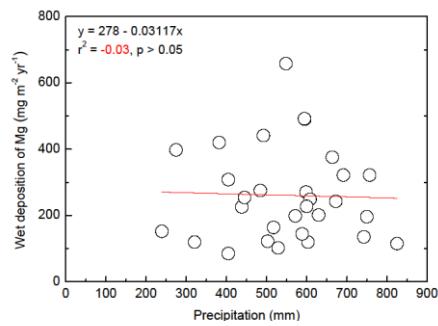
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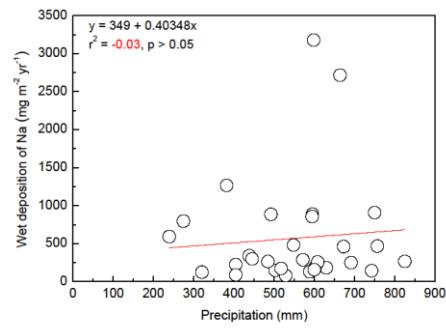
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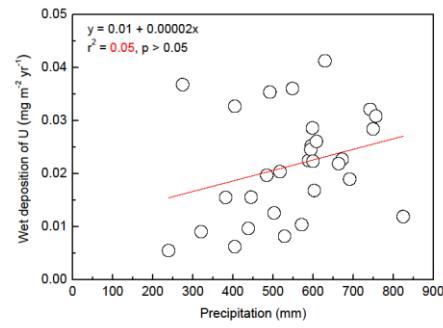
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Fig. S4 Annual wet deposition fluxes of trace elements vs. precipitation in Northern

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## References

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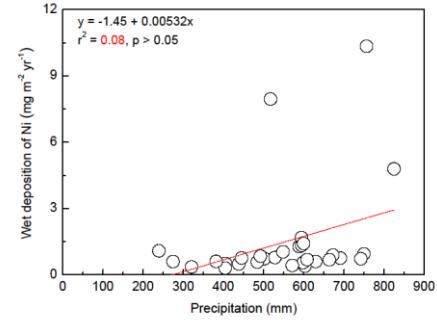
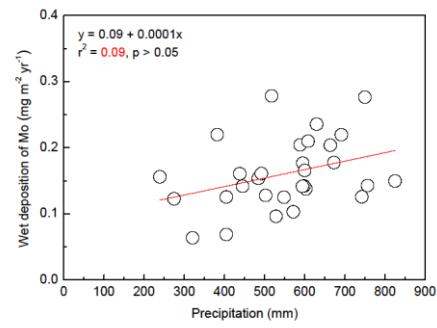
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