

Supplementary materials for

A Long-lasting Winter Haze Episode in Xiangyang, Central China: Pollution Characteristics, Chemical Composition and Health Risk Assessment

Huimeng Jiang¹, Han Xiao¹, He Song¹, Jian Liu¹, Tao Wang²,
Hairong Cheng^{1*}, Zuwu Wang^{1**}

¹ School of Resource and Environmental Sciences, Wuhan University, Wuhan 430079, China.

² Xiangyang Environment Protection Monitoring Station, Xiangyang 441021, China.

Contents:

Table S1. The corresponding percent change in TSV (%) as possible number of clusters are combined.

Table S2. The variables for human health risk assessment.

Table S3. The percentages of air-mass back-trajectory clusters and corresponding mass concentrations of PM_{2.5} and species during the sampling period.

Table S4. The comparison of reported No. of haze days, maximum and mean PM_{2.5} concentrations ($\mu\text{g m}^{-3}$), and prevailing meteorological conditions in Xiangyang and other cities around the world.

Table S5. The average daily exposure doses ($\text{mg kg}^{-1} \text{day}^{-1}$) of PM_{2.5}-bound metal(loid)s for three different exposure pathways at the four pollution levels.

Fig. S1. The pollution level days and PM_{2.5} concentrations at the four pollution levels in January from 2015-2018.

* Corresponding author.

Tel.; +86-027-68775543; Fax: +86-027-68778893

E-mail address: chenghr@whu.edu.cn

** Second corresponding author.

Tel.; +86-027-68775543; Fax: +86-027-68778893

E-mail address: zwwang@whu.edu.cn

Table S1. The corresponding percent change in TSV (%) as possible number of clusters are combined.

Possible number of clusters	Percent change in TSV (%)
15	37.84
9	35.38
5	108.13
3	33.06
2	90.99

Table S2. The variables for human health risk assessment.

Variable	Description	Unit	Value			
R_{Ing}^*	Ingestion rate	mg day ⁻¹	200, 100 for children and adults			
R_{Inh}^*	Inhalation rate	m ³ day ⁻¹	20, 7.6 for children and adults			
ABS^*	Dermal absorption factor	Unitless	0.030, 0.001, 0.01 for As, Cd and other metal(loid)s			
EF^*	Exposure frequency	days year ⁻¹	350 for both children and adults			
ED^*	Exposure duration	years	6, 24 for children and adults			
BW^*	Body weight	kg	15, 70 for children and adults			
AT^*	Averaging time	days	Non-carcinogenic substances: 2190, 8760 for children and adults; Carcinogenic substances: 25500 for both children and adults			
PEF^*	Particle emission factor	m ³ kg ⁻¹	1.36×10 ⁹			
SA^*	Skin surface area in contact with air	cm ²	2800, 5700 for children and adults			
AF^*	Adherence factor for air-borne particulates to skin	mg cm ⁻² day ⁻¹	0.25, 1 for children and adults			
CF^*	conversion factor	kg mg ⁻¹	10 ⁻⁶			
RfD^{**}	Reference dose	mg kg ⁻¹ day ⁻¹	Metal(loid)s			
			RfD_{Ing}			
			RfD_{Inh}			
			RfD_{Dermal}			
			As	3.00E-04	3.01E-04	1.23E-04
			Cd	1.00E-03	1.00E-03	1.00E-05
			Cr (III)	3.00E-03	2.86E-05	6.00E-05
			Co	2.00E-02	5.71E-06	1.60E-02
			Cu	4.00E-02	4.02E-02	1.20E-02
			Mn	4.60E-02	1.43E-05	1.84E-03
			Ni	2.00E-02	2.06E-02	5.40E-03
			Zn	3.00E-01	3.00E-01	6.00E-02
			Pb	3.50E-03	3.52E-03	5.25E-04
			Ag	5.00E-03	—	9.00E-04
			Al	1.00E+00	1.43E-03	1.00E-01
			Ba	7.00E-02	1.43E-04	4.90E-03
			Mo	5.00E-03	—	1.90E-03
Sb	4.00E-04	—	8.00E-06			
Sr	6.00E-01	—	1.20E-01			
U	6.00E-04	—	5.10E-04			
V	7.00E-03	—	7.00E-05			

Variable	Description	Unit	Value			
			Metal(loid)s	SF_{Ing}	SF_{Inh}	SF_{Dermal}
SF^{**}	Slope factor	mg kg ⁻¹ day ⁻¹	As	1.50E+00	1.51E+01	3.66E+00
			Cd	—	6.30E+00	—
			Co	—	9.80E+00	—
			Cr (VI)	5.00E-01	4.20E+01	—
			Ni	—	8.40E-01	—
			Pb	2.80E-01	8.00E-05	—

Note. *The values were obtained from the Exposure Factors Handbook: 2011 Edition by USEPA, (2011). **The values were obtained from obtained from Ferreira-Baptista and De Miguel, (2005), Hu et al., (2012) and Li et al., (2013).

Table S3. The percentages of air-mass back-trajectory clusters and corresponding mass concentrations of PM_{2.5} and species ($\mu\text{g m}^{-3}$) during the sampling period.

Cluster type (%)	PM _{2.5}	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	WSIIs	SNA	OC	EC	TC	OM	SOC	POC	TE
1 (62)	165.49	6.30	51.70	25.16	1.09	21.43	0.20	1.27	0.55	107.70	98.29	20.59	6.65	27.23	32.94	10.02	10.57	10.30
2 (24)	102.68	3.21	27.69	16.30	0.76	12.93	0.10	0.73	0.06	61.78	56.92	12.28	3.99	16.27	19.65	5.93	6.35	11.60
3 (14)	234.61	6.02	84.64	35.71	1.20	33.29	0.21	1.59	0.70	163.38	153.64	27.84	7.84	35.68	44.54	15.38	12.46	10.53

Table S4. The comparison of reported No. of haze days, maximum and mean PM_{2.5} concentrations ($\mu\text{g m}^{-3}$), and prevailing meteorological conditions in Xiangyang and other cities around the world.

Station (Country)	Site type	Sampling period	No. of haze days	Maximum	Mean \pm Standard deviation	Prevailing meteorological conditions	References
Chengdu (China)	Near-road	2018 winter	—	—	113.2 \pm 60.3	Low T and WS	Qu et al., (2019)
Shanghai (China)	Suburban	2018 haze days	21	189.0	92.9 \pm 44.4	Lowest average WS (10.48 m s ⁻¹) and RH (73.52%)	Wei et al., (2019)
Zhengzhou (China)	Urban	Jan. 11-23, 2018	18	349.4	188.2 \pm 52.4	RH: 55.8 \pm 1.1%, WS: 1.5 \pm 1.3 m s ⁻¹ , T: 2.2 \pm 3.7 °C	Wang et al., (2019)
Hefei (China)	Industrial	Oct. 2016-Jan. 2017	—	100	81.0	—	Xue et al., (2019)
Guilin (China)	Urban	Dec. 10-15, 2016 Jan. 4-7, 2017	6	176	144.0 \pm 28.5	T: 15.3 °C, RH:75%, WS: 0.96 m s ⁻¹ , WD: N and NW	Zhong et al., (2019)
Beijing (China)	Urban	Dec. 29, 2014.-Jan. 27, 2015	—	270	117.0	WD: NW	Shen et al., (2019)
Tianjin (China)	Urban	—	—	350	124.0	—	—
Shijiazhuang (China)	Urban	Dec. 15, 2015-Jan. 14, 2016	56	588.7	234.0 \pm 139.5	Lowest boundary layer height	Xie et al., (2019)
Wuhan (China)	Urban	Jan. 9, 2013-Feb. 6, 2013	25	257	159.5 \pm 66.3	RH:85%, WS: 2 m s ⁻¹ , WD: SE and NE	Liu et al., (2016)
King Fahd (Saudi Arabia)	Urban	2013.01	—	180	71.9	WS: 2 m s ⁻¹	Modaihsh et al., (2015)
Zonguldak (Turkey)	Urban	2007 winter	—	83.3	37.3	T: 1.3 °C. WS: 0.9 m s ⁻¹ , RH: 70%	Akyuz and Cabuk, (2009)
Delhi (India)	Near-road	2013.12-2014.01	—	—	293.1 \pm 36.7	—	Khanna et al., (2018)
Iasi (Romania)	Urban	Jan. 2016	—	—	23.4 \pm 11.7	—	Galon-Negru et al., (2018)
Xiangyang (China)	Urban	Jan. 13-24, 2018	12	347.7	169.3 \pm 57.0	T: 3.7 °C, RH:81%, WS: 3.02 m s ⁻¹ , WD: NW and NE	This study

Note. T: Temperature (°C), WS: Wind speed (m s⁻¹), WD: Wind direction, RH: Relative humidity (%).

Table S5. Average daily exposure doses (mg kg⁻¹ day⁻¹) of PM_{2.5}-bound metal(loid)s for the three different exposure pathways at the four pollution levels.

Pollution level	Metal(loid)s	Children				Adults			
		<i>ADED</i> _{Ing}	<i>ADED</i> _{Inh}	<i>ADED</i> _{Dermal}	<i>ADED</i>	<i>ADED</i> _{Ing}	<i>ADED</i> _{Inh}	<i>ADED</i> _{Dermal}	<i>ADED</i>
Mild pollution	As	7.57E-01	2.11E-05	7.95E-02	8.36E-01	1.00E-01	1.47E-05	1.72E-01	2.72E-01
	Cd	3.25E-04	9.07E-09	1.14E-06	3.26E-04	4.30E-05	6.33E-09	2.45E-06	4.55E-05
	Cr	4.76E-02	1.33E-06	1.66E-03	4.92E-02	5.31E-03	7.81E-07	3.03E-03	8.34E-03
	Co	2.28E+00	6.36E-05	7.97E-02	2.36E+00	3.02E-01	4.44E-05	1.72E-01	4.74E-01
	Cu	5.53E-02	1.55E-06	1.94E-03	5.72E-02	5.93E-03	8.71E-07	3.38E-03	9.30E-03
	Mn	2.42E-01	6.77E-06	8.48E-03	2.51E-01	2.60E-02	3.82E-06	1.48E-02	4.08E-02
	Ni	1.59E-02	4.44E-07	5.56E-04	1.64E-02	2.10E-03	3.09E-07	1.20E-03	3.30E-03
	Zn	2.30E+00	6.43E-05	8.06E-02	2.38E+00	2.47E-01	3.63E-05	1.41E-01	3.87E-01
	Pb	2.32E+00	6.48E-05	8.12E-02	2.40E+00	3.07E-01	4.52E-05	1.75E-01	4.83E-01
	Ag	1.43E-05	3.98E-10	4.99E-07	1.48E-05	1.53E-06	2.25E-10	8.71E-07	2.40E-06
	Al	1.33E+02	3.70E-03	4.64E+00	1.37E+02	1.42E+01	2.09E-03	8.10E+00	2.23E+01
	Ba	2.84E-01	7.92E-06	9.92E-03	2.93E-01	3.04E-02	4.47E-06	1.73E-02	4.77E-02
	Mo	4.95E-04	1.38E-08	1.73E-05	5.12E-04	5.31E-05	7.80E-09	3.02E-05	8.33E-05
	Sb	6.13E+02	1.71E-02	2.14E+01	6.34E+02	6.56E+01	9.65E-03	3.74E+01	1.03E+02
	Sr	2.66E-03	7.42E-08	9.29E-05	2.75E-03	2.85E-04	4.18E-08	1.62E-04	4.47E-04
	U	1.41E-06	3.95E-11	4.95E-08	1.46E-06	1.51E-07	2.23E-11	8.63E-08	2.38E-07
V	4.22E-03	1.18E-07	1.48E-04	4.37E-03	4.52E-04	6.65E-08	2.58E-04	7.10E-04	
Moderate pollution	As	1.31E-02	3.67E-07	1.38E-03	1.45E-02	1.74E-03	2.56E-07	2.97E-03	4.71E-03
	Cd	7.32E-04	2.04E-08	2.56E-06	7.34E-04	9.70E-05	1.43E-08	5.53E-06	1.03E-04
	Cr	4.46E-03	1.25E-07	1.56E-04	4.62E-03	4.98E-04	7.32E-08	2.84E-04	7.82E-04
	Co	3.29E-02	9.19E-07	1.15E-03	3.40E-02	4.36E-03	6.41E-07	2.48E-03	6.84E-03
	Cu	8.57E-03	2.39E-07	3.00E-04	8.87E-03	9.18E-04	1.35E-07	5.23E-04	1.44E-03
	Mn	1.61E-02	4.49E-07	5.63E-04	1.66E-02	1.72E-03	2.53E-07	9.82E-04	2.70E-03
	Ni	3.24E-03	9.06E-08	1.13E-04	3.36E-03	4.30E-04	6.32E-08	2.45E-04	6.75E-04
	Zn	5.87E-02	1.64E-06	2.05E-03	6.07E-02	6.28E-03	9.24E-07	3.58E-03	9.87E-03
	Pb	3.30E-02	9.22E-07	1.15E-03	3.42E-02	4.37E-03	6.43E-07	2.49E-03	6.87E-03
	Ag	1.02E-04	2.84E-09	3.56E-06	1.05E-04	1.09E-05	1.60E-09	6.21E-06	1.71E-05
	Al	2.89E-01	8.08E-06	1.01E-02	2.99E-01	3.10E-02	4.56E-06	1.77E-02	4.86E-02
	Ba	8.77E-03	2.45E-07	3.07E-04	9.08E-03	9.40E-04	1.38E-07	5.36E-04	1.48E-03
	Mo	4.63E-04	1.29E-08	1.62E-05	4.79E-04	4.96E-05	7.29E-09	2.83E-05	7.79E-05
	Sb	3.51E-01	9.81E-06	1.23E-02	3.63E-01	3.76E-02	5.53E-06	2.14E-02	5.91E-02
	Sr	2.27E-03	6.34E-08	7.94E-05	2.35E-03	2.43E-04	3.58E-08	1.39E-04	3.82E-04

Pollution level	Metal(loid)s	Children				Adults			
		<i>ADED</i> _{Ing}	<i>ADED</i> _{Inh}	<i>ADED</i> _{Dermal}	<i>ADED</i>	<i>ADED</i> _{Ing}	<i>ADED</i> _{Inh}	<i>ADED</i> _{Dermal}	<i>ADED</i>
		U	4.18E-05	1.17E-09	1.46E-06	4.32E-05	4.47E-06	6.58E-10	2.55E-06
V	2.03E-03	5.66E-08	7.09E-05	2.10E-03	2.17E-04	3.19E-08	1.24E-04	3.41E-04	
Heavy pollution	As	6.67E-03	1.86E-07	7.00E-04	7.37E-03	8.84E-04	1.30E-07	1.51E-03	2.40E-03
	Cd	7.90E-04	2.21E-08	2.76E-06	7.93E-04	1.05E-04	1.54E-08	5.97E-06	1.11E-04
	Cr	4.95E-03	1.38E-07	1.73E-04	5.12E-03	5.53E-04	8.13E-08	3.15E-04	8.68E-04
	Co	2.45E-02	6.86E-07	8.59E-04	2.54E-02	3.25E-03	4.78E-07	1.85E-03	5.11E-03
	Cu	7.53E-03	2.10E-07	2.63E-04	7.79E-03	8.06E-04	1.19E-07	4.60E-04	1.27E-03
	Mn	1.51E-02	4.22E-07	5.29E-04	1.56E-02	1.62E-03	2.38E-07	9.23E-04	2.54E-03
	Ni	3.70E-03	1.04E-07	1.30E-04	3.83E-03	4.91E-04	7.22E-08	2.80E-04	7.71E-04
	Zn	6.65E-02	1.86E-06	2.33E-03	6.89E-02	7.13E-03	1.05E-06	4.06E-03	1.12E-02
	Pb	5.64E-02	1.58E-06	1.97E-03	5.84E-02	7.47E-03	1.10E-06	4.26E-03	1.17E-02
	Ag	9.14E-05	2.56E-09	3.20E-06	9.47E-05	9.80E-06	1.44E-09	5.58E-06	1.54E-05
	Al	2.89E-01	8.06E-06	1.01E-02	2.99E-01	3.09E-02	4.55E-06	1.76E-02	4.85E-02
	Ba	1.27E-02	3.55E-07	4.45E-04	1.32E-02	1.36E-03	2.00E-07	7.77E-04	2.14E-03
	Mo	6.63E-04	1.85E-08	2.32E-05	6.87E-04	7.11E-05	1.05E-08	4.05E-05	1.12E-04
	Sb	2.32E-01	6.47E-06	8.11E-03	2.40E-01	2.48E-02	3.65E-06	1.41E-02	3.90E-02
	Sr	2.36E-03	6.59E-08	8.26E-05	2.44E-03	2.53E-04	3.72E-08	1.44E-04	3.97E-04
	U	2.98E-05	8.32E-10	1.04E-06	3.08E-05	3.19E-06	4.69E-10	1.82E-06	5.01E-06
	V	1.25E-03	3.50E-08	4.38E-05	1.30E-03	1.34E-04	1.97E-08	7.64E-05	2.11E-04
Severe pollution	As	5.10E-03	1.43E-07	5.36E-04	5.64E-03	6.76E-04	9.95E-08	1.16E-03	1.83E-03
	Cd	7.95E-04	2.22E-08	2.78E-06	7.98E-04	1.05E-04	1.55E-08	6.01E-06	1.11E-04
	Cr	8.07E-03	2.26E-07	2.82E-04	8.35E-03	9.01E-04	1.33E-07	5.14E-04	1.42E-03
	Co	1.25E-02	3.48E-07	4.36E-04	1.29E-02	1.65E-03	2.43E-07	9.42E-04	2.59E-03
	Cu	6.62E-03	1.85E-07	2.32E-04	6.85E-03	7.09E-04	1.04E-07	4.04E-04	1.11E-03
	Mn	1.24E-02	3.48E-07	4.35E-04	1.29E-02	1.33E-03	1.96E-07	7.60E-04	2.09E-03
	Ni	4.43E-03	1.24E-07	1.55E-04	4.58E-03	5.87E-04	8.63E-08	3.34E-04	9.21E-04
	Zn	5.68E-02	1.59E-06	1.99E-03	5.88E-02	6.09E-03	8.95E-07	3.47E-03	9.56E-03
	Pb	3.00E-02	8.39E-07	1.05E-03	3.11E-02	3.98E-03	5.85E-07	2.27E-03	6.25E-03
	Ag	6.89E-05	1.92E-09	2.41E-06	7.13E-05	7.38E-06	1.09E-09	4.21E-06	1.16E-05
	Al	1.89E-01	5.27E-06	6.60E-03	1.95E-01	2.02E-02	2.97E-06	1.15E-02	3.17E-02
	Ba	5.60E-03	1.56E-07	1.96E-04	5.79E-03	6.00E-04	8.82E-08	3.42E-04	9.42E-04
	Mo	4.91E-04	1.37E-08	1.72E-05	5.08E-04	5.26E-05	7.73E-09	3.00E-05	8.25E-05
	Sb	1.47E-01	4.11E-06	5.15E-03	1.52E-01	1.58E-02	2.32E-06	8.98E-03	2.47E-02
	Sr	1.45E-03	4.05E-08	5.07E-05	1.50E-03	1.55E-04	2.28E-08	8.84E-05	2.44E-04
	U	2.35E-05	6.56E-10	8.22E-07	2.43E-05	2.52E-06	3.70E-10	1.43E-06	3.95E-06
	V	8.79E-04	2.46E-08	3.08E-05	9.10E-04	9.42E-05	1.39E-08	5.37E-05	1.48E-04

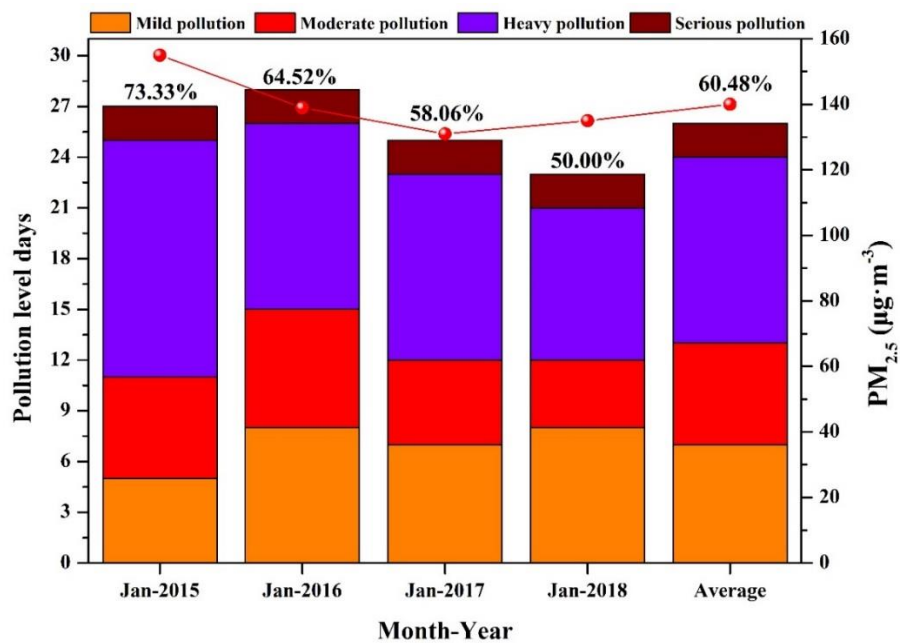


Fig. S1. The pollution level days and PM_{2.5} concentrations at the four pollution levels in January from 2015-2018.

References

- Akyuz, M. and Cabuk, H. (2009). Meteorological variations of PM_{2.5}/PM₁₀ concentrations and particle-associated polycyclic aromatic hydrocarbons in the atmospheric environment of Zonguldak, Turkey. *J. Hazard. Mater.* 170: 13-21.
- Ferreira-Baptista, L. and De Miguel, E. (2005). Geochemistry and risk assessment of street dust in Luanda, Angola: A tropical urban environment. *Atmos. Environ.* 39: 4501-4512.
- Galon-Negru, A.G., Olariu, R.I. and Arsene, C. (2018). Chemical characteristics of size-resolved atmospheric aerosols in Iasi, North-eastern Romania: Nitrogen-containing inorganic compounds control aerosol chemistry in the area. *Atmos. Chem. Phys.* 18: 5879-5904.
- Hu, X., Zhang, Y., Ding, Z., Wang, T., Lian, H., Sun, Y. and Wu, J. (2012). Bioaccessibility and health risk of arsenic and heavy metals (Cd, Co, Cr, Cu, Ni, Pb, Zn and Mn) in TSP and PM_{2.5} in Nanjing, China. *Atmos. Environ.* 57: 146-152.
- Khanna, I., Khare, M., Gargava, P. and Khan, A.A. (2018). Effect of PM_{2.5} chemical constituents on atmospheric visibility impairment. *J Air Waste Manag Assoc* 68: 430-437.
- Li, H., Qian, X., Hu, W., Wang, Y. and Gao, H. (2013). Chemical speciation and human health risk of trace metals in urban street dusts from a metropolitan city, Nanjing, SE China. *Sci. Total Environ.* 456-457: 212-221.
- Liu, J., Li, J., Vonwiller, M., Liu, D., Cheng, H., Shen, K., Salazar, G., Agrios, K., Zhang, Y., He, Q., Ding, X., Zhong, G., Wang, X., Szidat, S. and Zhang, G. (2016). The importance of non-fossil sources in carbonaceous aerosols in a megacity of central china during the 2013 winter haze episode: A source apportionment constrained by radiocarbon and organic tracers. *Atmos. Environ.* 144: 60-68.

- Modaihsh, A.S., Al-Barakah, F.N., Nadeem, M.E.A. and Mahjoub, M.O. (2015). Spatial and temporal variations of the particulate matter in Riyadh city, Saudi Arabia. *Journal of Environmental Protection* 6: 1293-1307.
- Qu, Y., Gao, T. and Yang, C. (2019). Elemental characterization and source identification of the near - road PM_{2.5} using edxrf in Chengdu, China. *X-Ray Spectrometry* 48: 232-241.
- Shen, R., Liu, Z., Chen, X., Wang, Y., Wang, L., Liu, Y. and Li, X. (2019). Atmospheric levels, variations, sources and health risk of PM_{2.5}-bound polycyclic aromatic hydrocarbons during winter over the North China Plain. *Sci. Total Environ.* 655: 581-590.
- USEPA (2011). *Exposure factors handbook: 2011 edition*. National Center for Environmental Assessment, Washington, DC.
- Wang, S., Yin, S., Zhang, R., Yang, L., Zhao, Q., Zhang, L., Yan, Q., Jiang, N. and Tang, X. (2019). Insight into the formation of secondary inorganic aerosol based on high-time-resolution data during haze episodes and snowfall periods in Zhengzhou, China. *Sci. Total Environ.* 660: 47-56.
- Wei, X.-Y., Liu, M., Yang, J., Du, W.-N., Sun, X., Huang, Y.-P., Zhang, X., Khalil, S.K., Luo, D.-M. and Zhou, Y.-D. (2019). Characterization of PM_{2.5}-bound PAHs and carbonaceous aerosols during three-month severe haze episode in Shanghai, China: Chemical composition, source apportionment and long-range transportation. *Atmos. Environ.* 203: 1-9.
- Xie, Y., Liu, Z., Wen, T., Huang, X., Liu, J., Tang, G., Yang, Y., Li, X., Shen, R., Hu, B. and Wang, Y. (2019). Characteristics of chemical composition and seasonal variations of PM_{2.5} in Shijiazhuang, China: Impact of primary emissions and secondary formation. *Sci. Total Environ.* 677: 215-229.

Xue, H., Liu, G., Zhang, H., Hu, R. and Wang, X. (2019). Similarities and differences in PM₁₀ and PM_{2.5} concentrations, chemical compositions and sources in Hefei city, China. *Chemosphere* 220: 760-765.

Zhong, S., Zhang, L., Jiang, X. and Gao, P. (2019). Comparison of chemical composition and airborne bacterial community structure in PM_{2.5} during haze and non-haze days in the winter in Guilin, China. *Sci. Total Environ.* 655: 202-210.