

1 **Measurement report: Crustal materials play an increasing role in elevating**  
2 **particle pH: Insights from 12-year records in a typical inland city of China.**

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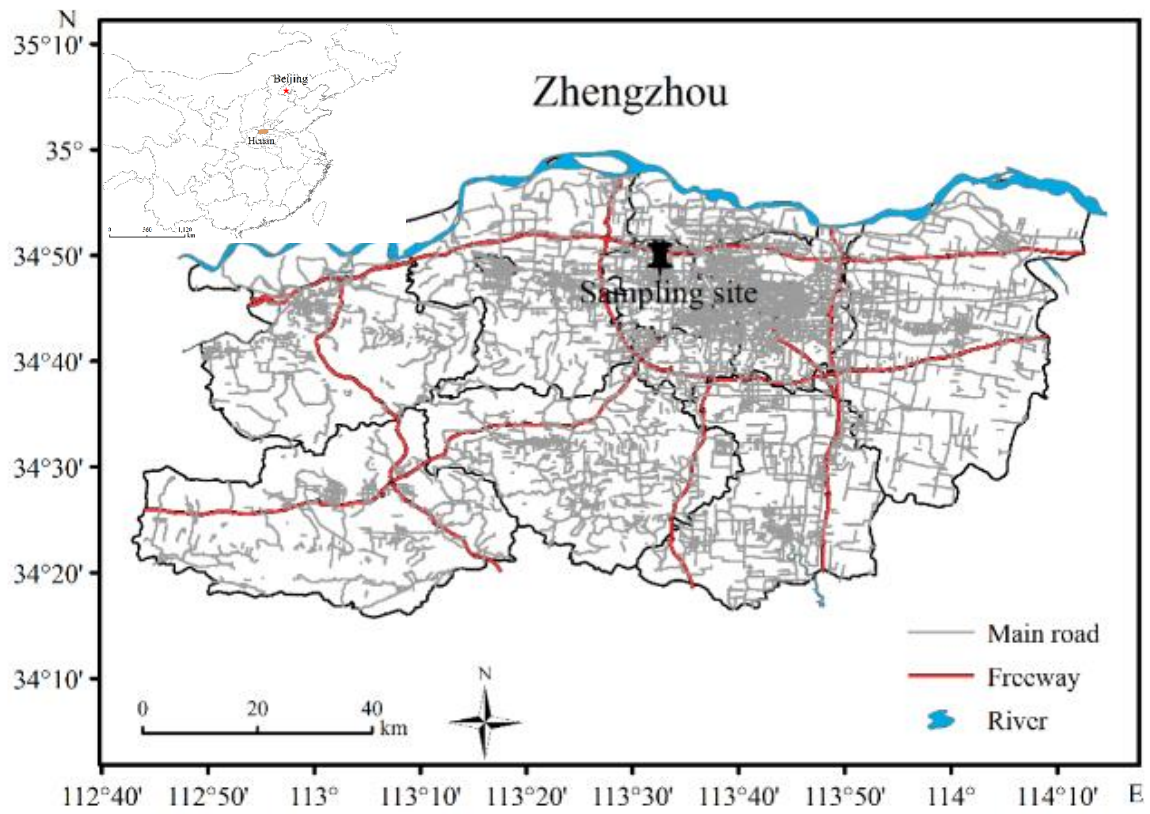
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14 **Figures**



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Figure S1. Sampling site in Zhengzhou, China. © 2019 National Geomatics Center of China. All rights reserved.

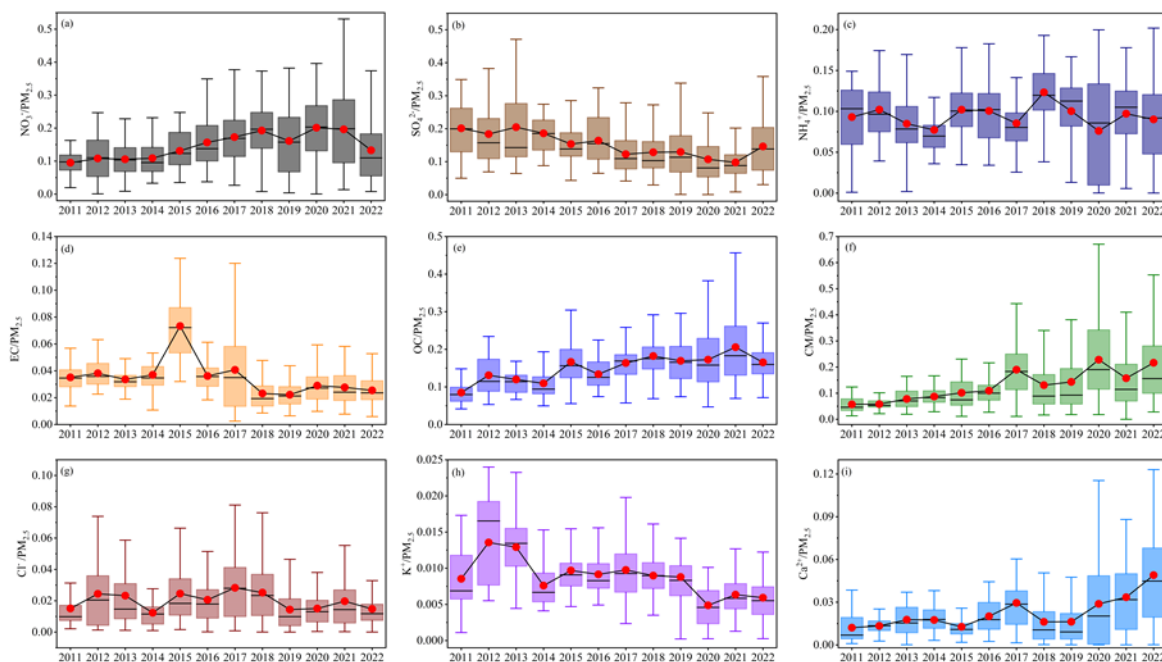


Figure S2. Trends in the proportions of chemical components in PM<sub>2.5</sub> from 2011 to 2022.

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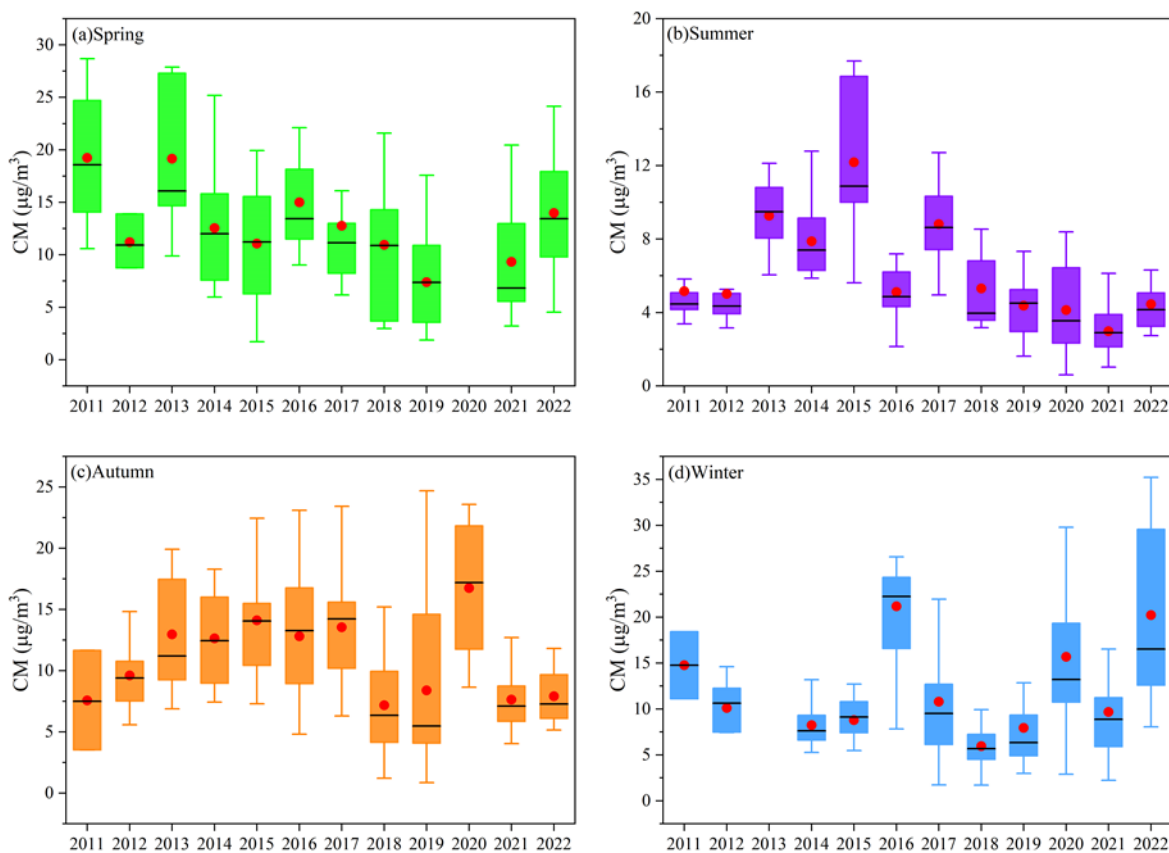
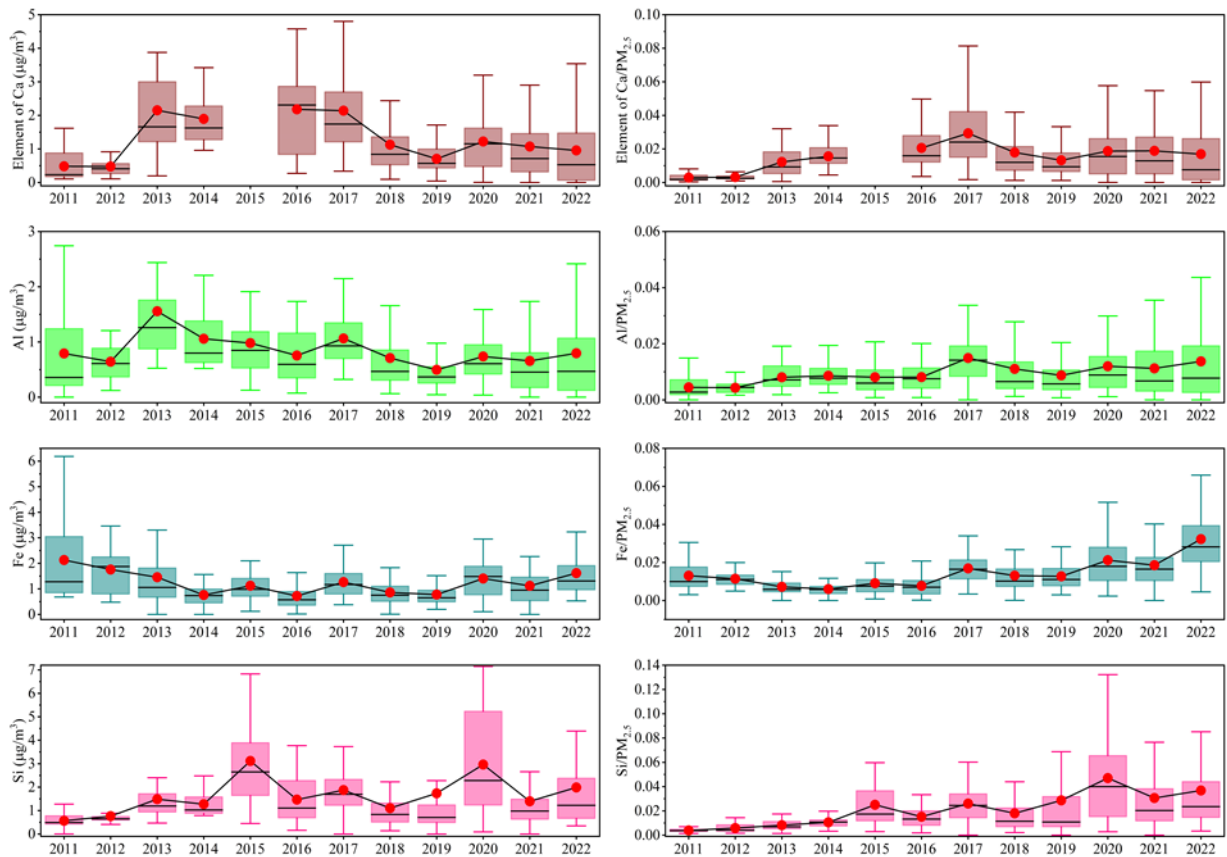


Figure S3. Trends in the CM concentrations in different seasons from 2011 to 2022.

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Figure S4. Trends in the concentrations of crustal elements and their proportions in PM<sub>2.5</sub> from 2011 to 2022.

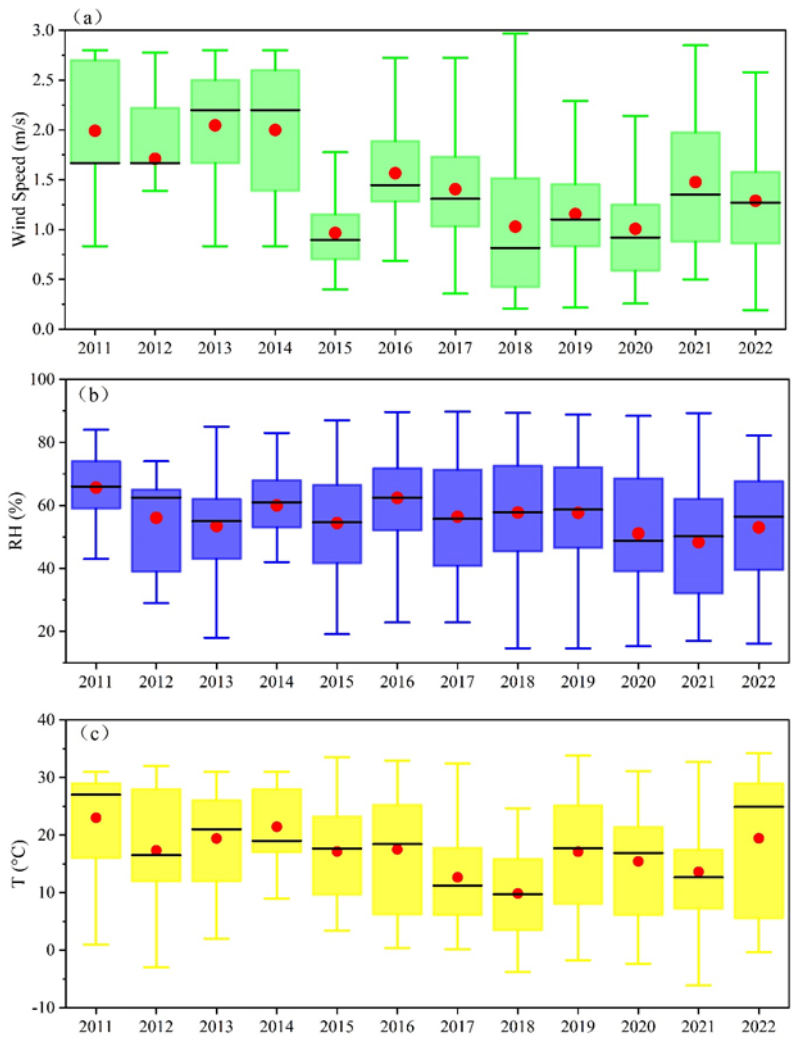


Figure S5. Trends in the meteorological parameters from 2011 to 2022.

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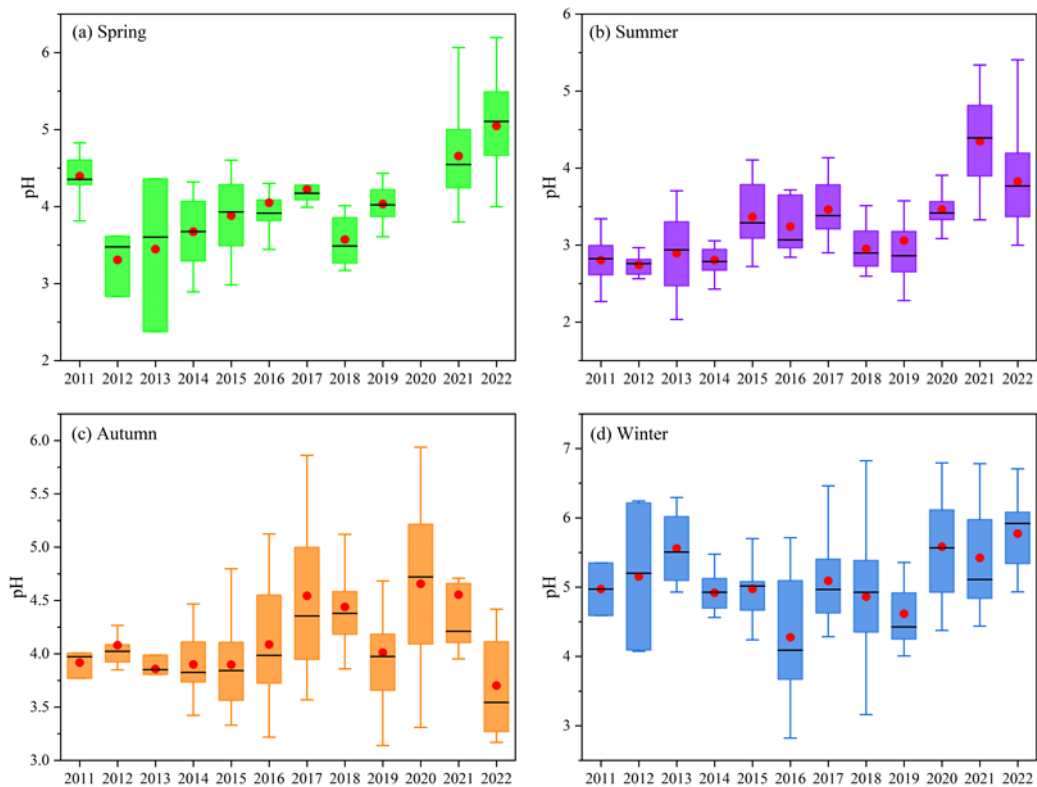


Figure S6. Trends in the particle pH in different seasons from 2011 to 2022.

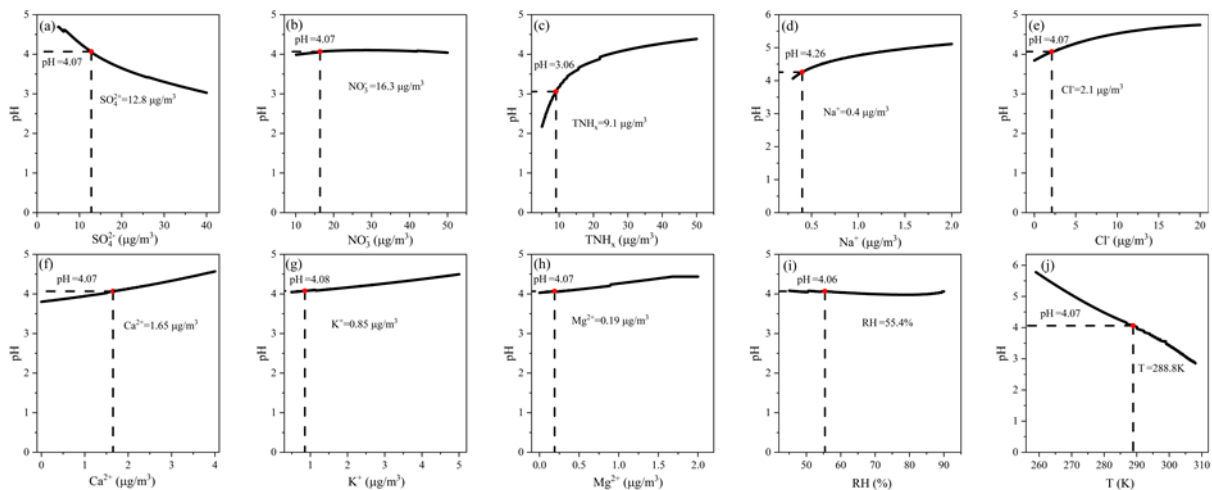
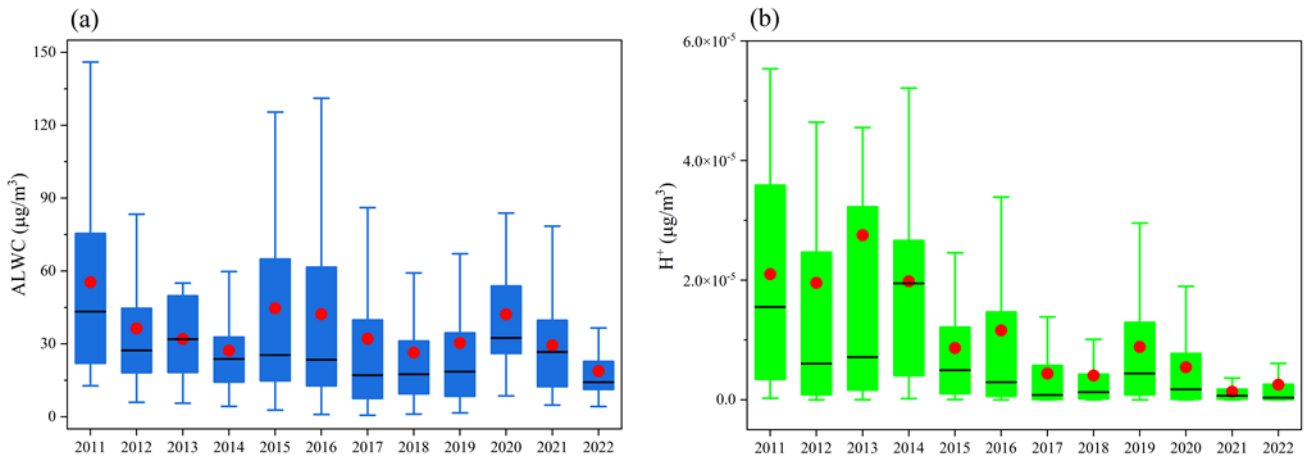


Figure S7. Sensitivity analysis of input parameters to particle pH. The dashed line represents the average of the observational data from 2011 to 2022.

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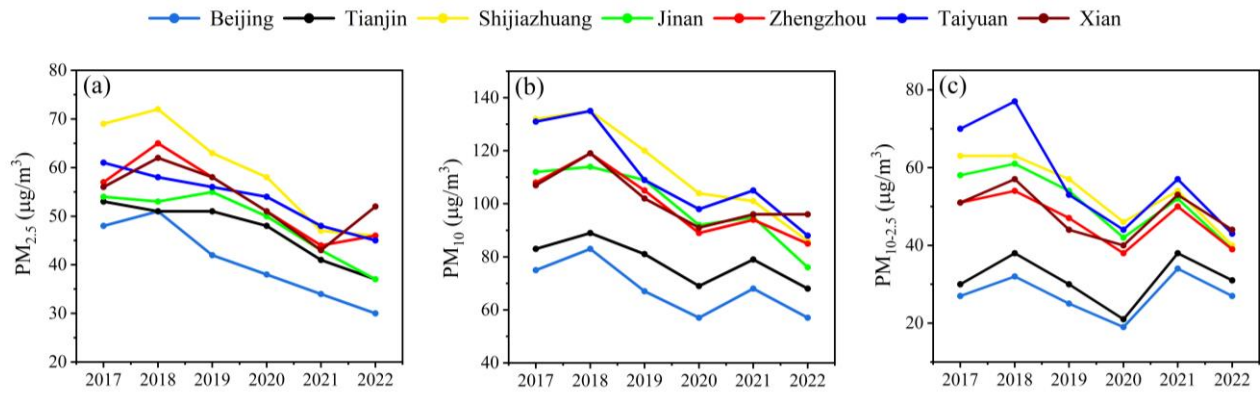


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Figure S8. Trends in aerosol liquid water content (ALWC) and  $H^+$  concentrations from 2011 to 2022



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Figure S9. Trends in the annual average concentrations of  $PM_{2.5}$ ,  $PM_{10}$ , and  $PM_{10-2.5}$  in provincial capitals in the North China Plain.

50 Table S1. Information on sampling date and numbers.

Years	Sampling date	The effective number of samples
2011	April 7–20 July 1–31 October 28–December 2 December 11–November 23	188
2012	February 25–26 April 21–May 6 July 22–August 2 October 17–November 1 December 8–25	140
2013	February 25–March 6 April 1–May 1 June 5–July 30 September 20–October 13 December 2–18	184
2014	April 1–May 5 June 18–July 20 October 7–24 December 30–31	180
2015	January 1–15 April 1–20 July 1–20 October 9–24	248
2016	January 6–22 April 8–30 July 9–31 October 1–20 December 29–31	252
2017	January 1–20 April 18–May 4 July 1–26 October 14–December 31	480
2018	January 1–31 April 1–30 July 1–31 October 9–December 31	600
2019	January 1–31 April 1–30 July 1–31 September 2–October 31 November 12–30 December 21–31	592



2020	January 1–20 June 5–July 31 October 6–November 13 December 15–31	332
2021	January 1–31 March 16–April 30 July 1–August 8 October 17–December 31	540
2022	January 1–4 April 1–May 3 July 1–August 11 September 5–October 11 December 10–31	492
Total		4228

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56 Table S2. Control measures for dust implemented by Henan Province and Zhengzhou government

Release time	Policies	Regulatory focus
2013.9	Regulations on Reducing Pollutant Emissions in Henan Province	Road, Construction
2014.8	Temporary Regulations on Dust Control Management at Construction Sites in Henan Province	Construction
2016.7	Implementation Plan for Controlling Dust Pollution in Henan Province	Road, Construction
2018.2	Regulations on the Prevention and Control of Atmospheric Pollution in Henan Province	Road, Construction, Piles
2019.4	Special Action Plan for Fine Management of Dust Pollution Prevention and Control at Construction Sites in Zhengzhou City, 2019	Construction
2019.8	Enhanced Action Plan for Intensive Dust Control at Construction Sites in 2019	Construction
2021.1	Special Governance Plan for Key Project Dust Pollution in Zhengzhou	Road, Construction, Piles

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Table S3. The ratios of Ca/Si in the source spectrum of different dust sources in China

Dust source	City	Ca/Si	Reference
Road dust	Xi'an	2.04	<a href="http://www.klaccp.ac.cn/wgPMzypfypk/ycy/201706/t20170610_375562.html">http://www.klaccp.ac.cn/wgPMzypfypk/ycy/201706/t20170610_375562.html</a>
	Yinchuan	2.48	
	Lanzhou	1.67	
	Beijing	1.25	
	Tianjin	1.03	
	Baoding	1.16	
	Shijiazhuang	1.98	
	Handan	1.83	
	Shenyang	1.81	
	Changsha	1.92	
	Chongqing	1.38	
	Chengdu	1.17	
	Kunming	1.94	
	Taiyuan	1.55	
	Nanjing	1.28	
Construction dust	Xi'an	1.69	<a href="http://www.klaccp.ac.cn/wgPMzypfypk/ycy/201706/t20170610_375562.html">http://www.klaccp.ac.cn/wgPMzypfypk/ycy/201706/t20170610_375562.html</a>
	Yinchuan	1.84	
	Lanzhou	2.33	
	Beijing	2.65	
	Tianjin	1.46	
	Baoding	1.58	
	Shijiazhuang	1.38	
	Handan	1.86	
	Shenyang	1.92	
	Changsha	2.30	
	Chongqing	2.52	
	Chengdu	2.15	
	Kunming	1.60	
	Taiyuan	1.92	
	Nanjing	2.26	
Piles dust	Xi'an	0.72	(Yang, 2016)
	Tianjin	0.57	(Zhang et al., 2018)
	Taiyuan	0.61	(Bi et al., 2007)
	Jinan	1.01	(Bi et al., 2007)
	/	0.65	<a href="http://www.nkspap.com:9091/Index.aspx">http://www.nkspap.com:9091/Index.aspx</a>
Soil dust	Nanchang	0.37	(Xu et al., 2019)
	Xi'an	0.27	(Yang, 2016)
	Jincheng	0.13	(Wang et al., 2016)
	Wuhan	0.52	(Gong and Luo, 2018)
	/	0.53	<a href="http://www.nkspap.com:9091/Index.aspx">http://www.nkspap.com:9091/Index.aspx</a>

Table S4. The difference between component concentrations ( $\mu\text{g}/\text{m}^3$ ) and meteorological parameters between adjacent years.

Years	$\text{NO}_3^-$	$\text{SO}_4^{2-}$	$\text{TNH}_x$	$\text{Na}^+$	$\text{Cl}^-$	$\text{K}^+$	$\text{Ca}^{2+}$	$\text{Mg}^{2+}$	RH (%)	T ( $^\circ\text{C}$ )
2012VS2011	4.0	-4.6	1.3	0.02	2.0	0.9	-0.2	0.04	-9.6	-5.7
2013VS2012	2.6	13.0	2.1	0.2	0.4	0.3	1.4	0.1	-2.6	2.1
2014VS2013	-7.3	-14.6	-6.9	-0.4	-3.4	-1.6	-1.1	-0.2	6.6	2.0
2015VS2014	5.2	-1.8	5.5	0.1	2.1	0.4	-0.6	0.6	-5.6	-4.2
2016VS2015	-0.2	-4.5	-3.7	-0.03	-0.1	-0.4	0.5	-0.7	8.0	0.3
2017VS2016	-2.9	-5.3	-3.6	-0.2	-0.3	-0.2	-0.1	0.1	-6.0	-4.9
2018VS2017	-0.8	-2.4	1.3	-0.1	-0.8	-0.2	-0.1	-0.1	1.4	-2.8
2019VS2018	-3.0	-0.8	-2.2	-0.04	-0.7	-0.03	-0.1	-0.01	-0.1	7.3
2020VS2019	4.9	-0.3	-0.9	0.1	0.1	-0.2	0.7	0.02	-6.6	-2.1
2021VS2020	-3.6	-2.3	0.2	-0.01	0.03	0.01	0.1	0.04	-2.8	-1.5
2022VS2021	-5.1	1.9	-1.4	0.03	-0.3	0.01	0.5	0.04	4.7	5.8

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