1	Supplementary Information for						
2	Fugitive road dust PM _{2.5} emissions and their potential						
3	health impacts						
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The FRD sample Collection and Measuring characteristics of road-deposited sediment

We collected FRD samples using a domestic vacuum cleaner (Philips FC6400) 38 from July to November 2017 during a dry weather period and wind velocities less 39 40 than 7.9 m s⁻¹. Samples were collected every 0.8 km along the length of roads longer than 2.4 km and at three random sample sites for roads less than 2.4 km (Figure S1a). 41 At each sampling location, a rectangular sampling grid was selected, with a width of 42 0.5 m and a length being the width of the road (Figure S1a and c). The handheld 43 44 cordless vacuum cleaner can collect road-deposited sediment conveniently (Figure S1b). Moreover, the vacuum cleaner has high efficiency to catch both fine and coarse 45 particulates with air filtration, dust bucket, the dust separator and the cyclone. The 46 FRD samples were preserved in numbered vacuum cleaner bags and dried at 35 °C 47 for 7 days (Figure S1d). 48

We measured the value of silt loading at each sampling site by a 200-mesh sieve (<75 μ m) an electronic weighting scale. And the silt loading (sL, units: g m⁻²) is calculated as follow:

$$SL = \frac{m_{total} - m_{75\mu \,\mathrm{m}}}{S} \tag{1}$$

52

53 Where m_{total} is the mass of total FRD samples; $m_{75\mu m}$ is the mass of FRD samples 54 lager than 75 μ m; S is sampling area.

The size distribution of FRD samples was measured in laboratory. A 10-mesh sieve (<2 mm) is used to screen out leaves, scree and cigarette butts. The gross samples were divided by using coning and quartering¹. To measure the size of dust particles more accurately, the remaining particles were further oxidized by using hydrogen peroxide solution and hydrochloric acid solution to remove potential contaminants (organic matter and calcium carbonate) from the FRD samples. And then the samples were tested by a laser particle sizer (Malvern Mastersizer, 2000) to determine the size distribution of the FRD.

63

64 Simulation of FRD PM_{2.5} concentrations based on the WRF-Chem model

65 1. WRF-Chem model

The Weather Research and Forecasting coupled with Chemistry (WRF-Chem) mode was investigated to simulate FRD PM_{2.5} concentrations in the study. Gas-phase chemical mechanisms, photolysis schemes, and aerosol schemes are coupled into the WRF-Chem model, which considers a variety of coupled physical and chemical processes such as aerosol emission, transport, deposition, aerosol interactions, chemical transport, and radiative forcing².

72 2. Model configuration

The key physical and chemical schemes used in simulations are listed in Table 73 S2. It is noted that Peking University (PKU) emission inventory has six sectors 74 including energy production, industry, transportation, residential & commercial, 75 agriculture and deforestation & wildfire for CO₂, CO, PM_{2.5}, PM₁₀, TSP, BC, OC, SO₂, 76 NOx, and NH₃, and polycyclic aromatic hydrocarbons³⁻⁸. PKU emission inventories 77 with 0.1 by 0.1 degree spatial resolution and monthly temporal resolutions in 2014 78 have been included in the WRF-Chem model in this study. And FRD emission 79 80 inventory is constructed in this study.

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In this study, we divided the study domain into the grid of 1450 square cells with

a horizontal grid interval of 500 m. The domain covered the whole urban Lanzhou in 82 China, as shown in Fig. S3. The model atmosphere was divided into 35 vertical layers, 83 84 and the top pressure of the model was 100 hPa. The simulation period was from December 15st, 2016–December 31st 2017. Only the results from the whole year in 85 86 2017 were used in this study. The initial and boundary meteorological conditions were constructed from the National Center for Environmental Prediction Final 87 Analysis (NCEP/FNL) data at a 6 h temporal interval and 1 degree horizontal 88 resolution. To produce a more realistic simulation, the modeled u- and v-wind 89 90 components and atmospheric temperatures were nudged towards the NCEP/FNL data 91 with a nudging timescale of 6 h.

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93 Ground monitoring PM_{2.5} Data

Daily $PM_{2.5}$ data from January to December 2017 in Lanzhou, China were obtained from the website of the China National Environmental Monitoring Center (http://113.108.142.147:20035/). Based on using the tapered elementoscillating microbalance to measure $PM_{2.5}$, the platform displays the real-time concentration of $PM_{2.5}$. This data has covered all cities at prefecture level since 2015 and has been widely used to investigate the acute health effects of ambient $PM_{2.5}^{8,9}$.

100

101 The diurnal cycle of traffic volume

The data of traffic volume is provided by Traffic Police Detachment of Lanzhou Public Security Bureau. The traffic volume is counted by monitors at each road intersection. Based on quality control, we get diurnal cycle of traffic volume on each road ("roads" refers to the road segment between intersections), including 45 main roads, 60 minor roads, and 55 branch roads. Compared with our observation, the data of traffic volume from monitor is reliable. 108 The diurnal cycle of traffic volumes on the major road, minor road, and branch 109 road are shown in Figure S2. The traffic volumes show the lowest at 5:00 local time with average value about 250, 100, 80 vehicles h⁻¹ on the major, minor, and branch 110 road, respectively, and maintained the high value during 9:00 to 23:00 local time (LT) 111 with dramatic increase after 7:00 local time and down at 23:00 LT. Especially, the 112 magnitude of traffic volumes is the highest on the major road, followed by the minor 113 road and branch road. The traffic volumes have a slight variation during the day time 114 115 mainly ranging from 1000 to 2500, 800 to 1400 vehicles h⁻¹ and 500 to 1000 vehicles h⁻¹ on the major road, minor road, and the branch road, respectively. The high periods 116 on three types of roads are all delayed about one or two hours on weekends compared 117 with those on weekdays. The traffic volumes change more significantly on major 118 roads than those on minor road, and the slight variations of traffic volumes occur on 119 branch roads. 120

121

122 FRD PM₁₀ emission.

123 The spatial distribution of FRD PM10 emission fluxes in Lanzhou is constructed in Figure S3a. The magnitude of FRD PM_{2.5} emission in Lanzhou is estimated to be 124 approximately 3216 kg d⁻¹. The FRD PM_{2.5} emission fluxes are enhanced over the 125 regions with large traffic volumes and high density of road network, predominantly in 126 the central of the DET and the eastern of the DIT, where the value is larger than 3×10^4 127 μg m⁻² d⁻¹. The FRD PM10 emission fluxes with comparatively lower values varying 128 from 11.8×10^4 µg m⁻² d⁻¹ to 7.5×10^4 µg m⁻² d⁻¹ are occurred in the UT and ID (Figure 129 S3a). The spatial distributions of the FRD PM₁₀ emission fluxes are found to be quite 130 similar to that of the PM_{2.5} emission fluxes. The FRD emissions with the PM_{2.5}/PM₁₀ 131 ratio of 0.35 can sufficiently increase the amount of fine particulate matters in urban 132 areas, which could be suspended in the ambient atmosphere over a longer time and be 133 more harmful for human health compared to its coarse fraction³. The FRD PM₁₀ 134 emission factors, as an indicator of the FRD emission ability, are sensitive factors in 135 the construction of emission inventory. The FRD PM10 emission factors are 136 approximately 3 times lager than the PM_{2.5} emission factors. The interaction of large 137

silt loading and small particle size causes high values of FRD PM₁₀ emission factors 138 in the DIT and ID, with average value of 1.13 and 0.96 g VKT⁻¹, respectively (Figure 139 S3b). And the magnitude of FRD PM10 emission in the different UFZs decrease in 140 the order DET (1188 kg d⁻¹) > DIT (1023 kg d⁻¹) > ID (693 kg d⁻¹) > UT (312 kg d⁻¹) 141 (Figure S3c). The diurnal cycle of FRD PM₁₀ emission is mainly consistent with 142 variation of traffic volumes, that is, the lowest FRD PM₁₀ emission occurs at 5:00 LT 143 with value as low as 19.98 kg h⁻¹ while rises dramatically to 165.42 kg h⁻¹ at 11:00 LT. 144 145 It maintains the high value from 8:00 to 23:00 LT accompanied by human activities, exposing citizen to high PM₁₀ level (Figure S3d). Moreover, meteorological 146 conditions also influence FRD emission as the monthly FRD PM₁₀ emission is the 147 largest $(8.5 \times 10^4 \text{ kg month}^{-1})$ in winter, followed by spring $(8.0 \times 10^4 \text{ kg month}^{-1})$ and 148 149 smallest $(7.8 \times 104 \text{ kg month}^{-1})$ in summer which aligns with the precipitation cycles (Figure S3e). 150

151 **References:**

Procedures for Laboratory Analysis of Surface/Bulk Dust Loading Samples; Appendix C:2; United States Environmental Protection Agency, 1993; www3.epa.gov/ttn/chief/ap42/appendix/app-c2.pdf.

- 155 2. Grell, G. A.; Peckham, S. E.; Schmitz, R.; McKeen, S. A.; Frost, G.; Skamarock,
- W. C.; Eder, B. Fully coupled "online" chemistry within the WRF model. *Atmos. Environ.* 2005, 39, 6957–6975, DOI 10.1016/j.atmosenv.2005.04.027, 2005.
- 158 3. Huang, Y.; Shen, H. Z.; Chen, H.; Wang, R.; Zhang, Y. Y.; Su, S.; Chen, Y. C.; Lin,
- 159 N.; Zhuo, S. J.; Zhong, Q. R.; Wang, X. L.; Liu, J. F.; Li, B. G.; Liu, W. X.; Tao,
- 160 S. Quantification of global primary emissions of pm2.5, pm10, and tsp from
- 161 combustion and industrial process sources. *Environ. Sci. Technol.* 2014, 48,
 162 13834-13843.
- 163 4. Meng, W. J.; Zhong, Q. R.; Yun, X.; Zhu, X.; Huang, T. B.; Shen, H. Z.; Chen, Y.

- 164 L.; Chen, H.; Zhou, F.; Liu, J. F.; Wang, X. M.; Zeng, E. Y.; Tao, S. Improvement
- ----

of a global high-resolution ammonia emissions inventory for combustion and
 industrial sources with new data from the residential and transportation sectors.
 Environ. Sci. Technol. 2017, 51 (5), 2821-2829.

- 168 5. Shen, H.; Huang, Y.; Wang, R.; Zhu, D.; Li, W.; Shen, G.; Wang, B.; Zhang, Y.;
- 169 Chen, Y.; Lu, Y.; Chen, H.; Li, T.; Sun K.; Li, B.; Liu, W.; Liu, J.; Tao, S. Global
- atmospheric emissions of polycyclic aromatic hydrocarbons from 1960 to 2008
 and future prediction., *Environ. Sci. Technol.* 2013, 47, 6415–6424.
- 172 6. Wang, R.; Tao, S.; Ciais, P.; Shen, H. Z.; Huang, Y.; Chen, H.; Shen, G. F.; Wang,
- 173 B.; Li, W.; Zhang, Y. Y.; Lu, Y.; Zhu, D.; Chen, Y. C.; Liu, X. P.; Wang, W. T.;
- 174 Wang, X. L.; Liu, W. X.; Li, B. G.; Piao, S. L. High-resolution mapping of
- combustion processes and implications for CO2 emissions. *Atmos. Chem. Phys.*2013, 13, 5189-5203.
- 177 7. Zhong, Q.; Huang, Y.; Shen, H.; Chen, Y.; Chen, H.; Huang, T.; Zeng, E.; Tao, S.
- 178 Global estimates of carbon monoxide emissions from 1960 to 2013. *Environ. Sci.*
- 179 *Pollut. Res.* **2017**, 24, 864-873.
- 180 8. Chen, R.; Yin, P.; et al. Fine Particulate Air Pollution and Daily Mortality: A
- 181 Nationwide Analysis in 272 Chinese Cities. Am. J. Resp. Crit. Care. 2017, 196(1),
- 182 73-81; DOI 10.1164/rccm.201609-1862oc.
- 183 9. Song, C.; He, J.; et al. Health burden attributable to ambient PM 2.5 in China.
- 184 *Environ. Pollu.* **2017**, 223, 575 586; DOI 10.1016/j.envpol.2017.01.060.
- 185 10. Philip, S.; Martin, R.; Snider, G.; et al. Anthropogenic fugitive, combustion and

industrial dust is a significant, underrepresented fine particulate matter source in
global atmospheric models. *Environ. Res. Lett.* 2017, 12(4); DOI
doi.org/10.1088/1748-9326/aa65a4.

- 11. Chen, F.; Dudhia, J. Coupling an advanced land surface-hydrology model with
 the Penn State-NCAR MM5 modeling system. Part I: Model implementation and
 sensitivity. *Mon. Weather Rev.* 2001, 129, 569-585; DOI
 10.1175/1520-0493(2001)129< 0569:CAALSH> 2.0.CO;2
- 193 12. Hong, S. Y.; Noh, Y.; Dudhia, J. A New Vertical Diffusion Package with an
- Explicit Treatment of Entrainment Processes. *Mon. Weather Rev.* 2006, 134,
 2318–2341; DOI 10.1175/MWR3199.1, 2006.
- 196 13. Morrison, H.; Curry, J. A.; Khvorostyanov, V, I. A New Double-Moment
- 197 Microphysics Parameterization for Application in Cloud and Climate Models. Part I:
- 198 Description. J. Atmos. Sci. 2005, 62, 1665–1677; DOI 10.1175/JAS3446.1.
- 199 14. Mlawer, E, J.; Taubman, S, J.; Brown, P, D.; Iacono, M.J.; Clough, S, A. RRTM, a
- validated correlated-k model for the longwave. J. Geophys. Res. 1997, 102,
- 201 16663–16682; DOI 10.1029/97JD00237.
- 202 15. Iacono, M, J.; Mlawer, E, J.; Clough, S, A.; Morcrette, J, J. Impact of an
- 203 improved longwave radiation model, RRTM, on the energy budget and
- thermodynamic properties of the NCAR community climate model, CCM3. J.
- 205 *Geophys. Res.* **2000**, 105, 14. http://dx.doi.org/10.1029/2000JD900091.
- 206 16. Zaveri, R, A.; Peters, L, K. A new lumped structure photochemical mechanism
- 207 for large-scale applications. J. Geophys. Res. 1999, 104, 30387–30330,30415; DOI

S8

208 10.1029/1999JD900876.

209 17. Zaveri, R, A.; Easter, R, C.; Fast, J, D.; Peters, L, K. Model for simulating aerosol

interactions and chemistry (MOSAIC). J. Geophys. Res. 2008, 113, D13204; DOI

- 211 10.1029/2007JD008782.

	Atmospheric Process	Model Option
Physics	Land surface	Noah
	Boundary layer	YSU
	Cumulus clouds	New Grell scheme
	Cloud microphysics	Morrison 2-mom
	Long-wave radiation	RRTMG
	Shortwave radiation	RRTMG
Chemistry	Gas-phase chemistry	CBM-Z
	Aerosol chemistry	MOSAIC
	Photolysis	Fast-J

213 Table S1. The WRF-Chem configuration in this study

UFZs ^a	Spring	Summer	Autumn	Winter	Annual
	(^b FRD/Total ^c)	(FRD/Total)	(FRD/Total)	(FRD/Total)	(FRD/Total)
DET	13.1/45.7	10.1/31.9	14.6/44.6	19.9/66.2	14.4/47.1
DIT	9.2/41.4	5.9/27.7	11.2/41.2	16.8/63.6	10.8/43.5
UT	6.7/38.9	3.7/25.4	7.5/37.6	12.1/58.9	7.5/40.2
ID	5.3/37.4	3.1/24.9	7.0/37.1	9.9/56.7	6.3/39.0

227 Table S2. The average of $PM_{2.5}$ concentrations (unit: $\mu g m^{-3}$)

^aUrban function zones: UFZs= urban function zones; DET=developed downtown; DIT=developing downtown; UT=university town; ID=industrial district; ^bFRD: the FRD PM_{2.5} concentration simulated by the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) Model; ^dTotal: the simulated PM_{2.5} concentrations including FRD, natural dust and anthropogenic sources

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229

Table 55. The key parameters for estimation of premature mortainly						
	parameters	$COPD^a$	LC ^a	ALRIª	IHD ^a	stroke ^a
	α	0.565	0.841	1.854	1.043	1.579
	γ	0.019	0.014	0.002	0.104	0.013
	δ	0.861	0.915	1.281	0.684	1.235
	C_{θ}	5.8	5.8	5.9	5.8	5.8
	Baseline	43.8 (CI:	23.4 (CI:	28.6 (CI:	105.7 (CI:	42.3 (CI:
	mortality ^b	40.4; 49.1)	17.3; 27.3)	25.5; 30.6)	98.8; 111.9)	39.6; 48.7)
	^a Disease: CO	PD= chronic d	bstructive puli	monary diseas	se; LC= lung co	ancer; ALRI=
	acute lower	respiratory in	fections; IHD	= ischemic	heart disease;	and stroke=
	cerebrovascul	ar disease; ^b	Baseline mort	tality: CI de	enotes the 95%	% confidence
	intervals					

230 Table S3. The key parameters for estimation of premature mortality



231

Figure S1. The process of gathering samples. (a) Sampling locations; (b) road-deposited

233 sediments were sampled by a vacuum cleaner; (c) measuring the areas of the sampling grid;

234 (d) FRD samples collected in vacuum cleaner bags.

235



236

Figure S2 The diurnal cycle of traffic volume on (a) major road; (b) minor road; (c) branch road. (Dashes in the boxes denote medians of traffic volume. Opening and closing of the boxes presents 25 and 75th percentiles for each dataset. The dotted line tops of the boxes are maximum and minimum, respectively).

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Figure S3. (a) The pattern of FRD PM_{10} emission fluxes (unit: $\mu g m^{-2} d^{-1}$); (b) emission factors (unit: g VKT⁻¹), (c) Total amount (unit: kg d⁻¹), (d) Diurnal variations (unit: kg h⁻¹), and (e) Monthly variations (unit: kg month⁻¹) of FRD PM_{10} emission in four UFZs.



246

247 Figure S4 The spatial distributions of simulated FRD PM2.5 concentrations in (a)

spring, (b) summer, (c) autumn and (d) winter based on the WRF-Chem model.