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and Their Health Impact



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Fugitive Road Dust PM_{2.5} Emissions and Their Potential Health Impacts

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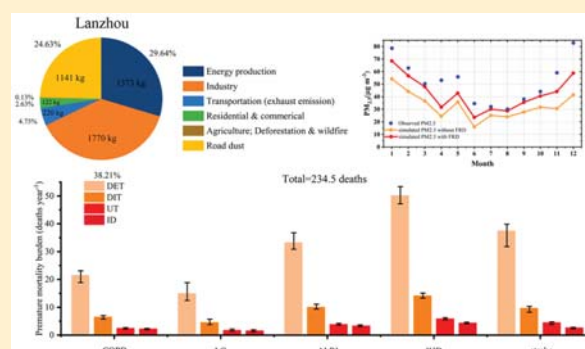
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Supporting Information

ABSTRACT: Fugitive road dust (FRD) particles emitted by traffic-generated turbulence are an important contributor to urban ambient fine particulate matter (PM_{2.5}). Especially in urban areas of developing countries, FRD PM_{2.5} emissions are a serious environmental threat to air quality and public health. FRD PM_{2.5} emissions have been neglected or substantially underestimated in previous study, resulting in the underestimation of modeling PM concentrations and estimating their health impacts. This study constructed the FRD PM_{2.5} emissions inventory in a major inland city in China (Lanzhou) in 2017 at high-resolution (500 × 500 m²), investigated the spatiotemporal characteristics of the FRD emissions in different urban function zones, and quantified their health impacts. The FRD PM_{2.5} emission was approximately 1141 ± 71 kg d⁻¹, accounting for 24.6% of total PM_{2.5} emission in urban Lanzhou. Spatially, high emissions exceeding 3 × 10⁴ μg m⁻² d⁻¹ occurred over areas with smaller particle sizes, larger traffic intensities, and more frequent construction activities. The estimated premature mortality burden induced by FRD PM_{2.5} exposure was 234.5 deaths in Lanzhou in 2017. Reducing FRD emissions are an important step forward to protect public health in many developing urban regions.



INTRODUCTION

Particulate matter (PM), a major environmental threat around the world, plays an important role in ambient air quality degradation as well as in acute damage to public health.^{1–4} Fugitive road dust (FRD) particles are those emitted from roads into the ambient atmosphere by traffic-generated turbulence.^{5–7} Sources of these particles include surrounding soils, mud carried by vehicles, demolition and construction, fly ash from asphalt, bioclastics, natural dust deposition, and other processes.^{8,9}

Over the recent decades, the contribution of FRD to the total PM has become increasingly important due in part to rapid growth of the global number of vehicles worldwide, i.e., approximately 4% per year from 2006 to 2013.^{7,10} This contribution is especially prominent in China, where the vehicle number has increased at a rate of 15% per year from 2015 to 2018.^{10–14} Global measurements indicated that FRD emissions accounted for 55% of the PM₁₀ (particles with

aerodynamic diameter < 10 μm) concentrations in Delhi, India, in 2010,¹⁵ 25.7% of PM₁₀ in Brazil, São Paulo, in 2014,¹⁶ and 14–48% of PM₁₀ in European urban areas from 2000 to 2009.¹⁷

FRD particles are an important carrier for high levels of harmful components such as heavy metal elements (i.e., Pb, Mn, Fe, Cu, Co, and others),^{18,19} polycyclic aromatic hydrocarbons (i.e., acenaphthene, anthracene, fluoranthene, and others),^{20,21} and other carcinogens,²² which exert a high potential health burden on cardiovascular, respiratory, and cancer diseases.²³ Therefore, understanding the magnitude and spatial and temporal distributions of FRD emissions is vital to better understand the interactions among FRD, air quality,

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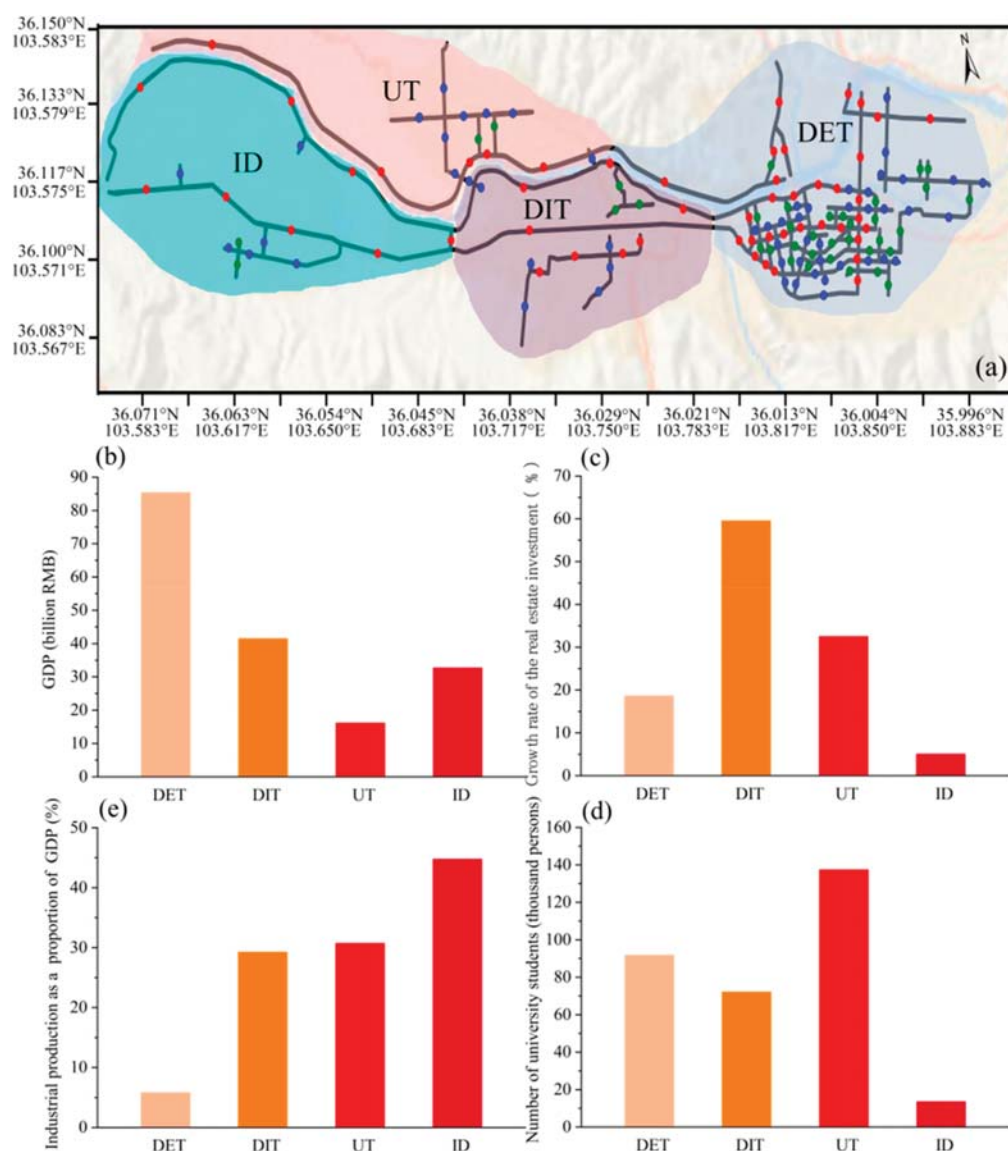


Figure 1. (a) Spatial distribution of sampling points in the study area (the red, blue, and green points represent the sampling points in the major roads, minor roads, and branch roads, respectively). And (b) GDP; (c) real estate investment growth rate; (d) number of university students (thousand persons); and (e) industrial production as a proportion of GDP in the four urban function zones (UFZs) of Lanzhou in 2017. DET = developed downtown; DIT = developing downtown; UT = university town; and ID = industrial district.

economic development, and public health. However, previous studies neglected or greatly underestimated FRD emissions,⁷ resulting in high uncertainties in model estimates of PM concentrations, especially for PM_{2.5}, and their environmental, health, and climatic effects.^{12,13,24,25} For example, FRD emissions are not included in current Chinese emission inventories,^{26–29} nor are they represented in many model simulations which use these inventories.^{30,31}

FRD emissions are particularly important in developing urban areas due to the large amounts of vehicles, dust sources, and inhabitants. The urban environment can be separated into several distinctive urban function zones (UFZs) differentiated by the intensity of social/economic activities and the characteristics of environmental pollution,^{32–34} such as downtown, residential, educational, and industrial zones.³⁵ In this study, we constructed a high-resolution (500 × 500 m²) gridded inventory for FRD PM_{2.5} emissions in 2017 in the

urban area of Lanzhou, a major inland city in China threatened by the heavy FRD pollution. And we further characterized the spatial and temporal variability of emissions across different UFZs of Lanzhou and estimated their potential health impacts. Our results provided evidence to help plan and implement emission control measures of FRD in developing urban regions.

EXPERIMENTAL SECTION

Study Area. Lanzhou, located in the center of mainland China (103.73°E, 36.03°N), is an important industrial city and transportation hub of northern China. Lanzhou has a unique river valley topography (Figure 1a), with high concentrations of PM in urban areas.^{36,37} The urban area of Lanzhou is 1088 km². The average annual temperature was 11.2 °C, and the annual precipitation was 341.3 mm in 2017.

Table 1. Cumulative Particle Volume Percentages and Revised Values of k for Each Type of Road in the Four UFZs

UFZ ^a	type of road	particle volume percentage ($\leq 2.5 \mu\text{m}$)	particle volume percentage ($\leq 15 \mu\text{m}$)	revised k (g vkt^{-1})
DET	major roads	3.561	13.44	1.46
	minor roads	4.803	17.57	1.50
	branch roads	3.651	14.16	1.42
DIT	major roads	4.206	15.18	1.52
	minor roads	4.5	16.72	1.48
	branch roads	3.199	12.33	1.43
UT	major roads	3.6	15.53	1.27
	minor roads	4.505	16.63	1.49
	branch roads	3.941	13.91	1.56
ID	major roads	5.99	19.78	1.67
	minor roads	10.12	33.82	1.65
	branch roads	8.874	31.82	1.53

^aUrban function zones: UFZs= urban function zones; DET = developed downtown; DIT = developing downtown; UT = university town; and ID = industrial district.

To characterize FRD emissions in different UFZs, we divided urban Lanzhou into four regions (Figure 1a). The developed downtown (DET) region, as the political, economic, and trade center of Lanzhou, is located in the east of the urban area and has the largest annual Gross Domestic Product (GDP) of \$14.6 billion among the four regions in 2017 (Figure 1b). The developing downtown (DIT) region, as a transport hub connected to each district, is located in the center of the urban area and has a rapidly increasing annual GDP of approximately \$6.7 billion in 2017. The real estate investment growth rate is 59.53%, indicating frequent demolition and construction activities (Figure 1b and c). The university town (UT), located in the northwest of urban area, has over 137 000 university students distributed among 17 universities and scores of primary and secondary schools. In the industrial district (ID), as a large petrochemical industrial base with several industrial factories, has a large share of industrial production in its annual GDP (44.76%). Coal and biomass burning are the dominant sources of anthropogenic aerosol emissions in the ID region.^{37,38} Our sampling points cover the four divided regions above, including 45 main roads, 60 minor roads, and 55 branch roads (i.e., road segments between intersections) (Figure 1a).

Revised AP-42 Method. There exist a few methods to quantify FRD emissions based on different hypotheses and measurements, including the AP-42 method,³⁹ Testing Re-entrained Aerosol Kinetic Emissions from Roads (TRAKER) method and upwind-downwind method.^{40–42} Our calculation was based on AP-42 but with updated parameters.

Especially, AP-42 was published in 1968 and has been gradually updated by the United States Environmental Protection Agency (USEPA). The method can be implemented with a standard sampling procedure suitable for constructing FRD emission inventories at a large scale. The FRD $\text{PM}_{2.5}$ emission factor (EF, g vkt^{-1} , where vkt represents the number of vehicle kilometers traveled) on each paved road is calculated as follows:

$$\text{EF} = k \times \left(\frac{sL}{2}\right)^{0.65} \times \left(\frac{W}{3}\right)^{1.5} \quad (1)$$

where k is a particle size multiplier for $\text{PM}_{2.5}$ smaller than $i \mu\text{m}$ (unit: g vkt^{-1}); sL is the silt loading, which is defined as the amount of PM less than $75 \mu\text{m}$ on the road surface per unit area (unit: g m^{-2}); and W is the average vehicle weight (unit:

tons). The values of sL and W were obtained from our actual sampling in Lanzhou.

In this study, we divided the study domain into the grid of 1450 square cells ($500 \times 500 \text{ m}^2$). Aggregated FRD $\text{PM}_{2.5}$ emissions (E , g d^{-1}) from paved roads are calculated as follows:

$$E = \left(1 - \frac{P}{4N}\right) \times \sum_{i,j=1}^n \text{EF}_{i,j} \times F_{i,j} \times L_{i,j} \quad (2)$$

where P is the number of days (unit: days) when the daily precipitation exceeds 0.254 mm during the study period, which was taken from National Oceanic and Atmospheric Administration (NOAA)-National Climatic Data Center Surface (NCDC) (<ftp://ftp.ncdc.noaa.gov/pub/data/gsod/>); N is the study period (unit: days); F is the traffic volume on road i in grid cell j provided by the Traffic Police Detachment of the Public Security Bureau in Lanzhou (unit: vehicle h^{-1}); and L is the length of road i in grid cell j (unit: km) obtained from the Open Street Map (<https://www.openstreetmap.org/#map=12/36.0781/103.7880>).

The particle size multiplier (k) is a function of particle size. The default values of k were conducted based on sampling in the U.S.A. Moreover, the differences are also attributed to different method of estimating FRD emission factor. Therefore, the default value of k in AP-42 could lead to large uncertainties on the quantities of the FRD $\text{PM}_{2.5}$ emissions in developing regions (such as Lanzhou). We revised the values of k based on sampling in Lanzhou as follows:

$$k = \frac{l_{2.5}}{l_{15}} \times k_{15} \quad (3)$$

where l is the mass percentage of particle mass with aerodynamic diameter less than 2.5 and $15 \mu\text{m}$, k_{15} is the recommended value from AP-42 guidance document for 5.5 g vkt^{-1} . The results are shown in Table 1.

Estimate of Premature Mortality Rate. We further simulated the FRD $\text{PM}_{2.5}$ concentrations based on the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) Model and further estimated their health impacts by a pollution-exposure model.

The traditional epidemiological relation approach has been unanimously recognized and widely used to estimate the premature mortality rate attributable to $\text{PM}_{2.5}$ exposure with respect to the international trade, residential, industrial,

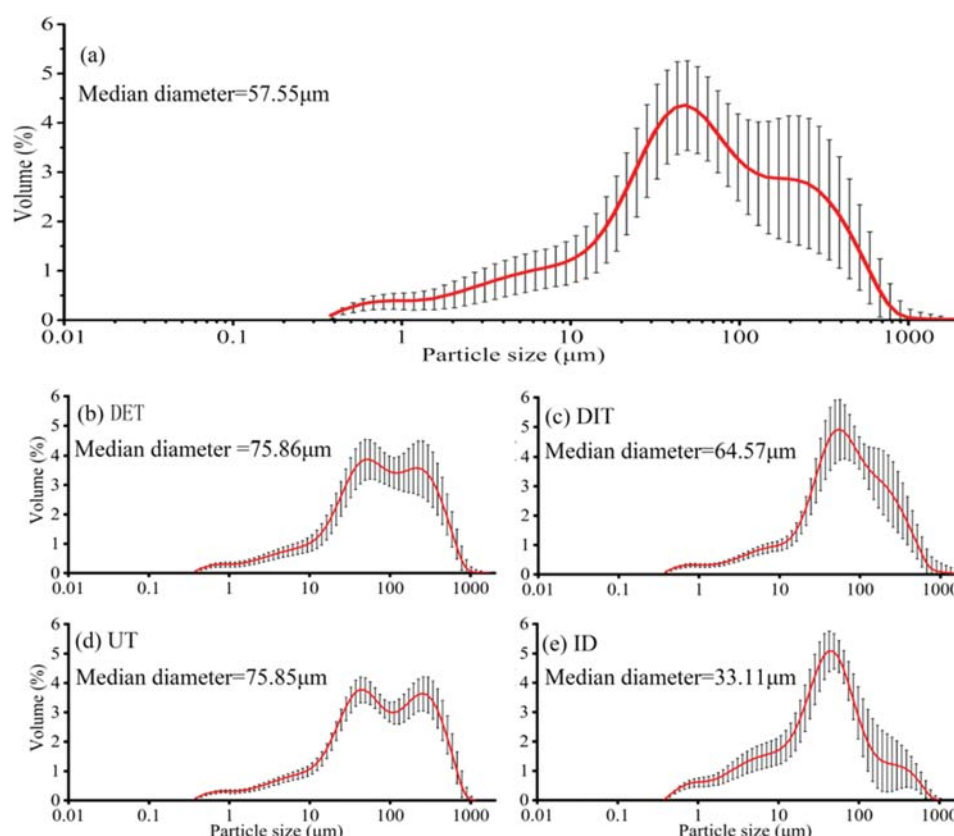


Figure 2. Average size distributions of road-deposited sediment in the study area (Figure 3a) and in the four UFZs including DET (Figure 3b), DIT (Figure 3c), UT (Figure 3d), and ID (Figure 3e). The error bar represents one standard deviation.

transportation and energy sectors.^{43–45} In this study, we calculated the premature mortality burden (M , deaths y^{-1}) attributable to FRD $PM_{2.5}$ for chronic obstructive pulmonary disease (COPD), lung cancer (LC), ischemic heart disease (IHD), cerebrovascular disease (stroke), and acute lower respiratory infections (ALRI) through eq 4:

$$\sum_{i,j=1}^N M_{i,j} = \sum_{i,j=1}^N IM_{i,j} \times \sum_{i,j=1}^N P_i \quad (4)$$

where P is the population in UFZ i ; and IM is the incidence mortality of the disease j in UFZ i attributable to exposure to FRD $PM_{2.5}$ (unit: deaths 10^{-5} population y^{-1}) calculated as follows:

$$\sum_{i,j=1}^N IM_{i,j} = \sum_{j=1}^N Y_j \times \frac{\sum_{i,j=1}^N RR_{i,j} - 1}{\sum_{i,j=1}^N RR_{i,j}} \quad (5)$$

Y is the baseline mortality for disease type j (unit: deaths 10^{-5} population y^{-1});⁴⁴ RR is the relative risk for the disease j in UFZ i , which is estimated by the integrated exposure–response (IER) function:²²

$$RR(C) = \begin{cases} 1 + \alpha[1 - \exp(-\gamma(C - C_0)^\delta)] & \text{for } C > C_0 \\ 1 & \text{for } C \leq C_0 \end{cases} \quad (6)$$

where C is the simulated FRD $PM_{2.5}$ concentrations based on the WRF-Chem model (see Table S2 of the Supporting

Information, SI); C_0 represents the theoretical minimum risk exposure level, α , γ , and δ are parameters specific to disease j .⁴⁵ The key parameters for calculations of premature mortality have shown in Table S3.

RESULTS

Size Distribution of Road-Deposited Sediment. The particle size of road-deposited sediment is critical to the assessment of FRD emissions and associated pollutant concentrations.¹⁸ For urban Lanzhou as a whole, the particle volume distribution of road-deposited sediment showed the bimodal pattern, where the first peak was more dominant than the second peak. The first mode peaked at 40–79 μm , and the second mode peaked at 159–316 μm (Figure 2a). The size distributions of road dust deposited in the four UFZs differed significantly (Figure 2b–e). The percentage contributions of particles smaller than 100 μm to the total road-deposited sediment were ordered as follows: ID (83.8%) > UT (65.0%) > DET (57.3%) > DIT (57.2%). The size distributions in the DET and UT both showed a bimodal pattern, with a large median size of 76 μm . In the ID and DIT, the road-deposited sediment samples had unimodal distributions with small median sizes ranging from 33 to 65 μm , because of a large amount of fly ash emitted from coal-fired factories and demolition and construction activities that greatly increase the proportion of fine particles.⁴⁸

Silt Loading. The silt loading in the four UFZs decreased in the following order: DIT (0.62 g m^{-2}) > ID (0.44 g m^{-2}) > UT (0.34 g m^{-2}) > DET (0.28 g m^{-2}). These values were

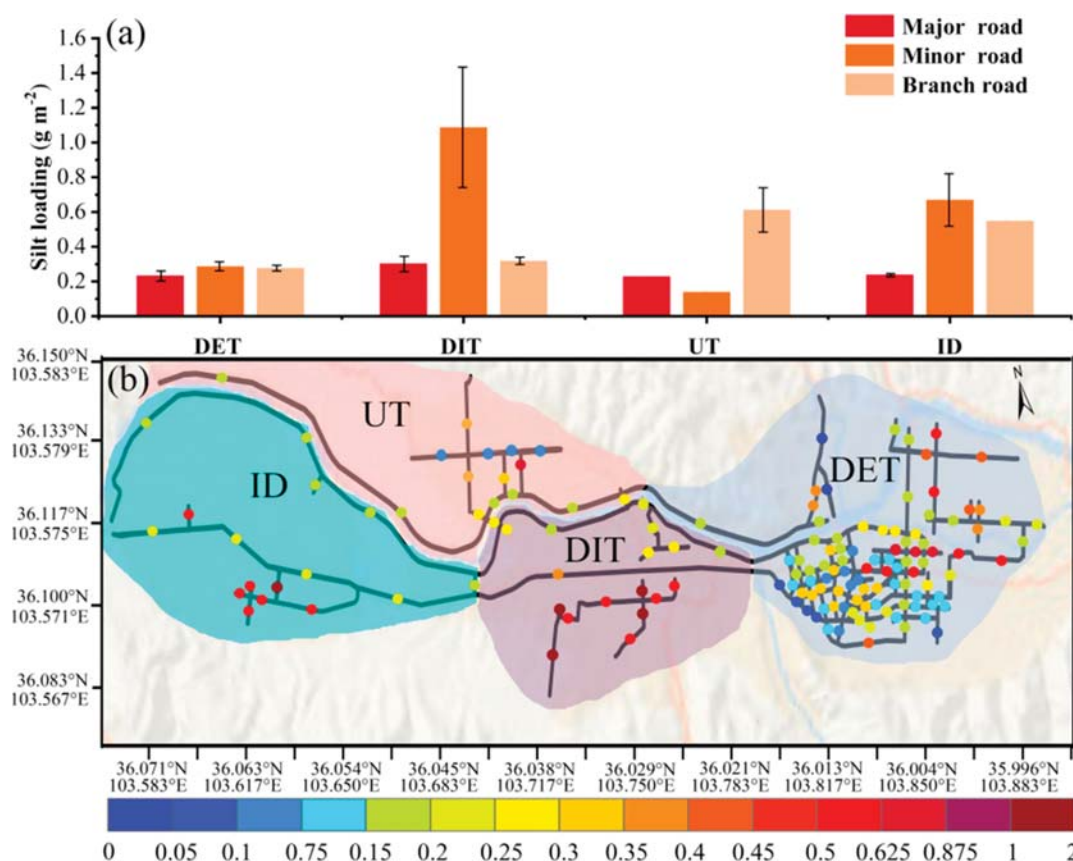


Figure 3. (a) Silt loading on three types of roads in each of the four UFZs (unit: g m^{-2} ; the error bar represents one standard deviation). (b) Spatial distribution of silt loading on sampling sites (unit: g m^{-2}).

averaged over major, minor, and branch roads. Among the types of roads and UFZs, the highest value of silt loading, $1.1 \pm 0.34 \text{ g m}^{-2}$, occurred in the minor roads of the DIT, which was almost 4 times higher than that in the DET ($0.3 \pm 0.02 \text{ g m}^{-2}$) with frequent street sweeping ($3\text{--}4 \text{ times day}^{-1}$). This highest value was in the DIT because of frequent construction, seldom sweeping, and thus high proportion of bare soil on the roads (Figure 3a). The UT had the fewest population and relatively few human activities, where the value of silt loading was small on major and minor roads and high on branch roads, with average values of 0.24, 0.15, and 0.62 g m^{-2} , respectively (Figure 3b). The ID had relatively high silt loading values of approximately 0.4 g m^{-2} on minor and branch roads due to the mud carried by vehicles. Averaged over all UFZs, the amount of the silt loading for the three types of road were ordered as follows: minor roads (0.54 g m^{-2}) > branch roads (0.33 g m^{-2}) > major roads (0.29 g m^{-2}).

Traffic Conditions. Lanzhou has about 0.9 million vehicles in 2017, and the resulting traffic intensity highly affects FRD emissions. Among the four UFZs, the DET had the highest traffic volume, with more than 34 000 vehicles day^{-1} on major roads and 10 000 vehicles day^{-1} on minor and branch roads together. The DIT, with the second highest traffic volume, supported more than 30 000 vehicles day^{-1} on major roads. The traffic volumes in the UT and ID ranging from 2500 to 32 500 vehicles day^{-1} were lower than that the DET traffic volume (Figure 4a and b).

Traffic volumes had an apparent diurnal cycle, with a minimum value in the nighttime, a rapid increase after 7:00,

and a more gradual decrease after 23:00 local time (LT = UTC + 8 h). The traffic volumes ranging from 600 to 1500 vehicles h^{-1} , are remained stable and high in the daytime, attributable to traffic congestion in urban areas caused by an imperfect transportation infrastructure. Further contrasting data in weekdays and in weekends shows that the high-traffic periods are delayed by approximately one to 2 h on weekends compared with those on weekdays.

FRD $\text{PM}_{2.5}$ Emissions. The daily FRD $\text{PM}_{2.5}$ emission inventory in Lanzhou was constructed with high-resolution ($500 \times 500 \text{ m}^2$) as shown in Figure 5a. Due to highly dense road networks and high silt loadings, a few locations in the central DET and eastern DIT have the highest FRD $\text{PM}_{2.5}$ emissions between 3×10^4 and $6 \times 10^4 \mu\text{g m}^{-2} \text{ d}^{-1}$. Relatively low FRD $\text{PM}_{2.5}$ emissions ($0.2 \times 10^4 \sim 1.2 \times 10^4 \mu\text{g m}^{-2} \text{ d}^{-1}$) occurred in the UT and ID zones (Figure 5a). The anthropogenic $\text{PM}_{2.5}$ emission fluxes were 7.2×10^4 , 5.7×10^4 , 0.5×10^4 , and $4.1 \times 10^4 \mu\text{g m}^{-2} \text{ d}^{-1}$ in the DET, DIT, UT, and ID, respectively, based on the PKU $\text{PM}_{2.5}$ inventory.³⁸ The value of $\text{PM}_{2.5}$ emissions from FRD was about two-fifths of that from combustion and industrial process sources in DET and DIT. Overall, the total FRD emission in urban Lanzhou was $1141 \pm 71 \text{ kg d}^{-1}$. The road dust accounted for 24.6% of urban $\text{PM}_{2.5}$ emissions (Figure 6a). Especially, the contributions of road dust to $\text{PM}_{2.5}$ emissions varied widely among four UFZs, ranging from 16.2% to 51.7% (Figure 6c–f). The magnitude of FRD $\text{PM}_{2.5}$ emissions in the four UFZs decreased in the following order: DET ($415 \pm 31 \text{ kg d}^{-1}$) > DIT ($367 \pm 43 \text{ kg d}^{-1}$) > ID ($162 \pm 15 \text{ kg d}^{-1}$) > UT ($114 \pm 10 \text{ kg d}^{-1}$).

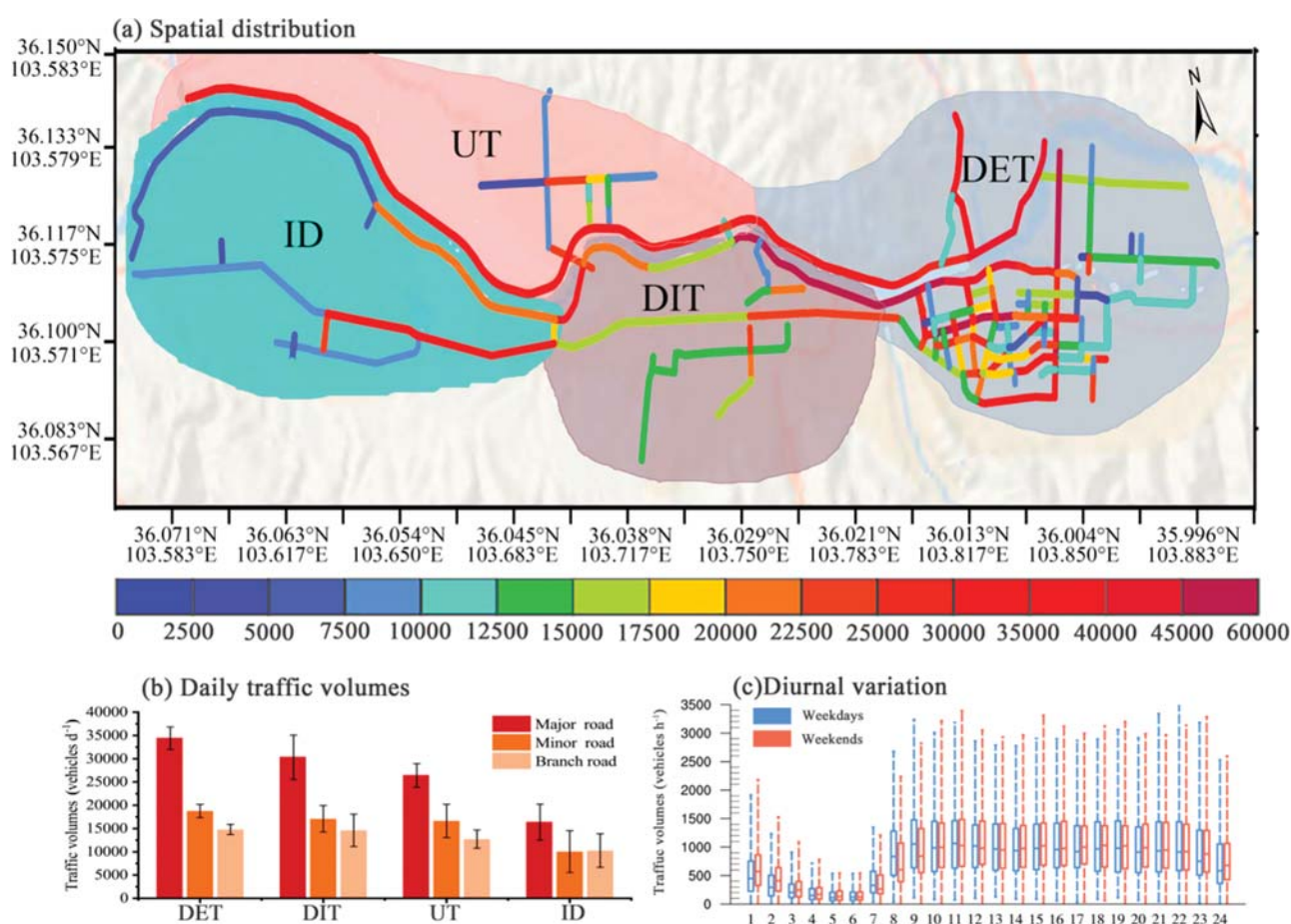


Figure 4. (a) Spatial distribution of daily traffic volume in Lanzhou (unit: vehicles d^{-1}); (b) average daily traffic volumes on three level roads in each UFZ; (c) diurnal cycle of traffic volume (unit: vehicles h^{-1}) on weekdays and weekends (dashes in the boxes denote the median traffic volumes; the lower and upper parts of the boxes represent the 25th and 75th percentiles for each data set, respectively. The dotted lines extending from the boxes indicate the maximum and minimum values).

d^{-1}) > ID ($250 \pm 62 \text{ kg d}^{-1}$) > UT ($109 \pm 13 \text{ kg d}^{-1}$) (Figure 5b).

The diurnal cycle of total FRD $\text{PM}_{2.5}$ emissions mainly follows the variations in traffic volume. Summed over urban Lanzhou, the lowest hourly emission with value of 7.11 kg h^{-1} occurred at 5:00 LT in urban Lanzhou, after which emissions rose dramatically to 58.63 kg h^{-1} at 11:00 LT. Emissions remained high from 8:00 to 23:00 LT, a period with intensive human activities (Figure 5d). Meteorological condition, especially precipitation, dominates the seasonal variation of FRD $\text{PM}_{2.5}$ emissions for urban Lanzhou (Figure 5c). Summed over urban Lanzhou, the monthly average FRD $\text{PM}_{2.5}$ emission was the highest in winter ($3.1 \times 10^4 \text{ kg month}^{-1}$), followed by spring and autumn (both $2.8 \times 10^4 \text{ kg month}^{-1}$) and summer ($2.7 \times 10^4 \text{ kg month}^{-1}$). The monthly variations of $\text{PM}_{2.5}$ concentration at Lanzhou from observation and WRF-Chem model were shown in Figure 6b. Overall, the WRF-Chem model without FRD reproduced the temporal variation of $\text{PM}_{2.5}$ well but always underestimated the observed $\text{PM}_{2.5}$ concentrations of approximately $20 \mu\text{g m}^{-3}$. The temporal variation of simulated $\text{PM}_{2.5}$ concentrations including FRD emissions are more consistent with that of observations especially in winter. However, the simulations always underestimated the observed $\text{PM}_{2.5}$ concentrations in spring due to

not covering all of the natural dust source regions in the simulated domain.

Estimate of Premature Mortality Rate Induced by FRD $\text{PM}_{2.5}$ Exposure. The premature mortality rate due to exposure to FRD $\text{PM}_{2.5}$ is estimated at 30.2 premature deaths 10^{-5} population y^{-1} in urban Lanzhou in 2017 (Confidence interval (CI) 95%: 27.4; 33.3), that is, 234.5 premature deaths for the total population of 2.5 million in urban Lanzhou. Of these deaths, 13.9% is related to COPD mortality, 9.7% to LC, 21.5% to ALRI, 31.8% to IHD, and 23.1% to stroke. The variation in the premature mortality rates for each disease is attributed to the difference in baseline mortality. In Lanzhou, IHD and stroke account for the most deaths, with values reaching 9.6 (CI 95%: 9.0; 10.1) and 6.9 (CI 95%: 6.4; 7.9) deaths 10^{-5} population y^{-1} , respectively (Figure 7a). The estimated mortality rate from FRD $\text{PM}_{2.5}$ varied substantially among the four UFZs, with relatively small values of 3.8 premature deaths 10^{-5} population y^{-1} in the ID (Figure 7b). Owing to the interaction of a larger population, higher baseline mortality, higher emission, the premature mortality burden was obviously large in the DET and the DIT with high value of 157.1 and 44.9 premature deaths, respectively (Figure 7c).

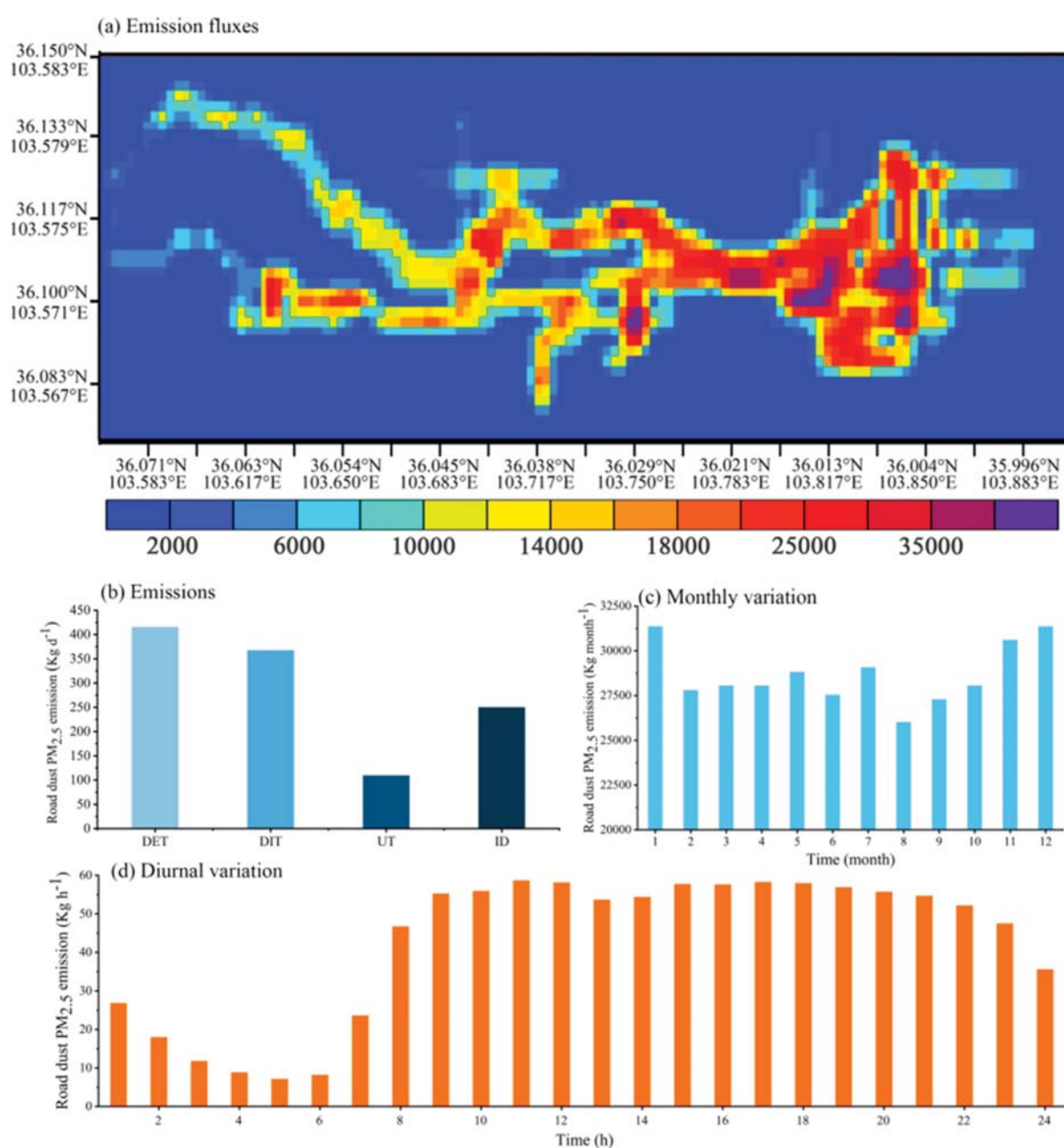


Figure 5. (a) Road dust PM_{2.5} emission fluxes (unit: $\mu\text{g m}^{-2} \text{d}^{-1}$); (b) the total amounts of road dust PM_{2.5} emission in the four UFZs (unit: kg d⁻¹); (c) monthly variations (unit: kg month⁻¹); and (d) diurnal variations (unit: kg h⁻¹) of road dust PM_{2.5} emissions.

DISCUSSION

In the process of urbanization, a large (sometimes the largest) fraction of urban PM_{2.5} comes from FRD, which cause ambient air quality degradation and acute damage to public health.^{15–17,23} UFZs, which are closely related to daily urban activities, are associated with distinctive characteristics of road-deposited sediment and FRD PM_{2.5} emissions.^{18,35} We constructed a high-resolution gridded FRD PM_{2.5} emission inventory based on a revised AP-42 method³⁹ to investigate the characteristics of FRD PM_{2.5} emissions in the four UFZs and their mortality impacts through a traditional epidemiological relation approach^{44–48} in the study.

The road-deposited sediment in Lanzhou predominantly comprises smaller particles (<100 μm , 57–84%), which can be easily released into atmosphere.¹⁸ In particular, the road dust particles deposited in the ID region are much smaller than those deposited in other regions, with a median size of 33.11 μm , because of a large amount of deposited fine particles originating from coal-fired factories and demolition and construction activities.^{37,38} Thus, these particles could be suspended in the ambient atmosphere over a longer period, contain more heavy metal contents, and be more harmful to human health in the ID region than in the other regions.^{18,49} Silt loading values are highly related to anthropogenic activities, e.g., frequent street sweeping reduces the silt

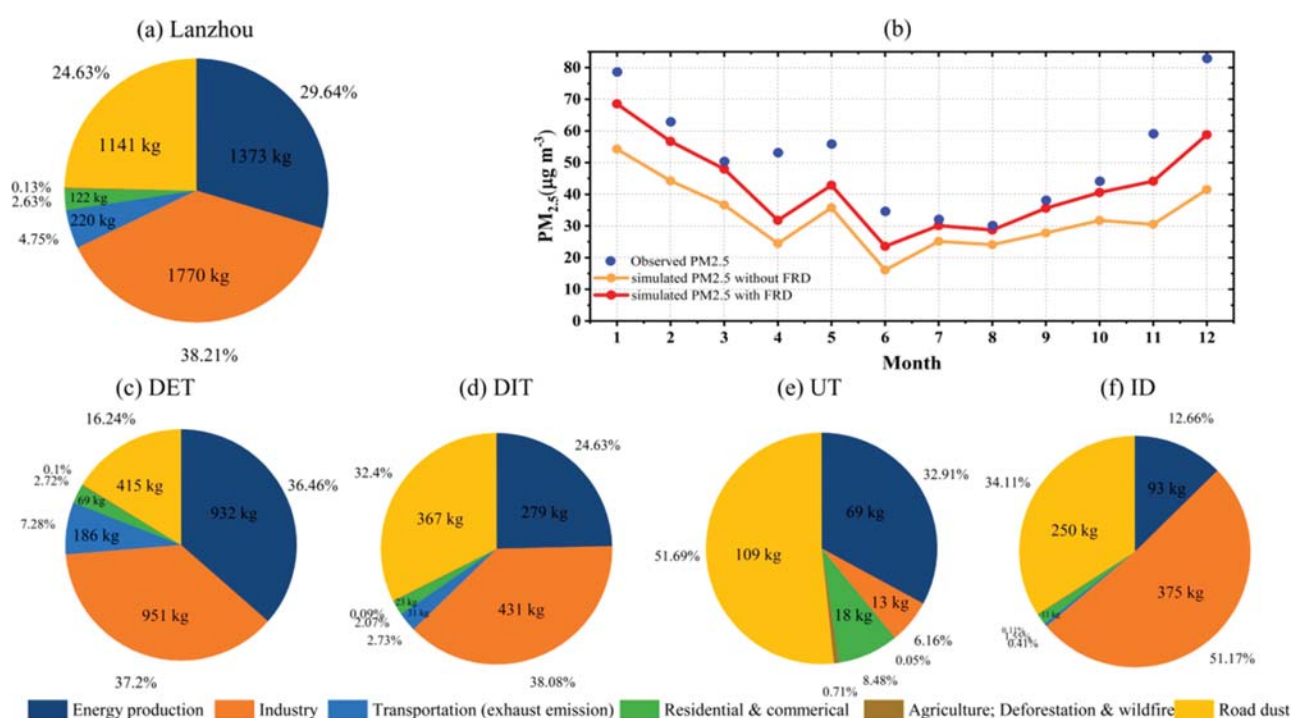


Figure 6. Source profiles for total emissions of $\text{PM}_{2.5}$ in (a) Lanzhou, (c) DET, (d) DIT, (e) UT, and (f) ID. And (b) monthly variations of $\text{PM}_{2.5}$ concentration (unit: $\mu\text{g m}^{-3}$) from observation (blue dots), simulation without FRD (yellow line), and with FRD (red line) during 2017.

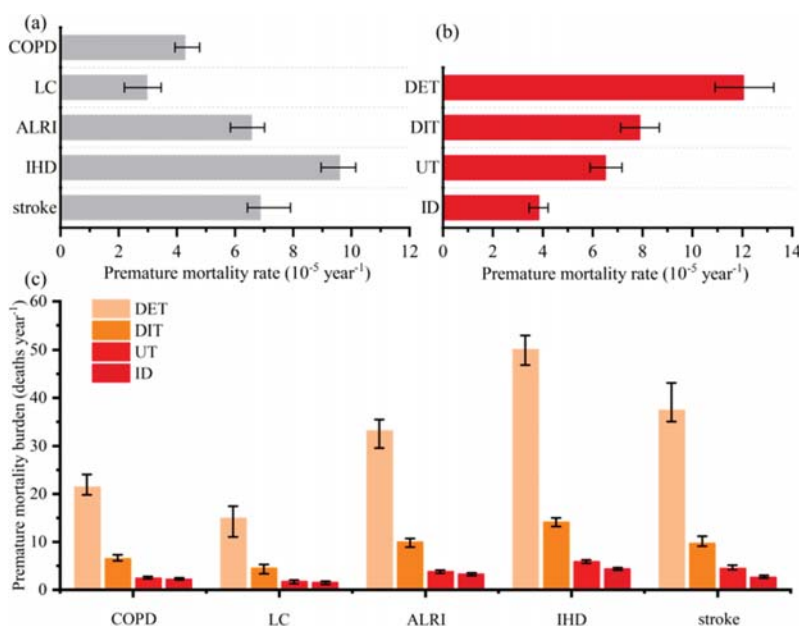


Figure 7. (a) Premature mortality rates in 2017 associated with FRD $\text{PM}_{2.5}$ exposure for chronic obstructive pulmonary disease (COPD), lung cancer (LC), acute lower respiratory infections (ALRI), ischemic heart disease (IHD), and cerebrovascular disease (stroke). (b) Premature mortality rates in the four UFZs. And (c) premature mortality burden in 2017 associated with FRD $\text{PM}_{2.5}$ exposure. The error bar denotes the 95% confidence intervals.

loadings, but transportation and construction activities lead to 2–10 times increases in silt loading.

The characteristics of FRD $\text{PM}_{2.5}$ emissions varied substantially among different UFZs. The FRD $\text{PM}_{2.5}$ emission is one of main contributor to urban $\text{PM}_{2.5}$ emissions, accounting for 16.2% to 51.7% of the $\text{PM}_{2.5}$ emission among four UFZs. The total FRD $\text{PM}_{2.5}$ emission in the study area

was approximately 1141 kg d^{-1} . The FRD $\text{PM}_{2.5}$ emission in the DET ranked the highest (415.35 kg d^{-1}) due to the largest traffic volume, despite the frequent street sweeping. The DIT, ranked second (367.27 kg d^{-1}), has the largest emission factor mainly due to the intensive construction activities. The spatial distribution of FRD $\text{PM}_{2.5}$ emissions demonstrates a significant relationship between FRD $\text{PM}_{2.5}$ emissions and human

activities: high emissions with values over $3 \times 10^4 \mu\text{g m}^{-2} \text{d}^{-1}$ are mainly concurrent with relatively infrequent street sweeping, frequent construction activities, and high density of road networks. Compared with observation, the WRF-Chem model with FRD reproduced the monthly variation of $\text{PM}_{2.5}$ better than that without FRD. It is noted that the revised AP-42 method used here only accounts for a limited number of factors including particle size of FRD, silt loading, vehicle weight, and precipitation. However, the previous studies pointed out that road characteristics (e.g., vehicle speed, types of vehicles, location, and topography),⁵⁰ other human activities (street cleaning activities and policies), and meteorological conditions (e.g., temperature, wind direction and wind speed, and relative humidity) also play important roles in FRD emissions.⁵¹ The AP-42 method should be improved by including more factors in the future.

The estimated premature mortality rate induced by FRD $\text{PM}_{2.5}$ exposure based on the traditional epidemiological relation approach was 234.5 (CI 95%: 212.2; 258.3) premature deaths in urban Lanzhou in 2017. Mortality due to FRD $\text{PM}_{2.5}$ varied substantially among four UFZs. Compared to mortality from the other UFZs, premature deaths rates were the largest in the DET with value of 12.0 (CI 95%: 10.9; 13.2) premature deaths 10^{-5} population y^{-1} owing to higher baseline mortality and larger dust emission. Note that the estimation of premature mortality induced by FRD exposure in the study may be underestimated due to ignoring the influence of the size distribution of particles, heavy metals, and polycyclic aromatic hydrocarbon.^{18,49,52}

As emission reduction measures are being implemented rapidly for fossil fuel burning, vehicle exhaust, and the power sector, FRD $\text{PM}_{2.5}$ emissions may represent a larger proportion of air pollution.⁷ FRD PM emissions abatement could significantly improve urban air quality and protect public health.⁴⁵ Effective FRD $\text{PM}_{2.5}$ mitigation measures may differ among the UFZs. Increasing the frequencies of street sweeping could reduce FRD $\text{PM}_{2.5}$ emissions in the DIT, in combination with dust control measures for construction activities.⁵¹ Moreover, use of dust suppressants with high hygroscopic and deliquescent properties is an effective mitigation measure in the DIT and ID, which areas have high silt loading and smaller particles.¹⁹ Moreover, the utilization of emission control technologies of factories should be promoted in the long run, such as implementing retirement of small and low-efficient power plants and applying end-of-pipe controls,^{53,54} to fundamentally reduce fine particles in FRD in the ID.

■ ASSOCIATED CONTENT

§ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b00666.

Details of road dust sampling and measuring characteristics of road-deposited sediment (Text S1; Figure S1); simulation of FRD $\text{PM}_{2.5}$ concentrations based on the WRF-Chem model (Text S2; Table S1, S2; Figure S4); source of ground monitoring $\text{PM}_{2.5}$ data (Text S3); the diurnal cycle of traffic volume in three types of roads (Text S3, Figure S2); and the characteristics of FRD PM_{10} emission (Text S4, Figure S3) (PDF)

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Supplementary Information for

Fugitive road dust PM_{2.5} emissions and their potential 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) from July to November 2017 during a dry weather period and wind velocities less 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). At each sampling location, a rectangular sampling grid was selected, with a width of 0.5 m and a length being the width of the road (Figure S1a and c). The handheld cordless vacuum cleaner can collect road-deposited sediment conveniently (Figure S1b). Moreover, the vacuum cleaner has high efficiency to catch both fine and coarse particulates with air filtration, dust bucket, the dust separator and the cyclone. The FRD samples were preserved in numbered vacuum cleaner bags and dried at 35 °C for 7 days (Figure S1d).

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

$$SL = \frac{m_{total} - m_{75\mu m}}{S} \quad (1)$$

Where m_{total} is the mass of total FRD samples; m_{75μm} is the mass of FRD samples larger than 75 μm; 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.

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

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.

2. Model configuration

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

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 China, as shown in Fig. S3. The model atmosphere was divided into 35 vertical layers, 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 2017 were used in this study. The initial and boundary meteorological conditions were constructed from the National Center for Environmental Prediction Final Analysis (NCEP/FNL) data at a 6 h temporal interval and 1 degree horizontal resolution. To produce a more realistic simulation, the modeled u- and v-wind components and atmospheric temperatures were nudged towards the NCEP/FNL data with a nudging timescale of 6 h.

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}.

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.

The diurnal cycle of traffic volumes on the major road, minor road, and branch 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 road, respectively, and maintained the high value during 9:00 to 23:00 local time (LT) with dramatic increase after 7:00 local time and down at 23:00 LT. Especially, the magnitude of traffic volumes is the highest on the major road, followed by the minor road and branch road. The traffic volumes have a slight variation during the day time 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 on three types of roads are all delayed about one or two hours on weekends compared with those on weekdays. The traffic volumes change more significantly on major roads than those on minor road, and the slight variations of traffic volumes occur on branch roads.

FRD PM₁₀ emission.

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 approximately 3216 kg d⁻¹. The FRD PM_{2.5} emission fluxes are enhanced over the regions with large traffic volumes and high density of road network, predominantly in the central of the DET and the eastern of the DIT, where the value is larger than 3×10⁴ µg m⁻² d⁻¹. The FRD PM₁₀ emission fluxes with comparatively lower values varying from 11.8×10⁴ µg m⁻² d⁻¹ to 7.5×10⁴ µg m⁻² d⁻¹ are occurred in the UT and ID (Figure S3a). The spatial distributions of the FRD PM₁₀ emission fluxes are found to be quite similar to that of the PM_{2.5} emission fluxes. The FRD emissions with the PM_{2.5}/PM₁₀ ratio of 0.35 can sufficiently increase the amount of fine particulate matters in urban areas, which could be suspended in the ambient atmosphere over a longer time and be more harmful for human health compared to its coarse fraction³. The FRD PM₁₀ emission factors, as an indicator of the FRD emission ability, are sensitive factors in the construction of emission inventory. The FRD PM₁₀ emission factors are approximately 3 times larger than the PM_{2.5} emission factors. The interaction of large

silt loading and small particle size causes high values of FRD PM₁₀ emission factors in the DIT and ID, with average value of 1.13 and 0.96 g VKT⁻¹, respectively (Figure S3b). And the magnitude of FRD PM₁₀ emission in the different UFZs decrease in the order DET (1188 kg d⁻¹) > DIT (1023 kg d⁻¹) > ID (693 kg d⁻¹) > UT (312 kg d⁻¹) (Figure S3c). The diurnal cycle of FRD PM₁₀ emission is mainly consistent with variation of traffic volumes, that is, the lowest FRD PM₁₀ emission occurs at 5:00 LT with value as low as 19.98 kg h⁻¹ while rises dramatically to 165.42 kg h⁻¹ at 11:00 LT. 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 conditions also influence FRD emission as the monthly FRD PM₁₀ emission is the largest (8.5×10⁴ kg month⁻¹) in winter, followed by spring (8.0×10⁴ kg month⁻¹) and smallest (7.8×10⁴ kg month⁻¹) in summer which aligns with the precipitation cycles (Figure S3e).

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Table S1. The WRF-Chem configuration in this study

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

226

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

UFZs ^a	Spring (^b FRD/Total ^c)	Summer (FRD/Total)	Autumn (FRD/Total)	Winter (FRD/Total)	Annual (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

^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

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

parameters	COPD ^a	LC ^a	ALRI ^a	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_0	5.8	5.8	5.9	5.8	5.8
Baseline mortality ^b	43.8 (CI: 40.4; 49.1)	23.4 (CI: 17.3; 27.3)	28.6 (CI: 25.5; 30.6)	105.7 (CI: 98.8; 111.9)	42.3 (CI: 39.6; 48.7)

^aDisease: COPD= chronic obstructive pulmonary disease; LC= lung cancer; ALRI= acute lower respiratory infections; IHD= ischemic heart disease; and stroke= cerebrovascular disease; ^bBaseline mortality: CI denotes the 95% confidence intervals

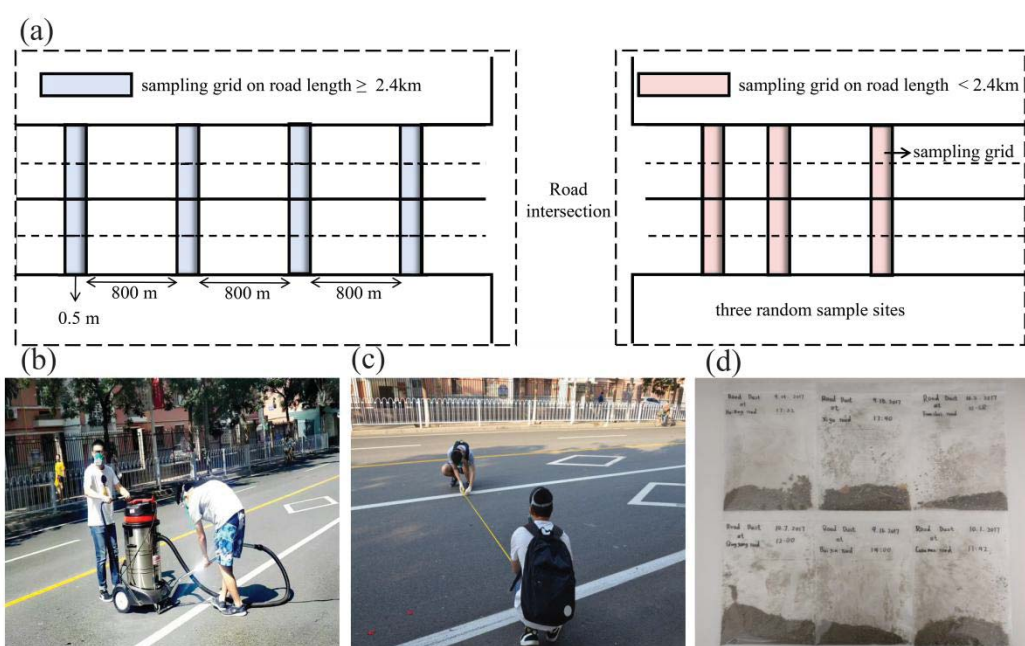


Figure S1. The process of gathering samples. (a) Sampling locations; (b) road-deposited sediments were sampled by a vacuum cleaner; (c) measuring the areas of the sampling grid; (d) FRD samples collected in vacuum cleaner bags.

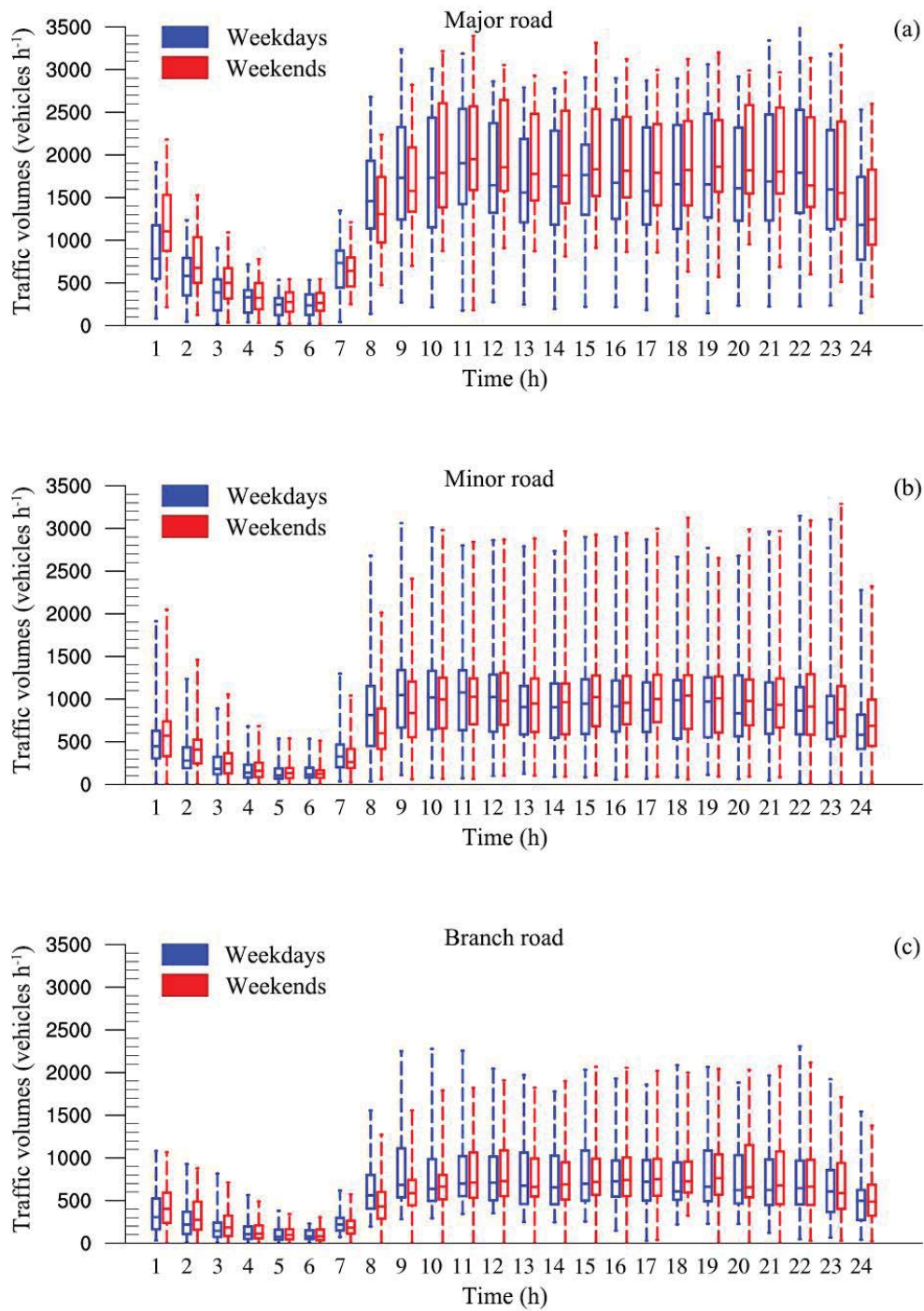


Figure S2 The diurnal cycle of traffic volume on (a) major road; (b) minor road; (c) branch road. (Dashes in the boxes denote medians of traffic volume. Opening and closing of the boxes presents 25 and 75th percentiles for each dataset. The dotted line tops of the boxes are maximum and minimum, respectively).

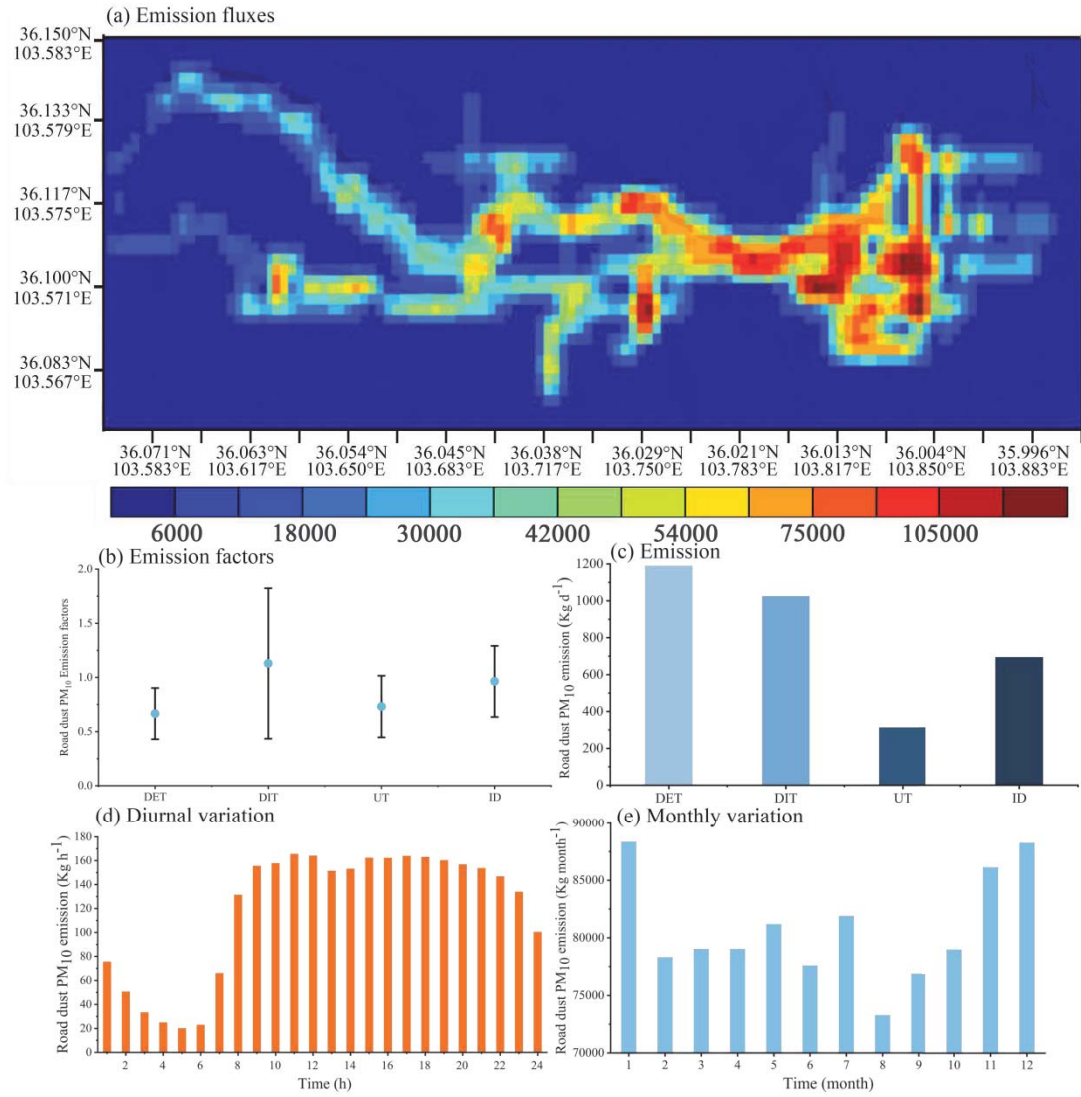
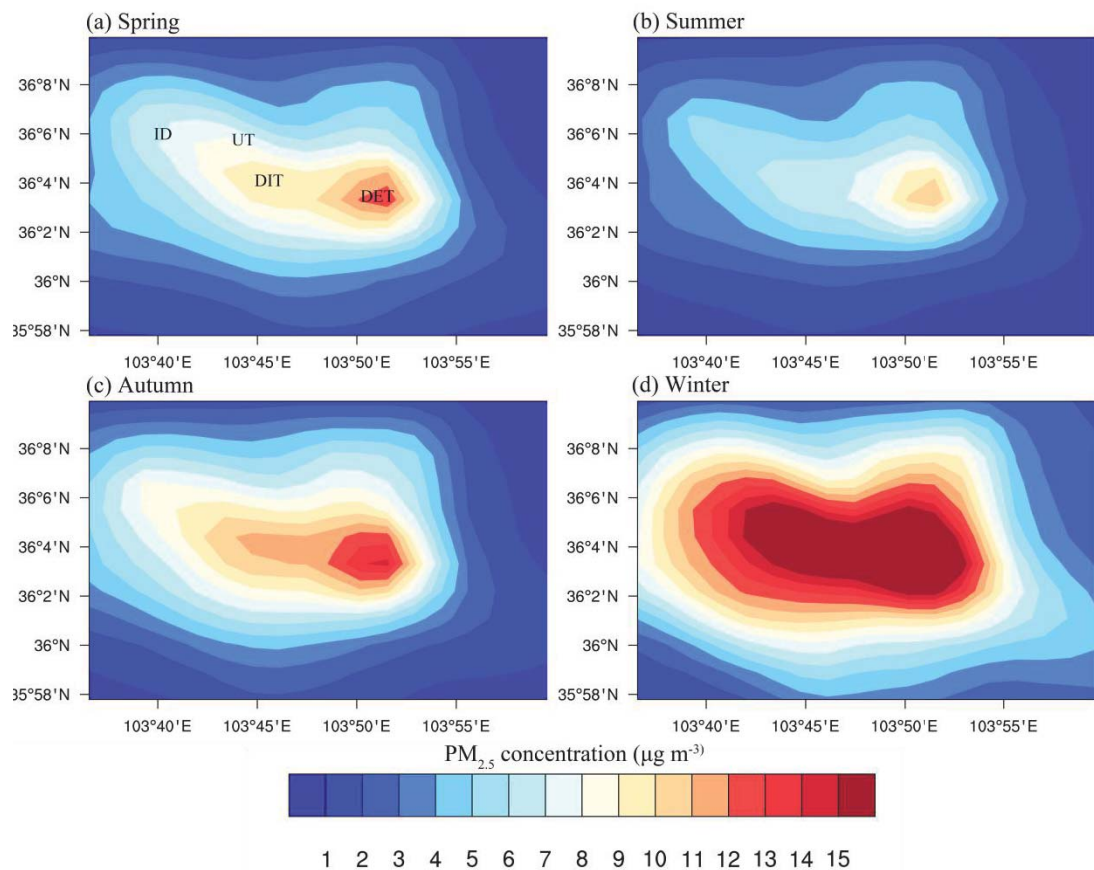


Figure S3. (a) The pattern of FRD PM₁₀ emission fluxes (unit: $\mu\text{g m}^{-2} \text{d}^{-1}$); (b) emission factors (unit: g VKT^{-1}), (c) Total amount (unit: kg d^{-1}), (d) Diurnal variations (unit: kg h^{-1}), and (e) Monthly variations (unit: kg month^{-1}) of FRD PM₁₀ emission in four UFZs.



246
 247 Figure S4 The spatial distributions of simulated FRD PM_{2.5} concentrations in (a)
 248 spring, (b) summer, (c) autumn and (d) winter based on the WRF-Chem model.