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Estimating emissions and concentrations of road dust aerosol over China using the GEOS-Chem model

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ABSTRACT

Paved road dust is one of the most important aerosols in China. The authors estimated road dust emissions using an empirical model (AP-42 model) developed by the U.S. Environmental Protection Agency, and simulated road dust concentrations over China for the years 2006–2011 using the GEOS-Chem model. The annual road dust emissions amount averaged over 2006–2011 is estimated to be 2331.4 kt, with much higher emissions in eastern China than in western China. Because of heavy traffic and a dense road network, emissions are high over Beijing–Tianjin–Tanggu (BTT), Henan Province, and Shandong Province. Meanwhile, emissions are calculated to be 459.1, 112.0, and 102.7 kt, respectively, over BTT, the Pearl River Delta (PRD) region, and the Yangtze River Delta (YRD). Due to the monthly variation of precipitation, road dust emissions over China are simulated to be highest in December and lowest in June. The highest annual mean road dust concentration is simulated to be 14.5 µg m⁻³ in Beijing. Over 2006–2011, because of the increases in road length and number of vehicles, annual road dust emissions for China as a whole, BTT, the PRD, and the YRD, are simulated to increase by 260%, 239%, 266%, and 59%, respectively, leading to 233%, 243%, 273%, and 100% increases in road dust concentrations in these regions, respectively. Our results have important implications for air pollution control in China.

商要

本文利用嵌套版本的GEOS-Chem和美国环保署推荐的AP-42经验公式,估算了2006至2011年中国地区的道路扬尘排放并模拟了中国地区的道路扬尘浓度。多年平均的道路扬尘年排放量为2331.4 kt,且在中国东部地区的排放量要大于西部。由于降水月变化的影响,中国地区道路扬尘月排放量在12月最大而在6月最小。由于道路长度和车辆保有量逐年增加,2006-2011年期间,中国、京津唐、珠江三角洲和长江三角洲地区道路扬尘年排放量分别增加了260%、239%、266%和59%,这使得道路扬尘浓度在这些区域分别增加了233%、243%、273%和100%。

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关键词 道路扬尘;空间分布; 时间变化;中国; GEOS-Chem

1. Introduction

Mineral dust aerosol is an important air pollutant with a radiative effect on climate (Miller and Tegen 1998; Bangalath and Stenchikov 2015). It is generated by wind erosion and comes from both natural and anthropogenic sources, contributing to more than half of the global aerosol burden (Ginoux et al. 2012). The anthropogenic sources of mineral dust include road dust and mineral dust from changes in land use (Tegen and Fung 1995; Ginoux et al. 2012).

Road dust emissions occur whenever vehicles travel over a road or parking lot. The resuspended particulate emissions from paved roads originate from the loose material present on the surface (road, tyres, brakes) (USEPA 2011). Several studies suggest that road dust is one of the main sources of urban aerosols (Han et al. 2007; Tian et al. 2014). Based on measurements of fine particulate matter in Beijing and the positive matrix factorization (PMF) approach, Song et al. (2007) reported that road dust contributed 7% to the PM $_{2.5}$ mass concentration in January 2004 and 8% in August 2004, and Yu (2013) found that road dust contributed 12.7% to the annual mean PM $_{2.5}$ mass concentration in 2010. Also using PMF, Amato et al. (2014) showed that road dust contributed to the concentration of PM $_{10}$ by 9%–22%, 17%–22%, 29%–34%, and 21%–35% at rural, urban-industrial, urban, and traffic sites at 11 locations in southern Spain from 2003 to 2010.

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Previous studies have also estimated road dust emissions using models, such as the TRAKER (Testing Re-entrained Aerosol Kinetic Emissions from Roads) system (Kavouras et al. 2016), the NORTRIP (Non-exhaust Road Traffic Induced Particle) emissions model (Norman et al. 2016), and the Air Pollutant Emission Factors (AP-42) empirical model developed by the U.S. Environmental Protection Agency. Using this latter model, Fan et al. (2009) showed a clear decline in road emissions in Beijing during the Olympic Games, and the PM₁₀ emissions of road dust were reduced by 70%, 40%, 55%, 65%, and 72% for freeways, major arterial roads, minor arterial roads, collector roads, and local roads, respectively. Based on the AP-42 model, the annual total suspended particle amount for road dust emissions in the central urban area of Tianjin was estimated to be 28.0 kt in 2010 (Xu and Zhou 2012), and the annual PM₁₀ road dust emissions amount in the city of Chengdu was approximately 48.7 kt in 2012 (Yang et al. 2015). Peng et al. (2013) reported that the total suspended particle amount for road dust emissions from different types of paved roads in the Pearl River Delta (PRD) region (21°-23.5°N, 112°-116°E) was 2755.0 kt in 2010. These previous studies, however, were focused on a single city or a small area, and did not show emissions and concentrations of road dust over the whole of China.

In the present study, we calculate road dust emissions using the AP-42 model and simulate the concentrations of road dust in China using the GEOS-Chem model driven by assimilated meteorological fields. The goals of the study are (1) to estimate the spatial distributions and monthly variations of emissions and concentrations of road dust over China, and (2) to understand the trends of changes in emissions and concentrations of road dust from 2006 to 2011.

The model and the method of calculation of road dust emissions are described in Section 2. The spatial distributions and temporal variations of road dust emissions and concentrations over China are presented in Section 3. Model uncertainties are discussed in Section 4.

2. Model, data, and method

2.1. Model description

The GEOS-Chem model is a global three-dimensional chemical transport model of tropospheric chemistry driven by assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling Assimilation Office (Bey et al. 2001). We use version 9-01-02 (http://acmg.seas.harvard.edu/geos/) driven by GEOS-5 assimilated meteorological fields. The nested domain for East Asia (11°S-55°N, 70°-150°E) has a horizontal resolution of 0.5° latitude by 0.667° longitude and 47 vertical layers up to 0.01 hPa. Tracer concentrations

at the lateral boundaries are provided by a global GEOS-Chem simulation at a horizontal resolution of 4° latitude by 5° longitude.

The GEOS-Chem model has fully coupled ozone-nitrogen oxides-volatile organic compounds chemistry, along with aerosols including sulfate, nitrate, ammonium (Pye et al. 2009), black carbon and organic carbon (Park et al. 2003), mineral dust (Fairlie, Jacob, and Park 2007) and sea salt (Alexander et al. 2005). The standard dust scheme in the GEOS-Chem is the DEAD (dust entrainment and deposition) mobilization scheme of Zender, Bian, and Newman (2003), which confines dust emissions to topographic depressions in arid and semi-arid areas (Ginoux et al. 2004), as described by Fairlie, Jacob, and Park (2007). Dust aerosol is distributed into four size bins (radii of 0.1-1.0, 1.0-1.8, 1.8–3.0, and 3.0–6.0 μ m) following Ginoux et al. (2004). Fine (PM_{2.5}) dust concentrations are estimated by summing the first size bin and 38% of the second size bin (Fairlie et al. 2010; Zhang et al. 2013). The calculation of road dust emissions is described below, and the size bins, transport, and deposition schemes of road dust follow those of mineral dust.

2.2. Emissions of road dust

We use the AP-42 model developed by the U.S. Environmental Protection Agency (USEPA 2011) to calculate road dust emissions from paved roads in China. The emission quantity in grid cell j, E, is calculated by (http:// www.epa.gov/ttnchie1/ap42/)

$$E_j = \sum_{i=1}^n A_{ij} \times EF_{ij},$$

where EF;; (grams of road dust per vehicle kilometer traveled, g VKT^{-1}) is the emissions factor for road type *i* in grid *j*; and n is 6, which represents six road types ('freeway', 'firstclass highway', 'second-class highway', 'third-class highway', 'fourth-class highway', and 'other highway'), according to the national highway levels of China (CCOT 2007–2012). A., (VKT) represents the traffic activity rate (vehicle mileage traveled) of road type *i* in grid *j*. The values of EF_{ij} and A_{ij} are calculated using data from the World Geodetic System 1984 (WGS84) Datum, the Hemispheric Transport of Air Pollution (HTAP_V2) inventory, the Year Book of China Transportation & Communication, and previous studies (Huang et al. 2006; Fan et al. 2009; Peng et al. 2013; Yang et al. 2015). A_{ii} is dependent on road length and vehicle number, which vary annually and are obtained from the Year Book of China Transportation & Communication. Over 2006–2011, the road lengths of 'freeway', 'first-class highway', 'second-class highway', 'third-class highway', and 'fourth-class highway' increased, whereas the road lengths of 'other highway' decreased. A more detailed description



of the calculation of road dust emissions is provided in the supplementary material.

2.3. Numerical experiments

We perform the following simulations of emissions and concentrations of road dust in China using the GEOS-Chem model driven by GEOS-5 meteorological fields:

- (1) CTRL: GEOS-Chem simulation of aerosols from 2006 to 2011 without road dust.
- (2) CTRL_RD: GEOS-Chem simulation of aerosols from 2006 to 2011 with road dust emissions, transport and deposition.

The emissions and concentrations of road dust are calculated as the differences between the CTRL_RD and CTRL simulations. The period 2006–2011 is chosen because of the availability of information on road types.

3. Results

3.1. Simulated horizontal distribution and monthly variation of emissions averaged over 2006–2011

The simulated annual emissions of paved road dust over China averaged over 2006–2011 are shown in Figure 1(a). The annual road dust emissions quantity is calculated to be 2331.4 kt over China. Road dust emissions are higher in eastern China than in western China, in agreement with there being more paved roads and busier traffic in eastern China (CCOT 2007–2012). Because of the heavy traffic and dense road network, emissions are especially high over Beijing-Tianjin-Tanggu (BTT; (35°-40°N, 114°-120°E)) and Henan and Shandong provinces. Over the heavily polluted regions of BTT, the PRD region (21°-23.5°N, 112°-116°E), and the YRD (29.5°-32.5°N, 118°-122°E), where emissions of other anthropogenic chemical species are already high, emissions of road dust are calculated to be 459.1, 112.0, and 102.7 kt, respectively. Higher road dust emissions in BTT are mainly due to its larger domain than that of the YRD or PRD, as shown in Figure 1(c). In addition, the emissions per unit area are also higher in BTT, as a result of less precipitation (Figure 1(g)).

Monthly variations of road dust emissions over China, BTT, the PRD, and the YRD are shown in Figure 1(e). The monthly variations in emissions are sensitive to monthly precipitation and traffic flow (Figure 1(g) and (h)), as described in Section 2.2 and the supplementary material. Accounting for the whole China domain, road dust emissions are lowest in June and highest in December, which can be explained by the largest number of precipitation days (12.7) being in June and the minimum number (4.7) in December, as averaged over 2006–2011. The second

lowest emissions quantity is in February, resulting from the lowest traffic flow being in that month (Figure 1(h)).

The monthly variations of road dust emissions in BTT, the PRD, and the YRD also show the highest emissions being in December, because of the low precipitation. For BTT and the PRD, the lowest road dust emissions are simulated for July and June, respectively, mainly due to the relatively high number of precipitation days. Over the YRD, the lowest emissions quantity is found in February, which is a result of the low monthly traffic flow.

The annual amounts of PM₁₀ road dust emissions in 2010 were reported to be 90, 5.4, and 67.3 kt in the cities of Guangzhou (22.4°–23.9°N, 113.0°–114.0°E) (Peng et al. 2013), Tianjin (39.12°–39.17°N, 117.15°–117.22°E) (Xu and Zhou 2012), and Nanjing (31.2°–32.8°N, 118.0°–119.7°E) (Wang et al. 2014), respectively. Our simulated emissions for 2010 are 82.7, 8.7, and 76.7 kt in Guangzhou, Tianjin, and Nanjing, respectively, which compare fairly well with the values in those studies.

3.2. Simulated horizontal distribution and monthly variation of concentrations averaged over 2006–2011

Figure 1(b) shows the simulated road dust concentrations over China averaged over 2006–2011. The horizontal distributions of concentration mimic those of emissions. The road dust concentration is simulated to be the highest in Beijing (39.9°N, 116.4°E), with a value of 14.5 μ g m⁻³. The fine road dust concentration (summing the first size bin and 38% of the second size bin) in Beijing is 5.8 μ g m⁻³, which contributes approximately 7% to the annual mean PM_{2.5} concentration of all aerosol species (81 μ g m⁻³), according to the value for the year 2015 reported by the Ministry of Environmental Protection of the People's Republic of China. This proportion is close to the values of 7%–8% reported in Song et al. (2007), 3.6%–6.2% in Cheng et al. (2013), and 9.7% in Wang et al. (2016).

Figure 1(f) shows the simulated monthly variations of road dust concentrations over China, BTT, the PRD, and the YRD. For all the regions considered, road dust concentrations are the highest in winter (December–January–February); the average wintertime value is 8.5 μ g m⁻³ in BTT, followed by that in the PRD (5.5 μ g m⁻³) and YRD (4.5 μ g m⁻³). The lowest monthly mean concentrations of 5.0 μ g m⁻³ in BTT and 3.6 μ g m⁻³ in the PRD occur in July and June, respectively, as a result of the largest precipitation amount occurring in these months (Figure 1(g)). Over the YRD, the lowest monthly mean concentration of 2.6 μ g m⁻³ is in September, due to the combined effect of low emissions and large quantities of wet deposition.

Few studies have reported measured road dust concentrations. All the concentrations we collected are listed in

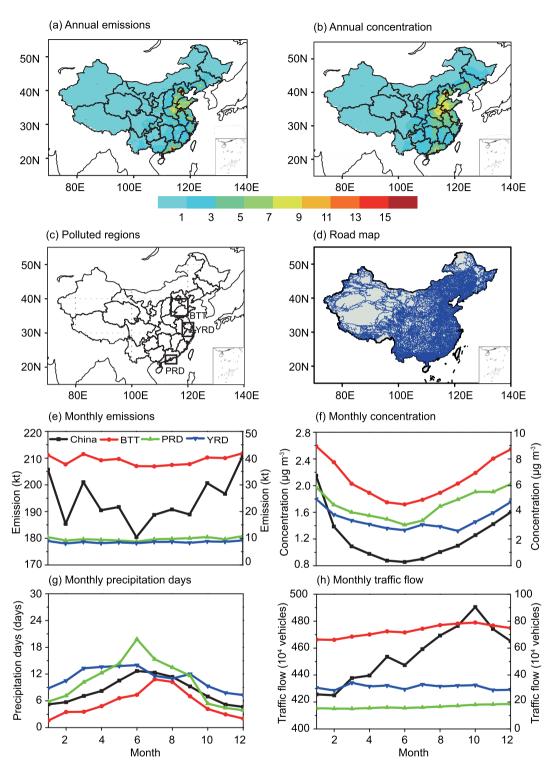


Figure 1. Road dust (a) emissions quantity (units: kt) and (b) concentration (units: μg m⁻³) in China averaged over 2006–2011. (c) Polluted regions examined in this study: Beijing–Tianjin–Tanggu (BTT; (35°–40°N, 114°–120°E)); Pearl River Delta region (PRD; (21°–23.5°N, 112°–116°E)); Yangtze River Delta (YRD; (29.5°–32.5°N, 118°–122°E)). (d) Road map over China from WGS84 Datum. Monthly variations in road dust (e) emissions (units: kt) and (f) concentration (units: μg m⁻³) over China, BTT, the PRD, and the YRD averaged over 2006–2011. (g) Monthly variations of the number of precipitation days over China, BTT, the PRD, and the YRD averaged over 2006–2011 (units: d/month). (h) Monthly variations of traffic flow over China, BTT, the PRD, and the YRD averaged over 2006–2011 (units: 10⁴ vehicles/month). Note: The left-hand axes are for China, and the right-hand axes are for BTT, the PRD, and the YRD.

Table 1 and are used to evaluate the simulated concentrations in Figure 2. Specifically, the simulated and observed seasonal mean road dust concentrations in different seasons and years for Beijing and Nanjing are compared. Although most observations are outside of our simulation period, the simulated concentrations, to some extent, capture the magnitude of the observed concentrations. In Beijing, the simulated seasonal road dust concentrations averaged over 2006-2011 show higher values in winter than in summer, which agree with the seasonal variation reported by Song et al. (2007). In Nanjing, the simulated seasonal mean concentration is highest in winter and lowest in summer, which is similar to the variation reported by

Table 1. Observed road dust concentrations used in Figure 2.

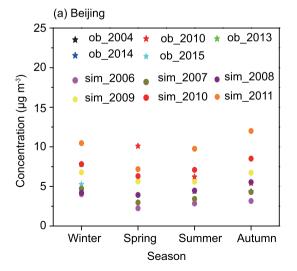
| | _ | | Concen- tration | |
|--------------------------------|------------------------------------|--------|-----------------------|-----------------------|
| Location | Time period | | (µg m ⁻³) | Reference |
| Beijing | Jan. 2004 | | 7.8 | Song et al. |
| (39.9°N, | Aug. 2004 | | 4.4 | (2007) |
| 116.4°E) ^a | 1 Jan. to 31 | Spring | 10.1 | Yu (2013) |
| | Dec. 2010 | Summer | 6.2 | |
| | | Autumn | 5.4 | |
| | | Winter | 4.7 | |
| | 11 Sep. to 2 Nov. 2014 | | 4.34 | Wang et al. (2016) |
| | 3 Nov. to 12 Nov. 2014 | | 3.33 | (2010) |
| | 13 Nov. 2014 to 31 Jan. 2015 | | 5.25 | |
| Nanjing, Gulou (32.1°N, | 24 Apr. to 5 May 2013 | Spring | 8.53 ± 5.96 | Li et al. (2016) |
| 118.8°E) | 11–28 Aug. 2013 | Summer | 2.71 ± 2.32 | (====, |
| | 10–18 Oct. 2013 | Autumn | 5.47 ± 3.32 | |
| | 2–12 Dec. 2013 | Winter | 16.0 ± 7.39 | |
| Nanjing, Xian- lin (32.1°N, | 24 Apr. to 5 May 2013 | Spring | 12.0 ± 4.92 | |
| 119.0°E) | 11–28 Aug. 2013 | Summer | 6.54 ± 3.77 | |
| | 10–18 Oct. 2013 | Autumn | 5.37 ± 3.01 | |
| | 2–12 Dec. 2013 | Winter | 24.0 ± 11.6 | |

^aCentral latitude and longitude of the city

Li et al. (2016), but the simulated values have low biases. The gap between simulation and observation in Beijing and Nanjing stems from the mismatch of the model grid resolution and the spatial scale of observation, as well as the mismatch of the modeled years and the observational period. In addition, road dust concentration measurements are usually carried out at specific locations near traffic and may not represent the average over the model grid cell.

3.3. Simulated variations of emissions and concentrations over 2006-2011

It is of interest to examine how road dust emissions and concentrations change over time. Figure 3(a) and (b) show the annual emissions and concentrations, respectively, of road dust, for the years 2006-2011, for the whole of China, for BTT, the PRD, and the YRD. The emissions of all the studied areas exhibit an increasing trend over 2006-2011; relative to the year 2006, the annual emissions in China, BTT, the PRD, and the YRD in the year 2011 increase by 260, 239, 266, and 59%, respectively. This increasing trend is mainly caused by the increased road length and number of vehicles (CCOT 2007-2012). Note that the change in climate, especially the change in precipitation, can also influence the increasing trend, but our sensitivity study with fixed precipitation days over 2006-2011 indicates that the changes in precipitation have a small impact on the increasing trend of road dust emissions (<5%). The annual road dust emissions quantity in China increases from 1103.2 kt in 2006 to 3974.0 kt in 2011, suggesting an increase to 5000 kt in 2013 if the increasing trend holds. Fu et al. (2016) reported a PM₁₀ road dust emissions quantity of 5242 kt in 2013 over China through use of an empirical model, which



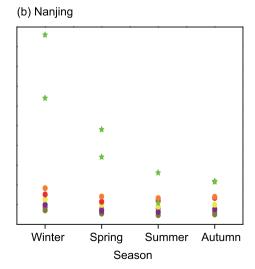


Figure 2. Comparison of the simulated concentrations (units: µg m⁻³; colored dots) of road dust with observations (units: µg m⁻³; colored stars) for (a) Beijing and (b) Nanjing.

Note: The measurements used here are listed in Table 1.

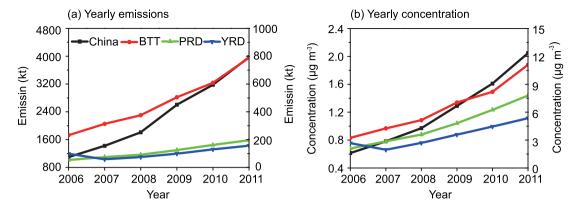


Figure 3. Yearly variation of (a) road dust emissions (units: kt) and (b) road dust concentration (units: μg m⁻³) over China, BTT (Beijing–Tianjin–Tanggu), the PRD (Pearl River Delta), and the YRD (Yangtze River Delta) from 2006 to 2011.

Note: The left-hand axes are for China and the right axes are for BTT, the PRD, and the YRD.

is close to our estimate. In the YRD, road dust emissions in 2007 are lower than in 2006, which can be explained by the reduced road lengths in 2007 of road types other than 'freeway', 'first-class highway', 'second-class highway', 'third-class highway', and 'fourth-class highway', according to the Year Book of China Transportation & Communication (CCOT 2007–2012).

Annual road dust concentrations in the studied regions for the years 2006–2011 show similar increasing trends to those of road dust emissions (Figure 3(b)). The annual concentration averaged over China increases from 0.6 $\mu g \, m^{-3}$ in 2006 to 2.0 $\mu g \, m^{-3}$ in 2011, with a percentage increase of 233% over 2006–2011. Compared to the year 2006, the annual concentrations in BTT, the PRD, and the YRD in the year 2011 increase by 243%, 273%, and 100%, respectively. Such large increasing trends in road dust emissions and concentrations indicate that road dust aerosol is set to become much more important in the future.

4. Conclusion and discussion

In this study, we estimate road dust emissions in China using the AP-42 empirical model developed by the U.S. Environmental Protection Agency, and examine the spatial distributions, monthly variations, and trends of changes in emissions and concentrations of road dust from 2006 to 2011. Emissions and concentrations of road dust are higher in eastern China than in western China, in agreement with there being more paved roads and busier traffic in eastern China (CCOT 2007-2012). Emissions are calculated to be 459.1, 112.0, and 102.7 kt, respectively, over the heavily polluted regions of BTT, the PRD, and the YRD. Monthly variations of emissions are mainly determined by monthly precipitation and traffic flow; emissions over China are highest in December and lowest in June, following the variation in precipitation, and second lowest in February as a result of the lowest traffic flow occurring in that month. The highest annual mean road dust concentration is

simulated to be 14.5 μ g m⁻³ in Beijing. Compared to the year 2006, annual mean concentrations of road dust in BTT, the PRD, and YRD in the year 2011 increase by 243%, 273%, and 100%, respectively, which have important implications for future air quality in China.

It is important to note that there are uncertainties in our estimates of road dust emissions. Firstly, the road emissions factor varies with location. For example, in our calculation, the factors in BTT, the YRD, the PRD, and the Sichuan Basin (SCB) are averaged for other areas without enough road information. Therefore, more accurate emissions factors are needed in future studies. Secondly, the physical properties of road dust aerosol are assumed in the model to be the same as those of mineral dust aerosol; this should be improved when direct measurements of the size distribution of road dust become available. And thirdly, more measurements of emissions and concentrations of road dust are needed for evaluating and improving simulations of road dust.

Disclosure statement

No potential conflict of interest was reported by the authors.

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