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The washout effects of rainfall on atmospheric particulate pollution in two Chinese cities[☆]

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ABSTRACT

Though rainfall is recognized as one of the main mechanisms to reduce atmospheric particulate pollution, few studies have quantified this effect, particularly the corresponding lag effect and threshold. This study aimed to investigate the association between rainfall and air quality using a distributed lag non-linear model. Daily data on ambient $PM_{2.5}$ and $PM_{2.5-10}$ (particulate matter with an aerodynamic diameter less than 2.5 μm and from 2.5 to 10 μm) and meteorological factors were collected in Guangzhou and Xi'an from 2013 to 2014. A better washout effect was found for $PM_{2.5-10}$ than for $PM_{2.5}$, and the rainfall thresholds for both particle fractions were 7 mm in Guangzhou and 1 mm in Xi'an. The decrease in $PM_{2.5}$ levels following rain lasted for 3 and 6 days in Guangzhou and Xi'an, respectively. Rainfall had a better washout effect in Xi'an compared with that in Guangzhou. Findings from this study contribute to a better understanding of the washout effects of rainfall on particulate pollution, which may help to understand the category and sustainability of dust-haze and enforce anthropogenic control measures in time.

1. Introduction

With increasing attention to dust-haze and related harmful health effects in China, air pollution has become a significant environmental concern for the public and policy makers (WHO, 2011; Rauhala, 2013; Xu et al., 2013). One of the major factors that causes dust-haze is atmospheric particles, especially $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm), which originates mainly from fuel combustion, construction dust and vehicle exhaust (Vingarzan and Li, 2006). The natural and anthropogenic processes for reducing atmospheric particle levels are important to improve air quality.

Rainfall is recognized as one of the main natural processes to improve air quality (Duhanyan and Roustan, 2011; Elperin et al., 2011), and it can greatly enhance the positive reductions achieved by anthropogenic control measures (Leung and Gustafson, 2005). Our previous study assessed the rainfall removal efficiency for total suspended particles (TSP) and found that artificial rain interventions may worsen air quality due to the lower removal of TSP without rainfall (Guo et al., 2014). However, there is limited information about the washout of particles and improvement in air quality following a single rain event and corresponding lag effects, as previous studies usually combined several rain events in one sample or focused on insufficient daily samples (Mattiot and Scafe, 1999; Encinas et al., 2004; He and Balasubramanian, 2009; Huang et al., 2009; Guo et al., 2014). Moreover, the results for analyzing washout of TSP are not applicable to a more narrowly defined specific size range of particle (e.g., $PM_{2.5}$), as the washout effects are variable among different sizes of particles (Baklanov and Sørensen, 2001; Wang et al., 2010).

Numerous studies have conducted experimental and theoretical assessments on scavenging coefficients; a microphysical approximation of the particle washout effect by raindrops (Slinn, 1984; Flossmann et al., 1985; Volken and Schumann, 1993; Mircea et al., 2000; Andronache, 2003; Laakso et al., 2003; Loosmore and

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Cederwall, 2004; Bae et al., 2006; Feng, 2007; Wang et al., 2010; Quérel et al., 2014a, 2014b; Wang et al., 2014). However, due to the complex collection of microphysical parameters (e.g., raindrop diameter, particle diameter, collision efficiency between raindrop and atmospheric particles, etc.) and huge computing resources required (Mircea et al., 2000; Bae et al., 2006; Wang et al., 2010), the scavenging coefficient is not easy to calculate, and therefore not suitable to evaluate air quality directly. Instead of linking air quality to scavenging coefficient, an approach is needed that links it to observable meteorological parameters such as rainfall conditions (Quérel et al., 2014b). Also, focusing on observable parameters may allow a more direct evaluation of the change in air quality following rain. For example, rainfall may worsen air quality following low amounts of precipitation (Levin and Cotton, 2008; Feng and Wang, 2012; Yuan, 2014) and identifying the critical threshold which corresponds to precipitation amount may help to discriminate and forecast changes in air quality.

In view of this knowledge gap, we assessed the removal effects of rainfall by collecting daily $PM_{2.5}$ and $PM_{2.5-10}$ (particulate matter with an aerodynamic diameter between 2.5 and 10 μm) concentrations and meteorological factors (wind speed and precipitation) in Guangzhou and Xi'an during 2013 and 2014. This study aims to provide a quantitative description of the rainfall washout effect using a new approach, which can enhance the knowledge for evaluating particulate pollution, and hence assist in implementing efficient control measures to cope with worsening air quality.

2. Materials and methods

2.1. Study setting and data collection

Two cities were selected to explore rainfall washout effects on particles. One is Guangzhou, a coastal city in South China with high amounts of precipitation, e.g., annual average precipitation of 2200 mm in 2014 (http://cdc.nmic.cn/home.do). The other is Xi'an, an inland city in Northwest China with low amounts of precipitation, e.g., annual average precipitation of 660 mm in 2014 (http://cdc.nmic.cn/home.do). The two cities have a similar annual wind speed (2.3 ± 0.97 km h^{-1} for Guangzhou and 2.3 ± 0.96 km h^{-1} for Xi'an in 2014) (http://cdc.nmic.cn/home.do) and different particulate pollution levels (Duan et al., 2005; Wang et al., 2013). Annual average concentrations of PM_{10} were 69 μg m^{-3} in Guangzhou and 118 μg m^{-3} in Xi'an in 2012 (Guangzhou Environmental Protection Bureau, 2012; Xi'an Environmental Protection Bureau, 2012).

Daily air pollution levels from 11 air monitoring stations in Guangzhou and 13 monitoring stations in Xi'an were obtained from relevant environmental monitoring Centers (Fig. 1 and Table S1 of Supplemental material; "S" indicates the tables and figures in the Supplementary material thereafter). Daily $PM_{2.5}$ and PM_{10} were collected at each station with a continuous dichotomous ambient particulate monitor 1405-D TEOM™ (Thermo Fisher Scientific Inc, Franklin, MA) during the study period from January 18th, 2013 to December 31st, 2014. The $PM_{2.5-10}$ concentration at each site was calculated as the difference in corresponding PM_{10} and $PM_{2.5}$ concentrations. Daily average values were calculated for all monitoring stations in both cities.

Corresponding meteorological data for daily rainfall (*RA*, mm), daily wind speed (*WS*, km h^{-1}), daily temperature (*T*, °C) and relative humidity (*RH*, %) were obtained from meteorological stations located in Luogang, Guangzhou and Jinghe, Xi'an (Fig. 1 and Table S1). All data were obtained from China's Meteorological Data Sharing Service System (http://cdc.nmic.cn/home.do).

2.2. Statistical analysis

As previous studies indicated that the relationship between rainfall and atmospheric particle concentrations might be nonlinear with a lag effect (Barmpadimos et al., 2011), a distributed lag non-linear model (DLNM) was utilized to describe the impacts of daily rainfall on particle concentrations at different lag days. Being a flexible model to simultaneously investigate non-linear affect–response dependencies and delayed effects, the DLNM has been widely used to investigate the health effects of day-to-day fluctuations of environmental factors (Gasparrini, 2011; Zeng et al., 2012; Wu et al., 2013).

The polynomial function and natural cubic spline function were used to represent the change in rainfall and lag structure in "cross-basis" function. Lags up to 14 days were selected to capture the full picture of the lag effects based on empirical values. The choice of potential confounding factors and degrees of freedom (*df*) to optimize model fit was based on Akaike Information Criterion (AIC) (Peng et al., 2006). Daily temperature and relative humidity were excluded as they were not statistically significant in the model (Figs. S1 and S2). Daily wind speed and month trend were controlled for in the model. The model can be expressed as:

$$\log[E(Y_t)] = \alpha + \beta RA_{t,l} + WS_t + NS(Month, 2) \quad (1)$$

where $E(Y_t)$ denotes the expected daily particle concentration on day t, α is intercept, $RA_{t,l}$ is "cross-basis" function, l refers to the number of lag days, WS_t refers to the daily wind speed on day t, *NS* represents the natural cubic spline function, *Month* represents the month trend, and its *df* is 2, the *df* for polynomial function is 1. The reference value was reported as relative effects (with 95% confidence intervals (CI)) of rainfall increase or decrease (1 mm) on particle concentration along certain lag days. To represent a consequence of rainfall washout, a relative effect greater than 1 represents an increase in particulate pollution from baseline, and vice versa. In particular, a relative effect equal to 1 represents the threshold.

Additional sensitivity analysis was conducted to test the robustness of models by changing the *df* for month trends (from 1 to 4) and wind speed (from 1 to 4). All statistical tests were two-sided and values of $p < 0.05$ were considered statistically significant. The "DLNM" package in R software Version 3.2.0 (R Development Core Team, 2014) was utilized to fit all the models.

3. Results

Average daily concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ in Guangzhou were relatively lower than those in Xi'an. In Guangzhou, concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ were 50 ± 26 μg m^{-3} (range: 9.3–160 μg m^{-3}) and 21 ± 9.9 μg m^{-3} (range: 0.53–66 μg m^{-3}), respectively (Table 1). In Xi'an, the concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ were 89 ± 77 μg m^{-3} (range: 9.6–900 μg m^{-3}) and 75 ± 56 μg m^{-3} (range: 4.1–640 μg m^{-3}), respectively (Table 1). The daily average rainfall was 6.0 ± 15 mm in Guangzhou and 1.5 ± 5.2 mm in Xi'an (Table 1).

Relative effects representing non-linear associations between rainfall and lag effects (lag0–14 days) of atmospheric particle levels in both cities are displayed in Fig. 2. Significant effects of rainfall were found on both $PM_{2.5}$ and $PM_{2.5-10}$ (Tables 2 and S2), with relative effects highly dependent on the precipitation amount. For example, with an increase in rainfall from 1 mm to 20 mm, the relative effect on $PM_{2.5}$ at lag0 in Guangzhou gradually decreased from 1.03 (95% CI: 1.02–1.04) to 0.93 (95% CI: 0.90–0.95). Moreover, relative effects were slightly lower for $PM_{2.5-10}$ compared to those for $PM_{2.5}$ in both cities for the same rainfall and lag days.