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Below-cloud scavenging of aerosol particles by precipitation in a typical valley city, northwestern China

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HIGHLIGHTS

- Scavenging effects of rain and snow on aerosol particles were evaluated.
- Snow scavenged particles in 20–1000 nm and 2000–10,000 nm more efficiently than rain.
- The thunderstorm rain more efficiently scavenged the particles in 500–1000 nm.
- Duration and raindrop velocity were main factors affecting aerosol particles.

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ABSTRACT

To fill the blank information for aerosol precipitation-scavenging research in north-west of China, the aerosol particle and raindrop size distributions were measured simultaneously during 1 September 2012 to 31 August 2013 in urban Lanzhou. The scavenging coefficients of thunderstorm and non-thunderstorm rain and snow events were studied and presented on the basis of nine selected precipitation cases including 3 snow and 6 rain events. The variation of scavenging coefficients of snowfall across the size distribution clearly exhibited a trough of lower values for particles of 1000 nm–2000 nm in diameter, while the particles smaller than 500 nm were scavenged efficiently by non-thunderstorm rain, and thunderstorm rain more effectively scavenged the particles in 500–1000 nm. The snow scavenging coefficients varied between $3.11 \times 10^{-7}\ s^{-1}$ and $1.18 \times 10^{-3}\ s^{-1}$ in the 10–10,000 nm size range. The scavenging coefficients of thunderstorm (non-thunderstorm) rain were between $8.25 \times 10^{-7}\ s^{-1}$ ($7.48 \times 10^{-6}\ s^{-1}$) and $1.23 \times 10^{-3}\ s^{-1}$ ($7.46 \times 10^{-4}\ s^{-1}$). Additionally, the number of particles in 10–50 nm was more sensitive to duration of snow, while snowfall intensity was more responsible for particle number concentrations in 50–100 nm and 100–1000 nm. The longer period of precipitation with lower raindrop velocity can more effectively scavenge the particles in the size range of 10–50 nm.

1. Introduction

Aerosol particles released or formed in the atmosphere will eventually be removed to the Earth's surface by wet scavenging or dry deposition. Wet scavenging is a major mechanism for maintaining a balance between the sources and sinks of aerosol particles and cleaning the polluted atmosphere. Depending on the vertical position of the aerosol particle, the scavenging process can be classified as in-cloud—particles serving as cloud condensation nuclei or undergoing capture by cloud water and as below-cloud—the removal of aerosol particles by precipitating raindrops or snow particles. Knowledge of wet scavenging of aerosol particles is required in a lot of fields, such as air quality modeling (Wang et al., 2005; Tost et al., 2006; Feng, 2007; Croft et al., 2009), environmental emergency responses, and estimation of deposition of airborne hazardous chemical materials (Chate, 2011). For example, Feng (2007) developed a 3-mode parameterization of below-cloud

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scavenging of aerosols for use in atmospheric dispersion models, and found the new parameterization was obviously superior to the traditional bulk parameterization, which represents the mean wet scavenging coefficient for the whole aerosol size range.

The capture of aerosol particles by falling raindrop takes place with Brownian and turbulent shear diffusion, inertial impaction, diffusiophoresis, thermophoresis and electrical charge effects. Detailed studies of these processes (Greenfield, 1957; Pilat, 1975; Wang et al., 1978; Slinn, 1983; Herbert and Behang, 1986) have revealed that there exists a minimum in the collection efficiency of aerosol particles between sizes ranging from 0.1 to 2 μm diameter, which is often called the "Greenfield gap". The size ranges of Greenfield gap in various environments exists some differences between studies (Laakso et al., 2003; Chate, 2005; Feng, 2007), which may be related to atmospheric conditions, precipitation type, rainfall intensity and particle size range (Chate, 2005). Inertial impaction is most important for particles with diameter more than about 2 μm, while for particles smaller than 0.1 μm Brownian diffusion is the main removal mechanism (Chate and Pranesha, 2004). Fraction of aerosols collected by raindrop or snow particles relative to initial number concentrations of aerosols in its path is called scavenging efficiency, which represents scavenging ability of precipitation. Scavenging coefficient (s^{-1}) for particle of different sizes is a rate of aerosols washout by precipitation which varies with collection efficiency, particles and hydrometers size distributions and their terminal velocities (Seinfeld and Pandis, 2006). There are some studies (Laakso et al., 2003; Chate, 2005; Kyrö et al., 2009) where scavenging coefficients have already been calculated and investigated for different size ranges and environment. For example, Kyrö et al. (2009) calculated snow scavenging coefficients using four years of particle number concentration spectra measurements together with meteorological measurements, and found experimental median scavenging coefficients varied between $8.7 \times 10^{-6}\ s^{-1}$ and $5.2 \times 10^{-5}\ s^{-1}$ in the 10 nm to 1 μm size range. Although there is some information on aerosol particle scavenging by falling raindrops (Laakso et al., 2003; Chate and Pranesha, 2004), scavenging by falling snowflakes and ice crystals is even less known both experimentally and theoretically. Due to the lack of suitable experiments only few field studies on scavenging by snow crystals have been reported (Jylhä, 2000; Kang et al., 2009; Kyrö et al., 2009). For example, in study of Jylhä (2000), the snow scavenging coefficient was estimated to be of the order of $10^{-6}\ s^{-1}$ or less. Their studies show that snow scavenging is an important removal mechanism for aerosol particles and snow can scavenge aerosol particles up to 50 times more efficiently than rain, when based on equal equivalent water content of the precipitation.

The effect of electrical forces is often assumed to be one of the major sources of uncertainty in the calculations of scavenging coefficients (Jaworek et al., 2002). Based on the fact, Chate (2005) studied scavenging of submicron-sized aerosol particles by thunderstorm rain events, and indicated scavenging coefficients for 0.013–0.75 μm particles were between $1.08 \times 10^{-5}\ s^{-1}$ and $7.58 \times 10^{-4}\ s^{-1}$. In addition, based on 6 years of outdoor measurements at a boreal forest site in Southern Finland, Laakso et al. (2003) showed median scavenging coefficients varied between $7 \times 10^{-6}\ s^{-1}$ and $4 \times 10^{-5}\ s^{-1}$ for aerosol particles having diameter between 10 and 510 nm, and their results indicated scavenging coefficient changed from 1×10^{-5} to $4 \times 10^{-5}\ s^{-1}$ when the rain intensity increased from 0.5 to 9 mm h^{-1}. In China, although there are some measurements of particle size distributions were operated in central eastern or coastal areas (Wu et al., 2008; Yue et al., 2010; Gao et al., 2009), there is very little research where the aerosol particle and raindrop size distributions were simultaneously measured (Kang et al., 2009). Several studies in northwestern China were mainly concentrated on particulate mass and coarse particle size distribution (0.5–20 μm) (Yu et al., 2010, 2011; Zhao et al., 2012), with little or no information on wet scavenging of aerosol particles. In this study high resolution particle size distributions in 10–10,000 nm and raindrop size distributions were simultaneously measured to better understand the scavenging effect of different types of precipitation on particle concentrations in different size ranges and changes in aerosol particle size distribution properties due to precipitation.

The objective of this study is to investigate the effect of different types of precipitation on urban air quality in a valley city, especially particle concentrations and their size distributions using *in situ* observations, and explore the main characteristic quantities of precipitation alleviating urban air pollution.

2. Methods

The basic equation for the change of aerosol particle number concentration $c(D_p)$ due to precipitation scavenging is given by (Seinfeld and Pandis, 2006)

$$\frac{dc(D_p)}{dt} = -\lambda c(D_p) \tag{1}$$

where D_p is aerosol particle diameter and λ is the scavenging coefficient given by

$$\lambda(D_p) = \int_0^{\infty} \frac{\pi}{4} D^2 U_t(D) E(D, D_p) N(D) dD \tag{2}$$

where D is the rain droplet diameter, U_t is the velocity of the falling droplet, $E(D, D_p)$ is the collision efficiency between the falling rain droplet and aerosol particle and $N(D)$ is the concentration of rain droplets as a function of droplet diameter. The integral is due to the fact that an aerosol particle of certain size D_p can be captured by rain droplets of any size. λ can be calculated theoretically if the collision efficiency and raindrop size distribution are known. However, usually collision efficiency is not known.

Our approach to the problem is similar to that presented by Chate and Pranesha (2004). In order to minimize the effect of diurnal variations of particle number concentrations (PNCs) in different size ranges on scavenging role of precipitation and assuming precipitation scavenging is the only aerosol source or sink, the change in concentration for non-interacting and non-growing particles $N(D_p)$ with diameter D_p and $D_p + dD_p$ after precipitation is related to corresponding monthly mean concentration $N_a(D_p)$, as

$$N(D_p) = N_a(D_p) \exp[-\lambda(D_p)t] \tag{3}$$

The scavenging coefficient $\lambda(D_p)$ is derived from Eq. (3) as given below:

$$\lambda(D_p) = \ln[N_a(D_p)/N(D_p)]/t \tag{4}$$

where t is duration of precipitation. Eq. (2) can be compared with Eq. (4) when the rain scavenging is the only particle source or sink.

In order to know the effect of precipitation on modal structure of particle number size distributions, the hourly mean particle number size distributions (PNSDs) at rainy or snowy day were fitted using multivariate logarithm normal distribution. The formula of lognormal distribution is given as follows: