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Cloud deposition of PAHs at Mount Lushan in southern China

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HIGHLIGHTS

- The site is in the high pollution emission area, having many sources of PAHs around.
- Mount Lushan is a unique site for cloud chemistry monitoring.
- Atmospheric PAHs mostly deposited and transferred into cloud water.
- Special case showed PAHs are more concentrated in cloud water than in rainwater.

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ABSTRACT

Cloud water samples were collected from Mount Lushan, a high alpine area of southern China, and analyzed using GC–MS to investigate the concentration levels, seasonal variations, particle-dissolved phase partitioning, ecological risk of PAHs and its relationship to the atmosphere and rainwater. The average concentration of total (dissolved + particle) PAHs in cloud water was 819.90 ng/L, which ranged from 2.30 ng/L for DbA to 295.38 ng/L for PhA. PhA (33.11%) contributed the most individual PAHs, followed by Flu (28.24%). Distinct seasonal variations in the total PAHs measured in this research had a higher concentration during the spring and a lower concentration during the summer. When cloud events occurred, the concentration of the atmospheric PAHs of the two phases decreased. The contribution from the gaseous phase of total PAHs in the air to the dissolved phase in cloud water was up to 60.43%, but the particulate phase in the air only contributed 39.57% to the total scavenging. The contribution of total PAHs from the atmosphere to clouds is higher in the gaseous phase than in the particulate phase. A comparative study of the concentrations of cloud water and the closest rain water revealed that the PAH concentration in rainwater was 1.80 times less than that of cloud water and that the dominant individual compounds in cloud water and rainwater were PhA and Flu. A total of 81.27% of the PAHs in cloud samples and 72.21% of the PAHs in rain samples remained in the dissolved phase. Ecological risk assessment indicated that PAHs in cloud water in spring and summer caused a certain degree of ecosystem risk and the mean ecosystem risk in spring was higher than that in summer.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), which are mutagenic and carcinogenic compounds to humans and animals, originate mainly from human activities, such as automobile traffic, domestic heating, thermal power stations and industrial emissions (Motelay-Masseia et al., 2003). They are ubiquitous in the environment and can be found in the air, precipitation, surface waters, sediments, soils, and in food (Grynkiewicz et al., 2002). Additionally, because of their persistent characteristics, PAHs undergo long-range atmospheric transport and can be found far from their place of production or use (Halsall et al., 2001; Carrera et al., 2001; Lammel et al., 2009; Garban et al., 2002), including remote mountains that have no emission sources (Kim et al., 2012; Li et al., 2010). Therefore, atmospheric transport plays a key role in transferring pollutants to regional areas and influencing the global distribution of PAHs. Many studies have shown that there is an increasing concern about the existence of PAHs in the environment (Ravindra et al., 2008; Callen et al., 2008; Li et al., 2009).

As a group of environmental carcinogens, PAHs enter the atmosphere through the incomplete combustion of fossil fuels and were redistributed between the gaseous and particulate phases of atmospheric aerosol because they are semi-volatile (Škrdlíková et al., 2011). Clouds play a vital role in the transport and redistribution of atmospheric constituents (Seinfeld and Pandis, 2006). Cloud studies focusing on organic matter and its characterization and sources started

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in the 1990s and increased concerns during the 2000s (Herckes et al., 2013). During the evolution of the cloud, the scavenging of soluble gases and particles is an important mechanism to remove PAHs from the atmosphere and is a source for terrestrial and aquatic systems (Simcik, 2004). Golomb et al. (2001) reported that wet deposition was a predominant role in more remote areas because cloud droplets accumulate gaseous and particulate PAHs during long distance transportation.

Some cloud water studies were performed in Asia for example China, Japan and Korea (Guo et al., 2012; Budhavant et al., 2014; Wang et al., 2011). Only limited information regarding PAHs in cloud water was obtained, particularly at high altitudes, even though increasing amounts of hazardous pollutants were found in high alpine areas because of long-range atmospheric transportation. In this paper, a Caltech Active Strand Cloud Water Collector (CASCC) was used to collect cloud water at Mount Lushan, China (a.s.l. 1165 m), a location far away from ground level pollution sources. It is located in southern China and is one of the most developed and highest pollution emission areas in China because of rapid industrialization and economic development. More than 190 foggy days are reported each year at Mount Lushan and cloud events are universal from spring to summer. The aim of this study was to determine the concentration levels and distribution characteristics of PAHs in cloud water at Mount Lushan. Additionally, to investigate the influence of atmospheric PAHs to clouds, the concentration variation of PAHs from the atmosphere was analyzed, and the relative contribution of PAHs from the atmosphere to clouds was calculated during the observation period. This study compared PAH characteristics of clouds and rain.

2. Materials and methods

2.1. Sampling site and procedures

Intensive field measurements of PAHs in cloud water were conducted at Mount Lushan (E115.98°, N29.58°, Fig. 1) at an altitude of 1165 m. The measurements were taken over a 300 km^2 area, located in the

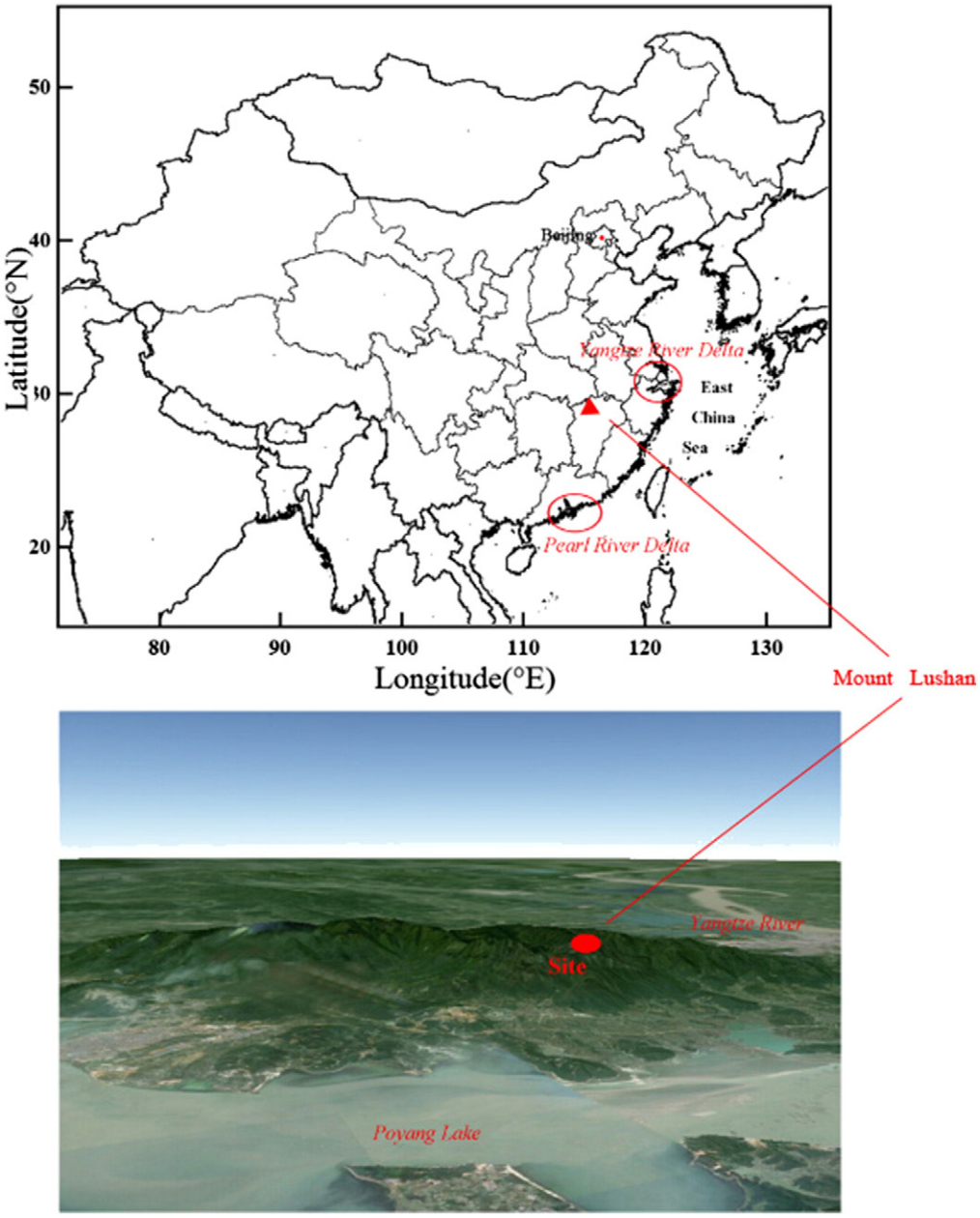

Fig. 1. The location of sampling site at the summit of Mount Lushan.