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Pb, Sr and Nd isotopic composition and trace element characteristics of coarse airborne particles collected with passive samplers

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ABSTRACT

Passive samplers for collection of coarse airborne particulate matter have been installed in and around the coal-mining town of Cam Pha, Quang Ninh Province (Vietnam). Analysis of Pb, Sr, and Nd isotope ratios and of major and trace element distribution patterns in atmospheric particulates collected at three stations allowed for the identification of four important dust components: (1) coal dust from an open-pit mine and fly ash particles from a coal-fired power station, (2) diesel soot, (3) traffic dust from metal, tire and pavement abrasion, and (4) limestone-derived dust. Outside of the coal-mining area, traffic-derived dust defines the atmospheric baseline composition of the studied environment.

1. Introduction

Airborne particulate matter (PM) of both natural and anthropogenic origin can affect air quality and human health (Kampa and Castanas, 2008; Murray et al., 2001; Petit et al., 2015). Among the anthropogenic PM sources, transportation, energy production and manufacturing industries emit large amounts of PM as well as persistent organic pollutants and heavy metal compounds (Cohen et al., 2010; Gieré and Querol, 2010; Gieré et al., 2006; Grobéty et al., 2010; Guéguen et al., 2012a,b,c, 2013; Lahd Geagea et al., 2007, 2008a, b; Négrel et al., 2015; Petit et al., 2015; Widory et al., 2010). In addition, the mining industry contributes significantly to the load of airborne PM in mining districts, especially through processes, such as open-pit mineral extraction and ore processing (Chaulya, 2004; Cohen et al., 2010; Ghose, 2004, 2007; Ghose and Majee, 2000a, b, 2007; Mukai et al., 2001; Widory et al., 2010). The impacts of such PM on environment and health depend, among other factors, on the chemical composition and physical properties of the individual particles (Gieré and Vaughan, 2013).

To monitor and, eventually, to improve air quality through implementation of dust-control policies, the sources of anthropogenic airborne dust must be identified. The isotopic ratios of certain elements (e.g., Pb, Sr, Nd, Zn) as well as major and trace element characteristics of PM can be used as powerful tools for identifying the source of airborne pollutants, such as coal-combustion plants, cement factories, metal smelters, steel plants, and waste incinerators (Grobéty et al., 2010; Guéguen et al., 2012a; Lahd Geagea et al., 2007; Mattielli et al., 2009).

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After studies upon the combination of Sr, Nd and Pb isotope systems as tracers of sources in soil forest (Stille et al., 2012), here, the goal of our investigation was to characterize coarse atmospheric PM and assess its sources in the coal-mining district of Quang Ninh Province, northeastern Vietnam. This region is known for its natural beauty (e.g., Halong Bay, a United Nations World Heritage Site), but also for extensive coal mining. The mining activities lead to considerable PM loads, which are further increased by particulate emissions from several other pollution sources; in order to characterize the different sources of atmospheric particulates, we have analyzed a set of representative samples with respect to their Pb, Sr, and Nd isotopic composition and their bulk major and trace element content.

2. Material and methods

2.1. Sampling sites

The study was conducted in Quang Ninh, a province in northeastern Vietnam (Fig. 1) that hosts vast reserves (~7.2 Gt) of high-quality anthracite. Of the overall coal production in Vietnam (11.3 Mt in the year 2000; USGS, 2000), production in Quang Ninh accounts for 85–95% and has increased significantly over past decades, with an estimated annual production of over 75 Mt by 2030 (HoangTrungHai, 2012; TranXuanHoa, 2010). Coal-fired power plants produce 56.4% of Vietnam's total electricity and will consume 171 Mt of coal by 2030 (Martens et al., 2011; NguyenTanDung, 2011; TranXuanHoa, 2010). The coal from Quang Ninh has a relatively high energy content and contains low amounts of ash and sulfur, but the mining activities have considerable impacts on the local environment (e.g., coal mine drainage, air pollution) and human health (Hoàng-Hòa et al., 2011, 2012; Martinez et al., 2013).

This study focuses on the coal-mining town of Cam Pha (Fig. 1) and its surroundings, where various sources of air pollution exist, including: several open-pits; coal-processing facilities; thermal power plants; a major harbor for coal export; waste-rock dumps; a cement factory; and road, train and ship traffic.

The *PM samples* were collected at three different locations: station 1 (ST1) was established on the roof of a private house (10 m above ground; 14 m above sea level), which is located at a distance of 20 m from a main road (# TL334) on Van Don island, ~10 km to the east of Cam Pha. Station 4 (ST4) was installed on the roof of a private house in the town of Cam Pha (14 m above ground; 20 m above sea level), ~2 km from the nearest open-pit coal mine, the Coc Sau mine (Fig. 1C). Station 5 (ST5) was installed in the village of Dong Trieu, at the curbside of the main access road to Cam Pha (~100 km to the west, Fig. 1B). ST5 was set up to obtain a PM sample representative of the local traffic emissions.

To characterize potential sources of airborne PM, we also collected samples of *geological and industrial materials*. These include coal from the Coc Sau open-pit mine (sample S16) and sandstone (sample C601), which represents a prominent type of host rock of the Coc Sau coal. All coal deposits in the Cam Pha area are hosted by the Triassic Hòn Gai formation, which consists mainly of sandstone, conglomerates, siltstone and shale, and which is up to 2400 m thick. During the large-scale mining operations, these sedimentary host rocks are trucked to waste-rock dumps (Fig. 1C), where samples C605 and S9 were collected. We further characterized a sample of the local Carboniferous to Permian limestone, which represents a dominant lithologic unit in Quang Ninh Province. This limestone is also mined extensively and transported by truck and conveyor belts from the quarries to the local cement-manufacturing plant (Cam Pha Cement Jsc.; Fig. 1C), where it is used as major raw material. A sample of the cement produced in this factory was collected and analyzed. We further sampled fly ash from one of the thermal power stations located next to the coal-processing facilities in Cam Pha (Fig. 1C), where 2.5 Mt of raw coal are washed and cleaned annually (USGS, 2000). Moreover, a soil sample (S7) was collected from a rice-field located between ST4 and the Coc Sau open-pit mine (Fig. 1C).

To evaluate a possible traffic contribution to the PM samples, we collected *diesel soot*, which was scraped off the exhaust pipes of various mobile sources:

- a large mining truck used for coal transportation in the Coc Sau mine (horizontal exhaust pipe near one of the wheels);
- two locomotive engines (samples DS01, DS02) of coal-transporting trains circulating inside and outside of the mining district (vertically arranged exhaust pipe on locomotives);
- two passenger cars (samples DS03, DS04) in Cam Pha;
- a ship (sample DS05; vertical chimney) from the coal-shipping Cua Ong harbor (Fig. 1C).

2.2. Sampling of airborne PM

Airborne PM was collected with a Sigma-2 passive sampler. This device, implemented by the German Meteorological Service, is primarily used for routine air quality measurements in German health and recreation resorts, which are licensed by state regulation based on specific air quality standards (VDI_3787, 2008). The Sigma-2 sampler with its specially designed wind-free interior allows particles to be deposited almost exclusively according to the sedimentation principle. Moreover, it largely avoids the collection of particles suspended in rain droplets (wet deposition), thus allowing for sampling of mainly dry deposition (VDI_2119, 2013). After deposition on a transparent adhesive collection plate, which is exposed for an appropriate time in the interior of the sampler, individual coarse particulates in the size range of 2.5–80 μm are analyzed by automated optical microscopy. Apart from this operational application, the Sigma-2 technique is also used for various air quality studies (e.g., Schleicher et al., 2011, 2012; Yu et al., 2011). The Sigma-2 sampler is a cost-effective device, which does not require a power supply and thus, can be installed in a wide range of environments. In a recent study, the Sigma-2 sampler has been used successfully with a new acceptor