Source apportionment of PM$_{2.5}$ in Baton Rouge, Louisiana during 2009–2014

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HIGHLIGHTS

- Seven sources were identified based on six-year PM$_{2.5}$ data in Baton Rouge, Louisiana using PMF.
- Secondary sulfate, industry and secondary nitrate were the top three sources.
- Industry and crustal dust contributions increased while traffic and sea salt decreased in recent years.
- Winds play significant role in contributions of different sources in different seasons.

GRAPHICAL ABSTRACT

Seasonal contributions of different sources to PM2.5 in Baton Rouge, LA
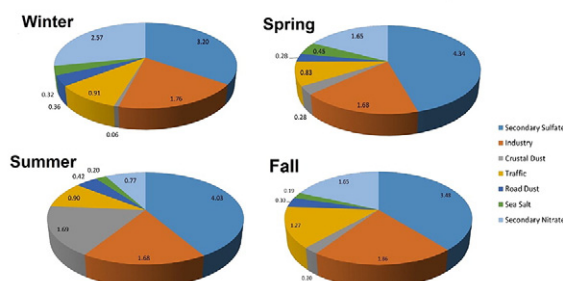
Secondary sulfate and nitrate would be partially from industry and traffic sources

ABSTRACT

Particulate matter with aerodynamic diameter <2.5 μm (PM$_{2.5}$) chemical composition data from the Speciation Trends Network (STN) site located in Baton Rouge, Louisiana were analyzed using the receptor Positive Matrix Factorization (PMF) model version 5.0. The PM$_{2.5}$ samples were collected every third day from January 2009 to December 2014. Seven sources were identified, including secondary sulfate, secondary nitrate, industrial emissions, traffic, crustal dust, road dust and sea salt. The contributions of these seven sources to PM$_{2.5}$ total mass were 38.4%, 17.6%, 18.7%, 11.5%, 6.1%, 4.2% and 3.6%, respectively. Secondary sulfate, industrial emissions and secondary nitrate were the top three sources. The contributions of industrial emissions and crustal dust have been rising in recent years while that of traffic and sea salt were decreasing. Secondary sources were higher than primary sources during the winter. The crustal and road dust were dominant during the summer, while traffic was more significant during the fall compared to other seasons. During summer, traffic emission and crustal dust were driven by northeast-north winds, traffic is also driven by northeast-north winds in winter, while industry emissions and sea salt were driven by prevailing west and northwest winds during other seasons. PM mass clearly showed the synergetic effects of local sources and distance sources. Thus, measurements and strategies should focus on not only local sources, but also regional transport. Attention should also be paid to industrial and traffic sources since they also account for secondary sources in addition to the primary contributions.

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1. Introduction

Particulate matter with aerodynamic diameter <2.5 μm (PM₂.₅) are usually microscopic liquid or solid particles suspended in the ambient air. It can be directly emitted from primary sources and secondarily formed by precursors through chemical and physical processes (Godri et al., 2009). In addition to its adverse effects on visibility (Ying et al., 2004), radiation, climate and ecosystems (Streets et al., 2006), PM₂.₅ also affects human health since the particles tend to penetrate into the gas exchange regions of the lung, among which the very small particles (<100 nm) may pass through the lungs to affect other organs (Valavanidis et al., 2008). Thus, long-term exposure to pronounced PM₂.₅ would increase morbidity and mortality (Sahu and Kota, 2016; Zheng et al., 2015). It is reported by the Global Burden of Disease Study that ambient particulate matter pollution accounted for over 3.1 million deaths around the world in 2010 (Lim et al., 2013).

To protect public health, the U. S Environmental Protection Agency (EPA) established the PM₂.₅ chemical Speciation Trend Network (STN) program to provide nationwide systematic and consistent data for assessing the trends (U.S. EPA, 1999). PM₂.₅ was also one of the criteria pollutants and the pollution level is shown as part of the Air Quality Index (AQI). It was reported that PM₂.₅ concentrations decreased by 27% from 2001 to 2010 in the U.S. (U.S. EPA, 2012, 2014), due to the significant reductions of gaseous sulfur and nitrogen oxides from coal-fired power plants and mobile sources (Chalbot et al., 2013). In 2006, the U.S. EPA reduced the 24-h PM₂.₅ standard to a level of 35 μg/m³ (F.R., 2007). The annual primary standard for protecting the health of sensitive populations for PM₂.₅ is 12 μg/m³ (U.S. EPA, 2013). It is also projected for 99% of the U.S. counties to fulfill the revised health standard by 2020, which is challenging for states currently having levels higher than the standard.

Design of effective air pollution control strategies of PM₂.₅ requires a clear and complete understanding of its sources, composition, size distribution, deposition, and spatial and temporal variations. The Positive Matrix Factorization (PMF) model developed by the U.S. EPA has been widely used around the world to understand the sources of PM₂.₅ (Heo et al., 2009; Hopke, 2016; Pekney et al., 2006; Song et al., 2001; Wang and Hopke, 2013). For example, Lee et al. (2006) studied the local emission sources at a supersite using wind direction and chemical components of PM₂.₅ and Pekney et al. (2006) studied the coupling of PMF results with surface wind direction data to identify local emission sources affecting a receptor site. Wang and Hopke (2013) studied the source apportionment of PM₂.₅ in San Jose on a ten-year basis and temporal variations of different sources were explored. Zhang et al. (2013) systematically studied the chemical characterization and source apportionment of PM₂.₅ in Beijing, China in a seasonal perspective, and showed the constituents and sources can largely vary with seasons. Chalbot et al. (2013) explored the coastal sources and transport of the sources, demonstrating significance of emission controls on SO₂ and NOx. Bressi et al. (2014) also suggested that more coordinated strategies among neighboring areas may be required to effectively control PM₂.₅ emissions.

Generally, the sources of PM₂.₅ may be classified into vehicle emission, either gasoline or diesel, wood or biomass combustion, power plants, industrial emission, crustal and sea salt and more. Commonly, PM₂.₅ particles consist of primary and secondary particles, primary particles refer to those emitted directly from a source such as construction dust and road dust, while secondary particles are those formed from the oxidation of primary gases. PM₂.₅ particles may also include organic matter, biogenic and anthropogenic, and oxidation of VOCs (Goldstein et al., 2009).

The Baton Rouge region in Louisiana is a major industrial and petrochemical center of the southern U.S., the inland port that is farthest to ocean on the Mississippi River to accommodate ocean-going tankers and cargo carriers. The area also straddles a main transportation corridor with Interstates 10 and 12. All these sources contribute to PM₂.₅ concentrations that threaten a population of 802 thousand. However, there are no previous studies focusing on both the concentrations and sources of PM₂.₅ in this region. In this study, the PMF model is used to identify the major sources of PM₂.₅ and their temporal/spatial variations in Baton Rouge.

2. Methods

2.1. Study area and the sampling site

Baton Rouge is the capital and the second-largest city of Louisiana in Southeast U. S with a humid subtropical climate from Köppen's climate classification, where winters are warm and summers are hot and humid (Peel et al., 2007). Spring is from March to May, summer is from June to August, fall is from September to November, and winter is from December to February in Baton Rouge. There are moderate to heavy rainfalls, strong winds throughout all year, and chances of hurricanes. The annual average temperature is 68.4 °F, and seasonal average temperatures are 53.4 °F in winter and 82.3 °F in summer. Baton Rouge is one of the top precipitation cities in the U.S. with an annual average precipitation (rainfall) of 60.6 in. Interstate 10, Interstate 12, Interstate 110, U.S. 61, and U.S. 190 are the primary freeways that run through Baton Rouge.

PM₂.₅ samples and meteorological data were collected at the Leesville Avenue STN site, also known as the Capitol site (site ID: 220,330,009, 30.46°N, 91.18°W). It is one of the four stations in the Baton Rouge area, but the only one with PM₂.₅ speciation data. Twenty-five PM₂.₅ components, including ions, trace elements and carbonaceous compounds, such as Al, As, Br, Ca, Cl, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, S, Si, Sr, Ti, V, Zn, SO_4^{3-}, NH_4^+, NO_3^-, element carbon (EC) and organic carbon (OC) are measured.

The site, as shown in Fig. 1, is located in the urban area, surrounded by a business district and is adjacent to highways and the Mississippi River. There are several industries located in the Baton Rouge region. This region is predominantly filled with the large petrochemical production and manufacturing industries. ExxonMobil's Baton Rouge Refinery complex in this region is the fourth largest oil refinery in the U.S. The Albemarle Corporation is also headquartered in Baton Rouge. They are both located to the north of the monitoring site. Additionally, the Entergy Louisiana station power plant is also located in the north.

2.2. Sample collection and analysis

The PM₂.₅ data were obtained from ambient air monitoring program of Louisiana Department of Environmental Quality (LDEQ). The PM₂.₅ samples were collected on Teflon, nylon and quartz filters with a spiral aerosol speciation sampler (U.S. EPA, 2014). The Teflon filters were used for the analysis of mass concentrations of 14 metal elements by the energy-dispersive X-ray fluorescence spectroscopy (XRF). The nylon filters were used for the analysis of 3 cations and 2 anions including NH_4^+, K^+, Na^+, NO_3^- and SO_4^{2-} (U.S. EPA, 2009a, b) by ion chromatography. The quartz filters were used for the analysis of organic carbon and element carbon by the thermal optical reflectance method The 24 h samples were collected every third or sixth day. Samples with 15 out of 25 species non-missing were only included in the analysis. Thus, 656 samples were analyzed at last from 2009 to 2014.

2.3. Positive matrix factorization

Various statistical and mechanistic methods have been developed for factor analysis and source apportionment studies in the past years. Receptor models have been developed and designated to analyze the pollutants, identify the sources as well as determine their contributions. Generally, these models are conducted on a site-by-site basis.

The Positive Matrix Factorization (PMF) is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices: factor contribution (G) and factor profiles (F) (Gary Norris, 2014). The PMF model reduces the number of variables in a data set,