



Effect of the shutdown of a large coal-fired power plant on ambient mercury species



Yungang Wang^a, Jiaoyan Huang^{b,1}, Philip K. Hopke^{c,*}, Oliver V. Rattigan^d, David C. Chalupa^e, Mark J. Utell^e, Thomas M. Holsen^{b,c}

^a Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

^b Department of Civil and Environmental Engineering, Clarkson University, Potsdam, NY 13699, USA

^c Center for Air Resource Engineering and Science, Clarkson University, Potsdam, NY 13699, USA

^d Division of Air Resources, New York State Department of Environmental Conservation, Albany, NY 12233, USA

^e Department of Environmental Medicine, University of Rochester Medical Center, Rochester, NY 14642, USA

HIGHLIGHTS

- ▶ A CFPP located in Rochester, NY was closed over 4-month period in early 2008.
- ▶ The ambient Hg concentrations significantly decreased after the CFPP closure.
- ▶ PMF results show Hg apportioned to the CFPP factor significantly decreased.
- ▶ CPF results show the greatest Hg reduction was with winds pointing toward the CFPP.
- ▶ These changes were clearly attributable to the closure of the CFPP.

ARTICLE INFO

Article history:

Received 9 August 2012

Received in revised form 14 December 2012

Accepted 7 January 2013

Available online 16 February 2013

Keywords:

Coal-fired power plant (CFPP)

Mercury

Positive Matrix Factorization (PMF)

Conditional probability function (CPF)

Gas-particle partitioning coefficient

ABSTRACT

In the spring of 2008, a 260 MWe coal-fired power plant (CFPP) located in Rochester, New York was closed over a 4 month period. Using a 2-years data record, the impacts of the shutdown of the CFPP on nearby ambient concentrations of three Hg species were quantified. The arithmetic average ambient concentrations of gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate mercury (PBM) during December 2007–November 2009 were 1.6 ng m⁻³, 5.1 pg m⁻³, and 8.9 pg m⁻³, respectively. The median concentrations of GEM, GOM, and PBM significantly decreased by 12%, 73%, and 50% after the CFPP closed (Mann–Whitney test, $p < 0.001$). Positive Matrix Factorization (EPA PMF v4.1) identified six factors including O₃-rich, traffic, gas phase oxidation, wood combustion, nucleation, and CFPP. When the CFPP was closed, median concentrations of GEM, GOM, and PBM apportioned to the CFPP factor significantly decreased by 25%, 74%, and 67%, respectively, compared to those measured when the CFPP was still in operation (Mann–Whitney test, $p < 0.001$). Conditional probability function (CPF) analysis showed the greatest reduction in all three Hg species was associated with northwesterly winds pointing toward the CFPP. These changes were clearly attributable to the closure of the CFPP.

© 2013 Elsevier Ltd. All rights reserved.

1. Introduction

Mercury (Hg) has been labeled as a Persistent Bioaccumulated Toxic (PBT) chemical by the US EPA since 1997 (US EPA, 1997). Hg is found in various compartments of an ecosystem including sediments where it is often in the form of methylated Hg²⁺ (Gilmour et al., 1992; Engstrom, 2007). There are three operationally defined

forms of Hg in the atmosphere, including gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate mercury (PBM) (Schroeder and Munthe, 1998). GEM is relative stable with an atmospheric residence time of 0.5–2 years because of its low reactivity and limited water solubility (Schroeder and Munthe, 1998). Vegetation uptake, dry deposition, and homogeneous/heterogeneous oxidation reactions are considered as significant sinks of GEM in the atmosphere (Schroeder and Munthe, 1998; Ericksen et al., 2003; Lin et al., 2006, 2007; Gustin, 2011). GOM consists of HgO, HgS, HgCl₂, HgBr₂, HgSO₄, Hg(OH)₂, and Hg(NO₃)₂ (Seigneur et al., 1994; Lindberg and Stratton, 1998; Schroeder and Munthe, 1998; Feng et al., 2004). GOM and PBM can be rapidly

* Corresponding author. Tel.: +1 315 268 3861; fax: +1 315 268 4410.

E-mail address: hopkepk@clarkson.edu (P.K. Hopke).

¹ Present address: Department of Natural Resources and Environmental Sciences, University of Nevada, 1664 N. Virginia Street, Reno, NV, USA.

removed by precipitation and dry deposition. Their atmospheric residence times range from days to weeks (Schroeder and Munthe, 1998; Zhang et al., 2009; Gustin, 2011).

Hg is emitted from both natural and anthropogenic sources. The combustion of coal in coal-fired power plants (CFPPs) represents the most important anthropogenic source of mercury released to the global atmosphere annually accounting for about 810 Mg year⁻¹, with an important contribution from Asian countries (nearly 50% of the total) (Pirrone et al., 2010). Source profiles including Hg forms, which are necessary to better understand Hg sources are not well documented, in-part because of limited long-term Hg measurements (Keeler et al., 2006; Lynam and Keeler, 2006; Liu et al., 2007; Huang et al., 2010; Gratz and Keeler, 2011). Previously Positive Matrix Factorization (PMF), Unmix, Quantitative Transport Bias Analysis (QTBA) have been used to identify Hg-related factors that influence Hg wet deposition (Keeler et al., 2006; Gratz and Keeler, 2011). Principle Component Analysis (PCA) also has been applied to explore the variations of atmospheric Hg concentration (Lynam and Keeler, 2006; Liu et al., 2007; Huang et al., 2010). These studies all reported an important coal combustion source that is related to S (or SO₂) and Hg. Results from these studies suggest that coal combustion is a common and significant Hg emission source in the Eastern US.

Between January and April 2008, a CFPP located in Rochester, New York was gradually shut down (Wang et al., 2011a). In this study, the impacts of the shutdown of the CFPP on local ambient Hg concentrations were quantified as a part of an extensive project evaluating Rochester air quality using a 2-years data record (Huang et al., 2010; Wang et al., 2011b).

2. Experimental

2.1. Sampling site

Measurements were performed in Rochester, New York, from December 2007 to November 2009. According to 2009 estimates, the population in Rochester is 207 294 (US Census Bureau, <http://www.census.gov/>), making it the third largest city in New York State. The New York State Department of Environmental Conservation (NYS DEC) maintains a monitoring site in Rochester, NY

(43°08'46"N, 77°32'54"W, Elevation = 137 m, US EPA site code 36-055-1007). This site is northeast and adjacent to the intersection (~300 m to the south of the site) of two major highways (I-490 and I-590). The 2005–2009 average annual average daily traffic (AADT) counts were 112 291 and 112 549 on I-490 and I-590, respectively. The percentages of buses and heavy-duty diesel vehicles (HDDVs) in the traffic on I-490 and I-590 were 21% and 14%, respectively (Wang et al., 2012).

A coal-fired power plant (estimated emissions ~42 kg-Hg year⁻¹) (US EPA, 2010) was located on the shores of Lake Ontario, ~12 km north of Rochester downtown area. This CFPP, built in 1948, was one of the state's oldest power plants. It included four coal-fueled power generating units with a total capacity of 260 MWe. A coal-fired steam generator plant (estimated emissions <5 kg-Hg year⁻¹) (US EPA, 2010) at Kodak Park is located in north-central Rochester. Its production and emissions substantially decreased during recent years. Huang et al. (2011) and Wang et al. (2011b,c) reported Hg emissions from winter residential wood combustion, which was mainly located to the south of the monitoring site. Fig. 1 shows the locations of the major roadways, the CFPP, the Kodak plant, and the monitoring site in Rochester, NY. The prevailing wind direction was southwesterly. A detailed description of the local meteorology was given by Wang et al. (2011c).

2.2. Instrumentation and data description

A Tekran speciation system (1130/1135/2537A, Tekran Instruments Corp., Ontario, Canada) was used for Hg measurements. An impactor was utilized in the inlet to remove particle larger than 2.5 μm. GOM and PBM were collected on a KCl-coated denuder followed by a quartz filter, respectively, in 2-h sampling intervals at 10 L min⁻¹ and sequentially desorbed (1-h). GEM concentrations were recorded every 5 min during each sampling period. In the desorption cycle, GOM and PBM were desorbed from their collection surface and converted to GEM, and qualified by the analyzer. A detailed description of this system was presented by Landis et al. (2002) and Choi et al. (2008), and the standard operation procedure of the Atmospheric Mercury Network (<http://www.nadp.sws.uiuc.edu/amn/>). The detection limit of GOM and PBM (3σ of flush blanks) were 0.46 pg m⁻³ and 1.10 pg m⁻³,



Fig. 1. Map of the locations of the sampling site, the major roadways, and the Hg emission sources in Rochester, New York.

Download English Version:

<https://daneshyari.com/en/article/6310397>

Download Persian Version:

<https://daneshyari.com/article/6310397>

[Daneshyari.com](https://daneshyari.com)