Atmospheric Pollution Research 4 (2013) 168-180

Atmospheric Pollution Research

www.atmospolres.com

Gaseous oxidized mercury dry deposition measurements in the Four Corners area and Eastern Oklahoma, U.S.A.

Mark E. Sather [1], Shaibal Mukerjee [2], Luther Smith [3], Johnson Mathew [4], Clarence Jackson [4], Ryan Callison [5], Larry Scrapper [5], April Hathcoat [5], Jacque Adam [5], Danielle Keese [5], Philip Ketcher [5], Robert Brunette [6], Jason Karlstrom [6], Gerard Van der Jagt [6]

[1] *Air Quality Analysis Section, U.S. Environmental Protection Agency (EPA) Region 6, 1445 Ross Avenue, Dallas, Texas 75202, USA*
[2] *National Exposure Research Laboratory, U.S. EPA (E205–03), Research Triangle Park, North Carolina 27711, USA*
[3] *Alion Science and Technology, Inc., POB 12313, Research Triangle Park, North Carolina 27709, USA*
[4] *Houston Laboratory, U.S. EPA Region 6, 10625 Fallstone Road, Houston, Texas 77099, USA*
[5] *Cherokee Nation Environmental Programs, 208 E. Allen Road, Tahlequah, Oklahoma 74464, USA*
[6] *Frontier Global Sciences, 11720 North Creek Parkway North, Suite 400, Bothell, Washington 98011, USA*

ABSTRACT

Gaseous oxidized mercury (GOM) dry deposition measurements using surrogate surface passive samplers were collected in the Four Corners area and eastern Oklahoma from August, 2009–August, 2011. Using data from a six site area network, a characterization of the magnitude and spatial extent of ambient mercury pollution in the arid Four Corners area was accomplished, which included the observation of a strong regional signature in the GOM dry deposition data set. GOM dry deposition rate estimates ranged from 0.4–1.0 ng/m^2 h at the six Four Corners area monitoring sites, while the GOM dry deposition rate estimate at the eastern Oklahoma monitoring site was lower at 0.2 ng/m^2 h. The highest GOM dry deposition estimates were recorded during the spring and summer while the lowest GOM dry deposition estimates were recorded during the fall and winter. During the second year of this study the highest annual GOM dry deposition estimate so far measured in the United States (U.S.) with smooth–edge surrogate surface passive samplers, 10 889 ng/m^2, was recorded at the Mesa Verde National Park site, a site at which the two–year cumulative GOM dry deposition estimate exceeded the mercury wet deposition estimate. GOM dry deposition estimates during the second year of the study were statistically significantly higher than the first year of the study at six of the seven sites. The data from this study provide a two–year baseline of GOM dry deposition data in the Four Corners area and eastern Oklahoma immediately before the current implementation of new U.S. power plant and boiler mercury control regulations which will significantly reduce mercury emissions from those two sectors of local and regional anthropogenic mercury emission sources.

Keywords: *Air pollution, arid area, surrogate surface passive sampling*

Corresponding Author:
Mark E. Sather
☎ : *+1-214-665-8353*
🖷 : *+1-214-665-6762*
✉ : *sather.mark@epa.gov*

Article History:
Received: 04 December 2012
Revised: 21 February 2013
Accepted: 22 February 2013

doi: 10.5094/APR.2013.017

1. Introduction

Ambient mercury pollution is a global concern (Slemr et al., 2003; Kim et al., 2005; Lindberg et al., 2007) and deposits to the earth in wet and dry processes. Wet deposition mercury measurements and core mercury measurements such as from ice and sediments have been collected for many years in North America (Schuster et al., 2002; Mast et al., 2005; Prestbo and Gay, 2009; Mast et al., 2010; National Atmospheric Deposition Program, 2011a), but there is a current dearth of dry deposition mercury measurements.

GOM consists of multiple oxidized mercury compounds such as $HgCl_2$ and $HgBr_2$ (Gustin and Jaffe, 2010). GOM has a short atmospheric lifetime and in part is associated with local/regional mercury emission sources (Schroeder and Munthe, 1998; Skov et al., 2007), such as from coal–fired power plants, boilers, and cities. It should be noted though that some of the GOM released from coal–fired power plant and other boiler plumes may also undergo reduction in the atmosphere downwind from the release points (EPRI, 2006). GOM is also formed through oxidation reactions of gaseous elemental mercury, especially in warmer seasons with higher photochemical activity (Lin et al., 2012). A significant amount of total dry deposition of mercury consists of GOM (Lin et al., 2012), and GOM, along with particle bound mercury, deposits faster to water, soils, and vegetation and is more water soluble than gaseous elemental mercury (Zhang et al., 2009). Surrogate surface sampling of mercury dry deposition, including GOM dry deposition, has been recently evaluated (Lyman et al., 2009; Huang et al., 2011; Lai et al., 2011), and also used to better understand spatial distributions of ambient mercury dry deposition (Huang et al., 2012). This paper discusses estimates of total mercury deposition in terms of mercury wet deposition estimates plus GOM dry deposition estimates only; gaseous elemental mercury dry deposition estimates or particle bound mercury dry deposition estimates are not included here. Thus, total mercury deposition estimates discussed in this paper should be viewed as conservative estimates (i.e. probably underestimating the "true" totals). There have only been a few published GOM dry deposition measurement studies of extended length in the U.S. (Caldwell et al., 2006; Lyman et al., 2007; Lyman et al., 2009; Castro et al., 2012; Huang et al., 2012; Peterson et al., 2012), and none outside of the U.S. This paper provides new information on GOM dry deposition estimates in two new areas in the south central U.S.

The Four Corners area of the U.S. consists of the region where four U.S. States (New Mexico, Colorado, Utah, and Arizona) come together (Figure 1, Inset 1). Two of the largest coal–fired power plants in the U.S. are located in the Four Corners area, and coal–fired power plants are the largest anthropogenic mercury emission source in the U.S., contributing about 50% of all stationary source mercury emissions (U.S. EPA, 2011a). Measurements from the Mercury Deposition Network (MDN) of the National Atmospheric Deposition Program (NADP) for 2010 showed elevated wet mercury deposition levels at Mesa Verde National Park (National Atmospheric Deposition Program, 2011a), and given the arid nature of this region, dry mercury deposition has been hypothesized to be a significant portion of the total mercury deposition in the Four Corners area (Lyman et al., 2007; Mountain Studies Institute, 2010; Huang and Gustin, 2012). Thus, a two year study to collect dry deposition mercury measurements in the Four Corners area was begun in August, 2009, to test the hypothesis that significant amounts of total mercury deposition in the Four Corners area are from dry atmospheric processes. Multiple air monitoring sites were established to study the spatial distribution of dry mercury deposition. This was the first attempt at such an effort in this area. An additional objective of the study was to estimate a dry mercury deposition baseline before mercury emission controls are implemented due to the U.S. 2011 Boiler/Incinerator (U.S. EPA, 2011b) and U.S. 2012 Power Plant (U.S. EPA, 2012) mercury rules. Only one of the power plants considered here (San Juan; see Figure S1 in the Supporting Material, SM) had installed mercury emission controls before the latter rule was issued. Both rules require affected industrial sources to install mercury emission controls during the 2012–2016 time–frame.

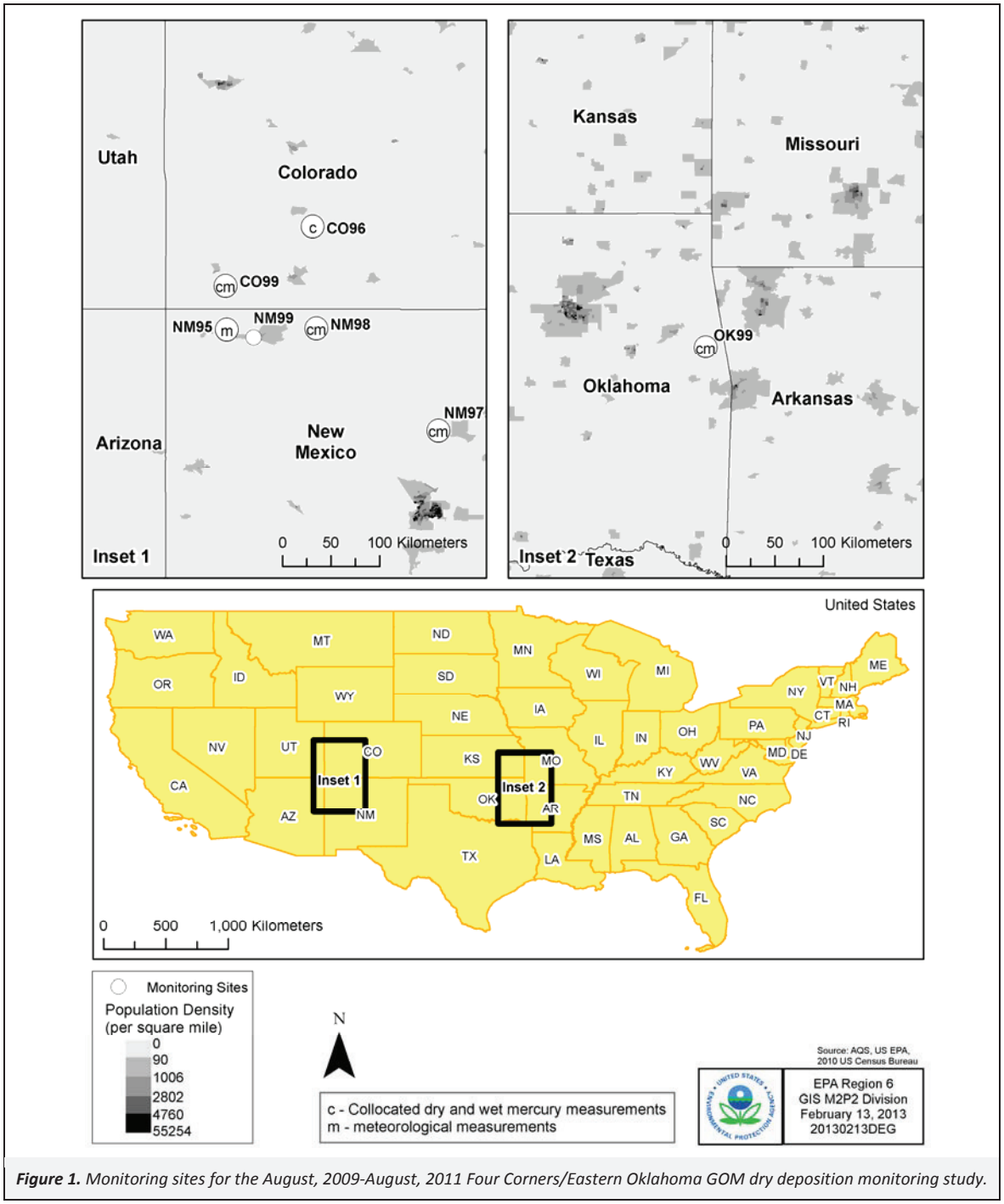

Figure 1. *Monitoring sites for the August, 2009-August, 2011 Four Corners/Eastern Oklahoma GOM dry deposition monitoring study.*