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Mercury species measured atop the Moody Tower TRAMP site, Houston, Texas

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ABSTRACT

Atmospheric mercury speciation was monitored within Houston, Texas, USA, August 6–October 14, 2006 as part of the TexAQS Radical and Aerosol Measurement Program (TRAMP). On average, all mercury levels were significantly elevated compared to a rural Gulf of Mexico coastal site. Concentrations varied from very clean to very dirty. Multi-day periods of stagnant or low-wind conditions brought elevated concentrations of all mercury species, whereas multi-day periods of strong winds, particularly southerly winds off the Gulf of Mexico, brought very low values of mercury species. Over the entire mercury measurement period, the daily averages of mercury species showed distinct and consistent relationships with the average planetary boundary layer dynamics, with gaseous elemental and particulate-bound mercury near-surface concentrations enhanced by a shallow nocturnal boundary layer, and reactive gaseous mercury concentration enhanced by midday convective boundary layer air entrainment transporting air aloft to the surface. Mercury concentrations were not significantly correlated with known products of combustion, likely indicating non-combustion mercury sources from the Houston area petrochemical complexes. On the morning of August 31, 2006 an observed emission event at a refinery complex on the Houston Ship Channel resulted in extremely high concentrations of aerosol mass and particulate-bound mercury at the TRAMP measurement site 20 km downwind.

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1. Introduction

The intensive measurement period of the TexAQS Radical and Aerosol Measurement Program (TRAMP) field study began August 14, 2006 and ended September 30, 2006. The study was conducted in Houston, Texas, USA and was part of the larger TexAQS 2006 intensive air-monitoring campaign. The purpose of the TRAMP program was to sample meteorological and pollutant concentration data, including secondary species such as ozone, to assist the Texas Commission on Environmental Quality (TCEQ) efforts to formulate a Strategic Implementation Plan (SIP) for meeting air quality goals.

Mercury (Hg) data were collected August 6, 2006–October 14, 2006, which included the nominal intensive measurement period. Similar to other measurements, the mercury data sets were collected by a suite of sensors on short towers and climate-controlled shelters on the rooftop of the Moody Tower dormitory building, 70 m above ground level, and within the University of Houston (UH) main campus. While the site is obviously highly urban, it is 2–4 km away from highways and industrial sources, and elevated enough above ground level (70 m) that we consider this to be an "urban background" site.

Mercury species were included to provide a more comprehensive research initiative to better understand the causes of all air pollutants, and their potential interactions in the Houston area. In particular, mercury speciation measurements were conducted to characterize local sources and possible heterogeneous reactions with sea salt aerosols. The study objectives, site selection, participating groups, measurement periods, and all measurements conducted are summarized in the TRAMP overview paper (Lefer and Rappenglück, in this issue)

2. Mercury species

During TRAMP, mercury species were continuously monitored atop the Moody Tower. The mercury speciation included gaseous elemental mercury (GEM, Hg(0)), reactive gaseous mercury (RGM, Hg(II,g)), and fine (PM2.5) particulate-bound mercury (FPM, Hg(II,p)), respectively. Each type of Hg has distinct properties:

- GEM has a long lifetime in the troposphere (6–12 months), has a northern hemisphere background level of ~1.5 ng m^{-3} (90% of the measurement period average), and has the characteristic of long-range transport making it difficult to distinguish local

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from distance sources. GEM is relatively insoluble, therefore is not wet deposited, and near-surface atmospheric concentrations are unaffected by rainfall or surface dew (condensation) events. GEM comprises ~97% of the total atmospheric mercury in the troposphere (e.g. Slemr et al., 2006) and has many natural and anthropogenic sources (volcanoes, enriched soils, coal combustion, biomass burning, etc.). It has been found that for combustion processes, like biomass burning, GEM usually correlated well with CO (e.g. Ebinghaus et al., 2007 and references therein).

- RGM is operationally defined as mercury collected by a KCl coated denuder tube. RGM is typically believed to be dominated by Hg(II) such as $HgCl_2$, HgClX and HgBrX. RGM is typically rare in the lower troposphere 1–2 pg m^{-3} (sub-parts per trillion levels) comprising <1% of total atmospheric mercury (Lindberg and Stratton, 1998). RGM has a high dry deposition rate and is rapidly removed from near-surface air (lifetime in the near-surface air is typically just 1–3 h; Skov et al., 2006). Near-surface RGM concentrations are generally highest when RGM is being actively mixed from aloft to ground level during afternoon boundary layer convection. RGM is also extremely water soluble and is readily removed from the lower troposphere during rain events. In the absence of rain, morning dew on vegetation also has been shown to remove RGM from the very-near-surface air (Malcolm and Keeler, 2002). This effect is absent from the TRAMP data where the measurement height was 70 m above ground level. With the exception of active volcanoes, RGM has negligible natural surface sources and is primarily emitted by coal combustion, waste incineration, cement manufacturing, and industrial processes. RGM can also be produced in-situ by the atmospheric oxidation of gaseous elemental mercury (Lindberg et al., 2002; Swartzendruber et al., 2006). RGM has the potential to convert to FPM in the presence of sea salts and other aerosols due to the high affinity of RGM and NaCl.
- FPM is comprised of oxidized mercury bound to fine (<PM2.5) particles. FPM has a low, but significant, dry deposition rate and, in the absence of rain, a significant lifetime in the near-surface air (2–5 days; Keeler et al., 1995). FPM from local sources concentrates in the near-surface air until it becomes convected out of the region, mixed (diluted) with air entrainment into a growing boundary layer, or, being water soluble, rained-out. FPM is the least studied and least measured form of atmospheric mercury. FPM is typically rare (~1–5 pg m^{-3}; Lu and Schroeder, 1999) in the near-surface air, but more common near the tropopause where rainout is minimal (Talbot et al., 2007). Potential mercury cycling between RGM and FPM has not been fully studied.

3. Sources and regulations

Mercury is released through coal burning, waste incineration, cement production, and, industrial and metallurgical processes. Nationwide, stationary fossil fuel combustion accounts for the majority of anthropogenic emissions (~66%) with coal-fired power plants the largest US source. Each individual source emits a characteristic distribution of GEM, RGM and particulate-bound mercury (PM). Coal-fired power plants, for example, are believed to emit on average 55% GEM, 42% RGM, and 3% PM. These percentages are altered by combustion temperatures, other contaminants in the coal, and, the use of wet and dry flue cleaners, and other emissions' controls. Estimates of total US anthropogenic atmospheric mercury emissions for 1999 are shown in Fig. 1. The percentages listed in Table 1 of local Houston sources are general percentages for that emission type and may not accurately reflect the actual individual source emissions.

Fig. 2 shows the geographical distribution of major Hg sources in the US and Canada. The highest density of mercury sources is found in the Eastern part of the US. Large emission sources are also located in East Texas, mostly associated with coal-fired electricity generation. In the greater Houston area other sources contribute, among them waste incineration and petrochemical processes. Major Houston point sources, from the US Environmental Protection Agency (EPA) National Emissions Inventory 2002, are shown in Fig. 3.

3.1. Regulation of mercury emissions

In 2005 EPA issued the Clean Air Mercury Rule (CAMR) to permanently cap and reduce atmospheric mercury emissions solely from coal-fired power plants. This rule forced limits on

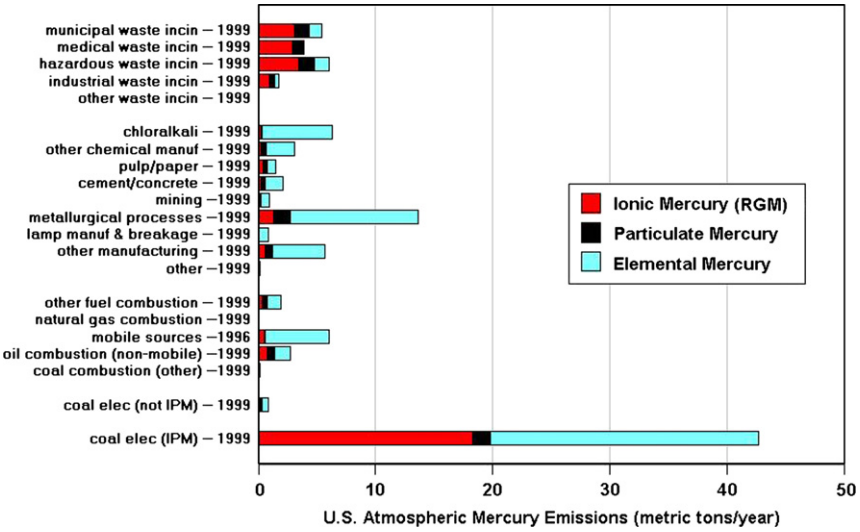

Fig. 1. Estimated 1999 US Atmospheric Anthropogenic Mercury Emissions and typical emission source strengths for the Hg species (USEPA National Emission Inventory 1999). Most sources emit significant amounts of gaseous elemental Hg. Only waste incineration emits mostly reactive gaseous Hg and particulate-bound Hg. The largest emissions of RGM and GEM are associated with coal-fired electricity generation.

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