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Atmospheric mercury emissions in Australia from anthropogenic, natural and recycled sources

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HIGHLIGHTS

- The first spatially resolved inventory of mercury emissions from Australia.
- The best estimate of anthropogenic emissions of mercury in 2006 was 15 ± 5 Mg.
- Natural and re-emitted sources comfortably exceed the anthropogenic emission.
- Emissions were ~8, 140 and 42 Mg pa from vegetation, soils and fires respectively.

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ABSTRACT

The United Nations Environment Programme (UNEP) has begun a process of developing a legally binding instrument to manage emissions of mercury from anthropogenic sources. The UNEP Governing Council has concluded that there is sufficient evidence of significant global adverse impacts from mercury to warrant further international action; and that national, regional and global actions should be initiated as soon as possible to identify populations at risk and to reduce human generated releases. This paper describes the development of, and presents results from, a comprehensive, spatially and temporally resolved inventory of atmospheric mercury emissions from the Australian landmass. Results indicate that the best estimate of total anthropogenic emissions of mercury to the atmosphere in 2006 was 15 ± 5 tonnes. Three industrial sectors contribute substantially to Australian anthropogenic emissions: gold smelting (~50%, essentially from a single site/operation), coal combustion in power plants (~15%) and alumina production from bauxite (~12%). A diverse range of other sectors contribute smaller proportions of the emitted mercury, but industrial emissions account for around 90% of total anthropogenic mercury emissions. The other sectors include other industrial sources (mining, smelting, and cement production) and the use of products containing mercury. It is difficult to determine historical trends in mercury emissions given the large uncertainties in the data. Estimates for natural and re-emitted emissions from soil, water, vegetation and fires are made using meteorological models, satellite observations of land cover and soil and vegetation type, fuel loading, fire scars and emission factors which account for the effects of temperature, insolation and other environmental variables. These natural and re-emitted sources comfortably exceed the anthropogenic emissions, and comprise 4–12 tonnes per year from vegetation, 70–210 tonnes per year from soils, and 21–63 tonnes per year from fires.

1. Introduction

Mercury is a naturally occurring metal found in small quantities throughout the environment in both the atmosphere and in aquatic and terrestrial ecosystems. While it is continuously released, transported, transformed and stored in and between these compartments, the atmosphere is considered to be the dominant transport medium of mercury in the environment (Fitzgerald et al., 1991; Lindquist et al., 1991; Pirrone and Mahaffey, 2005; Pirrone and Mason, 2008).

Previous monitoring and modelling studies, and a number of reviews which have summarised published data concerning the

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long-range atmospheric transportation of mercury from industrial areas, have concluded that there is scientific evidence of a linkage between anthropogenic mercury emissions and elevated mercury concentrations in remote areas (Lindberg et al., 2007; Pirrone and Mason, 2008). Measurements of mercury concentrations in ambient air support the conclusion that mercury deposited in remote areas may originate from anthropogenic sources far away. Thus, mercury becomes a global problem not only affecting local areas that are heavily industrialised, but also areas that are remote from emitting sources. In the view of some researchers, release of any mercury from anthropogenic sources which will lead to increases in the global pool, should be avoided since there is already evidence for significant impacts (Meili et al., 2003).

The environmental and human health impacts of mercury are a consequence of its ability to bio-concentrate in the food chain, and of its long persistence in environmental media. Exposure to mercury has deleterious impacts on human health, particularly on the developing nervous system (Pirrone and Mahaffey, 2005; Mergler et al., 2007) and on wildlife (Scheulhammer et al., 2007). There is substantial evidence that reductions in mercury exposure would bring significant socio-economic benefits to many people (Swain et al., 2007; Pacyna et al., 2010b; Sundseth et al., 2010).

In 2009 the UN Environment Programme's (UNEP) Governing Council agreed on a plan for a global approach to reduce population and ecosystem exposure to mercury. This landmark decision, taken by over 140 countries, sets the stage for the development of an international legally-binding instrument to reduce exposures and world-wide emissions and discharges of this pollutant.

In order to develop the UNEP instrument an accurate knowledge of the sources, transport and fate of mercury is essential. Recent, revised estimates (UNEP Chemicals Branch, 2008; Munthe et al., 2010; Pacyna et al., 2010a; Pirrone et al., 2010) of global anthropogenic emissions of mercury have been made. According to the report produced to support development of the UNEP legally-binding instrument (Munthe et al., 2010) these estimates reveal that:

- Total anthropogenic emissions were 1930 (range 1230–2890) tonnes in 2005.
- The largest sectoral source is the combustion of fossil fuels, largely coal. This sector accounts for a total of ~46% of emissions of mercury to atmosphere, about 25% from electrical power plants and 20% from industrial and residential heating.
- Emissions from gold production arises from both large scale industrial production (~6% of total global emissions) and from small scale and artisanal gold mining and production (~18%, and largely in developing countries).
- The mining, smelting and production of metals other than gold, and cement production each account for ~10% of global emissions.

Pirrone et al. (2010) reach similar conclusions, estimate total anthropogenic emissions of 2320 tonnes for 2008, and identify coal and oil combustion (35% of total anthropogenic emissions), artisanal gold production (17%), non-ferrous metal production (13%), cement production (10%) and waste disposal (8%) as the major anthropogenic sources.

The emission estimates are subject to large uncertainties, not only in the total amounts of mercury emitted, but also in its speciation between elemental and oxidised forms, and in partitioning between gas and particulate phases. Atmospheric chemistry, speciation and partitioning are of great importance as they control atmospheric and deposition processes, and hence the fate of mercury in the environment.

It is also recognized that there are very significant sources of emission of mercury from natural sources, such as volcanoes (Bagnato et al., 2010) and rocks (Pirrone and Mason, 2008; Pirrone et al., 2010), but similar, if not greater, uncertainty characterise these emissions. It is also clear that mercury emitted from any source and deposited in environmental media such as water, soil, and vegetation may be re-emitted due to the volatility of mercury. Mercury will also be released during wild fire events.

Recent estimates (Pirrone et al., 2010) of emissions from these natural and re-emitted sources suggest that they currently exceed anthropogenic sources by a factor of 2–3, amount to more than 5000 tonnes annually, and that the major sources are the oceans (52% of total non-anthropogenic emissions); vegetation and soil in a variety of environments (28%), and biomass burning (13%).

Development of an effective UN convention for mercury will clearly require accurate estimations of atmospheric emissions from anthropogenic sources; improved understanding of the partitioning, speciation and chemical transformation of mercury emissions; and ideally, estimates of mercury emissions from natural and re-emitted sources to provide a context for, and to establish effective management strategies and control measures for all emissions. In this paper we report estimates of Australian emissions for the year 2006 from anthropogenic sources; from natural sources and re-emitted sources of previously deposited mercury (biomass burning, soil, water bodies and vegetation); and the relative contributions of the different chemical forms of mercury (i.e. elemental, oxidised and particulate) in the sources. We have previously reported emissions from stationary sources (Nelson, 2007), but here extend that work to a complete inventory of emissions from Australian sources. Particular attention is given to the uncertainties in these estimates, and to the relative contributions of new anthropogenic emissions and those from natural/re-emitted sources.

2. Methodology

2.1. Anthropogenic emissions

The derivation of an inventory of Australian emissions of mercury from anthropogenic sources in 2006 was undertaken using a range of data sources. These included the National Pollutant Inventory (NPI, see www.npi.gov.au), and overseas protocols and emission factors (eg, those included in the *UNEP Toolkit for identification and quantification of mercury releases*, (UNEP, 2005)).

National Pollution Inventory (NPI) data was used where available and consistent with data reported elsewhere. Reporting of point source emissions in Australia has been mandated since 1998, under the National Pollutant Inventory (NPI) National Environment Protection Measure (NEPM). The NPI includes data on emissions of 93 substances to air, land and water. The NPI also includes estimation of some area sources, known as aggregated emission data. In 2007 the NPI NEPM was varied and the threshold for reporting mercury was reduced from 10 tonnes to 5 kg. This reduced threshold is mandatory for the 2007–08 NPI reporting year. Further details of the NPI program are given on the NPI website, www.npi.gov.au (DEH, 2006). As described above, in this study emission estimates were made for 2006. Examination of more recent NPI data (i.e. after the threshold variation discussed above) did not show any major changes in industrial source contributions.

Time series NPI data for the top 90% of reported mercury emissions were examined to determine data consistency in the period 2001–2007. If consistent, NPI data were used and are not further discussed here. However, for some potential sources, and most notably area and diffuse sources, NPI data are either not available, or were of inconsistent quality; these sources typically