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Historical releases of mercury to air, land, and water from coal combustion☆

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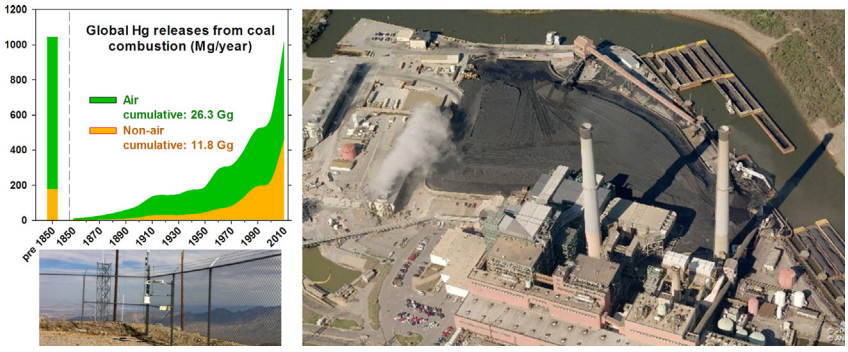
HIGHLIGHTS

- Total Hg released to the environment from coal combustion is estimated to be 38 Gg.
- 71% of this Hg was released into the atmosphere, while 31% went to land and water.
- Most of the Hg from coal combustion was released in Asia and Europe (32% each).
- The fraction of Hg released to the air as elemental Hg has steadily increased.
- In the year 2010 about 1 Gg of Hg was released worldwide to all media.

GRAPHICAL ABSTRACT



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ABSTRACT

Coal combustion is one of the largest contemporary sources of anthropogenic mercury (Hg). It releases geologically sequestered Hg to the atmosphere, and fly ash can contaminate terrestrial and aquatic systems. We estimate that coal combustion has released a cumulative total of 38.0 (14.8–98.9, 80% C.I.) Gg (gigagrams, 10^9 g or thousand tonnes) of Hg to air, land, and water up to the year 2010, most of which (97%) has occurred since 1850. The rate of release has grown by two orders of magnitude from 0.01 Gg yr^{-1} in 1850 to 1 Gg yr^{-1} in 2010. Geographically, Asia and Europe each account for 32% of cumulative releases and an additional 18% is from North America. About 26.3 (10.2–68.3) Gg, 71% of the total, were directly emitted to the atmosphere, mostly from the industrial (45%) and power generation (36%) sectors, while the remainder was disposed of to land and water bodies. While Europe and North America were the major contributing regions until 1950, Asia has surpassed both in recent decades. By 2010, Asia was responsible for 69% of the total releases of Hg from coal combustion to the environment. Control technologies installed on major emitting sources capture mainly particulate and divalent Hg, and therefore the fraction of elemental Hg in emissions from coal combustion has increased over time from 0.46 in 1850 to 0.61 in 2010. About 11.8 (4.6–30.6) Gg of Hg, 31% of the total, have been transferred to

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land and water bodies through the disposal or utilization of Hg-containing combustion waste and collected fly ash/FGD waste; approximately 8.8 Gg of this Hg have simply been discarded to waste piles or ash ponds or rivers.

1. Introduction

Mercury (Hg) contamination of the environment is a well-recognized global problem that poses risks to humans and wildlife (Mahaffey et al., 2011; Lavoie et al., 2013). The issue has been summarized in the Global Mercury Assessment (GMA) (UNEP, 2013) and accompanying documentation (AMAP/UNEP, 2013). Most emission estimates focus on a single year, on direct releases to the atmosphere, and on the Hg remaining at contaminated sites (e.g., UNEP, 2013). However, this is only a snapshot of the problem, because a large fraction of the water-soluble divalent Hg (Hg^{II}) deposited to terrestrial ecosystems and the ocean is reduced to gaseous elemental Hg (Hg^0) and evaded back into the atmosphere, prolonging its lifetime in the biosphere (Amos et al., 2013, 2015). This study focuses on one important component of the Hg picture, coal combustion, and its cumulative releases into the environment.

Some of the Hg liberated by coal combustion is emitted directly into the atmosphere during the combustion process, while the remainder is released to the land or nearby water bodies. The atmospheric lifetime of Hg is ~6 months (Corbitt et al., 2011; Horowitz et al., 2017), which allows for atmospheric transport on the hemispheric to global scale. Hg discarded in waste products to land and ponds can subsequently be transported into rivers and find its way into lakes and oceans (Amos et al., 2014; Fisher et al., 2012). Atmospherically deposited Hg cycles through the surface environment in oceans and soils and can be re-emitted to the atmosphere via reduction back to elemental Hg. Considerable mobilization and re-distribution of Hg therefore occurs over time. Eventually, Hg is buried in estuarine or ocean sediments or stable terrestrial reservoirs, but the time scale for this sequestration can be as long as millennia (Amos et al., 2013; Selin et al., 2008). Thus, Hg accumulates in the global environment, redistributing itself spatially and among different media, with continuous augmentation from human activities. Because of this persistence in the environment, it is important to know how much anthropogenic Hg has been liberated in total and what has been its fate. Single-year estimates and air-only estimates are insufficient to support biogeochemical modeling of the complete Hg cycle (Amos et al., 2013).

In this study, the cumulative amount of Hg released from coal combustion to all media (air, land, and water) is estimated. Hg is naturally enriched in any type of sulfidic ore and is thus contained as an impurity in many kinds of ores and fossil-fuel deposits. Coal is relatively enriched in Hg compared to other fossil fuels and contains 0.01–1 ppm Hg by mass (Mukherjee et al., 2008). The GMA (UNEP, 2013) reports that coal combustion released 474 Mg of Hg to air in the year 2010, 24% of total air emissions globally (1960 Mg), and was the second largest contributing source type after artisanal and small-scale gold mining (ASGM). Prior assessments for earlier years suggested that coal combustion was the largest anthropogenic source globally (Pirrone et al., 2010).

Because of its major contribution to Hg emissions in modern times, many countries have implemented emissions control technology for reducing emissions from coal combustion (Evers et al., 2016; Giang et al., 2015; Milford and Pienciak, 2009; Sunderland et al., 2016). The reduction and speciation of Hg in various combustion and control systems is an ongoing research topic (Wilcox et al., 2012). Coal combustion in power plants was the largest remaining emissions source of Hg in the U.S. by the mid-2000s (~40%), following reductions in emissions from municipal waste incinerators in the mid-1990s and the phase-out of Hg in many consumer products (Cohen et al., 2004). The U.S. Environmental Protection Agency (U.S. EPA) began developing regulations for Hg emissions from coal-fired power plants in the mid-1990s. This action was prompted by the Agency's finding that it was "appropriate and necessary" to regulate emissions, based on plausible human health risks associated with local exposures described in the Mercury Study Report to Congress (U.S. EPA, 1997). This culminated in the 2011 Mercury and Air Toxics Standard (MATS) that used a Maximum Available Control Technology (MACT) standard to propose reductions of more than 90% of emissions from this sector. Full compliance with this regulation was set for April 2016, although legal action related to this rule is ongoing (Sunderland et al., 2016).

Globally, the Minamata Convention on Mercury, signed by 128 countries in 2013, calls for worldwide control of anthropogenic releases of Hg, in which coal-fired power plants are listed under Article 8 as point sources requiring control (Giang et al., 2015). Both in the U.S. and in other countries, the recognition in the 1980s and 1990s of coal combustion as a major pollution source led to the implementation of a range of control technologies for SO_x and NO_x emissions that have achieved co-benefits for Hg control. In some respects, the rate of implementation of these controls worldwide, especially flue-gas desulfurization (FGD), charts the future pathway for Hg emissions from coal combustion (Streets et al., 2009).

2. Data and methods

This study combines the results of previous work on Hg releases (Streets et al., 2011, 2017; Amos et al., 2013; Horowitz et al., 2014; Wu et al., 2010), extends the dataset to the year 2010, updates the implementation of emission controls, and adds the media that were not calculated previously (land and water). It thus provides a complete quantification of total Hg releases from coal combustion from the beginning of human activity up to the year 2010, characterized by releases to different media, from different source types, and in different world regions. For the period 1850–2010, decadal estimates for 17 world regions are estimated and summarized for seven larger, continental-scale regions (Streets et al., 2011). For the period prior to 1850, single estimates of coal consumption and corresponding Hg emissions are developed and assigned to Europe where the majority of coal was consumed in historical times. This analysis also incorporates updates to the Hg content of coals, coal combustion amounts, and the use of FGD systems.

Coal use around the world and in past and present times is calculated for 70 distinct combinations of fuel type, combustion technology, and emission control technology, covering four sectors: residential (11 types), industrial (32), power generation (24), and transportation (3) (Bond et al., 2004, 2007; Streets et al., 2004). It is important to include older and simpler types of combustion and pollution controls in order to correctly estimate emissions in past times and in developing countries. A complete listing of all fossil-fuel alternatives was provided in Table 3 of Streets et al. (2004), and the 70 coal options are presented here as Appendix Table A.1, with the sole change that coal combustion by power plants has recently been split into those plants with and without FGD systems, in order to reflect the growing use of FGD and its significant uptake of Hg in the flue gas.

This study assumes that FGD is applied only to coal-fired power plants that use pulverized coal units with particulate control (see Appendix Table A.1). This is by far the largest Hg-emitting sub-sector in the energy industry today. Statistics on the fractions of coal-fired capacity that use FGD around the world have been gathered (e.g., Rubin et al., 2004; Smith et al., 2011; Soud, 1994; Srivastava and Jozewicz, 2001). The energy statistics for this category are then divided into two sub-categories for each region: coal combustion without FGD, in which no additional removal of Hg occurs, and coal combustion with FGD, in