science for a changing world

Mercury in U.S. Coal—Abundance, Distribution, and Modes of Occurrence

Introduction

In February 1998, The U.S. Environmental Protection Agency (EPA, 1998a,b) issued a report citing mercury emissions from electric utilities as the largest remaining anthropogenic source of mercury released to the air. EPA officials estimated that about 50 tons of elemental mercury are emitted each year from U.S. coal-burning powerplants, with lesser amounts coming from oil- and gas-burning units. According to EPA estimates, emissions from coal-fired utilities account for 13 to 26 percent of the total (natural plus anthropogenic) airborne emissions of mercury in the United States. On December 14, 2000, the EPA announced that it will require a reduction in mercury emissions from coal-fired powerplants, with regulations proposed by 2003 and final rules for implementation completed by 2004 (EPA, 2000).

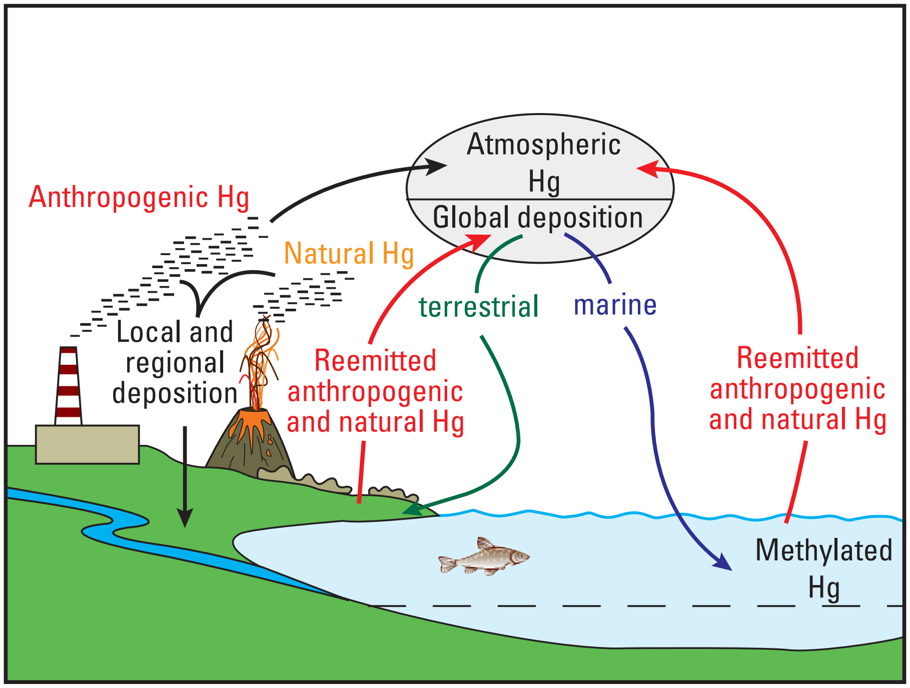

Figure 1. Simplified geochemical cycle of mercury (Hg).

Environmental Significance of Mercury

The mercury (Hg) directly emitted from powerplants generally is not considered harmful; however, in the natural environment, mercury can go through a series of chemical transformations that convert elemental mercury to a highly toxic form that is concentrated in fish and birds (fig. 1). The most toxic form of mercury is methylmercury, an organic form created by a complex bacterial conversion of inorganic mercury. Methylation rates (creation of methylmercury) in ecosystems are a function of mercury availability, bacterial population, nutrient load, acidity and oxidizing conditions, sediment load, and sedimentation rates (National Research Council, 1978).

Methylmercury enters the food chain, particularly in aquatic organisms, and bioaccumulates. Bioaccumulation is the enrichment of a substance in an organism and includes bioconcentration from environmental concentrations and additional uptake via the food chain. Cases of mercury poisoning have been documented in people who eat contaminated fish for prolonged periods, both in the United States and abroad. Pregnant women and subsistence fishermen are particularly vulnerable. Because high levels of mercury have been detected in fish, many U.S. States have issued advisories that restrict fishing.

Reduction in mercury emissions from U.S. coal-fired powerplants may help minimize or avoid health problems caused by exposure to excess mercury. There are several ways in which this reduction can be accomplished. One option to reduce the quantity of mercury in the atmosphere is to use high-rank coals. Generally, moisture in coal decreases and calorific value (thermal energy) increases as coal rank (degree of maturation) increases. Therefore, powerplants that burn high-rank coal in their boilers require less coal for a given thermal output. Thus, for coals having similar mercury concentrations, the higher rank coals will contribute less mercury to the environment. Additional options include selective mining of coal (avoiding parts of a coal bed that are higher in mercury content), coal washing (to reduce the amount of mercury in the coal delivered to the powerplants), switching from coal to natural gas, and postcombustion removal of mercury from the powerplant stack emissions. Information on the abundance, distribution, and forms of mercury in coal may be helpful in selecting the most efficient and cost-effective options for mercury reduction.

Abundance and Distribution of Mercury in Coal

The U.S. Geological Survey (USGS) has compiled a nationwide coal information database over the last 25 years. A subset of the data, called COALQUAL (Bragg and others, 1998) contains analyses of over 7,000 coal samples that have been collected or calculated to represent the entire thickness of a coal bed in the ground.

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Figure 2 is a histogram of the mercury values in the COALQUAL database for conterminous U.S. coal. Statistics for all analyses indicate a mean of 0.17 part per million (ppm), with a median and standard deviation of 0.11 ppm and 0.17, respectively. About 80 percent of the mercury concentrations in the database are less than 0.25 ppm. The maximum mercury database value for coal in the ground is 1.8 ppm, after deleting one higher value as a statistical outlier.

Table 1 shows the median and mean values for mercury concentrations (in ppm) and calorific values (British thermal units per pound (Btu/lb)), as well as the number of analyses, for selected coal-producing regions in the United States, using the COALQUAL database. The mercury data in table 1 have been calculated back to an as-received basis, approximately the mercury concentration of the coal in the ground.

Northern Appalachian area coal has the highest mean and median values for mercury, with coal from the southern Appalachian area having the second highest value and coal from the central Appalachian area slightly lower. Coal from these three areas has extremely high calorific values. Coal from the Uinta region has the lowest mean and median mercury values of all indicated areas. Some western U.S. coals are low in mercury but are also low in calorific value, because they are low in rank.

The concentration of mercury can also be presented on an equal-energy basis (input load) in pounds (lb) per trillion (10^{12}) Btu to provide a convenient unit of comparison between coal from different areas (fig. 3). This is a simple calculation, dividing as-received mercury ppm values by Btu/lb and expressing the value on a 10^{12} Btu basis. The data from COALQUAL used in this analysis yield a mean U.S. input load of 14 lb Hg/10^{12} Btu (with a median of 9.7 and a standard deviation of 15). The calculated input loads from individual samples were used to calculate a mean value for each of the selected coal-producing regions listed in table 1. Mean mercury input loads were divided into arbitrary 5-unit intervals and are color-coded in figure 3. According to the Energy Information Administration (EIA, 2001), U.S. coal production, which can be roughly correlated with usage, is similar between coal regions east and west of the Mississippi River (38 and 48 percent, respectively). About 14 percent of U.S. production comes from coal in the Interior areas.

On the basis of the information shown in figure 3, the Gulf Coast lignite may have the highest potential for mercury emissions, and the Green River coal from western Wyoming may have the lowest mercury emissions on an equal-energy basis. Of the two major bituminous coal-producing regions, samples from the Appalachian region contain higher mercury levels than those from the Eastern Interior. Samples from the Powder River Basin are slightly higher in mercury levels than the subbituminous coals of the San Juan River Basin.

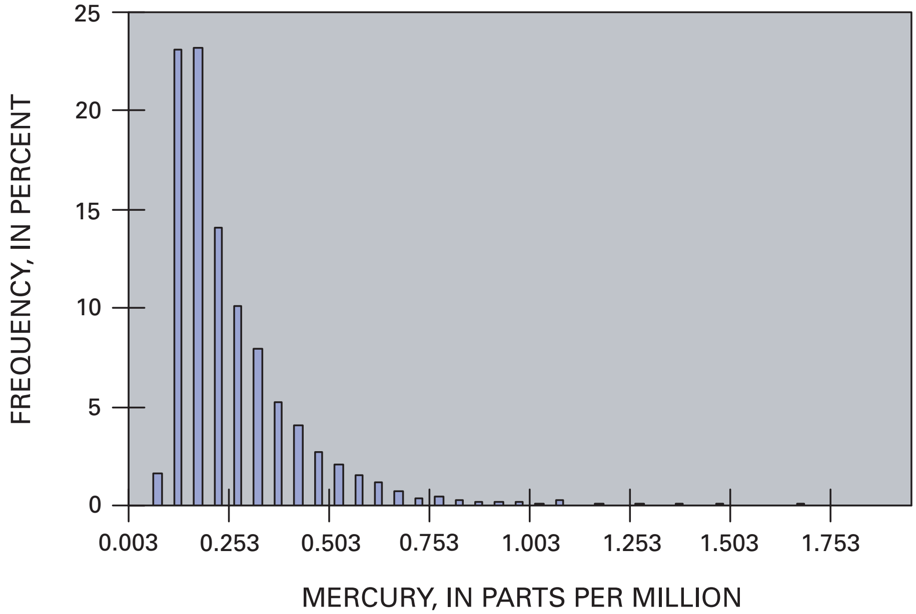

Figure 2. Histogram of mercury concentrations (remnant moisture, whole coal basis) for conterminous U.S. coal from the COALQUAL database.

Modes of Occurrence and Reduction of Mercury

The COALQUAL data set does not take into account the potentially substantial reduction of mercury by physical coal cleaning, because the analyses represent coal as it exists in the ground. The modes of occurrence of an element in coal can affect the way the element behaves during coal cleaning, combustion, and leaching.

Table 1. Median and mean values for mercury concentrations (in parts per million (ppm)) and calorific values (in British thermal units per pound (Btu/lb) on an as-received, whole coal basis for selected coal-producing regions in the United States.

[No. = number of analyses]

Coal-producing region	Mercury (ppm)			Calorific value (Btu/lb)		
	Median	Mean	No.	Median	Mean	No.
Appalachian, northern	0.19	0.24	1,613	12,570	12,440	1,506
Appalachian, central	.10	.15	1,747	13,360	13,210	1,648
Appalachian, southern	.18	.21	975	12,850	12,760	969
Eastern Interior	.07	.10	289	11,510	11,450	255
Fort Union	.08	.10	300	6,280	6,360	277
Green River	.06	.09	388	9,940	9,560	264
Gulf Coast	.13	.16	141	6,440	6,470	110
Pennsylvania Anthracite	.10	.10	51	12,860	12,520	39
Powder River	.06	.08	612	8,050	8,090	489
Raton Mesa	.05	.09	40	12,500	12,300	34
San Juan River	.04	.08	192	9,340	9,610	173
Uinta	.04	.07	253	11,280	10,810	226
Western Interior	.14	.18	286	11,320	11,420	261
Wind River	.08	.15	42	9,580	9,560	42

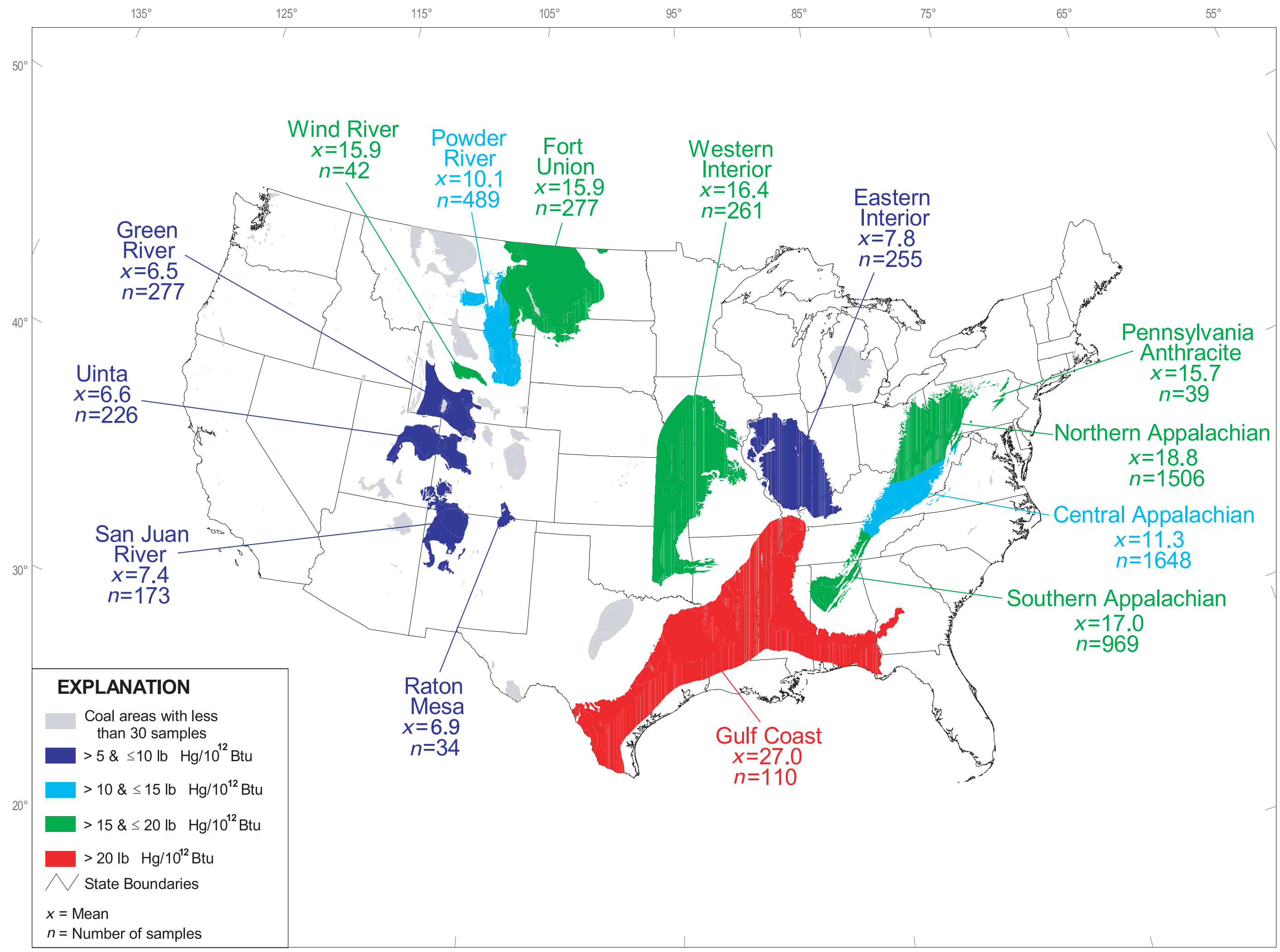

Figure 3. Mercury input loadings (in pounds of mercury per 10^{12} British thermal units (lb Hg/10^{12} Btu)) of in-ground coal for selected U.S. coal-producing regions.

Thus, the element's mode of occurrence has an important influence on its environmental and technological impacts. Because of the low concentrations (commonly less than 0.2 ppm) of mercury and its volatility, it is particularly difficult to determine the modes of mercury occurrence in coal. USGS research indicates that much of the mercury in coal is associated with pyrite, which generally forms after the coal is compacted (fig. 4). Other forms of mercury that have been reported in coal are organically bound, elemental, and in sulfide and selenide minerals (fig. 5).

The U.S. Geological Survey is collaborating on research to determine if the modes of occurrence of mercury in coal influence the formation of mercury species during the combustion process and thus the likelihood of mercury capture from the gas. The USGS has also collaborated with industry on research to assess the removability of mercury from coal by conventional physical coal-cleaning techniques. The results of these studies indicate that, on the average, 37 percent of the mercury is removed by coal cleaning (Toole-O'Neil and others, 1999). The information that the USGS is generating on mercury distribution and modes of occurrence is also relevant to mercury reduction by fuel switching, selective mining, and chemical coal cleaning. Flue gas controls on mercury (sorbent injection and hydrothermal treatment technologies) are also being evaluated by research organizations as possible economic solutions for mercury reduction.

Summary

The concentration of mercury in coal samples from the U.S. Geological Survey's COALQUAL database averages 0.17 ppm for in-ground coal in the conterminous United States. Mean values range from 0.07 ppm for coal samples from the Uinta region to 0.24 ppm for samples from the northern Appalachian coal-producing region. On an equal-energy basis, Gulf Coast coal samples have the highest input load values (27.0 lb Hg/10^{12} Btu), and the Green River region samples have the lowest values (6.5 lb Hg/10^{12} Btu).

The COALQUAL database is an extremely valuable source of information for raw or in-ground trace-element concentrations in U.S. coals and, if adjusted for the effect of coal cleaning in appropriate coals, can provide a first estimate of as-shipped mercury concentration in coal where data are not available. Physical coal cleaning is a viable method of reducing mercury that enters the combustion system and, therefore, reducing mercury that enters the atmosphere. The mean mercury concentration of eastern U.S. coals may be less than reported, if the impact of physical coal cleaning is considered.

—By Susan J. Tewalt, Linda J. Bragg, and Robert B. Finkelman

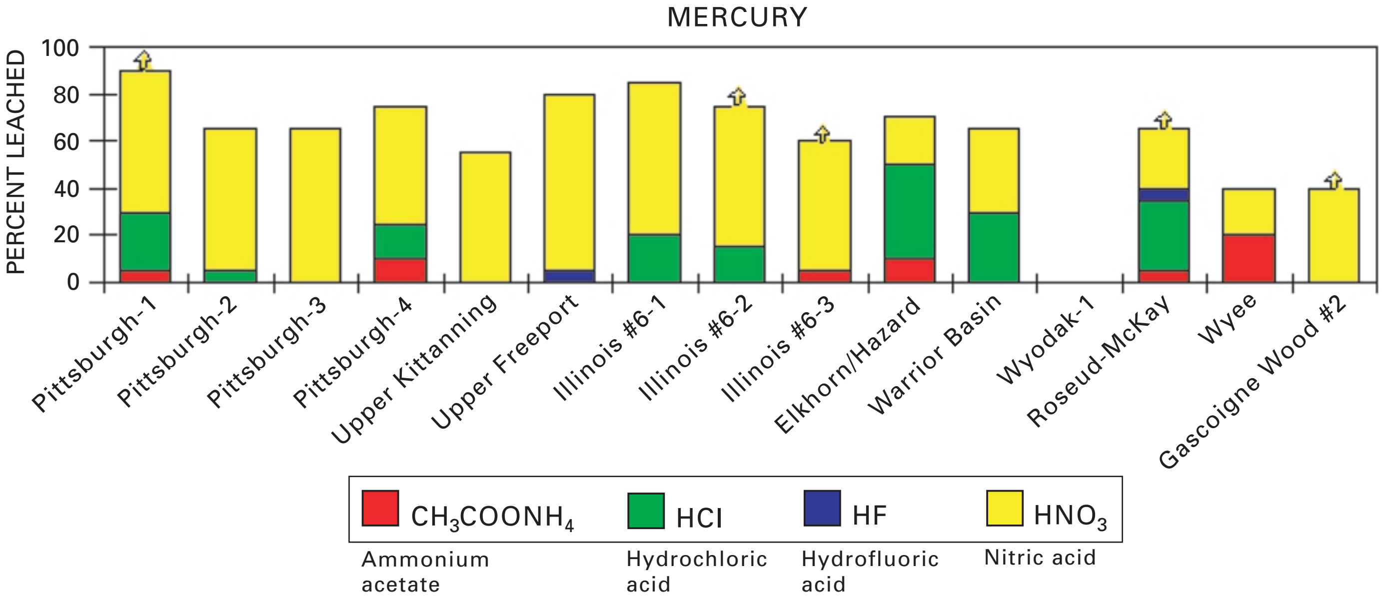

Figure 4. Selective leaching results for 15 coal samples (12 from the United States) (Palmer and others, 1998). The yellow bars indicate the proportion of mercury leached by nitric acid. This mercury is believed to be associated with the sulfide minerals, such as pyrite. Direct analysis of pyrite grains by a laser ablation mass analyzer indicated mercury concentrations consistent with selective leaching data. The green bars indicate the mercury leached by hydrochloric acid; much of this mercury may have come from oxidized pyrite. Arrows indicate minimum values.

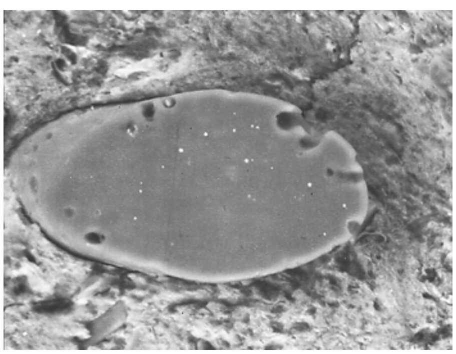

Figure 5. Scanning electron photomicrograph of a polished block of lignite from California. The minute (less than 1 micrometer) bright spots are rare grains of mercury selenide.

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