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# Mercury Emission Estimates from Fires: An Initial Inventory for the United States

CHRISTINE WIEDINMYER\* AND  
HANS FRIEDLI

National Center for Atmospheric Research,  
1850 Table Mesa Drive, Boulder, Colorado 80305

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Recent studies have shown that emissions of mercury (Hg), a hazardous air pollutant, from fires can be significant. However, to date, these emissions have not been well-quantified for the entire United States. Daily emissions of Hg from fires in the lower 48 states of the United States (LOWER48) and in Alaska were estimated for 2002–2006 using a simple fire emissions model. Emission factors of Hg from fires in different ecosystems were compiled from published plume studies and from soil-based assessments. Annual averaged emissions of Hg from fires in the LOWER48 and Alaska were 44 (20–65) metric tons  $\text{yr}^{-1}$ , equivalent to  $\sim 30\%$  of the U.S. EPA 2002 National Emissions Inventory for Hg. Alaska had the highest averaged monthly emissions of all states; however, the emissions have a high temporal variability. Emissions from forests dominate the inventory, suggesting that Hg emissions from agricultural fires are not significant on an annual basis. The uncertainty in the Hg emission factors due to limited data leads to an uncertainty in the emission estimates on the order of  $\pm 50\%$ . Research is still needed to better constrain Hg emission factors from fires, particularly in the eastern U.S. and for ecosystems other than forests.

## Introduction

Mercury (Hg) is a ubiquitous, persistent, and toxic environmental pollutant (1). Once released to the environment, elemental mercury can be converted in aqueous media into the highly toxic methyl mercury, which is accumulated efficiently in the aquatic food chain (2). It becomes concentrated in top vertebrates, for example, in predatory fish, birds, and mammals, where it poses a risk for wildlife and humans (3). Mercury enters the terrestrial and aquatic biosphere mostly by wet and dry deposition of atmospheric mercury species (4), which are operationally defined (5) as gaseous elemental mercury (GEM), gaseous ionic mercury called reactive gaseous mercury (RGM), and mercury associated with aerosols, designated particulate mercury (pHg). The three atmospheric mercury species have grossly different physical and chemical properties: the lifetime is short (days to weeks) for RGM and pHg; the lifetime of GEM is much longer (e.g., 1 year (4)), but spatially variable. Distant from point sources, GEM accounts for  $>95\%$  of the total atmospheric mercury, with concentrations less than 1 part per trillion by volume [ppt(v)].

Atmospheric Hg originates from three main sources: anthropogenic sources (e.g., fossil fuel combustion and waste incineration), natural sources (e.g., volcanoes and ocean vents), and re-emission of anthropogenic and natural mercury previously deposited onto terrestrial and water surfaces. Long-range transport of GEM, its conversion to RGM and pHg, and deposition of these species along a trajectory have caused widespread contamination of aquatic and terrestrial ecosystems (6, 7), even to those located far from industrial or natural sources.

The interaction of atmospheric Hg with the biogeosphere (8) leads to pools of Hg in vegetation and soil. When Hg is deposited to the earth, some is taken up (plant-specifically) by vegetation (9, 10); some collects from throughfall; most Hg collects as the litterfall of senesced leaves and needles on the soil surfaces, where the carbonaceous materials decompose and the mercury becomes complexed with reduced sulfur compounds in the organic soil (11). The rate of deposition is spatially variable. In the U.S., the highest wet deposition of Hg occurs in the Southeast, as indicated from the results of the Mercury Deposition Network (12).

During fires, some or all of the accumulated Hg is released (13). Mercury is concentrated in the ecosystem parts most vulnerable to combustion during fires: grasses, moss and lichen, leaves and needles, small branches, bark, and, predominantly, organic soil (14–16). In boreal and temperate forests,  $>90\%$  of the releasable mercury is contained in the organic soil horizons. Mercury is released from fires primarily as GEM, with up to 15% in the form of pHg (13, 17). Mercury released from fires can have local, regional, and global impacts.

In this paper, ecosystem-specific mercury emission factors are compiled from published reports and used within the North American fire emissions model developed by Wiedinmyer et al. (18) to arrive at mercury release estimates from biomass burning in all continental U.S. states for the period of 2002 through 2006. Spatial and seasonal changes, as well as variations of emissions from different fuel sources, are presented. The results are apportioned to National Inter-agency Coordination Center regions to serve the interests of the fire and air quality communities.

## Methodology

**Fire Emissions Modeling.** Fire emissions of Hg were estimated following the procedure outlined by Wiedinmyer et al. (18). This simple model calculates the daily fire emissions of Hg ( $E_{\text{Hg}}$ ):

$$E_{\text{Hg}} = A(x, t) \times B(x, t) \times \text{EF}_{\text{Hg}} \quad (1)$$

where  $A(x, t)$  is the area burned ( $\text{km}^2$ ) at location  $x$  and time  $t$ ,  $B(x, t)$  is the biomass burned ( $\text{kg m}^{-2}$ ) at location  $x$  and time  $t$ , and  $\text{EF}_{\text{Hg}}$  is an emission factor, or the mass of Hg that is emitted per mass of biomass burned ( $\text{g of Hg kg}^{-1}$ ).

The location and timing of fires were determined by observations from the MODIS instruments aboard the NASA Terra and Aqua satellites. Daily fire detections were processed by the U.S. Forest Service Remote Sensing Applications Center for 2002 through 2006 using the MODIS Active Fire data developed by the UMD Rapid Response team (19). Each fire pixel was treated as an individual fire.

The total fuel loading at each location was determined by the type of land cover underlying the fire pixel, as identified by the Global Land Cover 2000 (GLC2000) data set (20). GLC2000 identifies 29 different land cover classes in North and Central America at a  $1 \text{ km}^2$  resolution (20). For each

\* Corresponding author phone: (303)497-1414; e-mail: christin@ucar.edu.

**TABLE 1. Emission Factors (EFs) Used in the Emission Calculations**

reference	soil (S) or plume (P) <sup>a</sup>	emission factor (g Hg kg fuel burned <sup>-1</sup> ) × 10 <sup>6</sup>		notes	kg m <sup>-2</sup> burned <sup>b</sup>
		range	mean		
Boreal Forest					
Harden et al. (22)	S	0–138	69	Alaska, USA; coniferous forest, prescribed burn	2.5
Friedli et al. (13)	P	112–112	112	Quebec, Canada; pine forest, wildfire	2.5
Sigler et al. (27)	P	60–60	60	Quebec, Canada; pine forest, wildfire	2.4
Cinnerella and Pirrone (40)	P	62–12	87	Siberia, Russia; wildfire	5.6
Weiss-Penzias et al. (29)	P	136–278	207	Alaska, USA; wildfire	2.5
Turetsky et al. (15)	S	90–297	193	Alaska, USA; upland, wildfire	2.5
Turetsky et al. (15)	S	535–2417	1476	Alaska, USA; lowland, wildfire	2.9
<b>Mean</b>		<b>142–488</b>	<b>315</b>		
Temperate Forest					
Friedli et al. (17)	P	54–172	113	Washington, USA; mixed forest, wildfire	2.5
Brunke et al. (26)	P	78.7–163.4	121	South Africa, fynbos, wildfire	2.5
Engle et al. (23)	S	80–204	142	California, USA; coniferous forest, prescribed burn	2.5
Engle et al. (23)	S	88–196	142	Nevada, USA; coniferous forest, wildfire	2.5
Biswas et al. (30)	S	168–348	256	Washington, USA; mixed forest, wildfire	2.5
Biswas et al. (14)	S	296–1012	654	Wyoming, USA; coniferous forest, wildfire	2.5
Biswas et al. (14)	S	144–516	402	Wyoming, USA; aspen forest, wildfire	2.5
Woodruff et al. (41)	S	80–80	80	Minnesota, USA; prescribed burn	2.5
<b>Mean</b>		<b>124–336</b>	<b>239</b>		
Sage-Chaparral (Shrub)					
Engle et al. (23)	S	18.7–39.9	29.3	Nevada, USA; sage, wildfire	1.84
Cinnerella and Pirrone (40)	P	52.8–52.8	52.8	Mediterranean Region, wildfire	1.25
<b>Mean</b>		<b>35.8–46.4</b>	<b>41.1</b>		
Grasslands and Ag. Waste					
Friedli et al. (17)	S	38	38	Oregon, USA; wheat	3.06
Biswas et al. (14)	P	510	510	Wyoming, USA; meadow	0.4
<b>Mean</b>		<b>38–510</b>	<b>274</b>		

<sup>a</sup> Type of method used. <sup>b</sup> Used for EF calculation. When nothing else was available, 2.5 kg of fuel m<sup>-2</sup> burned was assumed (based on 23). All others are from respective references.

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classification, total biomass loading and the relative abundance of herbaceous and woody fuels have been assigned (18). The fraction of woody and herbaceous fuels at each fire pixel and the amount of biomass burned at each fire detected was calculated as a function of the total biomass loading and the amount of underlying forested and herbaceous cover (18). Vegetation cover was determined by the MODIS Vegetation Continuous Fields (VCF) product from the UMD Global Land Cover Facility ([www.landcover.org](http://www.landcover.org) (21)), scaled to 1 km<sup>2</sup>. The total possible area burned for each fire pixel was assumed to be 1 km<sup>2</sup>, on the basis of the nominal resolution of the MODIS instruments, and scaled to the amount of bare cover assigned at that spot by the VCF product.

The emissions model has several sources of error associated with the fire detections, the fuel loading, the area and amount of fuel burned, and the EFs. These uncertainties are difficult to quantify, and a detailed description about the uncertainties associated with the fire emissions model is presented by Wiedinmyer et al. (18). Here, the general model uncertainties are only briefly discussed.

The use of the MODIS fire detection product introduces uncertainty in the emission estimates, including missed potential fires due to cloud cover or the timing of the satellite overpass and multiple detections of a single fire since information from both the Aqua and the Terra satellites is currently considered in the model application. The data product does not include information on cloud cover or missing data. Additional uncertainties of the MODIS fire detections are described by Giglio and Descloitres (19). The assumed burn area (1 km<sup>2</sup> per fire detection, scaled to the

percent of bare cover) can lead to large errors in the estimated biomass burned per fire detection. Inaccuracies in the identification of land cover type, and the assigned fuel loadings, result in uncertainties in the amount of fire fuel burned and ultimately the Hg emitted from each land cover type. The difference in mercury release between low- and high-severity fires in coniferous forests can range from 20 to 80% (14, 22, 23). However, the model framework described here does not currently account for fire severity. Wiedinmyer et al. (18) propose that at least a factor of 2 uncertainty be assigned to the fire emission estimates.

**Mercury Emission Factors.** Mercury EFs from fires are estimated by two methods: plume-based and soil-based. EFs derived from the soil-based method are reported as grams of Hg released per hectare burned; EFs derived from the plume-based method have units of grams of Hg per kilogram of fuel burned. The conversion between EFs from the two methods requires fire-specific values for fuel burned per unit area (Table 1). For the most important contributors to mercury emissions, temperate and boreal forests, an average value of 2.5 kg m<sup>-2</sup> was used (17, 24). This compares reasonably with the average fuel burned calculated by the fire emissions model (e.g., 3.7 ± 1.1 kg m<sup>-2</sup> for the temperate and boreal forests in 2006).

For the plume-based method (13), measurements of Hg are collocated with measurements of carbon monoxide (CO) or carbon dioxide (CO<sub>2</sub>) in a fire plume. These concentrations can be used in combination with an enhancement ratio (ER, where ER = Δ[Hg]/Δ[CO]), to calculate emission factors for Hg (mass of Hg emitted per mass of biomass burned). The concentration of CO (or CO<sub>2</sub>) (adjusted by a factor of 1.05 to

**TABLE 2. Published Molar Enhancement Ratios (ER) Observed from Fire Plumes Worldwide**

location <sup>a</sup>	ER ( $\Delta[\text{Hg}]/\Delta[\text{CO}]$ )	reference
Washington state (ac)	$(0.79 \pm 0.04) \times 10^{-7}$	Friedli et al. (17)
Pacific NW (ground)	$(1.46 \pm 0.9) \times 10^{-7}$	Weiss-Penzias et.al. (29)
Alaska (ground)	$(1.57 \pm 0.67) \times 10^{-7}$	Weiss-Penzias et al. (29)
Quebec (ac)	$2.04 \times 10^{-7}$	Friedli et al. (13)
Quebec (ground)	$0.86 \times 10^{-7}$	Sigler et al. (27)
South Africa (ground)	$(2.1 \pm 0.21) \times 10^{-7}$	Brunke et al. (26)
South America (ac)	$(1.17 \pm 0.15) \times 10^{-7}$	Ebinghaus et al. (28)
South America (ac)	$(2.39 \pm 0.99) \times 10^{-7}$	Ebinghaus et al. (28)

<sup>a</sup> ac = aircraft measurements and ground = ground measurements.

account for coemitted methane, nonmethane hydrocarbons, and particulate matter) is used as a surrogate for fuel mass combusted (13), and fuel (dry weight) is assumed to contain 45% carbon. The CO/CO<sub>2</sub> ratio is measured or assumed to be 1:9 (v/v) (25). The EF for the most well-constrained fire (the Washington state temperate forest fire (17)) calculated by this method is  $(113 \pm 59) \times 10^{-6}$  g of Hg kg<sup>-1</sup> of fuel burned. The ratio of EF/ER is assumed to be constant for all fires. Therefore, the EFs for other U.S. plumes (Table 1) are calculated as the product of the EF from the Washington state fire with the ratio of their respective ERs.

To estimate a robust Hg budget, all three mercury species (pHg, GEM, and RGM) must be included in emission estimates. In nascent plumes, pHg is a significant fraction of the mercury budget, approximately 15% (13, 17). However, most studies report gaseous mercury only (26–28), which leads to underestimates of the total Hg released. Additionally, total Hg measurements far from emission sources are missing part or all of the pHg fractions that were deposited along the trajectory. For these two reasons, the EFs determined by these far field studies (26–29) must be considered as lower-limit estimates.

The variations in EFs caused by differences in ecosystems, fire severity, and mercury assessment methods are reflected in the variation in the reported ERs, where  $\text{ER} = \Delta[\text{Hg}]/\Delta[\text{CO}]$ . Published molar ER values are summarized in Table 2 (13, 17, 26–29).

The soil-based method of determining Hg released from fires determines the difference in the mercury pools in vegetation and soil in adjacent plots before and after a fire (14, 15, 22, 23, 30, 31). In temperate and boreal forests, these studies found that mercury resides mostly (>90%) in the organic soil (including litter). Contributions from the live under- and overstories (leaves and needles, small branches, bark, and bole wood) are minor, but their Hg content is largely released during fires (14, 16, 23, 32). Release of Hg from the underlying mineral layer is also negligible (23). Since most of the Hg resides in the organic soil, the Hg profile (16) in the soil and severity of the fires determine the extent of the Hg release. Therefore, the soil-based Hg EFs used in this work were determined from the difference between measurements of the Hg in the organic soil layer before and after a burn, and the mercury emitted from above-ground fuel was not included.

The Hg EF determined by the soil-based method is highly dependent on the depth of a burn. For most experiments (14, 22, 23), the burn depth is determined for a specific fire using carbon/nonvolatile metal measurements. In contrast, Turetsky et al. (15) assumed differing burn depths based on three different drought scenarios. These different approaches can lead to varied Hg EFs.

Table 1 summarizes the published Hg EFs, from both plume- and soil-based methods, used in this study. The average value from the reported EFs within a specific land cover type (boreal forest, temperate forest, shrublands, and grasslands/crops) was assigned to similar land covers in the

**TABLE 3. Annual Estimates of Hg from Fires for the Lower 48 and Alaska<sup>a</sup>**

year	Hg emissions (metric ton year <sup>-1</sup> )	
	lower 48	Alaska
2002	29 (14–41)	4 (2–6)
2003	36 (17–52)	3 (1–5)
2004	23 (11–34)	32 (14–51)
2005	28 (13–41)	22 (9–36)
2006	41 (20–59)	0 (0–1)
average	31 (15–45)	12 (5–20)

<sup>a</sup> The values in parentheses represent the annual estimates when the minimum and maximum emission factors are applied in the calculations.

fire emissions model (Supporting Information, Table S2). To better understand the uncertainty in these EFs, and to quantify the bounds of the emission estimates on the basis of this uncertainty, a minimum and a maximum EF<sub>Hg</sub> (based on the reported values) were also assigned to the general land cover types (Table 1). The emissions model was run with these EFs to produce a lower and an upper estimate of total emissions (Table 3).

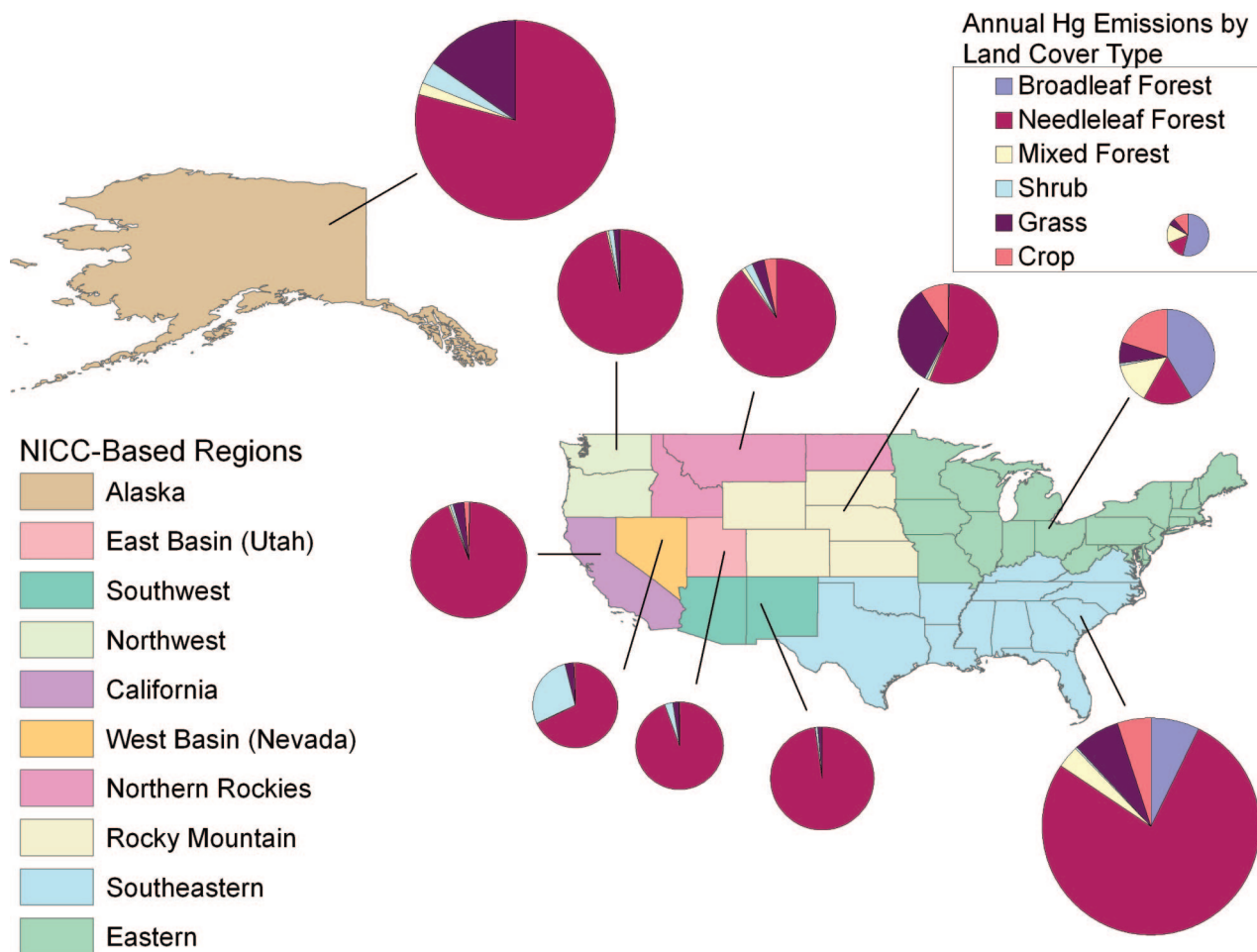
The uncertainty in the reported values of Hg EFs results from many contributions, including the influence of burn severity, inclusion of all mercury species, paucity of measurement for all fuel types (especially for grasses, shrubs, and agricultural waste products), and uncertainties in the emission factor estimation technique. The amount of fuel burned during a fire is highly variable among different ecosystems, and even within the same vegetation type (24, 33). The differences in fuel characteristics (species composition, stand density, and stand age) on one hand, and fire weather conditions on the other, result in fires with different degrees of fuel combustion and burn depth, and correspondingly variable Hg release.

One direct comparison of the two emission factor estimation techniques has been accomplished. Measurements in the fire plume from the Rex fire in Washington state were made from an aircraft during the burn (17), and soil measurements were made postfire in adjacent burned and unburned sites (31). The results from the soil measurements were expected to be higher than those from the aircraft measurements, since some of the pHg in the plume would be lost by deposition and, therefore, not measured from the aircraft. This was the case. The soil-based measurements were  $6.4 \pm 1.1$  grams of Hg ha<sup>-1</sup> compared to  $2.9 \pm 2.2$  grams of Hg ha<sup>-1</sup> estimated from the aircraft measurements. This comparison highlights the uncertainties in the reported emission factors of Hg from fires.

## Results and Discussion

Daily emissions of mercury from fires were estimated for North America for five years (2002–2006; Table 3). Annual





**FIGURE 1.** Regions for analysis (based on the NICC management region map, <http://www.nifc.gov/nicc/index.htm>). The pie charts indicate the fraction of annually averaged Hg emissions from the six general land cover types for each region. The size of each pie chart represents the relative fraction of the annual Hg emissions for each region.

estimates of mercury from fires in the lower 48 states (LOWER48) range from 23 tons (metric)  $\text{yr}^{-1}$  (2004) to 41 tons  $\text{yr}^{-1}$  (2006). For the five years investigated, the annual average Hg emission from fires in the LOWER48 is 31 tons  $\text{yr}^{-1}$  ( $\pm 7$  tons  $\text{yr}^{-1}$  standard deviation, sd). For Alaska, the total emissions vary substantially from year to year. Annual Hg emissions from fires in Alaska range from less than 1 ton  $\text{yr}^{-1}$  (2006) to 32 tons  $\text{yr}^{-1}$  (2004), with an average of 12 tons  $\text{yr}^{-1}$  ( $\pm 14$  tons  $\text{yr}^{-1}$  sd). Other published estimates for annual Hg fire emissions in the LOWER48 have similar ranges and uncertainties. Comparative (1995–2000 average) annual Hg emission estimates for the LOWER48 are 60–110 tons  $\text{yr}^{-1}$  (21), 11–64 tons  $\text{yr}^{-1}$  (23), and 19–64 tons  $\text{yr}^{-1}$  for wildfires (22).

For the LOWER48, the average annual emission of Hg from fires is 31 tons  $\text{yr}^{-1}$ . When the minimum (maximum) EFs are applied (Table 1), the annual average Hg emission from fires is 15 tons  $\text{yr}^{-1}$  (45 tons  $\text{yr}^{-1}$ ). Thus, the overall variation in the emissions estimates that results from the uncertainty associated with the EFs is +45% and –48%. In Alaska, the range in EFs leads to +67% and –42% uncertainties in the emission estimates.

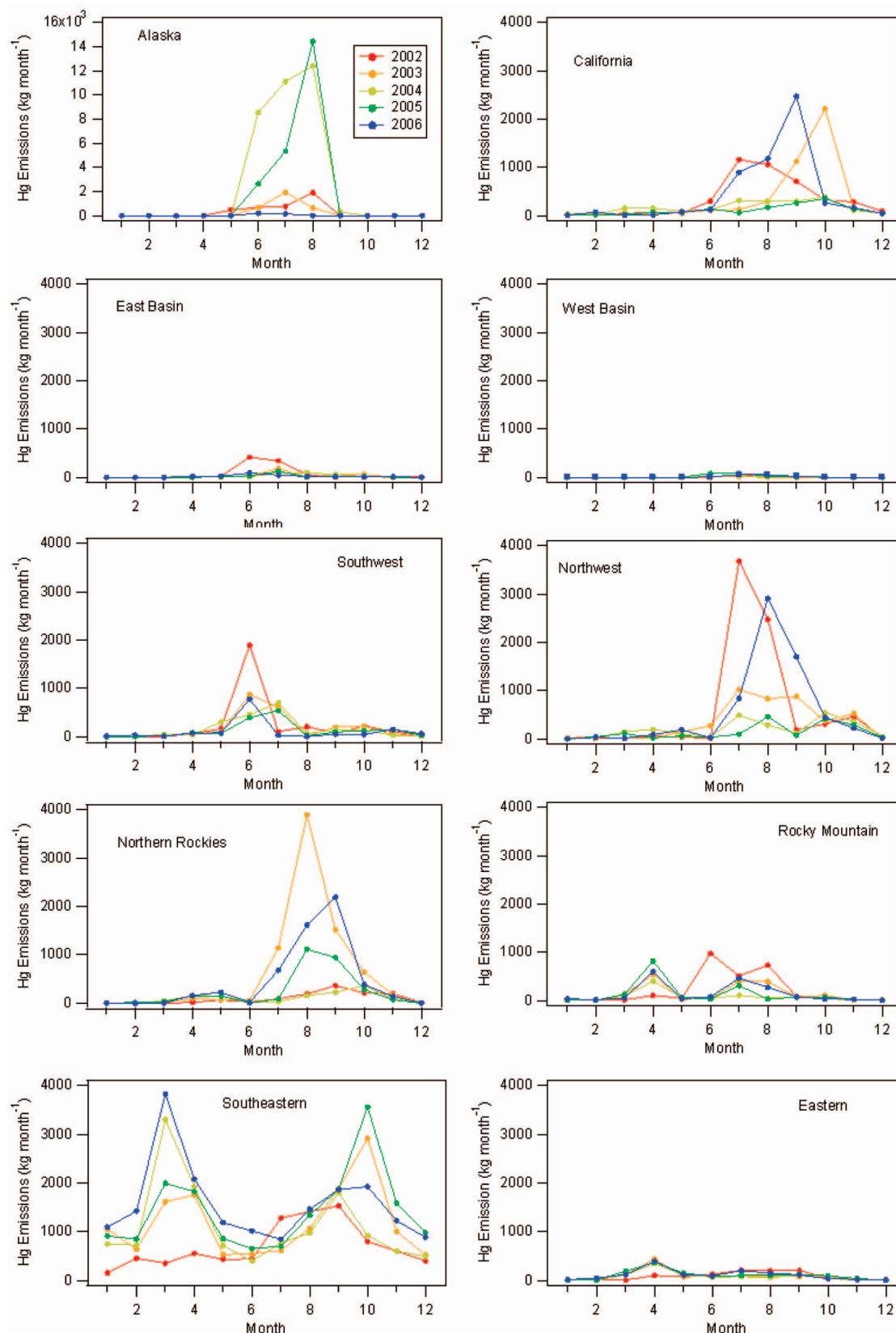
The regional and temporal variations in the fire emissions of mercury are highly dependent on the location within the U.S. (Supporting Information, Table S1). In the southeastern U.S. states (for example, Alabama), Hg emissions from fires are observed throughout the year, whereas emissions in the Rocky Mountain states (for example, Montana) have only significant fire emissions during the summer months. Alaska has the highest monthly averaged emissions, with a maximum average of 5.9 tons  $\text{month}^{-1}$  in August. Emissions of Hg in

Alaska during the summer months of 2004 and 2005 are 5–10 times higher than the those in the same months of 2002, 2003, and 2006. The temporal variability of the emissions is extremely large due to the stochastic nature of wildfires in that region.

In the LOWER48, the western states of California, Oregon, Montana, Idaho, Washington, and Arizona have the highest monthly averaged emissions of Hg from fires, with peak monthly emissions occurring in the summer months of June–September, concurrent with the western wildfire season (Supporting Information, Table S1). In many of the southeastern states (for example, Alabama, Florida, Georgia, Mississippi, and Louisiana), the fire emissions of Hg peak either in the spring (March) or in the fall (October) (Supporting Information, Table S1). These times coincide with the agricultural and prescribed burning that occurs in this region annually (Chris Geron, U.S. EPA, personal communications, April 2007).

**Regional Emissions.** To evaluate the emission estimates on a regional basis and serve the interests of the fire and air quality communities, the emission estimates have been apportioned to regions based on those used by the National Interagency Coordination Center (NICC; <http://www.nifc.gov/nicc/>) of the National Interagency Fire Center (Figure 1). Monthly Hg emissions have been totaled for each of these 10 areas of interest (Figure 2).

Regionally, the southeastern region has the greatest annual-averaged Hg fire emissions (14.4 tons  $\text{yr}^{-1}$ ). In this region, the emissions peak in the spring and again in the fall, indicative of the prescribed burning practices of that region



**FIGURE 2.** Monthly emissions of Hg from fires in each of the 10 analysis regions (Figure 1). (Note: the scales for all regions *except* Alaska are the same.)

(Chris Geron, U.S. EPA, personal communications, April 2007). However, some years show higher Hg emissions in the spring (2004 and 2006), while others have more emissions in the fall (2003 and 2005). This could be due to a variety of

factors, including the local meteorology and climate, as well as changes in management practices.

Other regions in the LOWER48 that contribute to the majority of the nationwide Hg fire emissions inventory are

the Northern Rockies, California, and the Northwest. However, year-to-year emissions are highly variable and not always consistent across regions. For example, large fire emissions of Hg were predicted for the Northwest in 2002 and 2006 (relative to all years studied). California also had high emissions during 2002 and 2006. There was also a late peak in fire Hg emissions during the fall of 2003 in California that is not observed in the emissions from the Northwest in 2003.

In addition to a summer peak in emissions, the Rocky Mountain region also shows a peak in emissions during the spring, suggestive of field burning in Kansas and Nebraska. The Southwest shows a peak in emissions earlier in the summer (June) than the regions to the north, presumably because this region warms and dries out earlier in the summer.

For each region, the amount of Hg emitted from each general land cover type [broadleaf forests, needleleaf forests, mixed (needleleaf and broadleaf) forests, shrubs, grasses, and crops] was determined (Supporting Information, Tables S2 and S3). Overall, Hg emissions from fires in needleleaf forests dominate the inventory, contributing to an average of 81% of the total Hg fire emissions from 2002 to 2006 for the LOWER48 and Alaska. Fires in areas identified as croplands only contribute an average of 3% of the total annual fire Hg emissions. This implies that, although a significant number of fires may occur in the various land cover types (including croplands), their overall contribution to the annual inventory is small compared to emissions from fires in forests.

In addition to the NICC-based regions used for the analysis, Figure 1 shows the relative contribution of fire Hg emissions from each of the six general land cover types in each region (Supporting Information, Table S3). Only in the eastern and the southeastern regions do fires in broadleaf forests have substantial emissions of Hg. Fires from croplands contribute 20% of the Hg emissions in the eastern region. In the Western Basin (Nevada), fires in shrublands emit 28% of the total annual Hg emissions from fires. Grasses and crops contribute 42% of the Hg emissions from the Rocky Mountain region, which includes the agriculture-dominated states of Kansas and Nebraska. In Alaska, 15% of the Hg emissions are estimated to come from fires in grasslands.

The emissions of Hg from fires in the different land cover types vary. In most instances, the emissions from the needleleaf forests are the greatest. However, for some seasons and regions, this is not the case. In the Rocky Mountain region, the needleleaf forests emit the most Hg emissions from fires during the summer months. In the spring, Hg emissions from fires in grasslands and croplands within that region emit more Hg than the forested land cover types. In the eastern region, the primary forest type, broadleaf forests, emits the most Hg from fires throughout the year, although mixed forests, needleleaf forests, and crops also produce Hg emissions from this region in the spring.

The magnitude of Hg emissions from fires is significant when compared to the U.S. National Emissions Inventory (NEI). For 2002, the U.S. EPA estimates annual anthropogenic emissions at 108 tons of Hg yr<sup>-1</sup>, with 46 tons of Hg yr<sup>-1</sup> from utility coal boilers (Anne Pope, U.S. EPA, personal communications, April 2007). The estimated annual average (2002–2006) emission of mercury from fires in the LOWER48 and Alaska is 44 tons yr<sup>-1</sup>. Thus, fires emit the equivalent of almost 30% of the total U.S. anthropogenic Hg emissions (and an amount equivalent to the emissions from coal-fired power plants), and more in years with large fire activity. Comparisons between the NEI and biomass burning are useful for regulatory considerations, but it must be recognized that full budgets also include natural sources, such as volcanoes, geothermal events, and biogeochemical exchanges (34, 35).

As future Hg emissions are contemplated, there are several factors to consider, and they require future study. Hg emissions from fires are a revitalization of previously deposited Hg from natural and anthropogenic sources. The amount of anthropogenic emissions in the U.S. has decreased 52% from 1990 to 2002 (Anne Pope, U.S. EPA, personal communications, April 2007). If anthropogenic emissions of Hg are decreasing in the U.S., it is possible that the accumulation of Hg will also slow, resulting in a decrease of Hg fire emission factors in the U.S. However, during the last 20 years, Asian sources of Hg have dramatically increased, keeping the atmospheric load and the deposition constant or increasing (36). Weiss-Penzias et al. (29) show that Hg emissions from Asia often impact at least the U.S. west coast (e.g., California, Washington, and Oregon). The magnitude of Hg transported from various sources and deposited in the U.S. is highly uncertain (37). More detailed studies about these processes and the impact of various sources on Hg accumulation in the U.S., and further, the emission factors, must be studied in much more detail in order to better understand the current Hg fire emissions and those expected in the future.

Changes in climate may also affect the future emissions of Hg from fires. The predicted extent of climate warming (38) at northern latitudes make the large mercury stocks in peat and permafrost in boreal forests available for large, more frequent high-severity wildfires (39) that would release large pulses of mercury to the atmosphere.

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## Supporting Information Available

A detailed table of monthly Hg emissions (averaged over 2002–2006) for each month in the continental U.S. is provided in Table S1. The emission factors and average area and biomass burned for the general vegetation classes are provided in Table S2. The percent contribution of the vegetation classes to the annual fire Hg emissions for each NICC region is given in Table S3. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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