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Impacts of the Minamata Convention on Mercury Emissions and Global Deposition from Coal-Fired Power Generation in Asia

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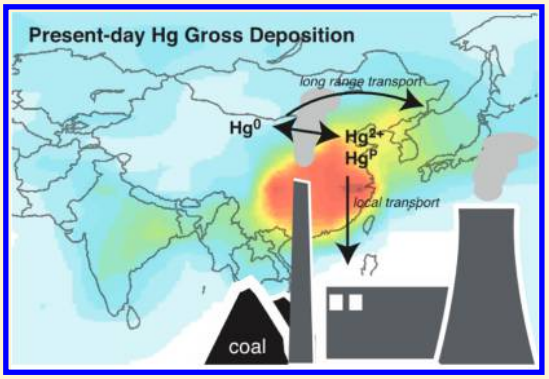
Supporting Information

ABSTRACT: We explore implications of the United Nations Minamata Convention on Mercury for emissions from Asian coal-fired power generation, and resulting changes to deposition worldwide by 2050. We use engineering analysis, document analysis, and interviews to construct plausible technology scenarios consistent with the Convention. We translate these scenarios into emissions projections for 2050, and use the GEOS-Chem model to calculate global mercury deposition. Where technology requirements in the Convention are flexibly defined, under a global energy and development scenario that relies heavily on coal, we project ~90 and 150 Mg·y^{-1} of avoided power sector emissions for China and India, respectively, in 2050, compared to a scenario in which only current technologies are used. Benefits of this avoided emissions growth are primarily captured regionally, with projected changes in annual average gross deposition over China and India ~2 and 13 μg·m^{-2} lower, respectively, than the current technology case. Stricter, but technologically feasible, mercury control requirements in both countries could lead to a combined additional 170 Mg·y^{-1} avoided emissions. Assuming only current technologies but a global transition away from coal avoids 6% and 36% more emissions than this strict technology scenario under heavy coal use for China and India, respectively.



INTRODUCTION

Atmospheric mercury emissions can travel short or long distances depending on chemical form, leading to both local and global mercury contamination.[1] In aquatic ecosystems, mercury poses risks to human and wildlife health as the potent neurotoxin methylmercury.[2−4] Human populations are typically exposed to methylmercury through fish and shellfish consumption, but also through rice and other food sources.[5,6]

Concern about mercury's global transport and human health impacts led to the 2013 adoption of the United Nations (UN) Minamata Convention on mercury. The Minamata Convention takes a life-cycle approach to regulating mercury and its compounds, with obligations for mining, use, emissions and releases, and disposal.[7] More than 50% of mercury emissions are estimated to be "byproduct": mercury is a trace impurity in raw coal, oil, and ores, released upon combustion or smelting.[8] Thus, efforts to control these emissions sources can interact with energy and development interests, as well as with traditional air quality priorities. Coal combustion is estimated to be the second largest global source of anthropogenic mercury emissions to air (24% of emissions in 2010).[8−10] For coal combustion, Asia is the largest regional contributor to global emissions, and these emissions are projected to increase with continued economic growth.[8,11−14] Historically, however, Europe and North America were major contributors, and this previously emitted mercury can continue to be re-emitted from soils and the ocean.[13] Existing projections suggest that emissions growth in Asia will drive the global trajectory until 2050.[14,15]

The 2013 UNEP Global Mercury Assessment estimated that China alone contributes approximately one-third of the global anthropogenic emissions total.[8] India is estimated to be the second largest national contributor, though the gap between India and China is large—in 2010, India contributed an estimated 7% of global emissions, compared to 29% from China.[16] For both countries, rapid economic development drives emissions: primarily, coal combustion for electricity

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generation and industry, but also other industrial and mining activity, such as cement production and non-ferrous metal smelting.[8,11]

Coal consumption in both countries is expected to grow substantially. The World Resources Institute estimates that China was responsible for 46% of world coal consumption in 2010—more than 3 times the next two largest consumers, the U.S. at 13% and India at 9%—and increases in coal electricity generating capacity are still planned.[17] Growth in India may be even steeper, as electrification is a major near-term government priority,[18] and close to 25% of the population may lack electricity access.[19,20] The Indian government plans to add ∼160 GW of additional capacity by 2022, and coal-fired power plants (CFPPs) will be a large part of this expansion.[18]

While the Minamata Convention contains obligations for regulating mercury emissions from coal, it does not specify quantitative emissions limits or require specific technologies. Instead, parties must "control, and where feasible, reduce" mercury emissions in new CFPPs by applying Best Available Techniques (BAT) and Best Environmental Practices (BEP) within five years of the treaty's entry into force. For existing plants, parties can choose between implementing BAT and BEP, quantified goals, emission limit values, multi-pollutant control strategies, or alternative reduction measures, within 10 years of the treaty's entry into force.[21] Given the flexibility of these requirements, what the Convention will mean in practice will differ by country. Specific guidance on BAT and BEP will be developed by the Convention's Conference of Parties, and will likely include a suite of approaches, with country representatives taking into account country-specific economic and technological considerations, as well as other pollutant controls.[21] Here, we explore how different technological approaches might affect the implications of the Minamata Convention for large, CFPP boilers in two major mercury-emitting countries in Asia: India and China. We develop technology scenarios consistent with the Convention, taking into account local political, technological, and geological factors. We examine the impacts of these technology scenarios on atmospheric mercury emissions from coal, and on mercury deposition worldwide. We also compare the magnitude of emissions and deposition benefits from increased use of control technology to that from a less carbon-intensive global energy trajectory.

METHODS

Technology Scenario Development. To better understand mercury-related policy, energy, and technology trends in India and China, we conduct an extensive literature review, including both peer-reviewed sources and technical reports from governments, and international agencies such as UNEP and the International Energy Agency (IEA). We supplement this literature review with semi-structured interviews with Convention negotiators, air pollution regulators, and coal and air pollution control experts. More information on these interviews and how interview data was used to support analysis is provided in the Supporting Information (SI). We combine insights from these sources to develop technology scenarios consistent with the Convention, focusing on how BAT and BEP are likely to be applied in China and India.

We consider three representative technology scenarios: a no additional control (NAC) scenario, a Minamata Flexible (MF) scenario, and a Minamata strict (MS) scenario. Under NAC, we specify technologies and techniques currently widely in use. We define the MF scenario as technologies and techniques consistent with existing domestic (not necessarily mercury specific) policy plans, and which could fall under a flexible definition of BAT. We define the MS scenario as technologies and techniques that represent a progression in stringency of mercury control beyond those specified in MF. For each scenario, we specify a representative, most likely suite of technologies and techniques for each country based on our review of the literature and interviews, and assume countrywide implementation. We use a representative scenario approach (in contrast to a plant-by-plant approach that would focus on variability in pollution control strategies) to better isolate the impacts of specific technological choices on emissions and transport.

For each technology scenario and country, we estimate most likely values for the mercury removal efficiency fraction, $f_{capture}$, and the resulting speciation of emissions among three chemical forms of mercury: gaseous elemental mercury (Hg^0), gaseous oxidized mercury (Hg^{2+}), and particulate-bound mercury (Hg^P). Additional analysis considering the reported range of $f_{capture}$ and emissions speciation for control technologies under each scenario is given in SI, section 6. Different pollution control technologies can alter emissions speciation, which has implications for atmospheric transport. Hg^0 has an atmospheric lifetime of six months to a year. In contrast, Hg^P and Hg^{2+} are readily deposited, leading to atmospheric lifetimes of days to weeks.[1] Thus, Hg^{2+} and Hg^P act as more regional pollutants, while Hg^0 is capable of global transport—though atmospheric chemical reactions can transform one species to another.[22,23] Mercury deposited from the atmosphere is not lost to a stable surface reservoir, however, but may be re-emitted and continue to cycle in surface atmosphere, terrestrial, and water reservoirs for decades to centuries.[1]

Where available, we use values for mercury capture efficiency and emissions speciation from measurements at Chinese and Indian plants. Where measurements are unavailable, we use the Process Optimization Guidance for Reducing Mercury Emissions from Coal Combustion in Power Plants[24] and the Interactive Process Optimization Guidance (iPOG) tool[25] to estimate removal efficiencies and speciation, based on average reported coal characteristics for each country. While we use deterministic estimates for these values in our baseline analysis, any given technology is likely to lead to a distribution of capture efficiencies and speciation fractions, due to variability in coal characteristics and implementation. As a result, we apply sensitivity analysis (discussed further below) to explore how variability in our estimates of $f_{capture}$ and emissions speciation affects our transport estimates.

Emissions Estimation. To estimate the impact of technology choices on emissions, we combine assumptions about the mercury removal efficiency and speciation effects of technologies with projections for future coal use. Following Streets et al.[15,26] and Wu et al.,[27] we calculate total mercury emissions from CFPPs in a given region as

$$E_{totHg} = Hg_c \cdot C \cdot f_{release} \cdot (1 - f_{capture} \cdot f_{uptake})$$

$$E_{Hg,i} = E_{totHg} \cdot f_{Hg,i}$$

where E_{totHg} represents total mercury emissions, Hg_c is coal mercury content, C is the amount of coal burned, $f_{release}$ is the fraction of mercury released to the gas phase when burning coal, $f_{capture}$ is the removal efficiency of the technology scenario,

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Table 1. Reported Mercury Reduction (%) of Control Strategies in China, India, and the U.S. (All Values Are for Pulverized Coal Combustion)

control strategy	device configuration	Hg reduction range (%)		
		China[24,73−79]	India[24,54]	United States[24,45,47,48,80,81]
plant efficiency improvements		0−7	0−7	0−7
pre-combustion controls				
coal cleaning		−	13−40[a]	0−78[b]
post-combustion controls				
PM control	CS-ESP	20−41[b]	19−73[a]	0−81[b]
				0−22[c]
	FF	10−80[b]	−	63−93[b]
				53−88[c]
PM + SO_2 control	CS-ESP + wFGD	13−74[b]	−	64−76[b]
				2−58[c]
	FF + wFGD	−	−	62−99[b]
	FF + SDA	−	−	97−99[b]
				0−48[c]
PM + SO_2 + NO_x control	SCR + CS-ESP + wFGD	−	−	>90[b]
	SCR + FF + SDA	−	−	94−99[b]
				0−47[c]
Hg specific control	CS-ESP + ACI[d]	−	−	up to 98[b]
				up to 62[c]

[a]Not specified. [b]Bituminous coal. [c]Sub-bituminous coal. [d]Capture depends on sorbent injection rate. Abbreviations: CS-ESP, cold-side electrostatic precipitator; FF, fabric filter; wFGD, wet flue gas desulfurization; SDA, spray dry absorber; SCR, selective catalytic reduction; ACI, activated carbon injection.

and f_{uptake} is the uptake rate of that technology suite. To calculate emissions of specific mercury species, $E_{Hg,i}$, where $i \in$ 0 (gaseous elemental), 2 (gaseous oxidized), P (particulate-bound), we then apply the speciation fraction $f_{Hg,i}$ to E_{totHg}. We apply values for Hg_c, C, and $f_{release}$ from Streets et al.,[15] who use forecasts based on the IPCC SRES scenarios from the IMAGE group.[28] We assume a constant f_{uptake} across scenarios of 0.95, the maximum control technology penetration rate under IMAGE assumptions.[15,28] Our baseline analysis uses the A1B SRES scenario, which is broadly consistent with Representative Concentration Pathway 6.0 and Shared Socio-economic Pathway 2, under the new scenario framework developed by the climate research community for future-looking global change modeling studies.[29] This scenario has been characterized as "business as usual", with continued growth in coal and limited global environmental cooperation.[15,29] To assess the relative effects of technology on mercury emissions compared to broader socio-economic development trends, we also consider energy use under the SRES B1 scenario.[29] This scenario is characterized by global cooperation on sustainable development, leading to transitions away from coal.[15,29]

Chemical Transport Modeling. To estimate worldwide mercury deposition, we use the GEOS-Chem global mercury model, version 9-02 (http://acmg.seas.harvard.edu/geos). The model includes a 3-D atmosphere from Holmes et al.[30] and Amos et al.,[31] and 2-D ocean and terrestrial slabs from Soerensen et al.[32] and Selin et al.[33] respectively. GEOS-Chem is driven by assimilated meteorology from the NASA Goddard Earth Observing System (GEOS-5). The model has been extensively compared in previous work to observed concentrations and wet deposition.[30−32,34−37] There are numerous uncertainties in modeling global mercury. Corbitt et al.[38] discuss uncertainties in quantifying source−receptor relationships, and identify mercury speciation and atmospheric reduction processes as particularly relevant. We quantitatively evaluate these uncertainties here through sensitivity analysis and an alternative chemistry scenario, described below.

The model includes gaseous elemental mercury, Hg^0, and gaseous and particulate-bound divalent mercury, Hg^{2+} and Hg^P. In GEOS-Chem, Hg^0 is emitted by natural and anthropogenic sources, while Hg^{2+} and Hg^P are emitted only by anthropogenic sources. Divalent mercury can deposit via wet and dry deposition, and Hg^0 can undergo dry deposition.[1] Re-emissions of mercury from terrestrial and aquatic reservoirs occur only as Hg^0.[1] Divalent mercury follows an empirical gas-particle partitioning relationship based on air temperature and aerosol concentration, following Amos et al.[31] Hg^0 oxidizes to Hg^{2+} by reaction with Br, and Hg^{2+} is photoreduced to Hg^0 in cloud droplets.[30]

Reduction of $Hg^{2+/P}$ to Hg^0 in power plant plumes has been hypothesized to occur, based on comparisons of measured speciation fractions from the stack and downwind, though its mechanism has not been identified.[23,39−41] Recent evidence suggests that the occurrence and extent of in-plume reduction (IPR) may depend on coal composition characteristics.[23] Implementing IPR in mercury transport models has improved correlations between modeled and observed wet deposition and concentrations within North America.[31,34,42] We therefore run model simulations both with and without IPR, where we use the without IPR scenario as our baseline. Based on measurement and experimental studies,[23,39,41,43] we implement IPR as a conversion of 70% of power sector $Hg^{2+/P}$ emissions to Hg^0.

Emissions gridded at 1°×1° from Corbitt et al.,[38] and scaled to 2050 projections from Streets et al.,[15] are scaled on a national basis for China and India given technology scenario totals constructed as described above. As the IMAGE 2.2 energy[28] and Streets et al.[15] emissions projections underlying the analysis are for East Asia and South Asia as regions (see SI, Figure S1), we apply these regional scaling factors to China and India, respectively.[38]

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Table 2. Technology Scenarios[a]

scenario	technologies and techniques	$f_{capture}$ (%)	f_{Hg0} (%)	$f_{Hg2/P}$ (%)	notes
China					
no additional control (NAC)	CS-ESP + wFGD	69	78	22	values from Wang et al.[74]
Minamata flexible (MF)	SCR + CS-ESP + wFGD	82	68	32	values from UNEP iPOG tool[25] with coal characteristics from UNEP China report[12]
Minamata strict (MS)	SCR + FF + wFGD	90	68	32	value for $f_{capture}$ based on interpretation of qualitative descriptions in Srivastava et al.[45] and limited test data from U.S. EPA;[80] in absence of speciation data, the same fractions as SCR + ESPc + wFGD are applied
India					
no additional control (NAC)	CS-ESP	42	67	33	values from UNEP India report[54]
Minamata flexible (MF)	efficiency gains + coal washing + CS-ESP	58	67	33	estimates for removal efficiency from washing and ESP from UNEP;[54] estimates for efficiency gains from UNEP[24] and India Central Electricity Authority[18]
Minamata strict (MS)	efficiency gains + coal washing + CS-ESP + wFGD	70.5	93	7	estimate from UNEP iPOG tool[25] with coal characteristics from UNEP[54]

[a]Coal characteristic assumptions are further described in Table S1 (SI).

Our GEOS-Chem simulations have a horizontal resolution of 4°×5° latitude−longitude, with 47 vertical layers. We simulate meteorological years 2007−2012, with 2050 emissions, using the first three years as initialization. We present 2010−2012 averages to account for inter-annual variability. We archive simulated total gross deposition as the sum of wet and dry deposition of all mercury species. We do not consider the effects of future meteorology in the present study to better isolate the effect of technology choices and emissions. Our GEOS-Chem simulations track the effect of primary anthropogenic emissions changes on mercury in surface reservoirs and resulting deposition. Additional deposition effects due to mercury in longer-lived soil and ocean pools (legacy pools) are addressed in the SI and Discussion.

To better understand the effects of the technology scenarios on mercury deposition patterns, we perform sensitivity analysis on the removal efficiency, and speciation of emissions for each country ($f_{capture}$ and f_{Hg0}). We perturb each of these variables ±20% from the MF scenario baseline, and evaluate the corresponding impacts on average total gross deposition flux over India, China, the U.S., and ocean basins. Because deposition responses to these perturbations are approximately linear in this range, we then calculate response ratios, α, for average deposition fluxes over India, China, and the U.S. (to illustrate long-range impacts). The response ratio represents the average percent change in average deposition flux due to a 1% change in the perturbed variable.

RESULTS

Synthesis of Available Mercury Control Technologies and Techniques. Mercury control measures for large CFPP boilers have been extensively reviewed elsewhere.[12,24,25,44,45] We assemble this information to apply to our technology scenarios, focusing on the performance of control measures in China and India specifically. Table 1 summarizes reported mercury reduction ranges for different control measures for India, China, and the U.S. (where measurements are the most abundant). A more in-depth review of this information is provided in the SI.

Table 1 summarizes three major categories of mercury reduction approaches for CFPPs: improvements to plant efficiency, pre-combustion controls, and post-combustion controls.[24,46]

Efficiency improvements can include upgrading equipment, optimizing combustion, minimizing short cycling and air leakages, and changing operations and maintenance. As shown in Table 1, these upgrades have been estimated to reduce emissions by up to 7%. Efficiency improvements can also have economic and climate benefits.

Pre-combustion controls include processing coal to improve efficiency and reduce unwanted trace elements, such as mercury and sulfur. Conventional coal cleaning targeting ash (which lowers the energy value of coal and combustion efficiency of boilers) and sulfur can also reduce mercury content by ∼30%.[24,27,47,48] However, a wide range of removal efficiencies has been reported (0−78%), as efficacy depends on coal source and characteristics.[24,27,47,48] Coal blending, additives, and beneficiation are discussed in the SI.

Post-combustion controls can be either mercury-specific, or aimed at other pollutants but with co-benefits for mercury. Substantial divalent, but not elemental, mercury reduction co-benefits can be achieved by controlling for particulate matter (PM), SO_2, and NO_x. Standard PM controls like electrostastic precipitators (ESP) and fabric filters (FF) are effective at capturing Hg^P, while controls for sulfur, such as wet flue gas desulfurization (wFGD) and spray dry absorber (SDA) systems, are effective at capturing mercury as Hg^{2+}. Selective catalytic reduction (SCR), used for NO_x control, increases the fraction of mercury in the stack gas as Hg^{2+}, facilitating removal by desulfurization measures. As with coal cleaning, the efficacy of these measures depends on coal characteristics such as moisture, ash, sulfur, halogen, and mercury content.[25] Table 1 presents reported capture ranges for selected combinations of cold-side ESP (CS-ESP), FF, wFGD, SDA, and SCR. The majority of these values are based on U.S. measurements, though measurements for CS-ESP are available for both India and China, and measurements for FF and wFGD are also available for China. The ranges among countries are consistent, though reported efficacy of PM and SO_2 control tends to be lower in China than in the U.S.

Post-combustion control technologies that specifically target mercury (particularly Hg^0) have been demonstrated to reduce

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emissions in the U.S. by greater than 90% and up to 98%.[24,45] However, these technologies may have higher installation and operating costs than co-benefit approaches.[44] The most common configurations involve sorbent injection, typically activated carbon (ACI), in conjunction with a PM control device.[24] While this technology is commercially available, application is not yet widespread, particularly in Asia.[11,49] However, recent stringent standards for mercury from utility boilers in the U.S. and Canada may result in installation of ACI at many plants.[49,50] Multi-pollutant systems specifically optimized to simultaneously capture mercury, SO_2, and NO_x are currently in development, however are not yet widely commercially available.[24,49]

Technology Scenarios. Table 2 presents the NAC, MF, and MS scenarios for China and India: technology configurations and values for $f_{capture}$, f_{Hg0}, and $f_{Hg2/P}$.

No Additional Control (NAC) Scenario. For China, under the NAC scenario, we assume countrywide use of CS-ESP and wFGD technologies, leading to a capture efficiency of 69%, and a 78% and 22% breakdown of power sector emissions between Hg^0 and $Hg^{2+/P}$. Installation of ESP technology at Chinese power plants has steadily grown since the 1980s, reaching ~95% of nationwide capacity by 2003.[51] Installation of FGD systems began approximately a decade later, with the most rapid increases in coverage occurring over the past 10 years (from approximately 14 to 86% of plants between 2005 and 2010), to meet SO_2 reduction targets in China's 11th Five Year plan.[49,51]

For India, the NAC scenario assumes countrywide CS-ESP only, with a capture efficiency of 42%, and speciation split of 67 and 33% between Hg^0 and $Hg^{2+/P}$. Currently, there is limited air pollution regulation of stationary combustion sources in India.[11,52,53] Indian coal is relatively low sulfur;[11,54] consequently, SO_2 controls have not been a regulatory priority, with the exception of requirements for stack height, which do not capture SO_2, but lessen its local impacts.[11,52] However, emissions limits for particulates have been in place since 1981, resulting in widespread installation of ESP systems.[11]

Minamata Flexible (MF) Scenario. Under the MF scenario, Chinese plants adopt SCR technology to control for NO_x, in addition to existing PM and SO_2 controls. Percent mercury reduction of control technology thus increases from 69% to 82%. There is a small shift toward divalent and particulate-bound emissions compared to NAC, as SCR promotes oxidation to these species. This scenario is based on existing policy. In 2011, the Chinese Ministry of Environmental Protection adopted the new Emission Standard of Air Pollutants for Thermal Power Plants (GB 13223-2011) for 2012–2017.[55] Under this standard, newly constructed plants in China face more stringent emissions limits for sulfur dioxide, nitrogen oxides, and particulates, with tighter standards for existing plants being imposed in 2014[55]—though enforcement remains a potential challenge.[56,57] Achieving the new standards will require almost all plants to install FGD, SCR, and increased use of ESPs and FFs.[12,49] Addressing mercury pollution was also a stated goal in China's 12th Five Year Plan (2011–2015). This is reflected in the Emission Standard, which limits mercury emission to 30 $\mu g/m^3$. This limit is an order of magnitude weaker than limits in the U.S. However, the stringency of requirements for SO_2, NO_x, and PM are likely to lead to large cobenefits for mercury.[12,49] Consequently, most plants will be well below the emission limit set for mercury without mercury-specific control technologies—though results will depend on coal attributes and plant processes, which can vary plant to plant.[49]

For India, the MF scenario assumes pre-combustion measures like plant efficiency gains and coal washing, in addition to ESP, increasing $f_{capture}$ from 42% to 58%. Since these measures are pre-combustion, we assume that the speciation breakdown does not change from NAC. In contrast to China, attributes of Indian coal and India's domestic power sector may make "end-of-pipe" mercury control more costly for India, with fewer opportunities for alignment with existing domestic environmental policies. Domestically sourced coal provides ~70% of India's heat and electricity generation.[11] Indian coal has a high reported ash content,[54] exacerbating already low plant efficiency and increasing mercury emissions per unit energy.[11] Moreover, the mercury content of Indian coal may be highly variable.[52−54,58]

Recent regulatory efforts for CFPPs have focused on plant efficiency improvements. The Indian Central Pollution Control Board has capped the ash content of coals used for thermal power generation at 34%, with all plants required to comply by 2016.[59] This requirement has led to ongoing increases in coal washing and blending. Plans for capacity expansion have also highlighted the need to increase unit sizes, and adopt supercritical and integrated gasification combined cycle technology in new installations—technologies that can offer efficiency gains of up to ~2 and 10%, respectively.[18]

Minamata Strict (MS) Scenario. Under the MS scenario, we assume a further increase from 82% to 90% capture efficiency in China, based on substituting FF for ESP. The actions currently being undertaken by U.S. CFPPs to comply with a recent U.S. rule targeting mercury emissions illustrate possible mercury control strategies beyond cobenefits from standard PM, SO_2, and NO_x control. To comply with the new rule, U.S. CFPPs are expected to double the application of FF, and increase ACI by 15-fold, compared to a baseline case including just air quality policies.[50] We focus on the substitution of FF for ESP for our MS scenario for China, given that it is less costly than ACI.[46]

For India, we assume an increase in mercury reduction from 58% to 70.5% through the introduction of FGD. With this technology, there is a large shift toward emissions in gaseous elemental form (93% of emissions). FGD is considered a logical next step in pollution control devices after those for PM control,[49,53] and this scenario, and its associated capture efficiency, is consistent with existing proposed mercury emissions reduction strategies for India.[53]

Emissions. Figure 1 shows projected emissions of total mercury (THg) under the three technology scenarios for the A1B (left) and B1 (right) energy scenarios. Colored bars represent emissions from the power sector, by species, while gray bars represent contributions from all other domestic anthropogenic sources (projected following Streets et al.[15]). Emissions for India are shown in red, while those for China are shown in blue.

For India, under MF with an A1B development scenario, projected emissions are 24% lower than under NAC (468 vs 619 $Mg{\cdot}y^{-1}$). Moving from MF to MS, emissions are an additional 26% (122 $Mg{\cdot}y^{-1}$) lower. All of these additional avoided emissions are as $Hg^{2+/P}$, with emissions of Hg^0 roughly constant between MF and MS. Under all scenarios, projected 2050 Indian power sector emissions are large increases over 2010 estimated emissions (49 $Mg{\cdot}y^{-1}$).[16] In China, projected emissions are 36% lower under MF compared to NAC (156 vs 247 $Mg{\cdot}y^{-1}$), and a further 33% (51 $Mg{\cdot}y^{-1}$) lower between

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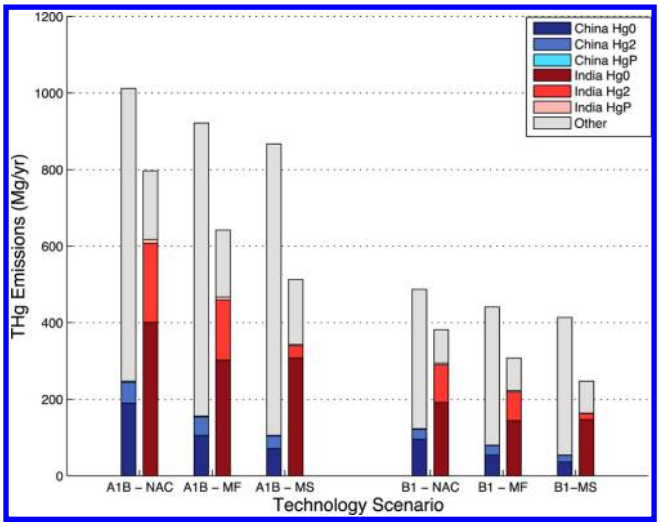

Figure 1. Projected 2050 emissions of THg (all species) under technology scenarios. Emissions for China (blue) and India (red) are presented under the three technology scenarios for the A1B and B1 development scenarios. Colored bars represent emissions from the power sector, by species. Gray bars represent contributions from all other domestic anthropogenic sources.

MF and MS. Projected 2050 Chinese power sector emissions exceed the 2010 estimate (97 $Mg \cdot y^{-1}$) for all technology scenarios,[16] though under MS the increase (+8 $Mg \cdot y^{-1}$) is within the uncertainty range of the current estimate (which ranges up to 139 $Mg \cdot y^{-1}$).

We also consider the same technology scenarios to 2050 under a B1 development scenario. Assuming no technological change (NAC) but a global transition away from coal, we project that power sector emissions are 61% (150 Mg) lower relative to A1B in China, and 60% (370 Mg) lower in India. The difference between 2050 A1B and B1 total emissions, including other sectors, under the NAC scenario is 525 Mg for China and 417 Mg for India.

Impacts on Deposition. Figure 2 shows modeled differences in annual gross deposition flux between technology scenarios (NAC − MF and MF − MS) for 2050 A1B. Spatially averaged values for the countries (total deposition mass/total area), along with their mass equivalents, are presented in SI, Table S2. Differences in average annual deposition flux over China and India between NAC and MF are approximately 4.6% (2.1 $\mu g \cdot m^{-2}$) and 42% (13.4 $\mu g \cdot m^{-2}$) of modeled present-day domestic deposition, respectively (shown in SI, Figure S2). The differences between MF and MS are 4.8% (2.2 $\mu g \cdot m^{-2}$) and 82% (26.1 $\mu g \cdot m^{-2}$) of modeled present-day deposition in China and India.

We also find changes in modeled deposition flux in other regions. Average annual deposition fluxes over both the U.S. and Europe change by approximately −5% of modeled present day annual average deposition between NAC and MS (−1.2 $\mu g \cdot m^{-2}$ and −0.7 $\mu g \cdot m^{-2}$, respectively). We also examine deposition to ocean basins, given that mercury exposure for many global populations is through marine fish, and it is the predominant U.S. exposure source.[60,61] Moving from the NAC to the MS scenario changes deposition to the North Pacific by −6.6% of modeled present day levels (−1.0 $\mu g \cdot m^{-2}$), and by

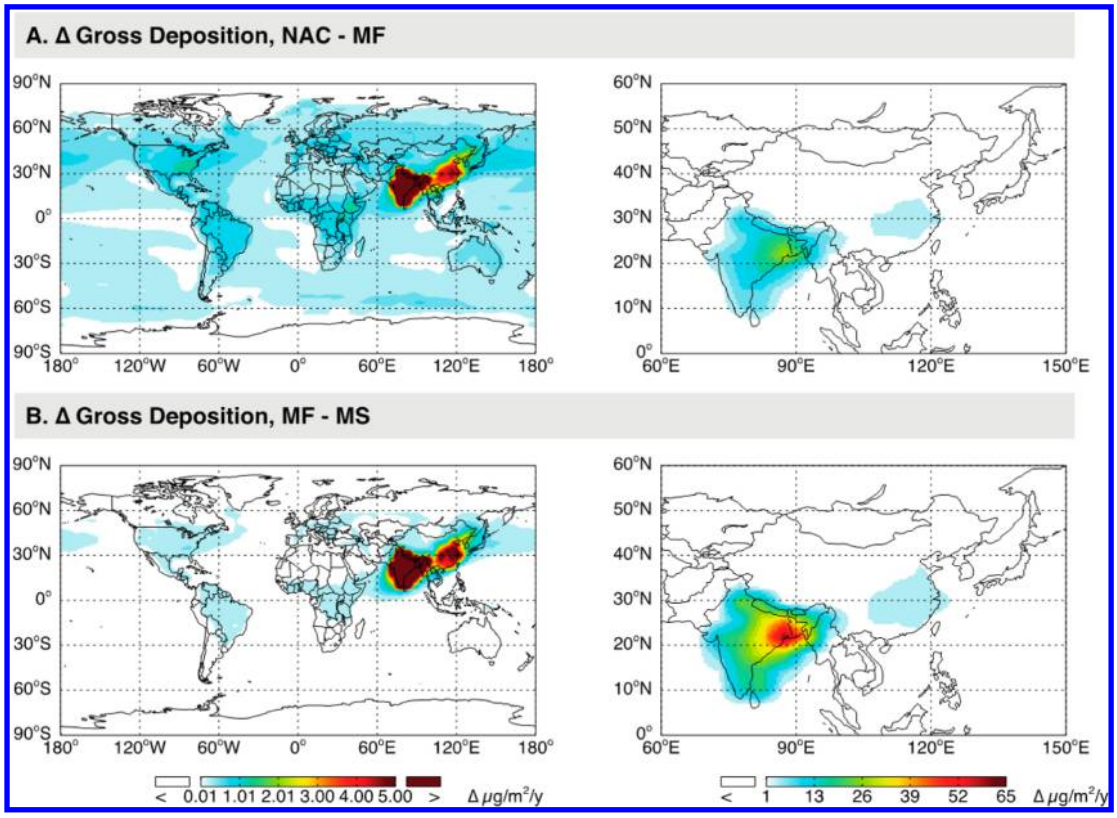

Figure 2. Modeled differences in annual gross deposition flux between technology scenarios for 2050 A1B. Note that the color bar for figures on the left-hand side, showing global extent, saturate at a maximum of 5 $\mu g \cdot m^{-2} \cdot y^{-1}$, while right-hand side figures, zoomed in on Asia, show a minimum of 1 $\mu g \cdot m^{-2} \cdot y^{-1}$. See SI, Figure S3, for a version with a log color scale.

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−9.6% (−0.8 $\mu g \cdot m^{-2}$) for the South Pacific/Indian Oceans. These source regions contribute the two largest shares to U.S. mercury intake from commercial fish.[62,63]

Assuming reduction of Hg^{2+} to Hg^0 in power plant plumes (IPR) results in simulated CFPP emissions contributing more strongly to extra-regional deposition. Simulated differences in annual gross deposition flux under the IPR assumption are shown in SI, Figure S4. With IPR, the deposition benefit of moving from NAC to MS decreases by 57% over India (from 39.5 to 17 $\mu g \cdot m^{-2}$), while the benefit increases by 25% over the U.S. (from 1.2 to 1.5 $\mu g \cdot m^{-2}$), compared to the without IPR case. For China, the benefit of moving from NAC to MS increases by 9% compared to the without IPR case (from 4.3 to 4.8 $\mu g \cdot m^{-2}$), because of reduced contributions from Indian emissions. The North Pacific and Atlantic oceans see larger benefits between NAC and MS compared to the without IPR case, while there is a small decrease in benefit for the South Pacific/Indian Ocean.

Sensitivity Analysis. We calculate the response ratio, α, the average percent change in mean deposition flux to a region due to a 1% change in capture efficiency or emissions speciation, based on ±20% perturbations from the MF scenario. Based on this analysis, changes in assumed domestic capture efficiency in the power sector lead to larger responses in domestic deposition flux for India than China (α = 0.561 and 0.382, respectively), reflecting India's larger share of total emissions from the power sector. We also find that speciation of power sector emissions plays a larger role in domestic deposition in India compared to China (α = 0.663 and 0.133), as most emitted Indian $Hg^{2+/P}$ is deposited domestically. For China, because Chinese emissions sources are predominantly in the east, and transport tends eastward, less of the benefit of reduced emissions is captured over the Chinese landmass itself. We find that a 1% change in Chinese and Indian power sector capture efficiencies lead to approximately equivalent responses in U.S. deposition. The larger influence of transpacific transport of Chinese emissions on U.S. deposition balances the fact that a 1% change in $f_{\text{capture India}}$ represents a larger absolute change in emissions. Additional discussion of sensitivity analysis results is provided in the SI. Tabulated α values are given in SI, Table S3. We also relate deposition responses by mass to the mass change in total mercury emissions in each country, ΔTHg, and the mass conversion of emissions from Hg^0 to Hg^{2+}, ΔHg0 → Hg2 in SI, Table S4.

■ DISCUSSION AND POLICY IMPLICATIONS

We project that the magnitude of avoided emissions and deposition from implementing more effective control technologies for mercury in Asian CFPPs is large (as a fraction of current emissions levels), even when considering a scenario where the Minamata Convention's requirements for mercury are consistent with existing, non-mercury-specific domestic pollution and energy policy plans (MF). Even such a flexibly designed Convention could lead to avoided emissions of 242 Mg in India and China in 2050 compared to a scenario with no additional technology, which is equivalent to ∼12% of the total global anthropogenic emissions in 2010.[8] More than 60% of these avoided emissions are from India, highlighting the importance of India's participation in any global requirements for CFPPs. Despite the fact that mercury reduction efficiencies in Chinese CFPPs are likely to be higher, the larger emissions avoided in India reflect the fact that power sector emissions contribute a larger fraction of total emissions in India across technology scenarios (67−78%) than in China (12−24%), where emissions from industrial processes represent the largest fraction of 2050 projected emissions.

The benefits of these avoided emissions in terms of avoided deposition are concentrated regionally in Asia, particularly in India, where the difference reaches a maximum of ∼30 $\mu g \cdot m^{-2}$. However, deposition differences between the NAC and MF technology scenarios in the U.S. and Europe, are also ∼5% of current deposition. Moreover, global benefits occur through avoided enrichment to oceans, particularly to the Pacific and Indian Oceans, from which many of the fish in the global seafood market are sourced.[60,62] Qualitatively, this distribution of benefits is robust to the IPR assumption (see SI, Figure S3), though regions outside of Asia receive a larger share of benefits.

The definition of BAT for CFPPs under the Convention is being discussed in a technical working group, and will also be discussed during future conferences of parties. We show that these definitions can have a substantial impact on environmental mercury: differences in stringency of required control technologies (MF vs MS) could result in emissions differences in India and China of 173 Mg combined in 2050, which is roughly the total estimated emissions from India in 2005.[9] Increased stringency in the definition of BAT could also avoid growth in power sector emissions over present-day levels in China, though this is unlikely for India. The majority of benefits of increased stringency in terms of deposition are captured by India and China, suggesting that there is a strong domestic incentive for these two countries to take further actions beyond a flexible BAT. In the model simulation, 73% by mass of the 173 Mg avoided emissions between MF and MS would have been deposited in India and China. This pattern is due to the speciation of modeled emissions reductions between MF and MS, as all Indian emissions reductions are in $Hg^{2+/P}$, forms that contribute most strongly to regional pollution.

For the power sector, the emissions gap between technology scenarios (A1B:NAC − A1B:MS) is comparable to, but smaller than, that between energy and development scenarios (A1B:NAC − B1:NAC), with the former being 94% of the latter for China, and 74% for India. This result underscores the importance of energy and development trajectories for mercury emissions. While moving toward more effective mercury control technologies in China and India can mitigate some of the emissions growth associated with aggressive increases in coal consumption for the power sector, avoiding coal consumption and transitioning toward less carbon-intensive energy sources is likely necessary for reducing emissions from present-day levels. Both Indian and Chinese governments have stated goals to increase renewable capacity, and to pursue demand-side management of electricity, for instance through energy efficiency targets.[18,64] A recent agreement between China and Russia on natural gas also suggests a shift toward Russian-exported gas-fired power plants in the future. Avoided consumption of coal could also be an important facet of a mercury emissions mitigation strategy.

Several assumptions made in the chemical transport modeling present opportunities for future exploration. Without locational data on Chinese and Indian power sector emissions, emissions were scaled uniformly across countries, based on 2005 spatial distributions.[65] This approach does not capture real spatial patterns, particularly as substantial new coal generation capacity will be built by 2050. The distribution of these new plants, particularly on the East−West axis, may have implications for transboundary transport to countries down-

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wind; however, the present analysis still offers insight into global distributional impacts, at the regional scale. Better spatial data could also provide further insight into the deposition patterns of divalent mercury within India and China.

Our projections suggest that under the Convention, Asian CFPP emissions will be avoided, but will likely increase from present-day, consistent with previous estimates that Minamata will result in avoided emissions increases;[66] however, total deposition benefits from these avoided emissions are likely to be larger than our estimates, which only take into account the impact of primary emissions changes. The mercury simulation used for this study does not completely account for the legacy impacts of anthropogenic emissions: primary anthropogenic emissions once deposited to terrestrial and aquatic ecosystems join a legacy pool of mercury that can continue to cycle through air, water, and land. Because primary emissions also enrich legacy pools in ocean and soil reservoirs, by 2050, re-emissions from these pools may contribute >50% of global deposition.[67,68] However, while the legacy anthropogenic contribution to total deposition by 2050 under an A1B scenario may be significant,[68] we address here the *difference* between technology scenarios. The influence of legacy emissions is less substantial in the difference calculation, which is small compared to projected overall changes in the global mercury budget. Using a global box model developed by Amos et al.,[67,69] we estimate that accounting for legacy effects could increase deposition differences between NAC and MF and MF and MS by ∼30% by mass (additional details are provided in SI, Table S5). Because legacy pools are likely to increase global background concentrations of mercury, their inclusion will not substantially change the modeled spatial patterns of deposition.

The effects of climate change could also have additional impacts on global mercury transport that we do not account for in this analysis. Climate change is likely to impact mercury biogeochemical cycling through increased volatilization from ocean and soil reservoirs (which will increase the influence from legacy mercury), increased plant respiration and wildfires, changes to oxidant concentrations, and changes to food webs, among other factors.[70−72] Exploring the coupled effects of direct anthropogenic mercury emissions changes and climate change will be critical for understanding to what extent there will be a "climate penalty" on policy efforts to reduce mercury pollution.[70] These interactions between future energy and development trajectories, climate change, and mercury biogeochemical cycling suggest that beyond technology choices for the power sector itself, a broader consideration of energy and development choices will be necessary to understand future mercury emissions trajectories and their impacts.

■ ASSOCIATED CONTENT

Supporting Information

Expanded background, methods, deposition results, and discussion on sensitivity analysis and legacy anthropogenic effects. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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