

Article

Trade-induced atmospheric mercury deposition over China and implications for demand-side controls

Long Chen, Jing Meng, Sai Liang, Haoran Zhang, Wei Zhang, Maodian Liu, Yindong Tong, Huanhuan Wang, Wei Wang, Xuejun Wang, and Jiong Shu

Environ. Sci. Technol., **Just Accepted Manuscript** • DOI: 10.1021/acs.est.7b04607 • Publication Date (Web): 12 Jan 2018

Downloaded from <http://pubs.acs.org> on January 29, 2018

Just Accepted

“Just Accepted” manuscripts have been peer-reviewed and accepted for publication. They are posted online prior to technical editing, formatting for publication and author proofing. The American Chemical Society provides “Just Accepted” as a free service to the research community to expedite the dissemination of scientific material as soon as possible after acceptance. “Just Accepted” manuscripts appear in full in PDF format accompanied by an HTML abstract. “Just Accepted” manuscripts have been fully peer reviewed, but should not be considered the official version of record. They are accessible to all readers and citable by the Digital Object Identifier (DOI®). “Just Accepted” is an optional service offered to authors. Therefore, the “Just Accepted” Web site may not include all articles that will be published in the journal. After a manuscript is technically edited and formatted, it will be removed from the “Just Accepted” Web site and published as an ASAP article. Note that technical editing may introduce minor changes to the manuscript text and/or graphics which could affect content, and all legal disclaimers and ethical guidelines that apply to the journal pertain. ACS cannot be held responsible for errors or consequences arising from the use of information contained in these “Just Accepted” manuscripts.

1 **Trade-induced atmospheric mercury deposition over China and implications for**
2 **demand-side controls**

3

4 Long Chen,^{1,2} Jing Meng,³ Sai Liang,⁴ Haoran Zhang,⁵ Wei Zhang,⁶ Maodian Liu,⁵

5 Yindong Tong,⁷ Huanhuan Wang,⁵ Wei Wang,^{1,2} Xuejun Wang,^{*,5} and Jiong Shu ^{*,1,2}

6

7 ¹ Key Laboratory of Geographic Information Science (Ministry of Education), East
8 China Normal University, Shanghai, 200241, China

9 ² School of Geographic Sciences, East China Normal University, Shanghai, 200241,
10 China

11 ³ School of Environmental Sciences, University of East Anglia, Norwich NR4 7TJ,
12 UK

13 ⁴ State Key Joint Laboratory of Environmental Simulation and Pollution Control,
14 School of Environment, Beijing Normal University, Beijing, 100875, China

15 ⁵ Ministry of Education Laboratory of Earth Surface Process, College of Urban and
16 Environmental Sciences, Peking University, Beijing, 100871, China

17 ⁶ School of Environment and Natural Resources, Renmin University of China, Beijing,
18 100872, China

19 ⁷ School of Environmental Science and Engineering, Tianjin University, Tianjin,
20 300072, China

21 **Abstract**

22 Mercury (Hg) is of global concern because of its adverse effects on humans and the
23 environment. In addition to long-range atmospheric transport, Hg emissions can be
24 geographically relocated through economic trade. Here, we investigate the effect of
25 China's interregional trade on atmospheric Hg deposition over China, using an
26 atmospheric transport model and multiregional input-output analysis. In general, total
27 atmospheric Hg deposition over China is 408.8 Mg yr^{-1} , and 32% of this is embodied
28 in China's interregional trade, with the hotspots occurring over Gansu, Henan, Hebei,
29 and Yunnan provinces. Interprovincial trade considerably redistributes atmospheric
30 Hg deposition over China, with a range in deposition flux from -104% to $+28\%$.
31 Developed regions, such as the Yangtze River Delta (Shanghai, Jiangsu, and Zhejiang)
32 and Guangdong, avoid Hg deposition over their geographical boundaries, instead
33 causing additional Hg deposition over developing provinces. Bilateral interaction
34 among provinces is strong over some regions, suggesting a need for joint mitigation,
35 such as the Jing-Jin-Ji region (Beijing, Tianjin, and Hebei) and the Yangtze River
36 Delta. Transferring advanced technology from developed regions to their developing
37 trade partners would be an effective measure to mitigate China's Hg pollution. Our
38 findings are relevant to interprovincial efforts to reduce trans-boundary Hg pollution
39 in China.

40 **1. Introduction**

41 Mercury (Hg), known as a global neurotoxic pollutant, is released into the
42 atmosphere from human activities, such as coal combustion, mining and commercial
43 waste.^{1, 2} The long atmospheric lifetime of elemental Hg⁰ results in its long-range
44 atmospheric transport. Eventually, Hg⁰ is oxidized to divalent Hg^{II}, which is easily
45 deposited in terrestrial and aquatic ecosystems.^{3, 4} Methylation and bioaccumulation
46 of Hg in food webs following deposition adversely affect humans and wildlife,
47 especially prenatally exposed children.⁵⁻⁸ Atmospheric deposition is a critical process
48 following emissions, enhancing the risk of exposure to humans and wildlife.^{9, 10}
49 Elevated Hg in the environment and its associated health impacts led to the launch of
50 the *Minamata Convention on Mercury*¹¹ to reduce global Hg emissions. Given 27% of
51 the global anthropogenic Hg emissions to the atmosphere,¹² Mainland China (termed
52 as China) plays an important role in the global Hg cycle.

53 The Chinese government has already made substantial efforts to reduce
54 atmospheric Hg emissions, mainly through measures related to improving energy
55 efficiency and end-of-pipe controls.¹³⁻¹⁵ For instance, the use of air pollution control
56 devices (APCDs) in coal-fired power plants, especially of electrostatic precipitators
57 (ESP) and fabric filters (FF), flue gas desulfurization towers (FGD), and selective
58 catalytic reduction (SCR), reached 100%, 92.1%, and 83.2% in 2014, respectively.¹⁶
59 In Zn smelters, the average Hg removal efficiency of APCDs reached 93% in 2014.¹⁶
60 As a consequence, around 1002 Mg of Hg from dominant emission sectors was
61 removed in 2010, while 73% of Hg from China's coal-fired power plants was

62 removed in 2014.¹⁶ However, the effects of existing production-side controls have not
63 achieved their desired goals, and Hg pollution is still a serious environmental issue in
64 China. Therefore, new demand-side controls are being proposed, having the same
65 importance as production-side controls. China has established standards for embedded
66 Hg concentrations in domestically consumed goods, such as sphygmomanometers and
67 fluorescent lamps.¹⁷ Meanwhile, the consumption of goods in specific sectors without
68 embedded Hg can lead to upstream Hg emissions throughout the economic supply
69 chains. Given this problem, new demand-side actions,^{18, 19} such as implementing
70 consumption taxes and transferring advanced technology and capital, are being
71 proposed to mitigate the embodied Hg.

72 Demand-side controls concern emissions embodied in the consumption of goods,
73 which are involved in interregional trade. Trade redistributes such emissions, and
74 geographically changes air pollution.²⁰ For instance, the production of trade goods
75 increases emissions locally, while reducing emissions in the consuming regions.
76 Recently, Hg emissions in China relocated by interregional trade have been
77 well-documented at different scales, such as city level,^{21, 22} national level,^{19, 23} and
78 global level.^{18, 24} In particular, a detailed and comprehensive view of the virtual
79 atmospheric Hg emission network among Chinese provinces has been provided by
80 Liang et al.^{19, 23} The previous studies shed light on the virtual transport of Hg
81 emissions via interregional trade. However, there still a gap in understanding how and
82 where the redistributed emissions exert direct risks on humans and wildlife through
83 atmospheric movement and deposition.

84 In this study, we combine an emission inventory with a multiregional input-output
85 model and an atmospheric transport model to simulate the atmospheric Hg deposition
86 embodied in China's interregional trade and investigate how and where the
87 redistributed emissions exert direct risks on humans and wildlife through atmospheric
88 movement and deposition (*SI S2*). We classify contributions from final consumers to
89 the deposition over receptors and assess the potential benefits of various demand-side
90 measures (e.g., implementing consumption taxes and transferring advanced
91 technology) for China. The main novelty of this study is to illustrate the effect of
92 interregional trade and the benefits of relevant demand-side measures on humans and
93 wildlife in deposited areas through atmospheric Hg deposition. Compared with
94 previous studies,^{18, 19, 21-24} this study is more closely connected with health risks on
95 humans and wildlife. Our results are relevant to interprovincial efforts to reduce
96 trans-boundary Hg pollution and provide a scientific basis for the implementation of
97 the *Minamata Convention on Mercury* within China.

98

99 **2. Materials and Methods**

100 **2.1. Production-based emission inventory**

101 Hg emissions from all sectors, including coal combustion, nonferrous metal
102 smelting, cement production, and other production activities, are estimated by
103 multiplying energy usage or product yields by their respective emission factors.
104 Multiplying these values again by Hg speciation profiles, we estimate the emissions
105 of Hg⁰, Hg^{II} and particulate Hg^P. Wu et al.¹⁶ used a technology-based approach to

106 compile the latest and comprehensive estimate of provincial emission factors and
107 associated speciation profiles for all primary anthropogenic Hg sources. Emission
108 factors for dominant sources were estimated based on the probability distribution of
109 Hg concentration in fuel/raw materials and Hg removal via pretreatment and APCDs.
110 We multiply the emission factors and speciation profiles by energy usage or product
111 yield, collected from the China Energy Statistical Yearbook 2011²⁵ and from relevant
112 industrial statistical yearbooks,²⁶⁻²⁹ to estimate provincial and sectoral Hg emissions.
113 In addition to primary anthropogenic Hg sources, its secondary emissions from the
114 disposal of waste/byproducts (i.e., the use of Hg-added products) are identified (102
115 Mg)¹⁸ and included in this study. In the atmospheric transport model, large coal-fired
116 power plants,³⁰ nonferrous metal smelter,³¹ cement plants,³² and iron and steel plants²⁸
117 are treated as point sources and assigned to the corresponding model grid in terms of
118 their productivity. The remaining emissions are regarded as nonpoint sources, which
119 are distributed with high spatial resolution in terms of gridded population density.³³ In
120 the multiregional input-output model, we use the emission inventory at a
121 province-level resolution, including both point and nonpoint sources. The direct Hg
122 emissions of various economic sectors in each province are given in *SI Dataset S3*.

123

124 **2.2. Multiregional Input-Output model**

125 The multiregional input-output (MRIO) model has been widely used for analyzing
126 environmental issues in the context of increasing interregional trade.^{19, 34-38} We use the
127 MRIO table for China in 2010 from Liu et al.,³⁹ to quantify Hg emissions embodied in

128 China's interregional trade. The MRIO table consists of the intermediate input/output
 129 for 30 economic sectors in 30 provinces (excluding Tibet, Taiwan, Hong Kong, and
 130 Macao), provincial final demand and international export, as well as provincial
 131 sectoral total monetary output. Moreover, we remove the column named "others"
 132 (regarded as the error of different statistics^{23, 40}) in the MRIO table. A brief
 133 introduction to the MRIO approach is given below, while more detailed descriptions
 134 of the approach can be found in previous studies.^{37, 38, 41} The MRIO approach assumes
 135 that

$$136 \quad \mathbf{X} = \mathbf{Z} + \mathbf{Y} = (\mathbf{I} - \mathbf{A})^{-1} \mathbf{Y}, \quad (1)$$

137 where \mathbf{X} is a vector of provincial sectoral total monetary output; \mathbf{Y} is a vector of the
 138 provincial sectoral final demand, including final consumption (\mathbf{F}) (i.e., urban
 139 household consumption, rural household consumption, government consumption and
 140 investment) and international export (\mathbf{E}); \mathbf{Z} is the intermediate input/output matrix;
 141 and \mathbf{A} represents the direct requirement coefficient matrix, whose element A_{ij} denotes
 142 the intermediate input from sector i to produce a unit output for sector j . \mathbf{A} can be
 143 used to characterize interregional economic interactions between sectors. $(\mathbf{I} - \mathbf{A})^{-1}$ is
 144 the Leontief inverse matrix, and \mathbf{I} denotes the unity matrix.

145 By multiplying these values by the direct emission intensity (Eq. 2), we obtain
 146 province- and sector-specific consumption-based emissions (\mathbf{C}) with the
 147 corresponding final consumption (\mathbf{F}). We also obtain sector-specific
 148 consumption-based emissions (\mathbf{C}) embodied in export to foreign countries, along with
 149 their corresponding international export (\mathbf{E}). The results are shown in *SI Dataset S3*.

$$150 \quad \mathbf{C} = \boldsymbol{\psi}(\mathbf{I} - \mathbf{A})^{-1} \mathbf{F}, \quad (2)$$

151 where $\boldsymbol{\psi}$ is a vector of direct emission intensity, including emission intensities of
 152 total Hg and three Hg species, which defines the atmospheric Hg emissions per unit of
 153 total output for each sector (Mg of Hg/RMB). Thus, $\boldsymbol{\psi}$ reveals Hg inputs from raw
 154 materials and the efficiency of end-of-pipe Hg controls during production in each
 155 provincial sector.

156 A further application of the basic input-output formula is to quantify emissions
 157 embodied in trade goods between trade partners, including emissions embodied in
 158 exports (EEE) and emissions embodied in imports (EEI). In this study, “export” and
 159 “import” refer to domestic trade or interprovincial trade unless noted, while
 160 “international export” refers to China’s exports to the rest of the world. For reference,
 161 the locations of Chinese provinces are shown in *SI Figure S1*. Hg emissions embodied
 162 in exports of goods from province i to province j (i.e., imports of goods in province j
 163 from province i) can be calculated from:

$$164 \quad \mathbf{C}_{ij} = \boldsymbol{\psi}_i(\mathbf{I} - \mathbf{A})^{-1} \mathbf{F}_j, \quad (3)$$

165 where $\boldsymbol{\psi}_i$ denotes a vector with direct emission intensity for province i but zero for
 166 all others, and \mathbf{F}_j is a vector of final consumption for province j . Using this basic
 167 formula, we calculate EEE and EEI values for each province (*SI S3*). The difference
 168 between EEI and EEE represents the net emission transfer in interprovincial trade,
 169 equal to the difference between consumption-based and production-based emissions.

170 Based on the relative size of emission intensities ($\boldsymbol{\psi}$) for one sector from different
 171 provinces, emissions avoided by imports (EAI) and emissions reduced by

172 technological migration (ERM) are defined. EAI is defined as the additional
173 emissions that a given province would have produced, if all imported goods
174 consumed by the province had been made locally. It can be used to investigate the
175 impact of interregional trade on atmospheric Hg deposition over China. Unlike
176 previous studies,^{40, 42} we assume that the production of the same goods releases
177 different amounts of Hg among provinces mainly due to the difference of
178 technologies. The total emissions avoided by imports for province i can simply be
179 expressed as:

$$180 \quad EAI_i = \sum C_{ij} \cdot \frac{\psi_i}{\psi_j} \quad (4)$$

181 In this study, ERM is defined as the reduced EEI for a given province, if the
182 province transfers a smaller ψ to its trade partners. It is assumed to represent the
183 impact on atmospheric Hg deposition over China of transferring advanced technology
184 from developed provinces to their trade partners. The reduced emissions embodied in
185 imports of province i from province j can simply be expressed as:

$$186 \quad ERM_{ij} = C_{ij} \cdot \begin{cases} 1 - \frac{\psi_i}{\psi_j} & \psi_i < \psi_j \\ 0 & \psi_i \geq \psi_j \end{cases} \quad (5)$$

187 The detailed calculation of EAI and ERM are outlined in *SI S3*.

188

189 **2.3. Atmospheric chemical transport model**

190 We simulate the atmospheric Hg deposition embodied in interregional trade over
191 China using the GEOS-Chem chemical transport model (version 9-02;
192 <http://geos-chem.org>). The model is a global 3-D atmospheric model, coupled to a

193 2-D surface slab ocean and a 2-D soil reservoir.^{43, 44} The model is driven by the
194 GEOS-5 assimilated meteorological fields, developed by the NASA Global Modeling
195 and Assimilation Office (GMAO). We implement a nested-grid capability with high
196 resolution over China within the GEOS-Chem model, based on the methods in Zhang
197 et al.⁴⁵ and Wang et al.⁴⁶ The nested model has $1/2^\circ \times 2/3^\circ$ horizontal resolution, and
198 47 vertical levels from the surface to 0.01 hPa. Three Hg species (i.e., Hg^0 , Hg^{II} , and
199 Hg^{P}) are tracked with oxidation of Hg^0 by Br atoms, photoreduction of Hg^{II} in
200 droplets and gas-particle partitioning between Hg^{II} and Hg^{P} .^{43, 47} We first conduct a
201 global $4^\circ \times 4.5^\circ$ simulation for the period 2008–2010 to determine lateral boundary
202 conditions for the nested model every 3 hours. Then we run the nested model for the
203 study year (2010), with an initial spin-up time spanning the last three months of 2009.
204 The global simulation is driven by the emission inventory of China and the
205 AMAP/UNEP (Arctic Monitoring and Assessment Programme/United Nations
206 Environment Programme) global anthropogenic emission inventory out of China.¹²
207 The outputs are archived monthly, and used to calculate average atmospheric Hg
208 deposition for the various regions of interest. We first conduct simulations with
209 production-based emissions to evaluate the nested model's performance against a
210 series of observations from published literature. Details of the evaluation are given in
211 *SI S4*.

212 To identify contributions from each source, an explicit tagging technique is added
213 to the GEOS-Chem model, adopted from previous applications used to estimate
214 source-receptor relationships for SO_2 , BC and $\text{PM}_{2.5}$ on continental and national

215 scales.^{34, 48, 49} Throughout this tagging approach, Hg is emitted from a series of
216 sources, is tagged and explicitly tracked using additional model variables in a single
217 model simulation. Breaking from traditional sensitivity approaches, this method
218 avoids modifying Hg emissions to maintain local integration of atmosphere and
219 climate. This method also avoids the assumption that the response to perturbations is
220 linear and saves computational costs by using a sensitivity approach.

221

222 **3. Results and Discussion**

223 **3.1. Atmospheric Hg emissions and consequent deposition embodied in trade**

224 China releases a total of 641.7 Mg of Hg to the atmosphere in 2010, of which 24.2
225 Mg of Hg is from residential coal combustion and use of Hg-added products, which is
226 not involved in the economic activities. Thus, 617.5 Mg of Hg is included in the
227 MRIO analysis herein. For consumption-based emissions, 81.9% (505.5 Mg) of Hg is
228 induced by domestic consumption, while the remaining amount is induced by foreign
229 consumption. A comparison of production-based and consumption-based Hg
230 emissions in 2010 for 30 Chinese provinces is shown in Figure 1A. The different
231 distribution patterns between the production-based and consumption-based emissions
232 observed in 2010 are similar to the results for 2007 presented in Liang et al.¹⁹ Some
233 developing provinces, such as Henan, Shandong, Gansu, Yunnan, Hebei and Inner
234 Mongolia, which mainly produce primary and semi-manufactured products (e.g.,
235 fossil fuels, metals, and nonmetallic mineral products), are located upstream of the
236 Chinese economic supply chains. These provinces have larger production-based

237 emissions than consumption-based emissions. For instance, Henan ranks first, with a
238 total of production-based emissions of 65.8 Mg, followed by Shandong (55.3 Mg),
239 Gansu (45.4 Mg), and Yunnan (43.7 Mg). In contrast, developed provinces, which are
240 located downstream of the Chinese economic supply chains, such as Beijing, Tianjin,
241 Shanghai, Jiangsu, Zhejiang, and Guangdong, have larger consumption-based
242 emissions. The difference between production-based and consumption-based
243 emissions reflects the transfer of emissions via trade. Figures 1B and 1C illustrate the
244 emissions embodied in exports and imports of goods and services for each province. It
245 shows that eleven provinces are net embodied Hg importers, while the other provinces
246 are net embodied exporters. Among the provinces, the largest importer is Zhejiang
247 (9.9 Mg), and the largest exporter is Henan (30.1 Mg). Similar to previous studies,^{19,}
248 ²³ we also find that emissions flow from the southeast coast to inland regions as a
249 result of interprovincial trade.

250 The total atmospheric Hg deposition over China simulated by the model is 408.8
251 Mg yr⁻¹, with 59% of this amount reflecting China's anthropogenic sources (*SI*
252 *Dataset S8*, Figure S3) and the remaining being from natural sources or foreign
253 anthropogenic sources. Thus, the Hg deposition related to China's anthropogenic
254 sources accounts for 60%–90% of the total Hg deposition over most of China (Figure
255 2A). China's anthropogenic sources also contribute to downwind deposition over the
256 Northwest Pacific Ocean and other East Asian countries. However, this deposition
257 decreases rapidly a certain distance away from mainland hotspots. Of the total
258 deposition over China, 48% is induced by domestic consumption, including

259 interprovincial trade, while 11% is induced by foreign consumption via international
260 export. This indicates the importance of domestic consumption in determining
261 atmospheric Hg deposition over China. Furthermore, 32% of the total deposition over
262 China is embodied in exports or imports of goods among China's trade partners (*SI*
263 *Dataset S8*), with hotspots occurring in Gansu, Henan, Hebei and Yunnan (Figure 2B).
264 Of the trade-induced deposition over China, the deposition embodied in
265 interprovincial trade (87.5 Mg yr^{-1}) is larger than the deposition embodied in
266 international export (43.6 Mg yr^{-1}) (*SI Dataset S8*). The largest hotspot occurs over
267 central provinces (e.g., Gansu, Henan, and Hebei) for interprovincial trade, but over
268 southern provinces (e.g., Yunnan, and Guangdong) for international export (Figures
269 2C and 2D). The substantial embodied atmospheric Hg deposition reveals there is a
270 profound influence of interregional trade and trans-boundary transport on atmospheric
271 Hg pollution in China.

272

273 **3.2. Redistribution of atmospheric Hg deposition by trade**

274 Using EAI, we investigate the impact of China's interregional trade on the
275 atmospheric Hg deposition over China, especially impacts of interprovincial trade.
276 Figure 3 shows that interprovincial trade considerably redistributes atmospheric Hg
277 deposition over China, with a range in deposition flux from -104% to $+28\%$.
278 Atmospheric Hg deposition over most central and eastern provinces is enhanced,
279 except over the Yangtze River Delta (i.e., Shanghai, Jiangsu, and Zhejiang), the
280 Beijing-Tianjin region and the southeast coast, where Hg deposition is diminished

281 (Figure 3A). In the case of southwestern and northwestern provinces, atmospheric Hg
282 deposition is diminished over Yunnan, Guizhou, Sichuan, Shaanxi, Ningxia and
283 Xinjiang, but considerably enhanced over Gansu. The provinces having reduced Hg
284 deposition show that a large decrease in deposition induced by their trade activities
285 can compensate for the increase induced by the trade activities of other provinces
286 (Figure 3B). Large EAIs for these provinces result in large reductions in deposition
287 over these provinces (*SI Dataset S5*). For developed regions (e.g., the Yangtze River
288 Delta and the Beijing-Tianjin region), large EEs result in large EAIs. For developing
289 provinces (e.g., Yunnan, Guizhou, and Sichuan), small EEs but less advanced
290 production technology result in large EAIs (Figure 3B).

291 The pattern emerging in Figure 3B shows aggregated decreased groups along the
292 diagonal, while increased groups away from the diagonal. The aggregated decreased
293 groups along the diagonal indicate that all provinces could avoid Hg deposition over
294 their geographic boundaries by importing goods. In the case of some provinces (e.g.,
295 Hebei, Zhejiang, and Gansu), their neighbors (e.g., Beijing, Shanghai, and Ningxia,
296 respectively) could also avoid Hg deposition due to their imports of goods. The
297 largest such decrease occurs between neighbors, Zhejiang and Shanghai (-8.0%). The
298 reduced deposition over their neighbors induced by their large EAIs could
299 compensate for the increased deposition over their neighbors induced by their EEs.
300 However, in the case of remote receptors, the reduced deposition induced by EAIs of
301 all provinces could not compensate for the increased deposition induced by their EEs.
302 In particular, the Yangtze River Delta and Guangdong both cause marked increases in

303 Hg deposition over other provinces. Importing goods by developed regions of China
304 could avoid large amounts of Hg deposition over their geographic boundaries, but
305 cause substantially additional Hg deposition over developing provinces, which reveals
306 a need for implementing demand-side measures in these developed provinces.

307 Figure 3C illustrates the variation related to trade-induced Hg deposition over the
308 whole country from each source province and shows two opposite patterns. Importing
309 goods by developed provinces, such as Beijing, Shanghai and Guangdong, causes
310 additional Hg deposition over the whole country, and Guangdong ranks first with an
311 increase of 1.3%. Importing goods by developing provinces, such as Henan, Yunnan
312 and Shaanxi, allows the whole country to avoid Hg deposition, and Yunnan ranks first
313 with a decrease of -1.5% . Thus, in terms of trade-induced deposition, the whole
314 country would benefit from cooperation between demand-side measures in developed
315 provinces and end-of-pipe measures in developing provinces.

316

317 **3.3. Implications for demand-side Hg controls**

318 Mercury controls have been the goal of both national and global policymakers.^{12,}

319 ⁵⁰ In addition to production-side measures, the Chinese government is suggested to
320 put demand-side Hg measures into action. Substantial embodied atmospheric Hg
321 deposition and associated profound redistributions of the deposition by trade indicate
322 there is a large potential for effective implementation of demand-side Hg controls to
323 reduce Hg pollution in China. Given this potential, we classify the source
324 contributions of atmospheric Hg deposition and investigate the effects under

325 substitution of emission intensity. In this way, we assess the potential benefits of
326 specific demand-side measures under consideration in China.

327

328 **3.3.1 Implications inferred from classification of source contributions**

329 By combining atmospheric movement and emission flows in trade, the source of
330 atmospheric Hg deposition can be classified according to its on-site emission location
331 and its final consumer of the relevant products. The classification of consumer
332 sources is informative for judging the priority of cooperative Hg mitigation. We
333 introduce changes in atmospheric Hg deposition in a receptor region resulting from a
334 unit of final consumption in a source (in $\% / (\text{trillion RMB} \cdot \text{yr}^{-1})$), as an indicator for
335 the classification of source contributions.

336 Figure 4A shows the emission flows in trade, while Figure 4B illustrates the
337 classification of source contributions to receptors' atmospheric Hg deposition. The
338 patterns in Figure 4A and 4B both show aggregated groups along the diagonal and
339 row-like distribution of eminent contributions. When discussing atmospheric
340 deposition, some southwestern provinces (e.g., Chongqing, Guizhou, and Yunnan) and
341 northwestern provinces (e.g., Gansu, and Ningxia) show significant local influence,
342 indicating local consumption and emissions, as well as a near-source regional
343 deposition. Some groups, such as Gansu-to-Ningxia ($37.5\% / (\text{trillion RMB} \cdot \text{yr}^{-1})$),
344 Gansu-to-Shaanxi ($9.9\% / (\text{trillion RMB} \cdot \text{yr}^{-1})$) and Guizhou-to-Chongqing
345 ($8.3\% / (\text{trillion RMB} \cdot \text{yr}^{-1})$), have small values in Figure 4A, but large values in Figure
346 4B. This indicates the influence of atmospheric movement from upwind provinces to

347 downwind provinces. Bilateral interaction among provinces is strong in some specific
348 regions, such as within the Jing-Jin-Ji region (Beijing, Tianjin, and Hebei) and the
349 Yangtze River Delta, because of their close geographical locations and frequent
350 economic trade (Figure 4B). These regions require joint control measures. The final
351 consumption of the Yangtze River Delta is the most effective driver for Hg deposition
352 over most of China, making a contribution of 2.0%/(trillion RMB·yr⁻¹). In contrast,
353 receptors, such as Tianjin, Shanghai, Sichuan, Inner Mongolia, Shaanxi and Qinghai,
354 have similar local contributions and contributions from one other province to Hg
355 deposition (i.e., in these cases: Hebei contributes to Tianjin, Zhejiang to Shanghai,
356 Chongqing to Sichuan, Ningxia to Inner Mongolia, and Gansu to Shaanxi and
357 Qinghai), indicating the need for interprovincial collaboration on Hg controls.

358 As a developing country undergoing rapid economic development, it is unrealistic
359 to reduce the consumption of all goods and services in China. However, changing
360 consumer behavior by shifting consumption of goods and services with large
361 embodied Hg emissions to consumption of those that have lower embodied Hg
362 emissions would be an effective way of reducing Hg deposition over China. A
363 promising method would be to implement consumption taxes based on embodied Hg
364 emissions. This would influence the consumption pattern of consumers, resulting in
365 less embodied Hg consumption and lower Hg emissions,¹⁸ consequently, reducing
366 atmospheric Hg deposition. Regional differentiation and collaboration in the
367 implementation of consumption taxes would need to be considered, according to our
368 results. In southwestern and northwestern provinces, measures taken in local regions

369 may primarily benefit local environment. However, in northern and eastern provinces,
370 measures taken in local regions will benefit the environment in both local and
371 neighboring regions equally (e.g., Hebei-to-Tianjin and Zhejiang-to-Shanghai). The
372 Yangtze River Delta, in particular, is a priority area, where demand-side measures
373 would effectively benefit most of China.

374

375 **3.3.2 Implications inferred from hypothetical substitution of emission intensity**

376 Developed provinces, such as Beijing, Tianjin, Shanghai, Jiangsu, Zhejiang, and
377 Guangdong, located downstream of the Chinese economic supply chains, would be
378 responsible for the provinces with greater influence from their trade activities.
379 Therefore, based on a hypothetical substitution of emission intensity, we introduce
380 ERM to investigate the impact of a hypothetical transfer of advanced technology from
381 developed provinces to their trade partners on atmospheric Hg deposition over China.
382 Figure 5 illustrates the spatial distributions of reductions in atmospheric Hg
383 deposition under such a transfer scheme for the six developed provinces. Though
384 transferring advanced technology to trade partners, developed provinces would cause
385 considerable reductions in atmospheric Hg deposition over China. However, the
386 spatial distributions of these reductions are different among the six provinces. The
387 measure from Beijing would cause reductions over the northern provinces, especially
388 Hebei (-3.3%) and the Beijing-Tianjin region (-2.2%). The impact of the measure
389 from Tianjin has a similar distribution to that of Beijing, but the reductions would be
390 smaller. Measures from the Yangtze River Delta (i.e., Shanghai, Jiangsu, and Zhejiang)

391 cause the most extensive reductions over China. In the case of Shanghai, the reduction
392 hotspot is over the central and eastern provinces, such as Henan (-2.6%) and Zhejiang
393 (-2.2%). In case of Jiangsu, the reduction hotspot is over the northwestern provinces,
394 such as Gansu (-3.7%). In addition to the northwestern provinces, the measure from
395 Zhejiang also causes a reduction over the southwestern provinces. Unlike the
396 Beijing-Tianjin region and the Yangtze River Delta, the measure from Guangdong
397 could reduce Hg deposition mainly over the southwestern provinces, such as Yunnan
398 (-4.8%). We also investigate the same measure for other provinces and find the
399 reductions of deposition induced by developed provinces are larger than developing
400 provinces (*SI S5*).

401 As the developed regions in China, the Beijing-Tianjin region, the Yangtze River
402 Delta and the Pearl River Delta appear to contribute large additional loads to Hg
403 deposition over developing provinces, and they are gave a priority need of
404 implementing demand-side measures (Figure 3). Furthermore, they have more
405 advanced technology than developing provinces. If they transferred advanced
406 technology to their trade partners, then atmospheric Hg deposition over these
407 developing regions would be considerably reduced. Therefore, importing goods
408 coupled with transfer of advanced technology in developed regions would be an
409 effective measure to mitigate China's Hg pollution. Given the different spatial
410 distributions of such reductions, we need to pay attention to measures in different
411 developed regions when mitigating Hg pollution over a specific developing region.

412

413 **3.4 Recommendations and uncertainties**

414 Aiming to improve economic competitiveness, China proposed the ambitious
415 *Made in China 2025* strategy to promote the development of high-end manufacturing
416 sectors.⁵¹ Increasing consumption of products from high-end manufacturing sectors
417 (e.g., equipment, machinery, and vehicles) which require substantial Hg-intensive
418 upstream inputs is predicted to increase Hg emissions in the future.¹⁸ In addition,
419 China has focused on building and developing urban agglomerations since the 11th
420 Five-Year Plan.^{52, 53} This rapid development of urban agglomerations calls for more
421 and more consumption and imports. Such increased consumption and imports
422 suggests that more and more Hg emissions will be exported from coastal developed
423 urban agglomerations to inland developing regions in the future. Above all, if no
424 measures were taken, Hg-related health risks would be aggravated. Reflecting both
425 interprovincial trade and atmospheric movement, inland developing regions would be
426 exposed to more Hg-related health risks than coastal developed regions. To control
427 these aspects of Hg pollution, both end-of-pipe Hg controls and demand-side Hg
428 controls are essential for China, especially the encouragement of interprovincial
429 collaboration on demand-side Hg controls.

430 Our emission and model results are subject to uncertainties from a variety of
431 sources. The calculations of consumption-based emissions, including trade-induced
432 emissions are subject to errors inherent in the production-based emissions and the
433 input–output table. A detailed error analysis for the consumption-based emissions is
434 presented in *SI S6*. We calculate an overall uncertainty of [–27%, 32%] for the

435 consumption-based emissions, including the trade-induced emissions in our study.
436 The atmospheric model results are subject to errors in emission inputs, as well as the
437 model representations of tropospheric chemical processes, especially Hg chemistry,
438 and meteorological processes. The model uncertainties are difficult to quantify and
439 are likely on the order of 30%.⁵⁴ Meanwhile, it is computationally prohibitive to
440 perform Monte Carlo simulations or sensitivity analyses that integrate all errors
441 associated with emissions, the input-output model and the chemical transport model.
442 A common practice to quantify model uncertainties is through an evaluation of the
443 model performance against observations. We outline such an evaluation in *SI S4*. In
444 Figures 2–5, we present our results as a percentage contribution, in which case, the
445 associated uncertainties may be reduced. The uncertainties in atmospheric Hg
446 deposition flux from various simulations may largely offset each other, when their
447 differences are normalized through calculation of a percentage. More field
448 measurements on production emission processes and atmospheric chemical processes
449 involving Hg in the future would reduce uncertainties in emission inventories and
450 chemical transport models, respectively. This would eventually enhance the accuracy
451 of results in this study.

452 Moreover, we use the MRIO database developed by Liu et al.,³⁹ to quantify Hg
453 emissions embodied in China's interregional trade. This makes it easy to compare our
454 results to existing studies^{19, 34, 55, 56} mostly based on the MRIO databases of Liu et al.³⁹
455 It is worth noting that there are also many other MRIO databases for China, including
456 the databases of Shi and Zhang,⁵⁷ Zhang and Qi,⁵⁸ and Wang et al.^{59, 60} It would be

457 important to compare and harmonize these MRIO databases in future studies.

458 In general, revealing embodied Hg deposition within economic supply chains
459 from a consumer perspective helps uncover the underlying trans-boundary drivers of
460 regional Hg pollution and assess the effects of proposed demand-side measures on the
461 mitigation of Hg pollution. Although this study only considers China, the framework
462 developed in this study could be applied to evaluate international collaboration on the
463 demand-side Hg controls between developed and developing countries. Finally, this
464 framework could be applied to other developing countries with large differences in
465 domestic regional development, such as the BRICS countries.

466

467 **Author Information**

468 **Corresponding Authors**

469 *(J.S.) Phone: +86-21-54341198; e-mail: jshu@geo.ecnu.edu.cn.

470 *(X.W.) Phone: +86-10-62759190; e-mail: xjwang@urban.pku.edu.cn.

471

472 **Acknowledgments**

473 The authors would like to thank the editor and anonymous reviewers for their
474 thoughtful comments. This study was funded by the National Natural Science
475 Foundation of China (41701589, 41271055, 41630748, 41571130010, 41471403), and
476 China Postdoctoral Science Foundation Grant (2017M611492). Sai Liang thanks the
477 financial support of the Interdiscipline Research Funds of Beijing Normal University
478 and the Start-up Funds of Beijing Normal University (312232104). All map images

479 are plotted by GAMAP (Global Atmospheric Model Analysis Package, Version 2.17;
480 <http://acmg.seas.harvard.edu/gamap/>). The computation was supported by the High
481 Performance Computer Center of East China Normal University.

482

483 **Supporting Information**

484 The Supporting Information provides additional text, tables, and figures supporting the
485 main text.

486 **References**

- 487 (1) Streets, D. G.; Devane, M. K.; Lu, Z.; Bond, T. C.; Sunderland, E. M.; Jacob, D. J.
488 All-time releases of mercury to the atmosphere from human activities. *Environ. Sci.*
489 *Technol.* **2011**, *45* (24), 10485–10491.
- 490 (2) Horowitz, H. M.; Jacob, D. J.; Amos, H. M.; Streets, D. G.; Sunderland, E. M.
491 Historical mercury releases from commercial products: Global environmental
492 implications. *Environ. Sci. Technol.* **2014**, *48* (17), 10242–10250.
- 493 (3) Lindberg, S.; Bullock, R.; Ebinghaus, R.; Engstrom, D.; Feng, X.; Fitzgerald, W.;
494 Pirrone, N.; Prestbo, E.; Seigneur, C. A synthesis of progress and uncertainties in
495 attributing the sources of mercury in deposition. *Ambio* **2007**, *36* (1), 19–33.
- 496 (4) Corbitt, E. S.; Jacob, D. J.; Holmes, C. D.; Streets, D. G.; Sunderland, E. M.
497 Global source-receptor relationships for mercury deposition under present-day and
498 2050 emissions scenarios. *Environ. Sci. Technol.* **2011**, *45* (24), 10477–10484.
- 499 (5) Mergler, D.; Anderson, H. A.; Chan, L. H. M.; Mahaffey, K. R.; Murray, M.;
500 Sakamoto, M.; Stern, A. H. Methylmercury exposure and health effects in humans: A
501 worldwide concern. *Ambio* **2007**, *36* (1), 3–11.
- 502 (6) Mahaffey, K. R.; Sunderland, E. M.; Chan, H. M.; Choi, A. L.; Grandjean, P.;
503 Mariën, K.; Oken, E.; Sakamoto, M.; Schoeny, R.; Weihe, P.; Yan, C.-H.; Yasutake, A.
504 Balancing the benefits of n-3 polyunsaturated fatty acids and the risks of
505 methylmercury exposure from fish consumption. *Nut. Rev.* **2011**, *69* (9), 493–508.
- 506 (7) Grandjean, P.; Satoh, H.; Murata, K.; Eto, K. Adverse effects of methylmercury:
507 Environmental health research implications. *Environ. Health Persp.* **2010**, *118* (8),
508 1137–1145.
- 509 (8) Karagas, M. R.; Choi, A. L.; Oken, E.; Horvat, M.; Schoeny, R.; Kamai, E.;
510 Cowell, W.; Grandjean, P.; Korrick, S. Evidence on the human health effects of
511 low-level methylmercury exposure. *Environ. Health Persp.* **2012**, *120* (6), 799–806.
- 512 (9) Harris, R. C.; Rudd, J. W.; Amyot, M.; Babiarz, C. L.; Beaty, K. G.; Blanchfield,

- 513 P. J.; Bodaly, R. A.; Branfireun, B. A.; Gilmour, C. C.; Graydon, J. A.
514 Whole-ecosystem study shows rapid fish-mercury response to changes in mercury
515 deposition. *Proc. Natl. Acad. Sci. U. S. A.* **2007**, *104*, (42), 16586–16591.
- 516 (10) Vijayaraghavan, K.; Levin, L.; Parker, L.; Yarwood, G.; Streets, D. Response of
517 fish tissue mercury in a freshwater lake to local, regional, and global changes in
518 mercury emissions. *Environ. Toxicol. Chem.* **2014**, *33* (6), 1238.
- 519 (11) United Nations Environment Programme (UNEP). *Minamata Convention on*
520 *Mercury* <http://www.mercuryconvention.org>.
- 521 (12) Arctic Monitoring and Assessment Programme and United Nations Environment
522 Programme (AMAP/UNEP). *Technical Background Report for the Global Mercury*
523 *Assessment*; AMAP/UNEP: Geneva, Switzerland, 2013.
- 524 (13) Ministry of environmental Protection (MEP). *Notice on the guidance opinions of*
525 *promoting the atmospheric pollution defense spreading work to improve regional air*
526 *quality*; MEP: Beijing, China, 2010.
- 527 (14) Ministry of Environmental Protection (MEP). *Notice about related issues of*
528 *support policy of ultra-low emission electricity price of coal-fired power plant*; MEP:
529 Beijing, China, 2015.
- 530 (15) Ministry of Industry and Information Technology (MIIT). *Specification*
531 *conditions on lead and zinc industry*; MIIT: Beijing, China, 2014.
- 532 (16) Wu, Q.; Wang, S.; Li, G.; Liang, S.; Lin, C. J.; Wang, Y.; Cai, S.; Liu, K.; Hao, J.
533 Temporal trend and spatial distribution of speciated atmospheric mercury emissions in
534 China during 1978–2014. *Environ. Sci. Technol.* **2016**, *50* (24), 13428–13435.
- 535 (17) Ministry of Environmental Protection (MEP). *Technical policy about mercury*
536 *pollution control*; MEP: Beijing, China, 2013.
- 537 (18) Hui, M.; Wu, Q.; Wang, S.; Liang, S.; Zhang, L.; Wang, F.; Lenzen, M.; Wang, Y.;
538 Xu, L.; Lin, Z. Mercury flows in China and global drivers. *Environ. Sci. Technol.*
539 **2017**, *51* (1), 222–231.

- 540 (19) Liang, S.; Chao, Z.; Wang, Y.; Ming, X.; Liu, W. Virtual atmospheric mercury
541 emission network in China. *Environ. Sci. Technol.* **2014**, *48* (5), 2807–2815.
- 542 (20) Guo, J. E.; Zhang, Z.; Meng, L. China's provincial CO₂ emissions embodied in
543 international and interprovincial trade. *Energy Policy* **2012**, *42* (C), 486–497.
- 544 (21) Jiang, W. Q.; Li, J. S.; Chen, G. Q.; Yang, Q.; Alsaedi, A.; Ahmad, B.; Hayat, T.
545 Mercury emissions embodied in Beijing economy. *J. Clean. Prod.* **2016**, *129*, 134–
546 142.
- 547 (22) Li, J. S.; Chen, G. Q.; Chen, B.; Yang, Q.; Wei, W. D.; Wang, P.; Dong, K. Q.;
548 Chen, H. P. The impact of trade on fuel-related mercury emissions in Beijing–
549 evidence from three-scale input-output analysis. *Renew. Sust. Energ. Rev.* **2017**, *75*,
550 742–752.
- 551 (23) Liang, S.; Xu, M.; Liu, Z.; Suh, S.; Zhang, T. Socioeconomic drivers of mercury
552 emissions in China from 1992 to 2007. *Environ. Sci. Technol.* **2013**, *47* (7), 3234–
553 3240.
- 554 (24) Li, J. S.; Chen, B.; Chen, G. Q.; Wei, W. D.; Wang, X. B.; Ge, J. P.; Dong, K. Q.;
555 Xia, H. H.; Xia, X. H. Tracking mercury emission flows in the global supply chains: A
556 multi-regional input-output analysis. *J. Clean. Prod.* **2017**, *140*, 1470–1492.
- 557 (25) National Energy Statistical Agency of China (NESA). *China Energy Statistical*
558 *Yearbook*; NESA: Beijing, China, 2011.
- 559 (26) National Statistical Bureau of China (NSB). *China Statistical Yearbook*; NSB:
560 Beijing, China, 2011.
- 561 (27) Nonferrous Metal Industry Association of China (NMIA). *Yearbook of*
562 *Nonferrous Metals Industry of China*; NMIA: Beijing, China, 2011.
- 563 (28) China Iron and Steel Industry Association (CISIA). *China Steel Yearbook*; CISIA:
564 Beijing, China, 2011.
- 565 (29) Chen, S. *China mining yearbook*; Seismological Press: Beijing, China, 2011.
- 566 (30) Editorial Board of China Electric Power Yearbook. *China Electric Power*

- 567 *Yearbook*; China Electric Power Press: Beijing, China, 2011.
- 568 (31) U.S. Geological Survey (USGS). *Minerals information* [http://](http://minerals.usgs.gov/minerals/pubs/commodity/)
569 minerals.usgs.gov/minerals/pubs/commodity/.
- 570 (32) China Cement Network. *Maps for cement plants* <http://hy.ccement.com/map/>.
- 571 (33) Center for International Earth Science Information Network (CIESIN), Columbia
572 University. *Gridded Population of the World (GPW), Version 3*
573 <http://beta.sedac.ciesin.columbia.edu/gpw/index.jsp>.
- 574 (34) Li, Y.; Meng, J.; Liu, J.; Xu, Y.; Guan, D.; Wei, T.; Huang, Y.; Tao, S.
575 Inter-provincial reliance for improving air quality in China: A case study on black
576 carbon aerosol. *Environ. Sci. Technol.* **2016**, *50* (7), 4118–4126.
- 577 (35) Lin, J.; Tong, D.; Davis, S.; Ni, R.; Tan, X.; Pan, D.; Zhao, H.; Lu, Z.; Streets, D.;
578 Feng, T.; Zhang, Q.; Yan, Y.; Hu, Y.; Li, J.; Liu, Z.; Jiang, X.; Geng, G.; He, K.;
579 Huang, Y.; Guan, D. Global climate forcing of aerosols embodied in international
580 trade. *Nat. Geosci.* **2016**, *9* (10), 790–794.
- 581 (36) Liang, S.; Wang, Y.; Cinnirella, S.; Pirrone, N. Atmospheric mercury footprints
582 of nations. *Environ. Sci. Technol.* **2015**, *49* (6), 3566–3574.
- 583 (37) Lenzen, M.; Moran, D.; Kanemoto, K.; Foran, B.; Lobefaro, L.; Geschke, A.
584 International trade drives biodiversity threats in developing nations. *Nature* **2012**, *486*
585 (7401), 109–112.
- 586 (38) Davis, S. J.; Caldeira, K. Consumption-based accounting of CO₂ emissions. *Proc.*
587 *Natl. Acad. Sci. U. S. A.* **2010**, *107* (12), 5687.
- 588 (39) Liu, W. D.; Tang, Z. P.; Chen, J.; Yang, B. *China's interregional input–output*
589 *table for 30 regions in 2010 (in Chinese)*; China Statistics Press: Beijing, China, 2014.
- 590 (40) Peters, G. P.; Weber, C. L.; Guan, D.; Hubacek, K. China's growing CO₂
591 emissions—a race between increasing consumption and efficiency gains. *Environ. Sci.*
592 *Technol.* **2007**, *41* (17), 5939–5944.
- 593 (42) Miller, R. E.; Blair, P. D. *Input-output analysis: foundations and extensions, 2nd*

- 594 *ed*; Cambridge University Press: Cambridge, UK, 2009.
- 595 (43) Weber, C. L.; Peters, G. P.; Guan, D.; Hubacek, K. The contribution of Chinese
596 exports to climate change. *Energy Policy* **2008**, *36* (9), 3572–3577.
- 597 (43) Holmes, C. D.; Jacob, D. J.; Corbitt, E. S.; Mao, J.; Yang, X.; Talbot, R.; Slemr, F.
598 Global atmospheric model for mercury including oxidation by bromine atoms. *Atmos.*
599 *Chem. Phys.* **2010**, *10* (24), 12037–12057.
- 600 (44) Soerensen, A. L.; Sunderland, E. M.; Holmes, C. D.; Jacob, D. J.; Yantosca, R.
601 M.; Skov, H.; Christensen, J. H.; Strode, S. A.; Mason, R. P. An improved global
602 model for air-sea exchange of mercury: High concentrations over the North Atlantic.
603 *Environ. Sci. Technol.* **2010**, *44* (22), 8574–8580.
- 604 (45) Zhang, Y.; Jaeglé, L.; van Donkelaar, A.; Martin, R. V.; Holmes, C. D.; Amos, H.
605 M.; Wang, Q.; Talbot, R.; Artz, R.; Brooks, S.; Luke, W.; Holsen, T. M.; Felton, D.;
606 Miller, E. K.; Perry, K. D.; Schmeltz, D.; Steffen, A.; Tordon, R.; Weiss-Penzias, P.;
607 Zsolway, R. Nested-grid simulation of mercury over North America. *Atmos. Chem.*
608 *Phys.* **2012**, *12* (14), 6095–6111.
- 609 (46) Wang, L.; Wang, S.; Zhang, L.; Wang, Y.; Zhang, Y.; Nielsen, C.; McElroy, M. B.;
610 Hao, J. Source apportionment of atmospheric mercury pollution in China using the
611 GEOS-Chem model. *Environ. pollut.* **2014**, *190*, 166–175.
- 612 (47) Amos, H. M.; Jacob, D. J.; Holmes, C. D.; Fisher, J. A.; Wang, Q.; Yantosca, R.
613 M.; Corbitt, E. S.; Galarnau, E.; Rutter, A. P.; Gustin, M. S.; Steffen, A.; Schauer, J.
614 J.; Graydon, J. A.; Louis, V. L. S.; Talbot, R. W.; Edgerton, E. S.; Zhang, Y.;
615 Sunderland, E. M. Gas-particle partitioning of atmospheric Hg(II) and its effect on
616 global mercury deposition. *Atmos. Chem. Phys.* **2012**, *12* (1), 591–603.
- 617 (48) Wang, H.; Rasch, P. J.; Easter, R. C.; Singh, B.; Zhang, R.; Ma, P. L.; Qian, Y.;
618 Ghan, S. J.; Beagley, N. Using an explicit emission tagging method in global
619 modeling of source-receptor relationships for black carbon in the Arctic: Variations,
620 sources and transport pathways. *J. Geophys. Res. Atmos.* **2014**, *119* (22), 12888–
621 12909.

- 622 (49) Rasch, P. J.; Barth, M. C.; Kiehl, J. T.; Schwartz, S. E.; Benkovitz, C. M. A
623 description of the global sulfur cycle and its controlling processes in the National
624 Center for Atmospheric Research Community Climate Model, Version 3. *J. Geophys.*
625 *Res. Atmos.* **2000**, *105* (D1), 1367–1385.
- 626 (50) Ministry of Environmental Protection (MEP). “*Twelfth Five-year*” plan for the
627 *comprehensive prevention and control of heavy metal pollution*; MEP: Beijing, China,
628 2011.
- 629 (51) State Council of the People’s Republic of China (SC). *Notice on printing and*
630 *distributing Made in China 2025*
631 http://www.gov.cn/zhengce/content/2015-05/19/content_9784.htm.
- 632 (52) State Council of the People’s Republic of China (SC). *The 11th five-year plan*
633 *(2006–2010)* <http://www.gov.cn/index.htm>.
- 634 (53) State Council of the People’s Republic of China (SC). *National new urbanization*
635 *plan (2014–2020)* http://www.gov.cn/zhengce/2014-03/16/content_2640075.htm.
- 636 (54) Lin, J. T.; Liu, Z.; Zhang, Q.; Liu, H. Modeling uncertainties for tropospheric
637 nitrogen dioxide columns affecting satellite-based inverse modeling of nitrogen
638 oxides emissions. *Atmos. Chem. Phys.* **2012**, *12* (24), 12255–12275.
- 639 (55) Feng, K.; Davis, S. J.; Sun, L.; Li, X.; Guan, D.; Liu, W.; Liu, Z.; Hubacek, K.
640 Outsourcing CO₂ within China. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110* (28), 11654–
641 11659.
- 642 (56) Wang, H.; Zhang, Y.; Zhao, H.; Lu, X.; Zhang, Y.; Zhu, W.; Nielsen, C. P.; Li, X.;
643 Zhang, Q.; Bi, J. Trade-driven relocation of air pollution and health impacts in China.
644 *Nat. Commun.* **2017**, *8* (1), 738.
- 645 (57) Shi, J.; Zhang, Z. *Inter-province input-output model and interregional economic*
646 *linkage in China (in Chinese)*; Science Press: Beijing, China, 2012.
- 647 (58) Zhang, Y.; Qi, S. *China multi-regional input-output models(in Chinese)*; China
648 Statistics Press: Beijing, China, 2012.

649 (59) Wang, Y.; Geschke, A.; Lenzen, M. Constructing a time series of nested
650 multiregion input-output tables. *Int. Reg. Sci. Rev.* **2017**, 40 (5), 476–499.

651 (60) Wang, Y. An industrial ecology virtual framework for policy making in China.
652 *Econ. Syst. Res.* **2017**, 29 (2), 252–274.

653 **Figure Captions**

654

655 **Figure 1.** Production-based and consumption-based Hg emissions (A), emissions
656 embodied in exports (EEE) and emissions embodied in imports (EEI) (B), and net
657 emissions transfer (C) for 30 Chinese provinces.

658

659 **Figure 2.** Contributions from China's total anthropogenic sources (A), total trade (B),
660 interprovincial trade (C), and international export (D) to atmospheric Hg deposition
661 over China.

662

663 **Figure 3.** Spatial redistribution of atmospheric Hg deposition by trade over China,
664 including the spatial redistribution of atmospheric Hg deposition (A), the deposition
665 variation over each provincial or marine receptor due to the trade activities of each
666 province (B), and the contributions of the provincial trade activities to total national
667 deposition (C). The abbreviations are HL, Heilongjiang; JL, Jilin; LN, Liaoning; BJ,
668 Beijing; TJ, Tianjin; HE, Hebei; SX, Shanxi; SD, Shandong; HA, Henan; HB, Hubei;
669 HN, Hunan; AH, Anhui; JX, Jiangxi; SH, Shanghai; JS, Jiangsu; ZJ, Zhejiang; FJ,
670 Fujian; GD, Guangdong; HI, Hainan; GX, Guangxi; CQ, Chongqing; SC, Sichuan;
671 GZ, Guizhou; YN, Yunnan; NM, Inner Mongolia; SN, Shaanxi; GS, Gansu; QH,
672 Qinghai; NX, Ningxia; XJ, Xinjiang; FN, foreign countries; Bs, Bohai Sea; Ys,
673 Yellow Sea; Es, East China Sea; Ss, South China Sea.

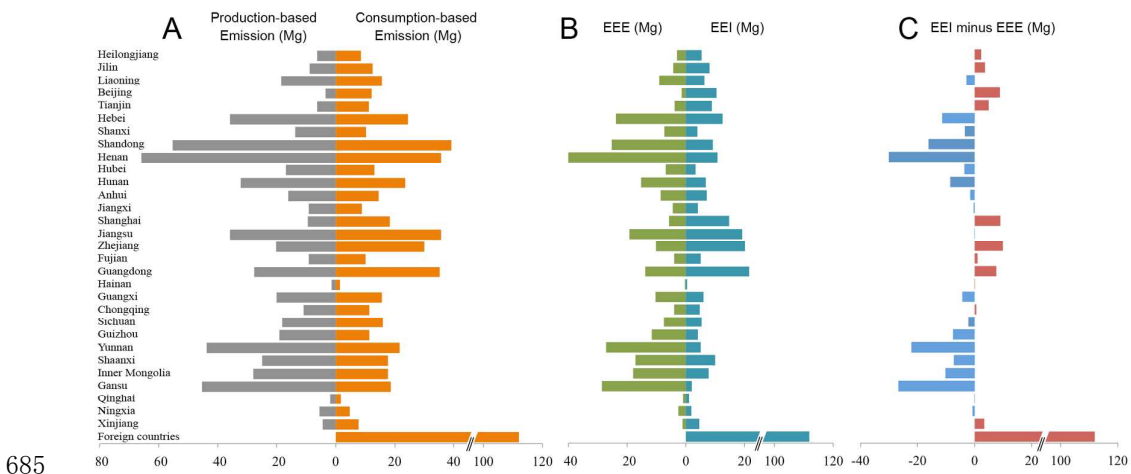
674

675 **Figure 4.** Source-receptor relationship between provincial final consumption and
676 on-site emission (A) and atmospheric Hg deposition (B). "Source" denotes the unity
677 final consumption of each province and foreign countries. "Receptor" denotes
678 percentage variation of on-site emission or atmospheric deposition over each receptor.
679 The abbreviations are the same as Figure 3.

680

681 **Figure 5.** Impact of the hypothetical substitution of emission intensity from
682 developed provinces to their trade partners on atmospheric Hg deposition over China.

683 The developed provinces include Beijing (a), Tianjin (b), Shanghai (c), Jiangsu (d),
684 Zhejiang (e), and Guangdong (f).



685

686

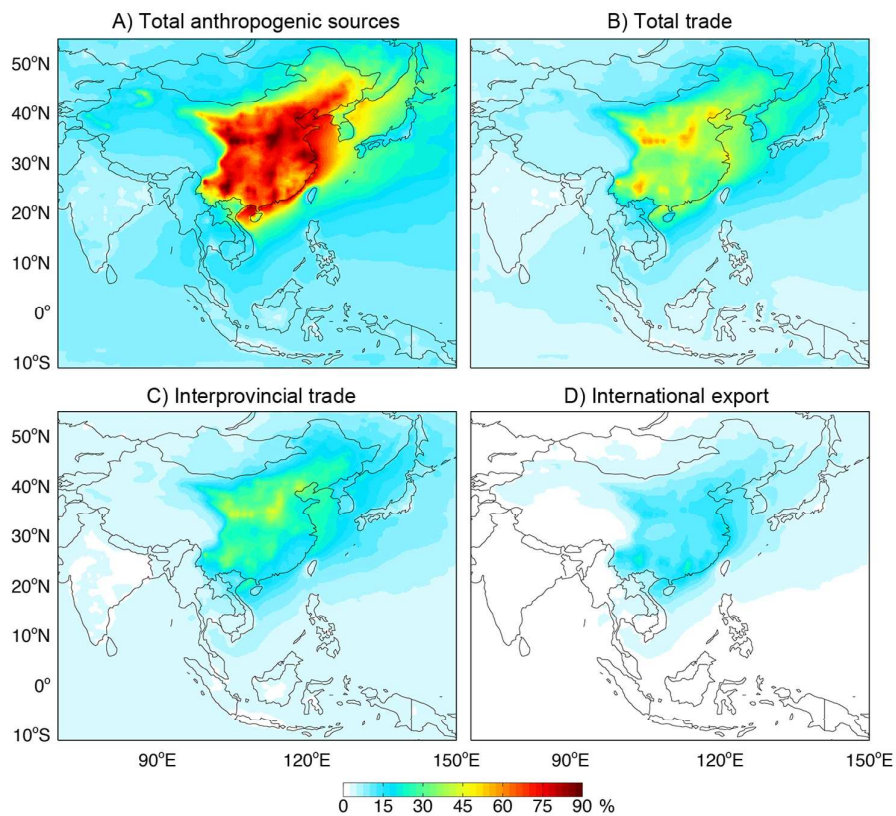
Figure 1. Production-based and consumption-based Hg emissions (A), emissions

687

embodied in exports (EEE) and emissions embodied in imports (EEI) (B), and

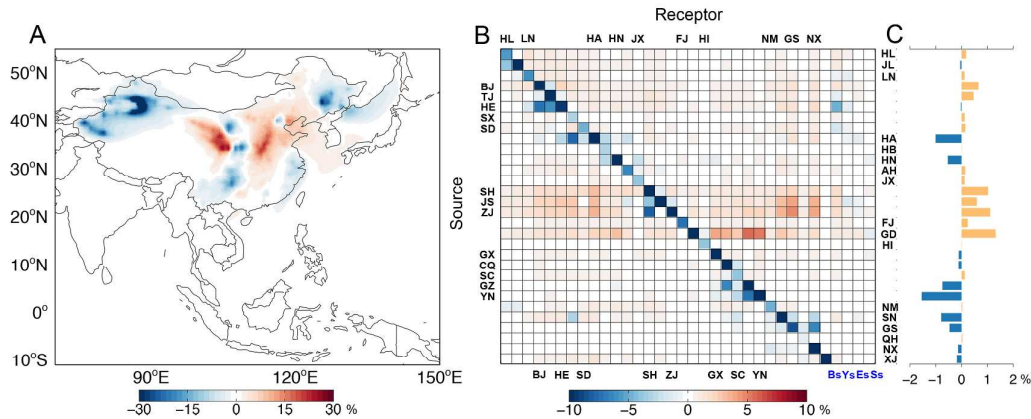
688

net emissions transfer (C) for 30 Chinese provinces.



689

690 **Figure 2.** Contributions from China's total anthropogenic sources (A), total trade (B),
691 interprovincial trade (C), and international export (D) to atmospheric Hg deposition
692 over China.



693

694 **Figure 3.** Spatial redistribution of atmospheric Hg deposition by trade over China,

695 including the spatial redistribution of atmospheric Hg deposition (A), the deposition

696 variation over each provincial or marine receptor due to the trade activities of each

697 province (B), and the contributions of the provincial trade activities to total national

698 deposition (C). The abbreviations are HL, Heilongjiang; JL, Jilin; LN, Liaoning; BJ,

699 Beijing; TJ, Tianjin; HE, Hebei; SX, Shanxi; SD, Shandong; HA, Henan; HB, Hubei;

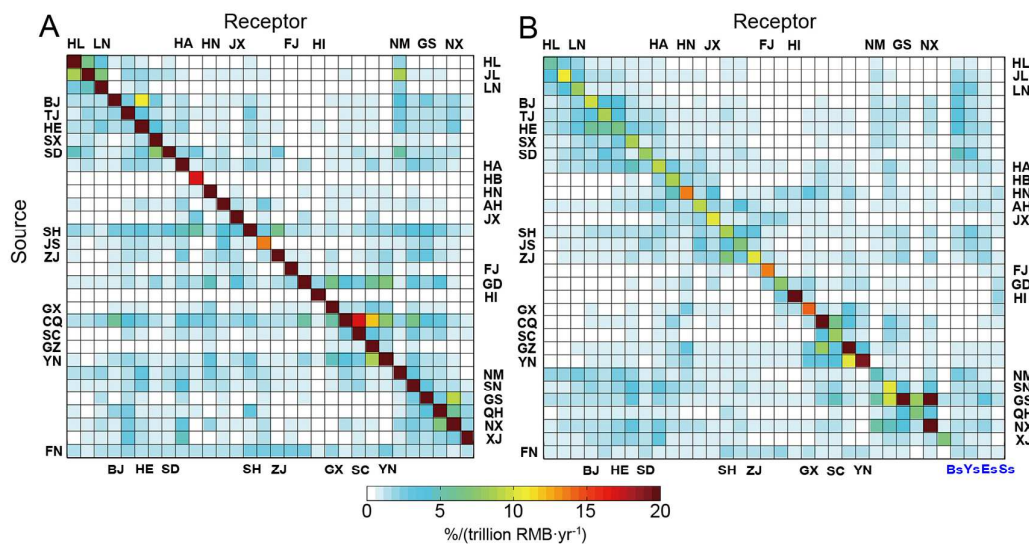
700 HN, Hunan; AH, Anhui; JX, Jiangxi; SH, Shanghai; JS, Jiangsu; ZJ, Zhejiang; FJ,

701 Fujian; GD, Guangdong; HI, Hainan; GX, Guangxi; CQ, Chongqing; SC, Sichuan;

702 GZ, Guizhou; YN, Yunnan; NM, Inner Mongolia; SN, Shaanxi; GS, Gansu; QH,

703 Qinghai; NX, Ningxia; XJ, Xinjiang; FN, foreign countries; Bs, Bohai Sea; Ys,

704 Yellow Sea; Es, East China Sea; Ss, South China Sea.



705

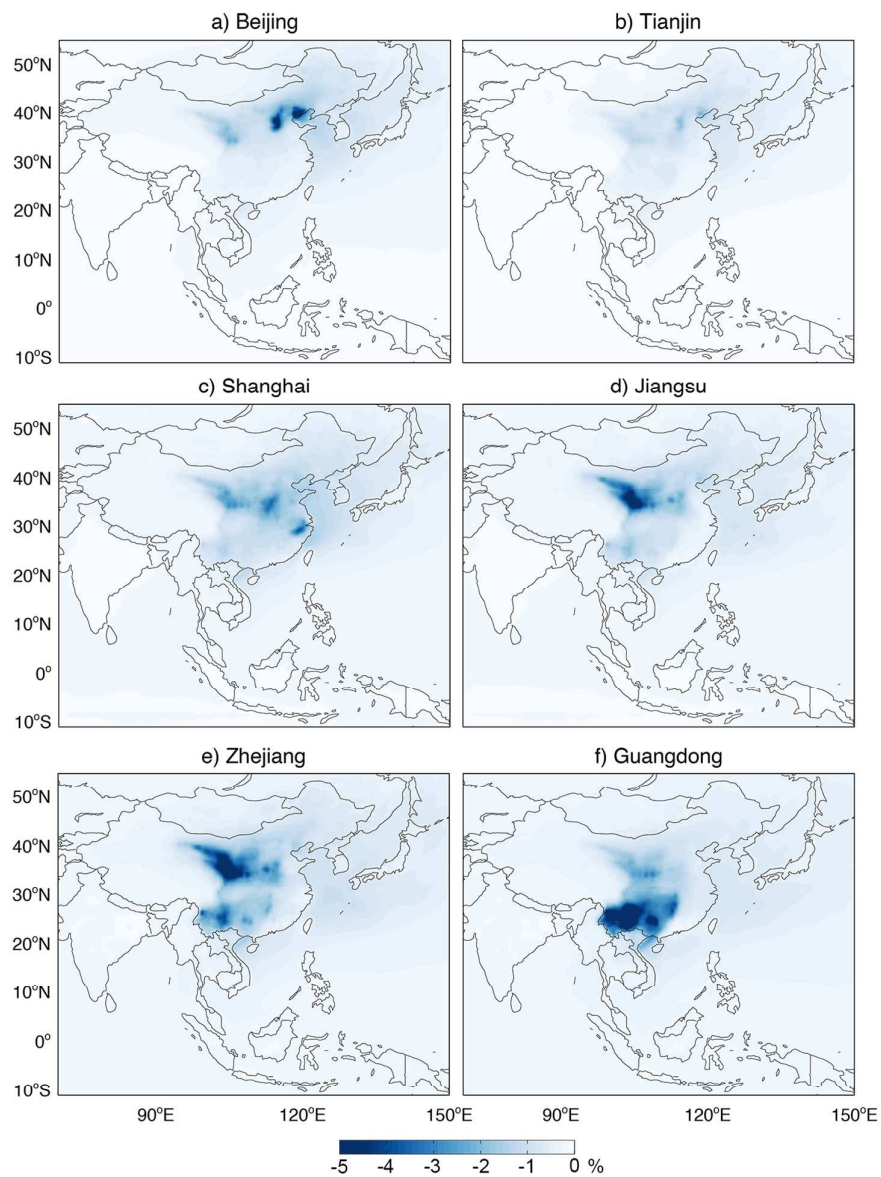
706 **Figure 4.** Source-receptor relationship between provincial final consumption and

707 on-site emission (A) and atmospheric Hg deposition (B). “Source” denotes the unity

708 final consumption of each province and foreign countries. “Receptor” denotes

709 percentage variation of on-site emission or atmospheric deposition over each receptor.

710 The abbreviations are the same as Figure 3.



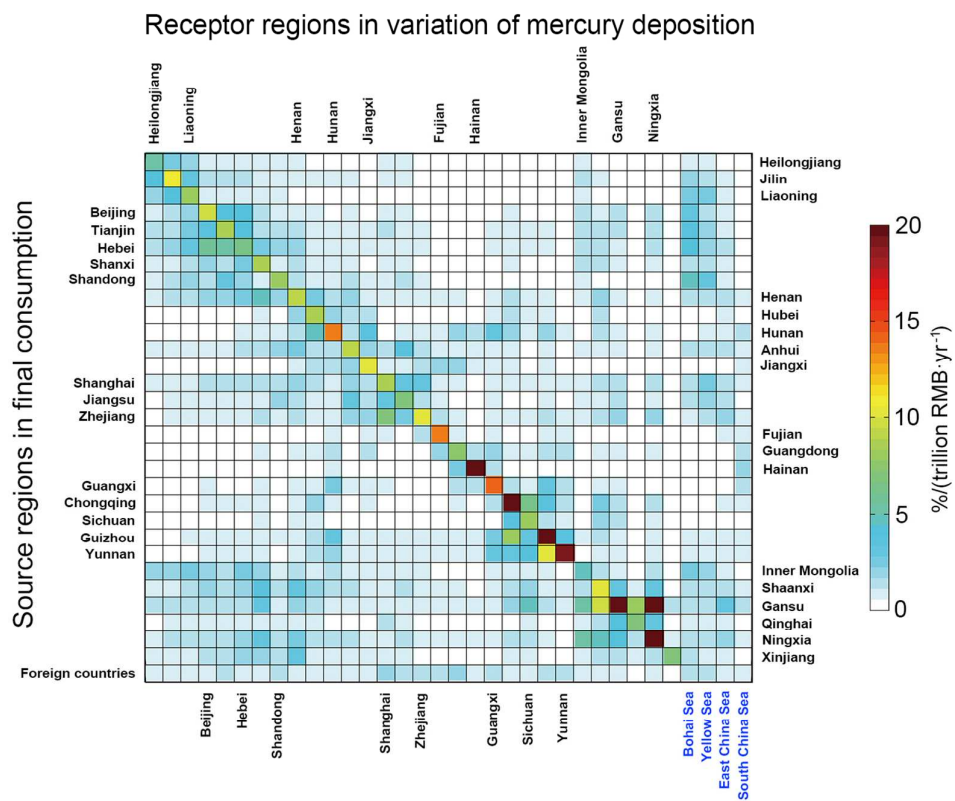
711

712 **Figure 5.** Impact of the hypothetical substitution of emission intensity from
 713 developed provinces to their trade partners on atmospheric Hg deposition over China.

714 The developed provinces include Beijing (a), Tianjin (b), Shanghai (c), Jiangsu (d),

715 Zhejiang (e), and Guangdong (f).

716 TOC/Abstract Art



717