

Supporting Information

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High-resolution inventory of Japanese anthropogenic mercury emissions

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Materials and methods

Advantage of stepwise spatial allocation of national mercury emissions

Japanese geographical information resources are generally characterized by a decline in the types of statistics available as geographical resolution is improved, while at the same time the details of other attribute information tends to decrease. In the methodology adopted in this study, mercury emissions from emission source category i at the national level are distributed over grid cells of approx. 1 km x 1 km using a stepwise allocation procedure, allocating them first to prefectures, then to municipalities, and only then to individual grid cells.

As a simple example, we here consider the relationship between the geographical resolution of statistics used to allocate emissions and the accuracy of the allocated emissions at each resolution, as illustrated in Figure S4. The national emission E in a particular source category includes an accurate emission E_k in prefecture k . When the emission E is allocated to prefecture k using statistics per prefecture, the accuracy of the allocated emission is inferior to that of the actual emission E_k by $(100 - X_k)\%$, where X_k (%) stands for how much the accuracy of the allocated emission to prefecture k is reduced in comparison to that of E_k (100%). Similarly, the national emission E in a certain source category includes an accurate emission E_m for municipality m in prefecture k . When emission E is allocated to municipality m using statistics by municipality, the accuracy of the allocated emission is inferior to the actual emission E_m by $(100 - X_m)\%$, with X_m (%) standing for how much the accuracy of the allocated emission to municipality m is reduced in comparison to that of E_m (100%). From the above relations we see that when the emission of prefecture k is allocated to municipality m using statistics by municipality, the accuracy of the allocated emission to municipality m is reduced by $(X_k - X_m)\%$ compared with the accuracy of the allocated emission to prefecture k . Here, the inequality $100 > X_k > X_m > 0$ is assumed.

Now consider the case of the national emission E first being allocated to prefecture k and then to municipality m . As mentioned above, by allocating from prefecture k to municipality m , the accuracy is reduced by $(X_k - X_m)\%$. In other words, an accuracy of $(100 - (X_k - X_m))\%$ is maintained for allocation from prefecture k to municipality m . The emission of prefecture k

now has an accuracy of $(100 - (100 - X_m)) = X_k \%$. Through the allocation from prefecture k to municipality m , the accuracy of allocated emission of municipality m thus becomes $X_k (100 - (X_k - X_m)) / 100 \%$.

Let us now compare this accuracy with that of the case where emission E is allocated directly to municipality m rather than via allocation to prefectures. Because the latter accuracy is $X_m \%$, we only have to confirm the sign of $X_k (100 - (X_k - X_m)) / 100 - X_m$. The equation $X_k (100 - (X_k - X_m)) / 100 - X_m$ can be transformed into $(100 - X_k)(X_k - X_m)$. Because of the relation $100 > X_k > X_m > 0$, the sign of $(100 - X_k)(X_k - X_m)$ is positive, which guarantees $X_k (100 - (X_k - X_m)) / 100 > X_m$. For this reason, it is considered that the stepwise spatial allocation method adopted in this study has the advantage of improving the accuracy of estimated emissions distribution.

Four spatial allocation methods

For the spatial allocation of emissions, one of four methods was used in this study. First, in Method 1, the starting point is $E_{src=i, nat=j}^{(N)}$, the total mercury emission in emission source category $src = i$ at the national level ($nat = j$), as shown in Figure S5, which is subsequently allocated step by step to the prefectural level, the municipal level and the grid-cell level. In this study the number of national levels is one (Japan), i.e. $nat = 1$, but the subscript nat was introduced for generality. First, $E_{src=i, nat=j}^{(N)}$ is multiplied by the spatial allocation factor $f_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$ to allocate the national emission to the geographical attribute $pref = k$ (referring to prefectures). A spatial allocation factor here is a coefficient of geographic disaggregation of an emission in a given area into an emission in a smaller part of that area, expressing the ratio of the emission in the detailed area to the emission in the larger area. The factor is thus used to increase the geographical resolution of an emission inventory.

Method 1: Nation $\langle N \rangle \rightarrow$ Prefecture $\langle P \rangle \rightarrow$ Municipality $\langle M \rangle \rightarrow$ Grid cell $\langle G \rangle$

Superscript $\langle N \rightarrow P \rangle$ in $f_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$ signifies allocation from the nation (N) to the prefecture (P) and indicates the ratio of $E_{src=i, nat=j}^{(N)}$ allocated to prefecture $pref=k$. Then,

$E_{src=i, pref=k}^{(P)} = E_{src=i, nat=j}^{(N)} \times f_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$, the emission allocated to prefecture $pref=k$, is multiplied by the spatial allocation factor $f_{src=i, nat=j, pref=k, mun=l}^{(P \rightarrow M)}$ to allocate emissions to the geographical attribute mun (municipality). Superscript $\langle P \rightarrow M \rangle$ represents allocation from prefecture (P) to municipality (M). Furthermore, $E_{src=i, mun=l}^{(M)} = E_{src=i, pref=k}^{(P)} \times f_{src=i, pref=k, mun=l}^{(P \rightarrow M)}$, the emission of $mun = l$, is allocated to grid cells (G). In parallel fashion, this emission is multiplied by the spatial allocation factor $f_{src=i, mun=l, grid=m}^{(M \rightarrow G)}$ to determine $E_{src=i, grid=m}^{(G)} = E_{src=i, mun=l}^{(M)} \times f_{src=i, mun=l, grid=m}^{(M \rightarrow G)}$, the emission from the geographical attribute $grid$ (grid cell) $= m$. In other words, the emission of $grid=m$ is yielded by Equation (S1).

The spatial allocation factor $f_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$ is now calculated from Equation (S2):

$S_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$ in Equation (S2) uses a proxy variable for the emission in source category i in $pref = k$, taking an activity volume assumed to be proportional to the emission in question. This is normalized to the sum total of the activity volumes of all the prefectures making up $nat = j$ to obtain $f_{src=i, nat=j, pref=k}^{(N \rightarrow P)}$. Similarly, the spatial allocation factors $f_{src=i, pref=k, mun=l}^{(P \rightarrow M)}$ and $f_{src=i, mun=l, grid=m}^{(M \rightarrow G)}$ are calculated from Equations (S3) and (S4).

$$E_{src=i, grid=m}^{(G)} = E_{src=i, nat=j}^{(N)} \times f_{src=i, nat=j, pref=k}^{(N \rightarrow P)} \times f_{src=i, pref=k, mun=l}^{(P \rightarrow M)} \times f_{src=i, mun=l, grid=m}^{(M \rightarrow G)} \quad (S1)$$

$$f_{src=i, nat=j, pref=k}^{(N \rightarrow P)} = \frac{S_{src=i, nat=j, pref=k}^{(P)}}{\sum_{pref \in nat=j} S_{src=i, nat=j, pref}^{(P)}}, \text{ where } \sum_{pref \in nat=j} f_{src=i, nat=j, pref}^{(N \rightarrow P)} = 1 \quad (S2)$$

$$f_{src=i, pref=k, mun=l}^{(P \rightarrow M)} = \frac{S_{src=i, pref=k, mun=l}^{(M)}}{\sum_{mun \in pref=k} S_{src=i, pref=k, mun}^{(M)}}, \text{ where } \sum_{mun \in pref=k} f_{src=i, pref=k, mun=l}^{(P \rightarrow M)} = 1 \quad (S3)$$

$$f_{src=i, mun=l, grid=m}^{(M \rightarrow G)} = \frac{S_{src=i, mun=l, grid=m}^{(G)}}{\sum_{grid \in mun=l} S_{src=i, mun=l, grid}^{(G)}}, \text{ where } \sum_{grid \in mun=l} f_{src=i, mun=l, grid}^{(M \rightarrow G)} = 1 \quad (S4)$$

As illustrated by the placement of $grid$ (cell) $= m+1$ in Figure S5, in some cases several different municipalities may be juxtaposed within a particular grid cell. In this case,

$E_{src=i, grid=m+1}^{(G)}$, the emission of $grid = m+1$ is defined as in Equation (S5) as the sum of the

emissions allocated from the emissions of the two municipalities $E_{src=i, mun=l}^{(M)}$ and $E_{src=i, mun=l+1}^{(M)}$.

$$E_{src=i, grid=m+1}^{(G)} = E_{src=i, mun=l}^{(M)} \times f_{src=i, mun=l, grid=m+1}^{(M \rightarrow G)} + E_{src=i, mun=l+1}^{(M)} \times f_{src=i, mun=l+1, grid=m+1}^{(M \rightarrow G)} \quad (S5)$$

$\bar{S}_{src=i, grid=m+1}^{(G)}$, the proxy variable for the emission of the total activity of $grid = m+1$ is now divided into $S_{src=i, mun=l, grid=m+1}^{(G)}$, the activity belonging to $mun = l$, and $S_{src=i, mun=l+1, grid=m+1}^{(G)}$, the activity belonging to $mun = l+1$, based on Equations (S6) and (S7), to set the two spatial allocation factors. $W_{src=i, mun=l, grid=m+1}^{(G)}$ and $W_{src=i, mun=l+1, grid=m+1}^{(G)}$ are the weighting factors to divide $\bar{S}_{src=i, grid=m+1}^{(G)}$ between $mun = l$ and $mun = l+1$ for $src = i$ in $grid = m+1$. Here, the relation $W_{src=i, grid=m+1}^{(G)} = W_{src=i, mun=l, grid=m+1}^{(G)} + W_{src=i, mun=l+1, grid=m+1}^{(G)}$ holds.

For example, suppose there are two emission sources (e.g. plants) belonging to $src = i$ in grid cell $(m+1)$ in which the two municipalities $mun = l$ and $mun = l+1$ are juxtaposed. If such information is available, the locations (latitude and longitude) of the plants in question can be used to identify which municipality each facility contributes to, and if the production outputs to be used as a proxies for the plant emissions are available from the relevant websites, these outputs can be used for the weighting factors $W_{src=i, mun=l, grid=m+1}^{(G)}$ and $W_{src=i, mun=l+1, grid=m+1}^{(G)}$.

In general, however, it is no easy matter to obtain this kind of information for direct use in calculating weighting factors. In such cases, the areas of the portions of $grid = m+1$ belonging to $mun = l$ and $mun = l+1$ can be applied for the weighting factors $W_{src=i, mun=l, grid=m+1}^{(G)}$ and $W_{src=i, mun=l+1, grid=m+1}^{(G)}$, and $\bar{S}_{src=i, grid=m+1}^{(G)}$ can then be apportioned based on these weighting factors as given by Equations (S6) and (S7). Here, $W_{src=i, grid=m+1}^{(G)} = W_{src=i, mun=l, grid=m+1}^{(G)} + W_{src=i, mun=l+1, grid=m+1}^{(G)}$ becomes the area of $grid = m+1$.

$$S_{src=i, mun=l, grid=m+1}^{(G)} = \bar{S}_{src=i, grid=m+1}^{(G)} \times \frac{W_{src=i, mun=l, grid=m+1}^{(G)}}{W_{src=i, grid=m+1}^{(G)}} \quad (S6)$$

$$S_{src=i, mun=l+1, grid=m+1}^{(G)} = \bar{S}_{src=i, grid=m+1}^{(G)} \times \frac{W_{src=i, mun=l+1, grid=m+1}^{(G)}}{W_{src=i, grid=m+1}^{(G)}} \quad (S7)$$

Method 2: Nation <N> → Prefecture <P> → Grid cell <G>

In Method 2, allocation is performed from $\langle N \rangle$ to $\langle P \rangle$ and then directly to $\langle G \rangle$. When no appropriate statistics are available on the assumption of $S_{src=i, pref=k, mun=l}^{(M)}$ as the municipality-based proxy value, the allocation is made directly to the grid cells without going through municipalities. In this case, the emissions $E_{src=i, grid=m}^{(G)}$ are defined by Equation (S8), and the allocation factor $f_{src=i, pref=k, grid=m}^{(P \rightarrow G)}$ from prefecture ($pref = k$) to grid cell ($grid = m$) forms Equation (S9). When several prefectures are juxtaposed within a given grid cell, the procedure described under Method 1 is again adopted.

$$E_{src=i, grid=m}^{(G)} = E_{src=i, nat=j}^{(N)} \times f_{src=i, nat=j, pref=k}^{(N \rightarrow P)} \times f_{src=i, pref=k, grid=m}^{(P \rightarrow G)} \quad (S8)$$

$$f_{src=i, pref=k, grid=m}^{(P \rightarrow G)} = \frac{S_{src=i, pref=k, grid=m}^{(G)}}{\sum_{grid \in pref=k} S_{src=i, pref=k, grid}^{(G)}}, \text{ where } \sum_{grid \in pref=k} f_{src=i, pref=k, grid}^{(P \rightarrow G)} = 1 \quad (S9)$$

Method 3: Nation $\langle N \rangle \rightarrow$ Municipality $\langle M \rangle \rightarrow$ Grid cell $\langle G \rangle$

In Method 3, allocation is performed directly from $\langle N \rangle$ to $\langle M \rangle$. If no statistics appropriate for the prefecture-based proxy value $S_{src=i, nat=j, pref=k}^{(P)}$ are available, alternatively if a proxy variable of the same type as that for prefectures is available for municipalities, then allocation to the prefectures is intentionally cancelled. In this case, the emission $E_{src=i, grid=m}^{(G)}$ is defined by Equation (S10), and the allocation factor $f_{src=i, nat=j, mun=l}^{(N \rightarrow M)}$ from the national level ($nat = j$) to municipalities ($mun = l$) forms Equation (S11).

$$E_{src=i, grid=m}^{(G)} = E_{src=i, nat=j}^{(N)} \times f_{src=i, nat=j, mun=l}^{(N \rightarrow M)} \times f_{src=i, mun=l, grid=m}^{(M \rightarrow G)} \quad (S10)$$

$$f_{src=i, nat=j, mun=l}^{(N \rightarrow M)} = \frac{S_{src=i, nat=j, mun=l}^{(G)}}{\sum_{mun \in nat=j} S_{src=i, nat=j, mun}^{(G)}}, \text{ where } \sum_{mun \in nat=j} f_{src=i, nat=j, mun}^{(N \rightarrow M)} = 1 \quad (S11)$$

Method 4: Nation $\langle N \rangle \rightarrow$ Grid cell $\langle G \rangle$

Finally, in Method 4 allocation is performed from $\langle N \rangle$ directly to $\langle G \rangle$. This method is used when the number of facilities in emission source category i is limited at the national level and some type of proxy variable $S_{src=i, nat=j, grid=m}^{(G)}$ is available for each of the grid cells in which the facilities in question are present. The emission $E_{src=i, grid=m}^{(G)}$ is defined by Equation (S12), and

the allocation factor $f_{src=i,nat=j,grid=m}^{(N \rightarrow G)}$ from the national level ($nat = j$) to grid cells ($grid = m$) forms Equation (S13).

$$E_{src=i,grid=m}^{(G)} = E_{src=i,nat=j}^{(N)} \times f_{src=i,nat=j,grid=m}^{(N \rightarrow G)} \quad (S12)$$

$$f_{src=i,nat=j,grid=m}^{(N \rightarrow G)} = \frac{S_{src=i,nat=j,grid=m}^{(G)}}{\sum_{grid \in nat=j} S_{src=i,nat=j,grid}^{(G)}}, \text{ where } \sum_{grid \in nat=j} f_{src=i,nat=j,grid}^{(N \rightarrow G)} = 1 \quad (S13)$$

Two approaches for determining spatial allocation factors

The procedure for setting a spatial allocation factor is first to determine the location of the process from which mercury is emitted to the atmosphere and then to identify an appropriate, quantitative activity to serve as a proxy variable for the emission volume. In this study, wherever possible we chose proxies anticipated to directly express an activity volume proportional to the emission volume at the location where the emission actually occurs. While quantitative mercury emissions are obviously not necessarily exactly proportional to the activity volume of the emission source, we have opted for such proportional allocation, given that the author of the national inventory himself assumes a linear relationship between activity volume and emission and that data suitable for defining a nonlinear function are currently unavailable. Wherever possible, then, the activity volume adopted as a proxy in each emission source category was the same as that used in estimating the national inventory. However, given the nature of Japanese geographical data resources, described earlier, as spatial resolution increases, the type of activity volume data used in the national inventory becomes unavailable, so that other activity volumes sometimes had to be adopted as proxy causes for mercury emissions in order to calculate allocation factors.

In the case of the emission source category “thermal power generation (coal)” ($src = 1$), for instance, the locations of the mercury emissions were simply taken to be the sites of coal-fired power plants. We therefore collected data on the location of each plant, using its output of electricity as the proxy variable. The national emissions inventory, on the other hand, uses coal consumption as the activity volume for estimating national emissions from thermal power generation (coal). In our approach, in which spatial allocation is based on the output of

each plant, emissions can be allocated far more precisely to individual plant sites.

In the case of “incineration of medical waste” ($src = 6$), on the other hand, the actual location of the mercury emissions will be the incineration plant where the waste is burned. The ideal way to determine actual emission volumes at the grid-cell level would therefore be to adopt the same volume activity used in the national inventory, i.e. the volume of medical waste actually incinerated in each grid cell. Such statistics are not currently available, however, nor even the volumes by prefecture or by municipality.

In this study we therefore used the number of sickbeds in hospitals as a proxy to develop an allocation factor for prefectures (in Japan, the number of hospitals and sickbeds are only available by prefecture), which seems a reasonable way to allocate the volume of medical waste generated. This means that the national-level mercury emission from medical waste was allocated to prefectures according to where the activities indirectly generating emissions occur, but not the emissions themselves. In the next two steps, municipal-level allocation was performed using the number of workers in industrial waste disposal in the municipality, and grid-cell level allocation using the number of incineration facilities for medical waste in the grid cell (data on the amount of waste treated in each facility itself is unavailable) .

However, combining spatial allocation to a location indirectly related to the causes of mercury emissions (to “hospitals” at the prefecture level) with allocation to the actual emission location (to “incineration facilities” at the grid-cell level) might lead to major uncertainties in grid-cell allocation of emissions. If all the medical waste from the hospitals in a given prefecture is disposed of at incineration plants located in the same prefecture, even if an allocation factor for a prefecture based on the number of sickbeds is used, it would be possible to arrive at an estimate emission in the prefecture close to the realistic emission. In reality, though, not all the medical waste from the hospitals in a given prefecture is necessarily treated in the same prefecture, with some waste possibly being disposed of in facilities in other prefectures. This causes large uncertainties in the emissions allocated.

Considering that such allocation factors related indirectly to the causes of mercury emissions need to be used for spatial allocation of national emissions to prefectures and municipalities, wherever possible this study used proxy variables linked directly to the locations of emission sources to calculate grid-cell allocation, even when high accuracy could not be achieved.

Table S1 specifies the proxy variables used in this study to calculate the spatial allocation factors for each emission source category.

Method and data used for spatial allocation of emissions by source category

(S1) Thermal power generation (coal)

Allocation method 4 was applied. Emissions were disaggregated to individual electric power companies based on the coal consumption of each company (t) [1]. These company emissions were then further allocated to grid cells, identified from the address of the coal-fired power plants of each company and based on plant output (kWh) [1].

(S2) Industrial boilers (coal)

Allocation method 2 was applied. The activity volume used for allocating emissions from industrial boilers (coal) to prefectures was coal consumption (t) per prefecture per sector as defined in the national energy consumption statistics [2]. The sectors covered were ‘chemicals, synthetic fibers and paper pulp’, ‘steel, nonferrous metals, ceramics, stone and clay products’, ‘machinery’ and other sectors included under the heading ‘manufacturing industry’.

Allocation to grid cells used product shipment value (yen) per industry per grid cell [3] as the activity volume. Because the industry classification in the industrial statistics [3] differs from the sectoral classification in the energy consumption statistics [2] used for allocation to prefectures, the equivalences shown in Table S2 below were used for allocation.

(S3) Thermal power generation (petroleum)

Allocation method 4 was applied. Following the same method as for thermal power generation (coal) ($src = 1$), emissions were disaggregated to individual electric power companies based on their petroleum consumption (t) [1]. The bulk of the petroleum burned is crude oil and fuel oil, with oil of other types scarcely being used. Consumption of crude oil and fuel oil was therefore used for allocation. Besides oil-fired power plants, LNG (Liquid Natural Gas)-fired power plants also consume a non-negligible amount of crude oil and fuel oil for power generation. Allocation to grid cells consequently used emissions from LNG-fired power plants in addition to oil-fired power plants.

(S4) Industrial boilers (petroleum)

Allocation method 2 was applied. The activity volume used for allocating industrial boilers (petroleum) to prefectures was fuel oil product consumption (kl) per prefecture per sector [2]. The sectors covered were ‘manufacturing’, ‘non-manufacturing’, and ‘offices’. Allocation to grid cells used product shipment value per industry as the activity volume for ‘manufacturing’ [3] and the number of workers per industry as the activity volume for ‘non-manufacturing’ and ‘offices’ [4, 5]. Table S3 reports the correspondence between sectors and industries in the statistics.

(S5) Incineration of general waste

Allocation method 3 was applied. Because the total volume of general waste incinerated annually per municipality was available, national-level emissions were allocated to municipalities in proportion to this volume (t) [6]. Subsequent allocation to grid cells used the address and annual incineration volume (t) of each incineration plant reported by the Japanese Environment Ministry [7]. However, there were cases in which although the national-level emissions were allocated to a municipality, no facility in the municipality was reported. This meant the emissions in the municipality could not be allocated further to the grid-cell level. In such cases it was assumed that the general waste generated in the municipality with no reported incineration facility was incinerated at the nearest facilities to the municipality, with emissions being allocated to the grid cells containing those facilities.

In such cases, the emissions of the municipality differ from the sum of the emissions from the grid cells attributed to the municipality. This study regarded the later summed emissions as the emissions of the municipality, not the former emissions initially allocated from the national level.

(S6) Incineration of medical waste

Allocation method 1 was applied. Incineration of medical waste is categorized into that of infectious waste and non-infectious waste. In the national mercury emissions inventory, however, the associated mercury emissions are not broken down and so these had to be estimated, as follows. The data relating to incineration facilities used for allocation to grid cells permit distinction between facilities burning infectious waste and those burning non-infectious waste. Assuming, therefore, that the difference between the two types can be

considered at the level of grid-cell emissions, the national emissions were divided into those from infectious and non-infectious waste materials based on the ratio of incineration in 2005 (assumed to be the same as 2002), as reported in Table S4, followed by spatial allocation.

The activity volume used for allocation to prefectures was the number of sickbeds per prefecture [8], while that for allocation to municipalities was the number of workers in the industrial waste disposal industry per municipality [9].

Allocation to grid cells used the locations of incineration plants accepting infectious waste and those accepting non-infectious waste [10]. Data on the incineration capacity of each plant and the annual volume treated was not available, however. Emissions were therefore allocated equally to all facilities. In some cases of allocation to municipalities, those first regarded as having mercury emissions (with a non-zero number of workers in the industrial waste disposal industry) might not actually have any incineration facilities that accept medical waste. In such cases, the emissions of municipalities without incineration facilities were summed within the prefecture and re-allocated based on the ratios of emissions in the municipalities that do have incineration facilities. The municipality in which medical waste is produced and that in which mercury emissions arise from incineration may differ. The ultimate emissions per municipality were therefore recalculated from the emissions per grid cell.

(S7) Incineration and dissolution of sewage sludge

Allocation method 1 was applied. Allocation of emissions from the incineration and dissolution of sewage sludge to prefectures used the volume of concentrated sewage sludge generated per prefecture (m^3) [11]. Allocation to municipalities used the number of people connected to a sewage system, as calculated from the sewerage coverage ratio [12] and population [13]. Allocation of mercury emissions per municipality to grid cells used two different methods, described below.

Despite being industrial waste, sewage sludge is treated differently from other types of industrial waste. It is collected for treatment in each area serviced by sewer companies, with four possible patterns:

(A) The sewer company has its own incineration facilities, where the waste is incinerated.

- (B) The sewage sludge is transported from several sewer companies for treatment in a single, dedicated sludge incineration plant ('sludge center', etc.).
- (C) The sewage sludge is transported to a general waste incineration plant and incinerated with general waste.
- (D) An industrial waste disposal company (IWDC) is contracted for disposal.

Sewer companies treating very large volumes of sewage sludge in densely populated urban areas are likely to conform to (A) or (B) above. The treatment capacities of sewage sludge incineration facilities in Tokyo and other ordinance-designated cities were identified from the websites of sewer companies and other sources of the respective local governments. Subsequently, the municipality-based mercury emissions of Tokyo and other ordinance-designated cities were allocated to each sewage sludge incineration facility according to its treatment capacity [14].

Meanwhile, other medium-sized and smaller cities and suburban areas are more likely to conform to (C) or (D). In this case, the form of disposal is not necessarily independent in each municipality, but might include two or more municipalities in a business area, as exemplified by the so-called 'supramunicipal' sewage works associations. Because this study was not sufficiently extensive to examine such broad-based waste disposal, the municipality-based emissions were allocated to grid cells using the locations of the incineration facilities accepting sludge [10] on the assumption of (D) above (disposal contracted to an IWDC). In some cases of allocation to municipalities, those initially regarded as having mercury emissions might not actually have any incineration facilities within them. In such cases, the procedure used for medical waste incineration described above was adopted.

(S8) Industrial waste (waste plastics)

Allocation method 1 was applied. Allocation of industrial waste (waste plastics) to prefectures used the estimated emissions (t) per waste type (here: waste plastics) per prefecture [11] as the activity volume. Allocation to municipalities used the number of workers in the industrial waste disposal industry by municipality [9]. Allocation to grid cells used the locations of IWDC facilities (limited to those carrying out incineration and also accepting waste plastics) [10].

In some cases of allocation to municipalities, those first regarded as having mercury emissions might not actually have any incineration facilities within them. In such cases, the procedure used for medical waste incineration (*src* = 6) described above was adopted.

(S9) Industrial waste (wastepaper)

Allocation method 1 was applied. Allocation was performed using the same method as for industrial waste (waste plastics) (*src* = 8), but with ‘wastepaper’ now taken as waste type.

(S10) Industrial waste (waste wood)

Allocation method 1 was applied. Allocation was performed using the same method as for industrial waste (waste plastics) (*src* = 8), but with ‘waste wood’ now taken as waste type.

(S11) Industrial waste (waste textile)

Allocation method 1 was applied. Allocation was performed using the same method as for industrial waste (waste plastics), but with ‘waste textile’ now taken as waste type.

(S12) Industrial waste (waste rubber)

Allocation method 1 was applied. Allocation was performed using the same method as for industrial waste (waste plastics) (*src* = 8), but with ‘waste rubber’ now taken as waste type.

(S13) Industrial waste (other sludge)

Allocation method 1 was applied. Allocation was performed using the same method as for industrial waste (waste plastics) (*src* = 8), but with ‘sludge’ now taken as waste type.

The estimated emissions (t) by waste type (‘sludge’) per prefecture in the *Investigation of the Actual State of Industrial Waste Treatment* (Environment Ministry) used as the activity volume for allocation to prefectures include ‘other sludge’ as well as ‘sewage sludge’. Therefore, the value obtained by subtracting the amount of concentrated sludge per prefecture provided in the above report was used as the activity volume for allocation of emissions related to ‘other sludge’.

(S14) Industrial waste (shredder dust)

Allocation method 1 was applied. Allocation was performed using the same method as for

industrial waste (waste plastics) (*src* = 8), but with ‘waste glass’ now taken as waste type.

(S15) Steel and iron manufacturing

Allocation method 4 was applied. Emissions from steel and iron manufacturing were allocated to individual steel and iron manufacturers based on pig iron output (10^4 t) [15] per manufacturer. Allocation to grid cells used the locations of the steelworks and the crude steel output (10^3 t) [16] of each manufacturer.

(S16) Nonferrous metal (zinc)

Allocation method 4 was applied. National emissions from nonferrous metal (zinc) were allocated directly to grid cells using the location and smelting capacity (10^3 t) [17] of each zinc smelter. The distinction between primary and secondary smelting at each plant was unclear. Allocation according to these processes was therefore omitted.

(S17) Nonferrous metal (copper)

Allocation method 4 was applied. National emissions from nonferrous metal (copper) were allocated directly to grid cells using the location and electrolytic refining capacity (10^3 t) [17] of each copper refinery.

(S18) Nonferrous metal (lead)

Allocation method 4 was applied. National emissions from nonferrous metal (lead) were allocated directly to grid cells using the location and smelting capacity (10^3 t) [17] of each lead smelter.

(S19) Nonferrous metal (nickel)

Allocation method 4 was applied. National emissions from nonferrous metal (nickel) were allocated directly to grid cells using the locations of nickel and nickel compound manufacturing establishments [18].

(S20) Cement manufacturing

Allocation method 4 was applied. National emissions from cement manufacturing were allocated directly to grid cells using the plant locations of each manufacturer’s clinker production plants (t) [19].

(S21) Quicklime and slaked lime manufacturing

Allocation method 1 was applied. The activity volume used for allocating quicklime and slaked lime manufacturing to prefectures was the volume of quicklime and slaked lime shipped (t) [20] per prefecture. Allocation to municipalities used the value (yen) [21] of the raw materials used for ceramics, stone and clay product manufacturing in each municipality as the proxy indicator. Allocation to grid cells used the number of facilities [22] by grid cell, derived from the locations of quicklime and slaked lime plants.

In some cases of allocation to prefectures or municipalities, those first regarded as having mercury emissions did not actually have any quicklime and slaked lime plants. In such cases, because the method of re-allocation within prefectures (as in the case of medical waste incineration ($src = 6$)) could not be adopted and because there were no other appropriate alternatives available, those prefectures (or municipalities) without the plants in question were excluded from allocation.

(S22) Carbon black manufacturing

Allocation method 4 was applied. National emissions from carbon black manufacturing were allocated directly to grid cells using the plant locations and the carbon black output (10^3 t) [23] of each manufacturer.

(S23) Coke manufacturing

Allocation method 4 was applied. Coke manufacturing is pig iron manufacturing; the products manufactured in the steelworks are regarded as nearly the same. The same allocation method as that used for steel and iron manufacturing was therefore applied.

Allocation to prefectures was not performed, but was substituted by allocation to individual coke manufacturers using pig iron output per manufacturer (10^4 t) [15] as the activity volume. Allocation to grid cells used the locations of steelworks and the crude steel output (10^3 t) [16] of each manufacturer.

(S24) Pulp and paper manufacturing

Allocation method 1 was applied. The activity volume used for allocating pulp and paper

manufacturing to prefectures was the value (yen) [24] of the raw materials used for pulp manufacturing per prefecture. The proxy used for allocation to municipalities was the value (yen) [21] of the raw materials used for the manufacture of pulp, paper and paper-worked products per municipality. Allocation to grid cells used the number of pulp manufacturing facilities [25] in each grid cell as the activity volume.

As in the case of quicklime and slaked lime manufacturing, those prefectures (or municipalities) with no plants were excluded from allocation.

(S25) Chlor-alkali industry

Spatial allocation was omitted because of zero mercury emissions from the chlor-alkali industry, as estimated in the national mercury emissions inventory.

(S26) Battery manufacturing

Allocation method 2 was applied. The activity volume used for allocating battery manufacturing to prefectures was the value (yen) [24] of the raw materials used for primary battery (dry-cell and wet-cell) manufacturing per prefecture. Allocation to grid cells used the shipment value (yen) [3] of the products of electrical machinery and appliance manufacturing per grid cell as the activity volume.

(S27) Electric switch manufacturing

Allocation method 2 was applied. The indicator used for allocating electric switch manufacturing to prefectures was the value (yen) [24] of the raw materials used for connectors, switches and relay manufacturing per prefecture. Allocation to grid cells used the shipment value (yen) [3] of the products of electronic component and device manufacturing per grid cell as the activity volume.

(S28) Fluorescent light manufacturing

Allocation method 2 was applied. The indicator used for allocating fluorescent light manufacturing to prefectures was the value (yen) [24] of the raw materials used for light bulb manufacturing per prefecture. Allocation to grid cells used the shipment value (yen) [3] of the products of electrical machinery and appliance manufacturing per grid cell as the activity volume.

(S29) Cremation

Allocation method 3 was applied. As the activity volume for allocation, the amount of cremation was calculated on the basis of the elderly population [13], which shows a high correlation with annual mortality. Precise data for the population by age of each municipality are available. Allocation to prefectures was therefore omitted and municipality-based emissions were calculated directly from national emissions. Allocation to grid cells used the number of cremation facilities [26] per grid cell per municipality.

As in the case of general waste incineration ($src = 6$), however, municipalities first regarded as having mercury emissions in the allocation to municipalities might not actually have any cremation facilities. For this reason, those municipalities with no such facilities of their own were assumed to use the facilities closest to them. In such cases, because the municipality in which a person dies and that in which cremation is assumed to take place may differ, the ultimate emissions per municipality were recalculated from the emissions per grid cell.

(S30) Collection and crushing of fluorescent lights

Spatial allocation was omitted because of the extremely low emissions indicated in the national mercury emissions inventory.

(S31) Dental service (amalgam)

Allocation method 2 was applied. Allocation of dental amalgam to prefectures used the number of patients [27] at dental clinics per prefecture as the activity volume. For allocation to grid cells the number of workers at dental clinics per grid cell [4] was used.

(S32) Transportation (gasoline)

Allocation method 2 was applied. The activity volume used for allocation of transportation (gasoline) to prefectures was gasoline consumption (kl) [2] in the transportation sector (passenger cars) per prefecture.

The activity volume used for allocation to grid cells was the travel distance (unit km/y) by fuel type (gasoline) by grid cell. This travel distance was estimated according to the following procedure, based on a combination of traffic statistics and geographical data on the road

network:

(Step 1) Calculate length of main and secondary road network per grid cell

Using a digital road map [28] (Japan Digital Road Map Association (DRM)), the road grid was divided into main roads (those included in the *Road Traffic Census*) and other, secondary roads. The former correspond to the road sections defined in the *General Traffic Volume Survey of the Road Traffic Census 2005* (Ministry of Land, Infrastructure, Transport and Tourism (MLIT)) [29]. Subsequently, the geographic information system (GIS) assigned roads in the digital road map to grid cells. For the main roads, the length (km) of road per road section of the *Census* by grid cell was calculated. For the secondary roads, the road area (km²) per grid cell was calculated.

(Step 2) Calculate travel distances on main and secondary roads

Travel distance on main roads: The travel distance (unit km/y) on main roads per car type per grid cell was calculated using the road length per road section of the *Road Traffic Census* by grid cell obtained in Step 1 above and the traffic volume (vehicle units) [29] by car type per road section of the *Census*.

Travel distance on secondary roads: For each car type, the ratio of main-road to total-road travel in a prefecture was calculated by dividing the travel distance on main roads by car type per prefecture provided in the *Census* by the total travel distance (unit km/y) [30] by car type per prefecture from the *Land and Transport Statistics 2005* (MLIT).

The travel distance on secondary roads per car type per prefecture was then estimated using the following relationship:

$$\begin{aligned} &\text{travel distance on secondary roads [units km/y]} = \\ &\frac{\text{travel distance on main roads [units km/y]}}{\text{share of main roads}} \times (1 - \text{ratio of main roads}) \end{aligned}$$

This travel distance on secondary roads was allocated to grid cells according to the area (km²) of road per obtained in Step 1, and the annual travel distance (unit km/y) on secondary roads per car type per grid cell was calculated.

(Step 3) Calculate travel distance per fuel type

Using the ratio between fuel types reported in the fuel consumption (kl) [30] per car type data in the *Land and Transport Statistics 2005* (MLIT), the travel distance (unit km/y) calculated in Step 2 above was broken down by fuel type to obtain the travel distance of gasoline-fueled cars. Additionally, the travel distance (unit km/y) per grid cell per prefecture was calculated by summing the travel distances on main and secondary roads.

(S33) Transportation (jet fuel oil)

Spatial allocation was omitted because of zero mercury emissions from transportation (jet fuel oil) as estimated in the national mercury emissions inventory.

(S34) Transportation (kerosene)

Spatial allocation was omitted because of zero mercury emissions from transportation (kerosene) estimated in the national mercury emissions inventory.

(S35) Transportation (diesel fuel)

Allocation method 2 was applied. Emissions from transportation (diesel fuel) were allocated using the same method as for transportation (gasoline) (*src* =32). Allocation to grid cells used the annual travel distance (km) per fuel type (diesel fuel) per grid cell.

(S36) Transportation (fuel oil)

In most cases, emissions from transportation (fuel oil) are likely to derive from shipping. Spatial allocation of mercury emissions from shipping would require development of spatial allocation factors for individual harbors and routes, which would be very difficult. This study consequently omitted spatial allocation of transportation (fuel oil).

(S37) Volcanoes

Mercury emissions caused by volcanoes, as natural activities, were excluded from spatial allocation because of difficulties in the development of spatial allocation factors for this purpose.

Specifications of the mercury emission inventory database

The grid-cell mercury emissions developed in this study are attached to the SI as an electronic database, with the data saved in CSV (comma separated value) format. The data are annual mercury emissions for 2005 per source category and per grid cell of approx. 1 km × 1 km for the whole of Japan. They can be used to aggregate emissions by prefecture or by municipality.

The headers given include “**EmissionSourceCode**,” “**PrefCode**,” “**MunCode**,” “**GridCode**,” “**MercuryEmission(g/y)**,” “**Maximum(g/y)**,” “**Minimum(g/y)**,” and “**StandardDeviation**.” In addition, “**SwEdgeOfGridCode_latitude**” and “**SwEdgeOfGridCode_longitude**” give the latitude and longitude of the lower left (southwest) corner of the rectangular grid cell given by the GridCode (a rectangle of 45 s longitude and 30 s latitude).

The administrative codes (PrefCode, MunCode) follow the *Standard Codes for Areas of Prefectures and Municipalities for Statistical Use* [30]. They were made consistent with the administrative areas as of October 1, 2006. GridCode follows the *Standard Regional Grid and Mesh Codes for Statistical Use* [31]. It should be noted that the geodetic reference system to which the mesh codes conform was developed using a code system based on the Japanese geodetic system (Tokyo Datum); because of restrictions on the mesh statistics used for this study, it was not based on the world geodetic system that is currently in use (JGD 20000).

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*Note) In census statistics certain values such as shipment volume, value of raw materials used and value of products shipped are not revealed when the figures relate to a single company or two companies, only being reported, in aggregated form, when three or more companies are concerned. In this study these concealed values were estimated using data on the number of workers, which are reported in the census.

Table S1:
Methods and proxy variables used to calculate spatial allocation factors per emission source category

Source code	Emission source category	Allocation method	Proxy variable used for calculation of spatial allocation factors					
			Allocation to prefectural level	Ref.	Allocation to municipal level	Ref.	Allocation to grid-cell level	Ref.
S1	Thermal power generation (coal)	Method 4	-		-		Coal consumption per power company (t)	[1]
							Electrical output per power plant (kWh)	[1]
S2	Industrial boilers (coal)	Method 2	Coal consumption per sector (t)	[2]	-		Shipment value of products per industry (yen)	[3]
S3	Thermal power generation (petroleum)	Method 4	-		-		Petroleum (crude oil and fuel oil) consumption per power company (t)	[1]
							Electrical output per power plant (kWh)	[1]
S4	Industrial boilers (petroleum)	Method 2	Fuel oil product consumption per sector (kl)	[2]	-		Number of workers per industry	[4][5]
							Product shipment value per industry (yen)	[3]
S5	Incineration of general waste	Method 3	-		Volume of general waste directly incinerated(t)	[6]	Volume of waste incinerated by general waste incineration facilities (t)	[7]

S6	Incineration of medical waste	Method 1	Number of sickbeds	[8]	Number of workers in industrial waste disposal	[9]	Infectious waste: number of “infectious waste” incineration plants operated by IWDCs (industrial waste disposal companies) Non-infectious waste: number of incineration plants operated by “all IWDCs”	[10]
S7	Incineration and dissolution of sewage sludge	Method 1	Volume of concentrated sewage sludge (m ³)	[11]	Sewerage coverage ratio (%) and population	[12] [13]	Ordinance-designated cities: processing capacity per sewage sludge incineration plant, etc. Other cities: number of “sludge” incineration plants operated by IWDCs	[14] [10]
S8	Industrial waste (waste plastics)	Method 1	Estimated emissions from “waste plastics” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “waste plastics” incineration plants operated by IWDCs	[10]
S9	Industrial waste (wastepaper)	Method 1	Estimated emissions from “wastepaper” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “wastepaper” incineration plants operated by IWDCs	[10]
S10	Industrial waste (waste wood)	Method 1	Estimated emissions from “waste wood” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “waste wood” incineration plants operated by IWDCs	[10]
S11	Industrial waste (waste textile)	Method 1	Estimated emissions from “waste textile” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “waste textile” incineration plants operated by IWDCs	[10]

S12	Industrial waste (waste rubber)	Method 1	Estimated emissions from “waste rubber” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “waste rubber” incineration plants operated by IWDCs	[10]
S13	Industrial waste (other sludge)	Method 1	Estimated emissions from “total sludge excluding sewage sludge” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “sludge” incineration plants operated by IWDCs	[10]
S14	Industrial waste (shredder dust)	Method 1	Estimated emissions from “waste glass” (t)	[11]	Number of workers in industrial waste disposal	[9]	Number of “waste glass” incineration plants operated by IWDCs	[10]
S15	Steel and iron manufacturing	Method 4	-		-		Pig iron output per manufacturer (10 ⁴ t)	[15]
							Crude steel output per steelworks (1000 t)	[16]
S16	Nonferrous metal (zinc)	Method 4	-		-		Smelting capacity per zinc smelter (6 sites) (10 ³ t)	[17]
S17	Nonferrous metal (copper)	Method 4	-		-		Electrolytic refining capacity per copper refinery (7 sites) (10 ³ t)	[17]
S18	Nonferrous metal (lead)	Method 4	-		-		Smelting capacity per lead smelter (6 sites) (10 ³ t)	[17]
S19	Nonferrous metal (nickel)	Method 4	-		-		Number of nickel and nickel compound manufacturing sites	[18]
S20	Cement manufacturing	Method 4	-		-		Clinker output per plant site (t)	[19]
S21	Quicklime and slaked lime manufacturing	Method 1	Volume of quicklime and slaked lime shipped (t)	[20]	Value of raw materials used for ceramics, stone and clay product manufacturing	[21]	Number of quicklime and slaked lime production sites	[22]

S22	Carbon black manufacturing	Method 4	-	-	Carbon black output per plant (13 sites) (10 ³ t)	[23]	
S23	Coke manufacturing	Method 4	-	-	Pig iron output per manufacturer (10 ⁴ t) Crude steel output by steelworks (10 ³ t)	[15] [16]	
S24	Pulp and paper manufacturing	Method 1	Number of employees in “pulp manufacturing” (value of raw materials used (yen))	[24]	Value of raw materials used for manufacturing of pulp, paper and paper-worked products	[21] [25]	
S25	Chlor-alkali industry	-	-	-	-		
S26	Battery manufacturing	Method 2	Value of raw materials used for “primary battery (dry cell and wet cell batteries) manufacturing” (yen)	[24]	-	Product shipment value of “electrical machinery and appliance manufacturing”, a medium-sized industry (yen)	[3]
S27	Electric switch manufacturing	Method 2	Value of raw materials used for “connector, switch and relay manufacturing” (yen)	[24]	-	Product shipment value of “electronic component and device manufacturing”, a medium-sized industry (yen)	[3]
S28	Fluorescent light manufacturing	Method 2	Value of raw materials used for “light bulb manufacturing” (yen)	[24]	-	Product shipment value of “electrical machinery and appliance manufacturing”, a medium-sized industry (yen)	[3]
S29	Cremation	Method 3	-	Elderly population	[13]	Number of cremation facilities	[26]

S30	Collection and crushing of fluorescent lights	-	-	-	-	-
S31	Dental services (amalgam)	Method 2	Number of patients at dental clinics	[27]	-	Number of workers at dental clinics [4]
S32	Transportation (gasoline)	Method 2	Gasoline consumption of transportation sector (passenger cars) (kl)	[2]	-	Length of main and secondary road network (km) [28] Traffic volume (vehicle units) by car type (4 types) and by workday or holiday [29] Travel distance (1000 km) by car type and fuel consumption (kl) [30]
S33	Transportation (jet fuel)	-				
S34	Transportation (kerosene)	-			-	-
S35	Transportation (diesel)	Method 2	Diesel consumption of transportation sector (passenger cars) (kl)	[2]	-	Length of main and secondary road network (km) [28] Traffic volume (vehicle units) by car type (4 types) and by workday or holiday [29] Travel distance (1000 km) by car type and fuel consumption (kl) [30]
S36	Transportation (heavy oil)	-	-		-	
S37	Volcanoes	-				

Table S2:

Statistics used for spatial allocation of emissions from industrial boilers (coal) and corresponding relations between sectors and industries in the statistics

Subcategory Code	Subcategory	Stats by pref. Fuel code	Stats by pref. Fuel	Stats by pref. Sector code	Stats by pref. Sector	Grid Stat. category	Grid stats	Grid stats Industry code	Grid industry
4	Industrial boilers (petroleum)	250B	Fuel oil products	6100A	Agriculture, forestry and fisheries	3	National Census 2005	1001	Agriculture
								1002	Forestry
								1003	Fisheries
				6100B	Construction, mining	2	Establishment and Enterprise Census 2006	1004	Mining
								1005	Construction
				6500A	Chemicals, synthetic fibers, paper pulp	1	Manufacturing Census 2005	11	Textile industry (excl. clothing and other textile products)
								15	Manufacturing of pulp, paper, and paper-worked products
								17	Chemical industry
				6500B	Steel, nonferrous metals, ceramics, stone and clay products			22	Ceramics, stone, and clay product manufacturing
								23	Steel industry
				6500C	Machinery			24	Nonferrous metal manufacturing
								26	General machinery and appliance manufacturing
								27	Electrical machinery and appliance manufacturing
								28	Communication equipment and device manufacturing
								29	Electronic component and

									device manufacturing
								30	Transportation equipment and device manufacturing
								31	Precision machinery and appliance manufacturing
				6500E	Other industries and small and medium-sized manufacturers			9	Food product manufacturing
								18	Petroleum and coal product manufacturing
								22	Ceramics, stone, and clay product manufacturing
								32	Other manufacturing businesses
				7500	Offices	2	Establishment and Enterprise Census 2006	1007	Power, gas, and heat, and water supply
								1008	Transportation and communication businesses
								1009	Wholesale, retail, and restaurants
								1010	Financial and insurance services
								1011	Real estate
								1012	Services
								1013	Public services

Table S3:

Statistics used for spatial allocation of emissions from industrial boilers (petroleum) and corresponding relations between sectors and industries in the statistics

Subcategory Code	Subcategory	Stats by pref. Fuel code	Stats by pref. Fuel	Stats by pref. Sector code	Stats by pref. Sector	Grid Stat. category	Grid stats	Grid stats Industry code	Grid industry
2	Industrial boilers (coal)	100	Coal	6500A	Chemicals, synthetic fibers, paper pulp	1	Manufacturing Census 2005	11	Textile industry (excl. clothing and other textile products)
				6500B	Steel, nonferrous metals, ceramics, stone and clay products			15	Manufacturing of pulp, paper, and paper-worked products
								17	Chemical industry
								22	Ceramics, stone, and clay product manufacturing
								23	Steel industry
								24	Nonferrous metal manufacturing
				6500C	Machinery			26	General machinery and appliance manufacturing
								27	Electrical machinery and appliance manufacturing
								28	Communication equipment and device manufacturing
								29	Electronic component and device manufacturing
								30	Transportation equipment and device manufacturing
								31	Precision machinery and appliance manufacturing

				6500E	Other industries and small and medium-sized manufacturers			9	Food product manufacturing
								18	Petroleum and coal product manufacturing
								22	Ceramics, stone, and clay product manufacturing
								32	Other manufacturing businesses

Table S4:**Estimated mercury emissions from infectious and non-infectious waste materials**

Types of medical waste	Amount incinerated in 2002 [Gg/y]		Amount incinerated in 2005 [Gg/y]		Mercury emissions in 2005 [Mg/y]	
	Min.	Max.	Min.	Max.	Min.	Max.
Medical waste (total)	944	1337	1095	1371	0.57	1.68
Infectious waste	120	197	139	202	0.072	0.248
Non-infectious waste	824	1140	956	1169	0.498	1.432

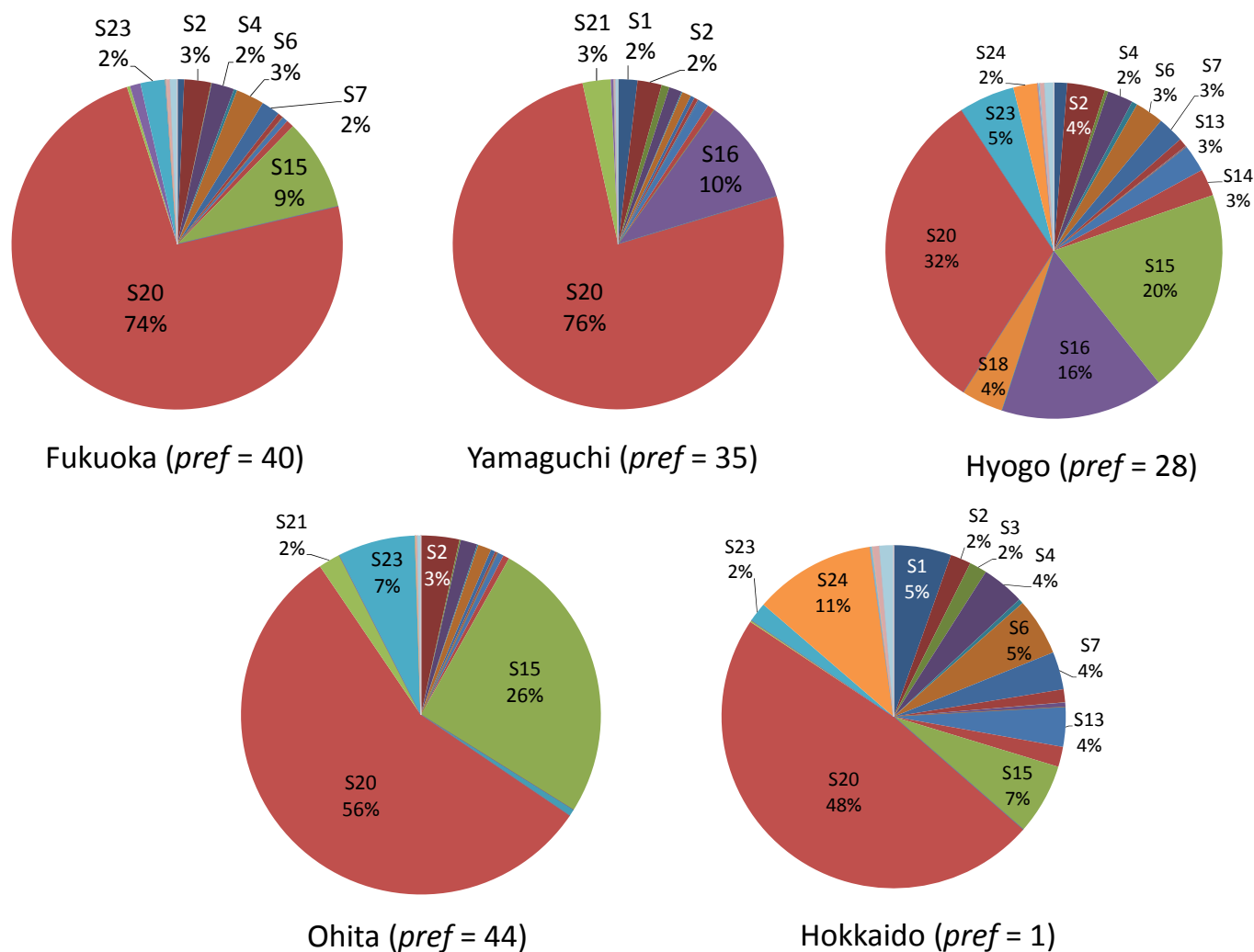


Figure S1: Breakdown of emissions of the five prefectures with the highest mercury emissions in 2005 by emission source category, as specified in Table 1. The ‘PrefCode’ codes of Japanese prefectures follows the *Standard Codes for Areas of Prefectures and Municipalities for Statistical Use* [27].

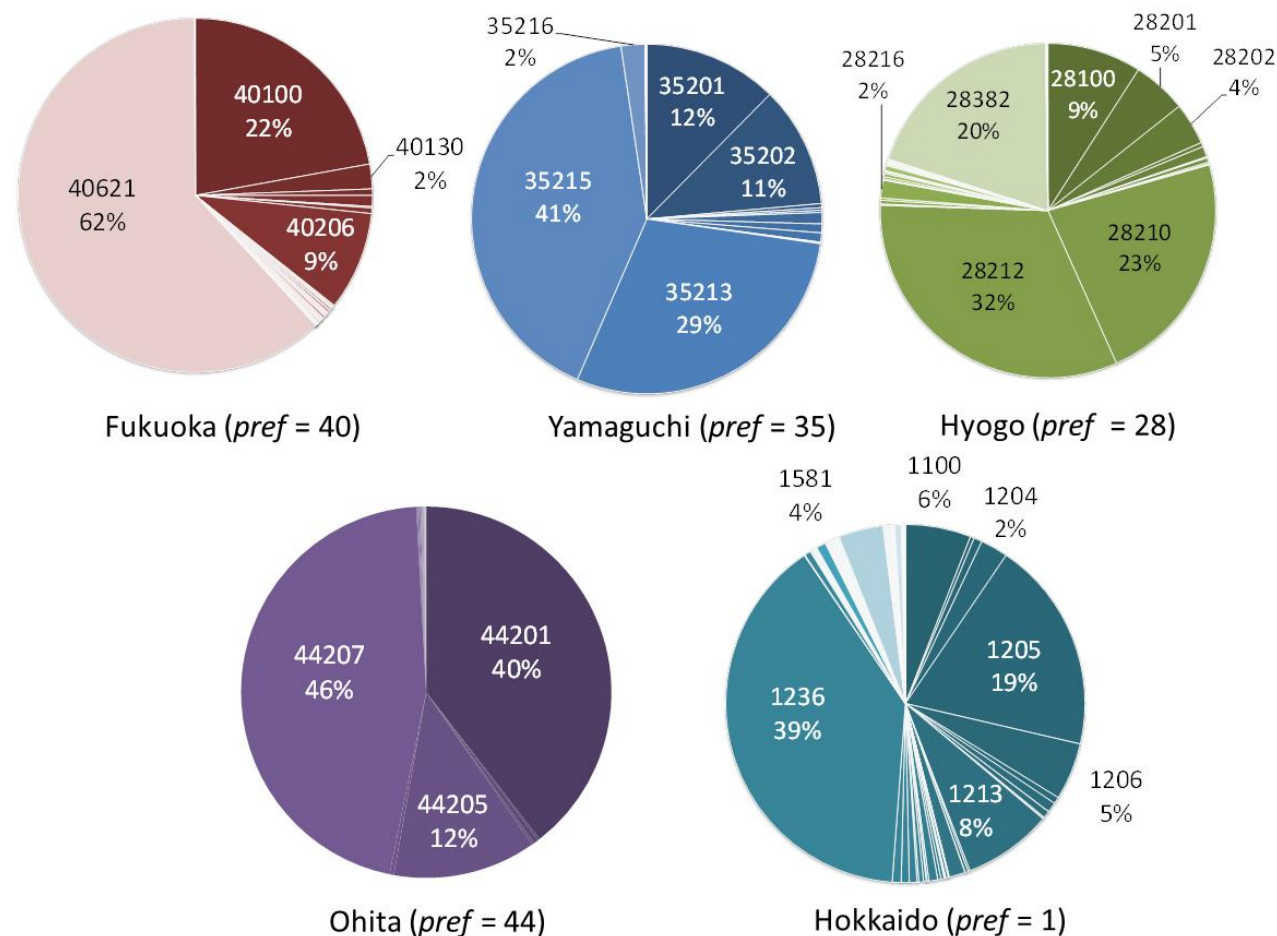


Figure S2: Breakdown of emissions of the five prefectures with the highest mercury emissions in 2005 by contributing municipality. The 'MunCode' codes of Japanese municipalities in the pie charts follow the *Standard Codes for Areas of Prefectures and Municipalities for Statistical Use* [27].

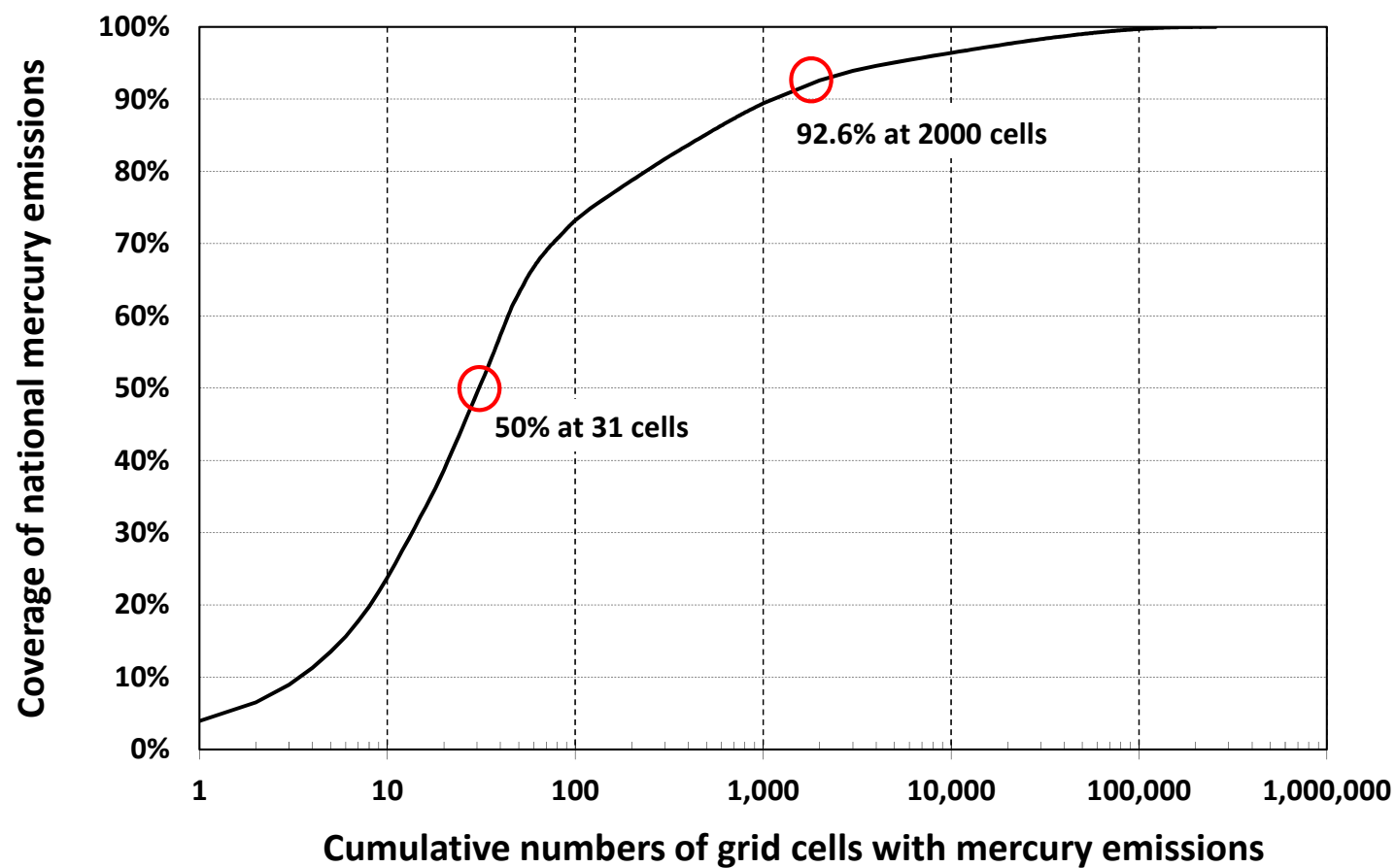


Figure S3: Relationship between cumulative number of grid cells in this study and their coverage of national mercury emissions.

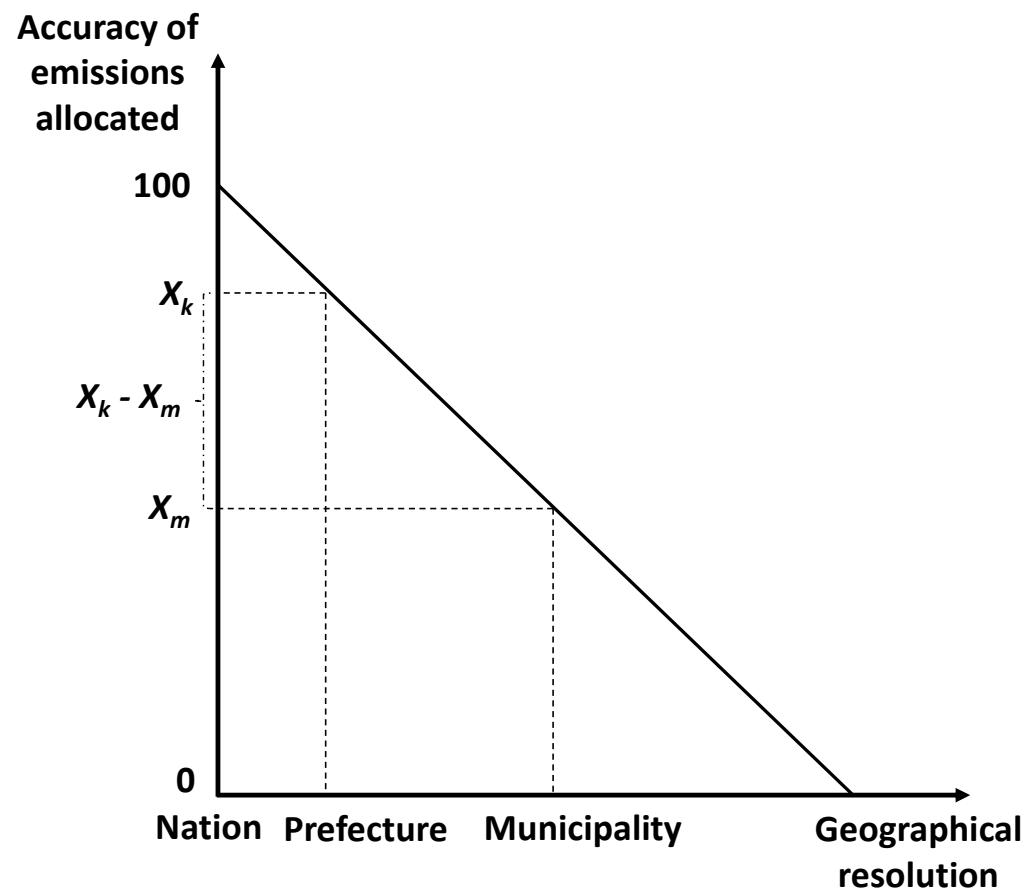


Figure S4: Hypothetical relationship between the geographical resolution of statistics used to allocate emissions and the accuracy of the allocated emissions at each resolution.

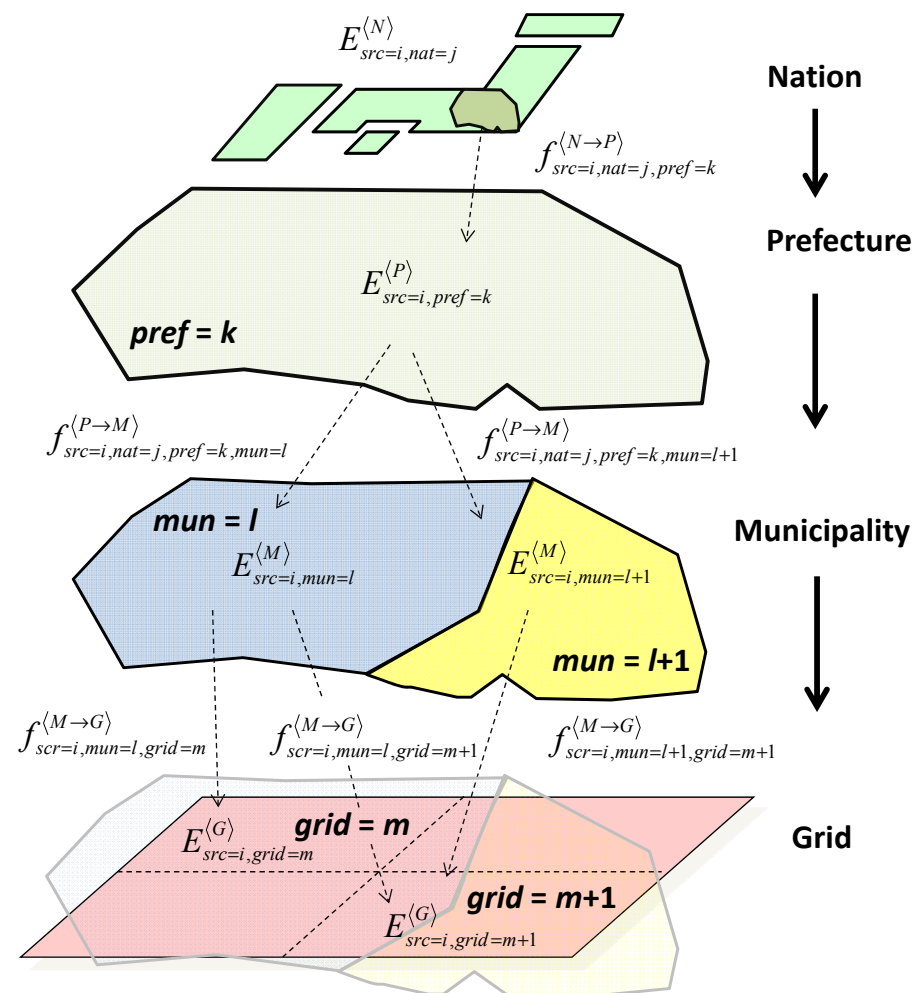


Figure S5: Conceptual scheme of stepwise spatial allocation of national mercury emissions.