

## Supplementary Information for “Two decades of changing anthropogenic mercury emissions in Australia: inventory development, trends, and atmospheric implications”

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### Contents

1. Text S1: Test of the delayed disposal assumptions
2. Table S1
3. Text S2: Description of the supplemental figures
4. Figures S1-S3
5. Table S2
6. References

### S1. Test of the delayed release assumptions

As described in the main text, when calculating the emissions from commercial product waste, we assumed a delay between consumption and disposal for wiring and measuring devices. We did not assume any delay for lamps, batteries, or medical devices on the basis that lifetimes for these product categories are thought to be 10 years or less and our estimates were developed using decadal-scale data (see Appendix 1). Here we test the implications of this assumption and show that the resulting change in waste mercury emissions is much smaller than the 300% uncertainty on emissions from this sector (Section 2.1.4).

For this test, we make the following assumptions about delays between consumption and disposal:

1. Following Cain et al.<sup>1</sup>, we assume 100% of lamps are disposed of 5 years after consumption.
2. For batteries, Jasinski<sup>2</sup> assumed all batteries were disposed of after 2 years, while a report to the Arctic Council<sup>3</sup> suggested 60% are disposed of after 1 year, 20% after 2 years and 10% after 3 years. We find both methods yield similar Australian consumption estimates and so use the more recent Arctic Council assumptions.
3. For medical devices, Floyd et al.<sup>4</sup> reported lifespans of 5 years for thermometers and 10 years for sphygmomanometers. Without further data on the breakdown between thermometers and sphygmomanometers in the “medical devices” category from our input dataset, we assumed 50% disposal after 5 years and 50% disposal after 10 years.

To apply sub-decadal delays required estimating sub-decadal consumption. For lamps this was straightforward as the original Oceania consumption data was available at 5-year intervals from the sources cited in Table A5. The 5-yearly data was sufficient in this case given the 5-year disposal assumption described above. For batteries, we again used the original 5-yearly Oceania consumption data (from the sources cited in Table A5) and then applied a linear interpolation to arrive at annual consumption estimates to which we applied

the 1-3 year delayed disposal assumptions described above. For medical devices, the original data were only available at 10-year intervals. We interpolated between decadal values to estimate 5-yearly consumption estimates to which we applied to the 5-10 year delayed disposal assumptions described above. From here, we converted from Oceania totals to Australian totals and applied the air emissions factors as described in Appendix 1.

The resultant product waste emissions are compared to the original emissions in Table S1. The table shows that incorporating delayed disposal for lamps, batteries, and medical devices has only a small effect (~200 kg) on emissions from the waste sector. The new modified estimate is ~200kg lower in 2000 and ~200kg higher in 2010, slightly dampening the trend. There is no difference between the two assumptions in 2020. At all points in time, the change is significantly smaller than the 300% uncertainty on emissions from this sector (Section 2.1.4).

**Table S1.** Air emissions from disposal of mercury in products in Australia, in Mg Hg emitted to air.

	2000	2010	2020
<b>Original delay assumptions <sup>a</sup></b>	2.8	1.4	1.5
<b>Modified delay assumptions <sup>b</sup></b>	2.6	1.6	1.5

<sup>a</sup> Incorporates delay between consumption and disposal for wiring and measuring devices only.

<sup>b</sup> Incorporates delay between consumption and disposal for all commercial products (wiring and measuring devices, lamps, batteries, and medical devices).

## S2. Description of the supplemental figures

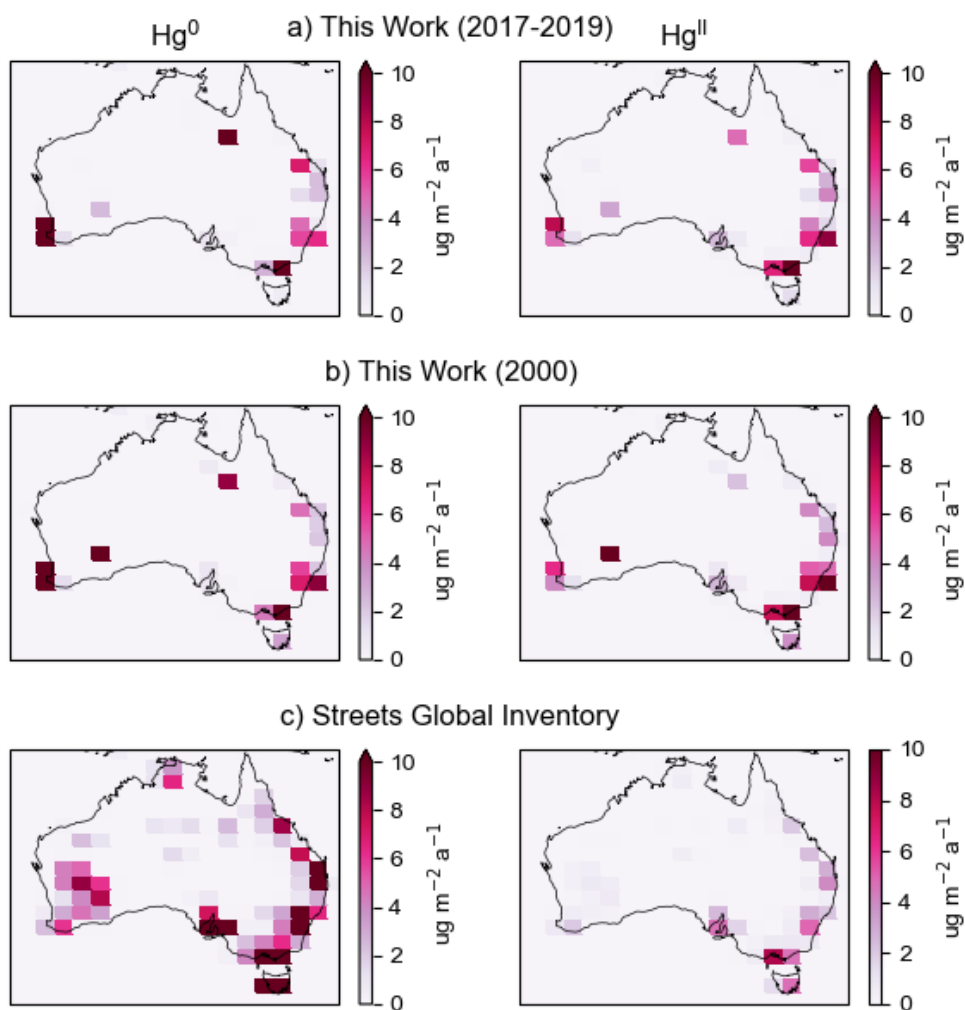
Figure S1 shows the anthropogenic emissions gaseous elemental mercury ( $\text{Hg}^0$ ) and oxidised mercury ( $\text{Hg}^{\text{II}}$ ) as used in the GEOS-Chem simulations described in the main text. Figures S1a and S1b display emissions from the new inventory described in this work, for present day (2017-2019) and the year 2000, respectively. Comparing the two panels shows changes consistent with those described in Section 3.1, most notably the large decrease in present-day emissions from the Kalgoorlie facility (inland southwestern Australia), relative to 2000 emissions. Smaller decreases are seen in urban and industrialised regions around Melbourne/Latrobe Valley and Sydney/Hunter Valley. Emission increases are seen at Mount Isa (inland northern Australia). Figure S1c shows the same emissions but for the Streets global inventory for the year 2015 (the most recent year). Comparing Figures S1a and S1c shows significant differences in both the spatial distribution and magnitude of the emissions, as well as the speciation between  $\text{Hg}^0$  and  $\text{Hg}^{\text{II}}$ . These differences and their impacts are discussed in more detail in the main text.

Figure S2 provides a regional perspective as to how mercury concentrations in surface air compare between the different model simulations described in the text. The figure is the same as Figure 5 in the main text, except for the regional bounds and the ranges used in the colour scales. Differences that can be directly attributed to changes in anthropogenic mercury emissions are shown in Figure S2c (change over time) and S2d (change of inventory). These two panels show that the impacts of changes in Australian anthropogenic emissions are localised to the Australian continent, with downwind changes in surface air concentrations of only a few percent at most.

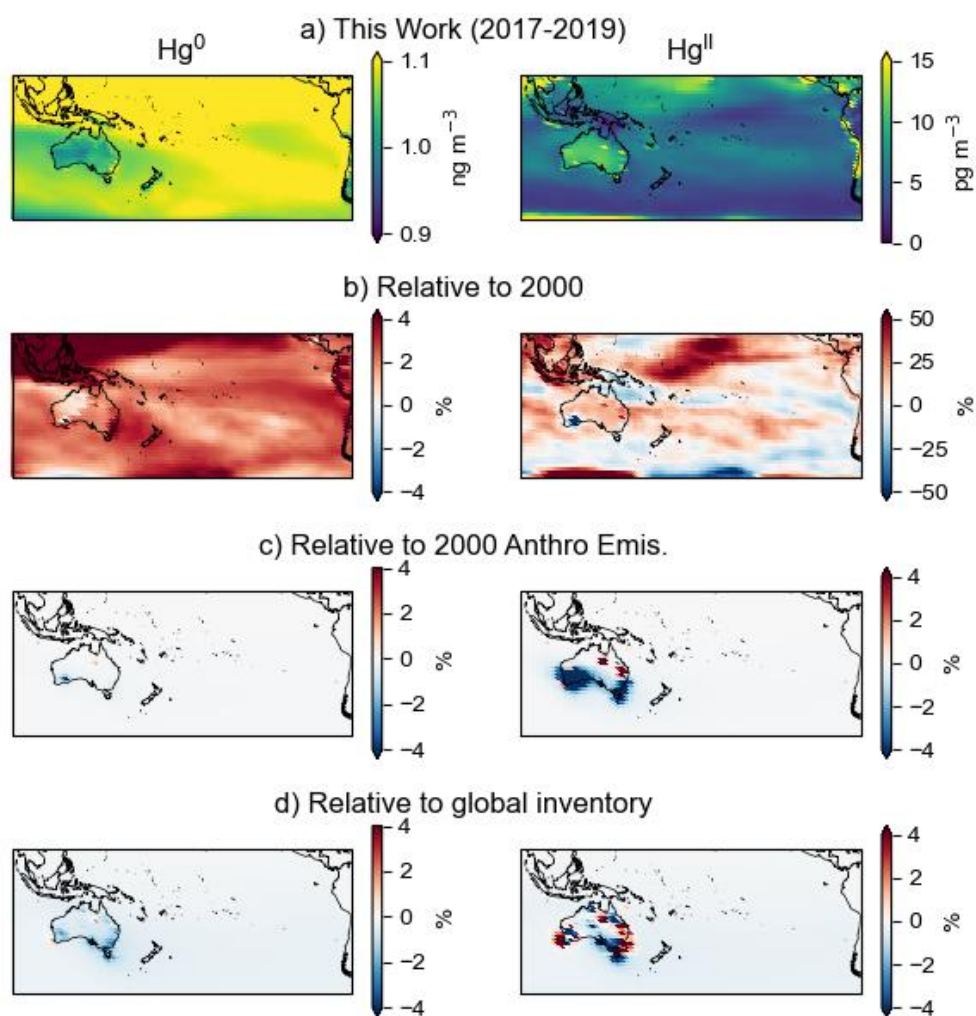
Figure S3 provides the same comparisons as Figure S2, but for mercury deposition. This is the same as Figure 6 in the main text, except for the regional bounds and the ranges used in

the colour scales. As seen for the surface air mercury concentrations, the changes in mercury deposition that can be directly attributed to changes in anthropogenic emissions are localised to Australia, with changes downwind of no more than a few percent.

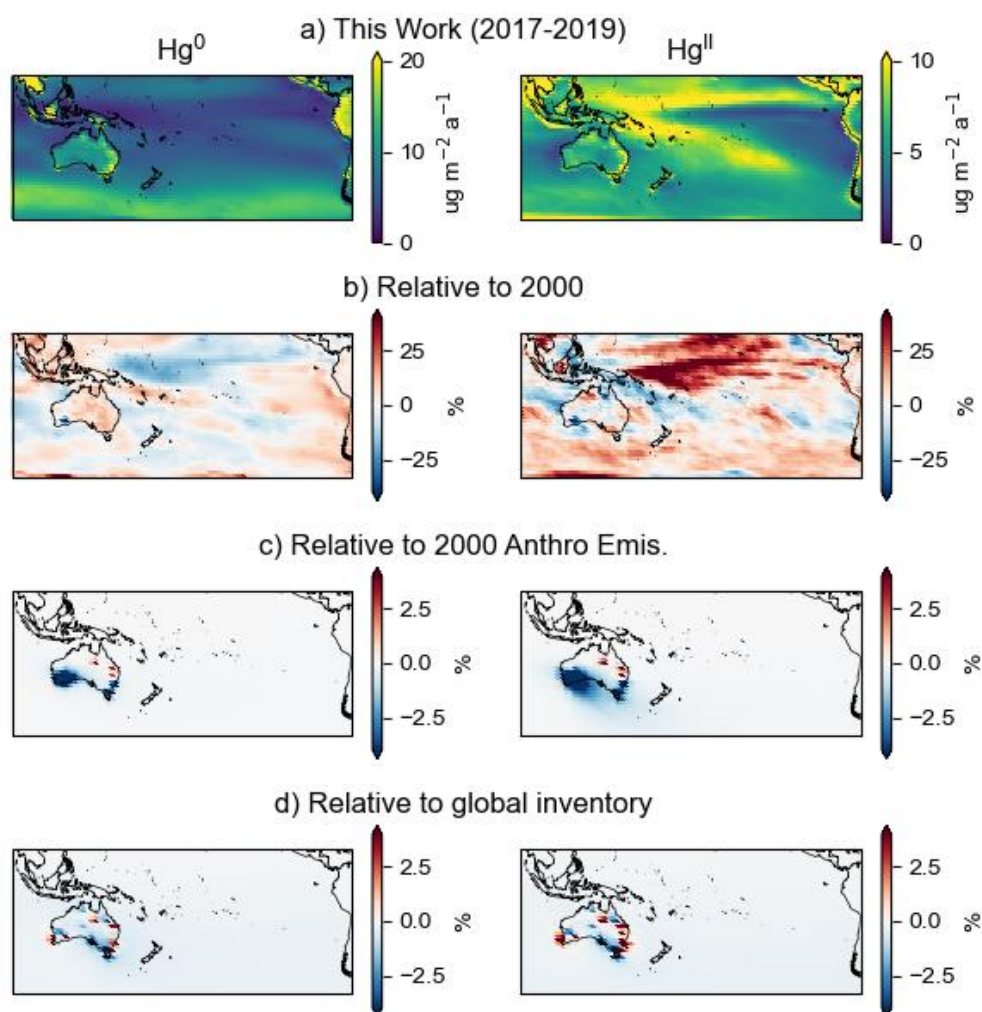
Table S2 compares the  $\text{Hg}^{\text{II}}$  speciation fractions between our inventory and the Streets global inventory. The table shows that similar speciation values are used for most sectors. The exceptions are the production of cement, which is more weighted towards  $\text{Hg}^{\text{II}}$  in our work (76%) than in the Streets inventory (49%) and most non-ferrous metals, with  $\text{Hg}^{\text{II}}$  speciation in our work versus Streets: of 68% vs 20% (gold), 50% vs 36% (copper), 61% vs 26% (lead), and 45% vs 27% (zinc).



**Figure S1.** Anthropogenic mercury emissions at the resolution of the GEOS-Chem model for elemental mercury ( $\text{Hg}^0$ , left) and oxidised mercury ( $\text{Hg}^{\text{II}}$ , right) from (a,b) the new Australian anthropogenic emissions inventory derived in this work for 2017-2019 (a) and 2000 (b) and from (c) the Streets global inventory. Note that the most recent year of emissions data available in the Streets inventory is 2015.



**Figure S2.** Same as Figure 5 in the main text (mean mercury concentration in surface air), but for a greater regional extent.



**Figure S3.** Same as Figure 6 in the main text (mercury deposition), but for a greater regional extent.

**Table S2.** Differences in speciation between our inventory and the Streets global inventory, shown as the fraction of emissions that are emitted as oxidised mercury ( $\text{Hg}^{\text{II}}$ ) for each sector.

Sector	This work $\text{Hg}^{\text{II}}$ %	Streets $\text{Hg}^{\text{II}}$ %
Black Coal	47	47
Brown Coal	42	47
Cement	76	49
Steel	68	55
Iron	68	60
Gold	68	20
Copper	50	36
Lead	61	26
Zinc	45	27
Other Non-Ferrous Metals	20	n/a
Refined Petroleum Products	50	50
Waste	80	78-80

## References

- 1 A. Cain, S. Disch, C. Twaroski, J. Reindl and C. R. Case, Substance Flow Analysis of Mercury Intentionally Used in Products in the United States, *Journal of Industrial Ecology*, 2007, **11**, 61–75.
- 2 S. M. Jasinski, *The Materials Flow of Mercury in the United States*, United States Department of the Interior, 1994.
- 3 Russian Federal Service for Environmental, Technological and Danish Environmental Protection Agency, *Assessment of Mercury Releases from the Russian Federation*, Arctic Council, 2005.
- 4 P. Floyd, P. Zarogiannis, M. Crane, S. Tarkoswki and V. Bencko, *Risks to health and the environment related to the use of mercury products*, Risk & Policy Analysts Limited, 2002.