

Global Biogeochemical Cycles

Supporting Information for

A Mass Budget for Mercury and Methylmercury in the Arctic Ocean

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Introduction

The Supporting Information contains two texts, nine tables, and four figures.

Rate constants

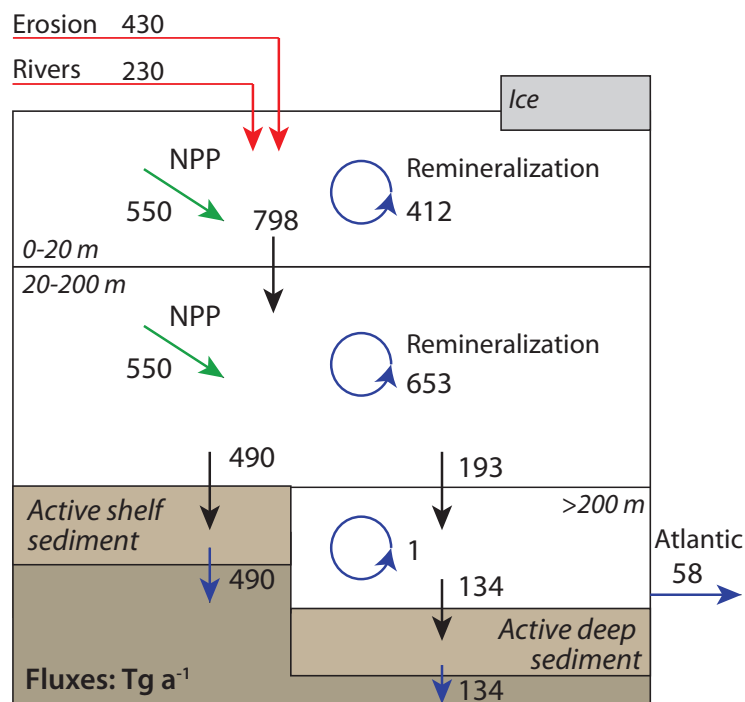
Table S1. Compilation of measured rate constants for Hg^{II} methylation and MeHg demethylation in seawater.

| | avg±stdv (d ⁻¹) | median (d ⁻¹) | min-max (d ⁻¹) | avg±stdv (m ² E ⁻¹) | Reference |
|---------------------------------------|-----------------------------|---------------------------|----------------------------|--------------------------------------------|----------------------------|
| Methylation | | | | | |
| Unfiltered (dark) | 0.0068±0.0039* | 0.0050* | 0.0006 – 0.013* | | [Lehnherr et al., 2011] |
| Unfiltered (light) | 0.0179±0.0197 | 0.0085 | <0.0002 – 0.060 | | [Monperrus et al., 2007] |
| Unfiltered (dark) | 0.0154±0.0119 | 0.0130 | <0.0002 – 0.038 | | [Monperrus et al., 2007] |
| Filtered (light) | 0.0072±0.0072 | 0.0040 | <0.0002 – 0.017 | | [Monperrus et al., 2007] |
| Filtered (dark) | 0.0056±0.0046 | 0.0060 | <0.0002 – 0.013 | | [Monperrus et al., 2007] |
| Unfiltered (dark) | | | <0.0001-0.001 | | [Sharif et al., 2014] |
| Unfiltered (light) | | | <0.0001-0.004 | | [Sharif et al., 2014] |
| Filtered (light) | | | <0.0001-0.003 | | [Sharif et al., 2014] |
| Filtered (dark) | | | <0.0001 | | [Sharif et al., 2014] |
| Unfiltered (dark) | 0.0025 | | Max 0.004 | | [Schartup et al. 2015] |
| n/a | 0.13 | | | | [Mason and Sullivan, 1999] |
| Demethylation | | | | | |
| Unfiltered (dark) | 0.36±0.09* | | 0.23 – 0.59* | | [Lehnherr et al., 2011] |
| Filtered (light) | 0.04 ^a * | | | 1×10 ⁻³ * | [Lehnherr et al., 2011] |
| Filtered (light) | 0.2 ^b * | | | | [Lehnherr et al., 2011] |
| n/a | | | <0.09 – 0.43 | | [Whalin et al., 2007] |
| Unfiltered (light) | 0.16±0.05 | 0.159 | 0.048 – 0.245 | | [Monperrus et al., 2007] |
| Unfiltered (dark) | 0.06±0.03 | 0.059 | <0.015 – 0.109 | | [Monperrus et al., 2007] |
| Filtered (light) | 0.09±0.04 | 0.090 | 0.033 – 0.139 | | [Monperrus et al., 2007] |
| Filtered (dark) | 0.05±0.03 | 0.064 | <0.015 – 0.088 | | [Monperrus et al., 2007] |
| Unfiltered (light) | | | | 3.2(±1.0) ×10 ⁻³ | [Black et al., 2012] |
| Unfiltered (light) | | | | 9.9(±26.0) ×10 ⁻³ | [Black et al., 2012] |
| Unfiltered (dark) | | | 0.07-0.21 | | [Sharif et al., 2014] |
| Unfiltered (light) | | | 0.06-0.55 | | [Sharif et al., 2014] |
| Filtered (light) | | | 0.08-0.29 | | [Sharif et al., 2014] |
| Filtered (dark) | | | <0.02-0.13 | | [Sharif et al., 2014] |
| Dimethylation | | | | | |
| Unfiltered (dark) | | | nd-0.0016* | | [Lehnherr et al., 2011] |
| Me₂Hg decomposition | | | | | |
| Light | | | | <1×10 ⁻³ | [Black et al., 2009] |
| n/a | | | 0.0002 | | [Mason et al., 1995] |
| Light | | | 0.32 – 1.64 | | [Mason and Sullivan, 1999] |
| Dark | | | 0.16 – 0.22 | | [Mason and Sullivan, 1999] |

* Arctic data, ^a 9 days incubation, ^b 24 hours incubation

Budgets

A) Solids budget



B) Hydrologic budget

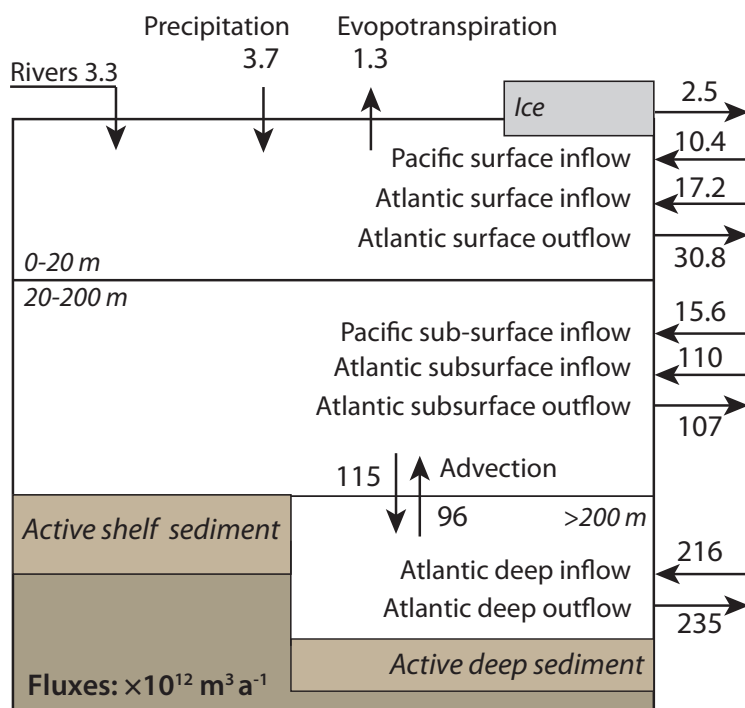


Figure S1. A: Solids budget for the Arctic Ocean in Tg a^{-1} . Red arrows denote external sources, green arrows denote internal sources, black arrows denote internal fluxes and blue arrows represent losses. The fraction of solids from rivers and NPP remaining after remineralization at each depth is 50% in the PML, 30% in the subsurface water and 1% in the deep ocean [Cai *et al.*, 2010; Moran *et al.*, 1997; Rachold *et al.*, 2004]. Solids from erosion are assumed not to remineralize. B: Hydrologic budget for external input and losses and internal circulation in the Arctic Ocean ($\times 10^{12} \text{ m}^3 \text{ a}^{-1}$).

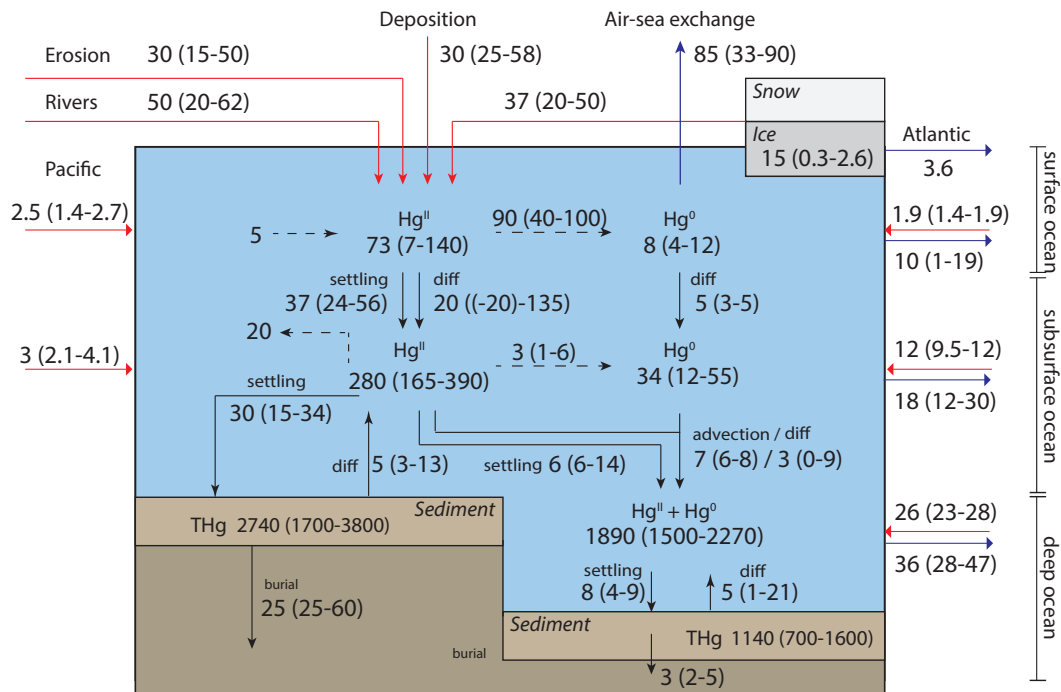


Figure S2. Empirical mass budget for the Arctic Ocean for inorganic Hg species including uncertainty ranges for masses and fluxes not given in Table S2. Red arrows are external sources, black arrows are internal fluxes and blue arrows are losses. Punctuated lines describe net fluxes. Masses are in Mg and fluxes in Mg a^{-1} .

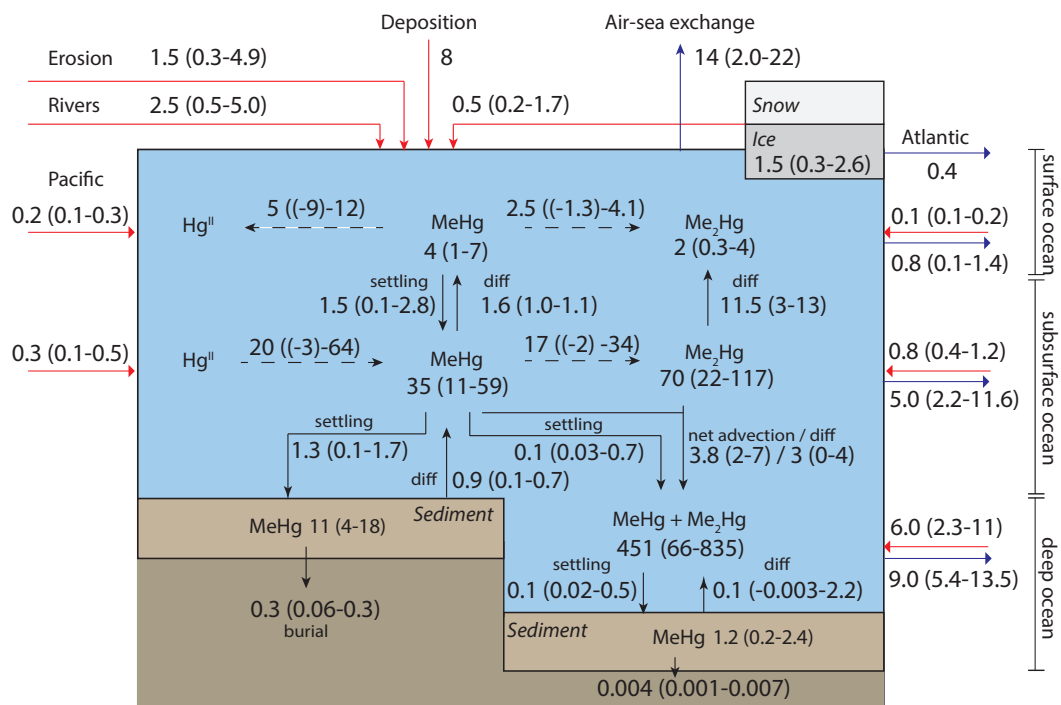


Figure S3. Empirical mass budget for the Arctic Ocean for organic Hg species including uncertainty ranges for masses and fluxes not given in Table S2. Red arrows are external sources, black arrows are internal fluxes and blue arrows are losses. The punctuated lines describe the net flux of biotic methylation/demethylation. Masses are in Mg and fluxes in Mg a^{-1} .

Observations

Text S1. Information on North Atlantic inflow data

Total inorganic Hg and methylated Hg concentrations from the CLIVAR A16N Cruise Track in the North Atlantic for stations occupied in August 2013. Measurements and analytical details were the same as Sunderland et al. (2009). The same depth intervals as in the Arctic were used to calculate average depth concentrations. Data are from station 007 (61.6 N; 20.0 W) and station 015 (58.0 N; 20.0W)(n=24).

| | | |
|---------------------------|------------------|-------------------------|
| 0-20 meter, surface: | 0.48±0.08 pM Hg, | 42±25 fM methylated Hg |
| 20-200 meter, subsurface: | 0.50±0.06 pM Hg, | 38±18 fM methylated Hg |
| >200 meter, deep ocean: | 0.6±0.06 pM Hg, | 153±99 fM methylated Hg |

Text S2. Canadian Arctic Archipelago Seawater Samples Collected by I. Lehnherr et al.

Seawater samples were collected at 17 stations across the Canadian Arctic Archipelago (Figure 1), in 2006, 2007 and 2010, between 7-August and 16-October while aboard the *CCGS Amundsen* icebreaker as part of the ArcticNet research program (N = 22, 5 stations were sampled in two separate years). At each station, 12 L Teflon-lined Niskin bottles (General Oceanics) mounted on the ship's rosette system were used to obtain samples from various depths, generally corresponding to the surface (~2-4 m), the subsurface chlorophyll maximum (SCM, the depth of maximum chlorophyll *a* fluorescence and hence algal biomass) and the lower oxycline (a zone of high heterotrophic respiration and organic carbon remineralization) and the bottom of the water column (~5-10 m above the seafloor). Subsamples were collected from the Niskin bottles into pre-cleaned amber Boston round bottles (Environmental Sampling Supplies) using the “clean hands-dirty hands” Hg sampling protocol. All samples were processed and preserved immediately after collection for analysis at a later date. Methylated Hg (MeHg + Me₂Hg) samples were preserved by acidification using trace-metal grade concentrated H₂SO₄ (equivalent to 0.4% of sample volume), the resulting low pH leading to the quantitative conversion of Me₂Hg to MeHg [Black *et al.*, 2009]. Total Hg samples were preserved with the addition of trace-metal grade concentrated HCl (0.2% of sample volume). MeHg samples were preserved by freezing. In 2006, Me₂Hg was collected using the purge and trap technique whereby seawater collected in a custom-made 1 L glass vessel was purged with ultra-high purity (UHP) N₂ gas and the Me₂Hg thus displaced from solution was collected in traps consisting of glass columns packed with Carbo trap. For the 2007 and 2010 data, Me₂Hg was calculated from the difference between methylated Hg and MeHg. Particles (seston) were collected in 2010 by filtering 5-9 L of seawater through 0.2 mm cellulose nitrate membranes. Filter membranes were frozen immediately after sample collection and freeze-dried once back on shore. Sample analysis for methylated Hg, MeHg and total Hg was carried out as described in Lehnherr *et al.* [2011]. MeHg was extracted from particles, by distilling the complete membrane filter, with Me²⁰¹Hg added as a species-specific internal standard prior to distillation to correct for procedural recoveries, similar to the procedure described for sediments in Lehnherr *et al.* [2012]. The distillate was then analyzed in the same fashion as with seawater samples.

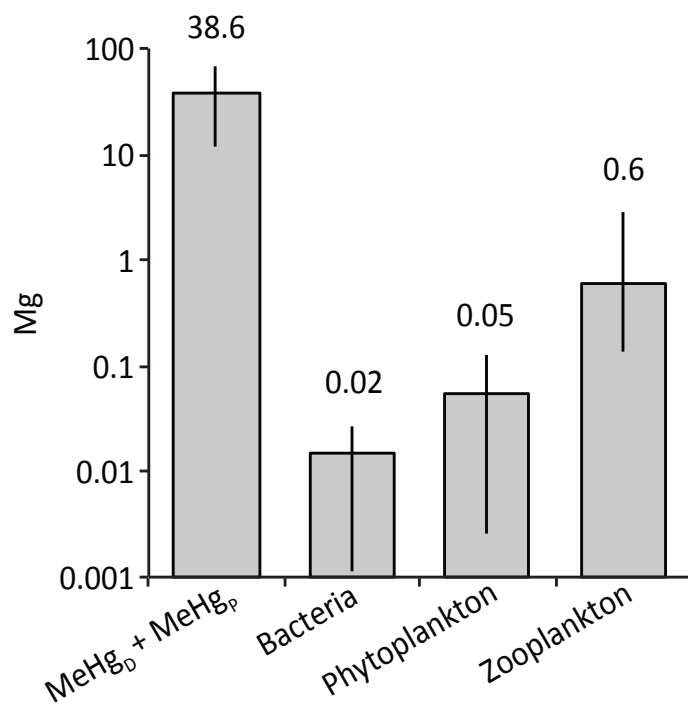
Biota

Table S2. Arctic zooplankton MeHg concentrations

| MeHg (ng g ⁻¹ dw) | MeHg:THg (%) | Reference |
|---------------------------------|-----------------|-----------------------------|
| <i>Mixed zooplankton</i> | | |
| 4±3 ^A | 70±38 | [Campbell et al., 2005] |
| 10±1 | 28 | [Loseto et al., 2008] |
| 95±19 | 75 | [Loseto et al., 2008] |
| 28±0.8 ^A | 100 | [Campbell et al., 2005] |
| <i>Copepods</i> | | |
| 7±1 | 28 | [Loseto et al., 2008] |
| 11±1 | 34 | [Loseto et al., 2008] |
| 7±1 | 51 | [Pucko et al., 2014] |
| 40±9 | 84 | [Pucko et al., 2014] |
| 5-28 | 10-60 | [Stern and Macdonald, 2005] |
| <1.5 ^A | | [Jaeger et al., 2009] |
| 2±1 ^A | 8 | [Campbell et al., 2005] |
| <i>Themisto libellula</i> | | |
| 66±2 | 75 | [Loseto et al., 2008] |
| <1.5 ^A | | [Jaeger et al., 2009] |
| <i>Themisto abyssorum</i> | | |
| 88±10 | 58 | [Pucko et al., 2014] |
| <i>Chaethognats</i> | | |
| 17±4 | 74 | [Pucko et al., 2014] |

^A ng g⁻¹ wet weight

Figure S4. Reservoirs of MeHg in plankton and bacteria (note log scale). Note that in the Arctic phytoplankton and zooplankton often has an inverted biomass pyramid and that the zooplankton biomass therefore is higher than that of phytoplankton [ACIA, 2005].



Model parameter description

Table S3. Atmospheric deposition and melt water flux from snow and ice

| | | | |
|----------------------------------|---------------------------------------------------------------------------------------|------------------------------------------------------------------------|----------------------------------------------------------------|
| <i>Deposition</i> | | | |
| $M_{dep-HgII}$ ($Mg\ a^{-1}$) | Hg ^{II} deposition to open ocean | 30 | [Dastoor and Durnford, 2014; Fisher et al., 2012] |
| $M_{dep-MeHg}$ ($Mg\ a^{-1}$) | MeHg deposition to open ocean based on fast redeposition of evaded Me ₂ Hg | $M_{eva-Me2Hg} \times F_{open-ocean-summer} \times F_{evasion-summer}$ | ($F_{open-ocean-summer}$; $F_{evasion-summer}$: Table 3) |
| <i>Meltwater</i> | | | |
| $M_{melt-HgII}$ ($Mg\ a^{-1}$) | Hg ^{II} from meltwater | $C_{melt-HgII} \times Prec \times (1 - F_{open-ocean})$ | ($F_{open-ocean}$; Prec: Table 3, $C_{melt-HgII}$: Table 4) |
| $M_{melt-MeHg}$ ($Mg\ a^{-1}$) | MeHg from meltwater | $C_{melt-MeHg} \times Prec \times (1 - F_{open-ocean})$ | ($F_{open-ocean}$; Prec: Table 3, $C_{melt-MeHg}$: Table 4) |
| <i>Multi-year sea-ice loss</i> | | | |
| $M_{ice-HgII}$ ($Mg\ y^{-1}$) | Hg ^{II} from multi-year sea-ice loss | 0.42 | [Beattie et al., 2014] |
| $M_{ice-MeHg}$ ($Mg\ y^{-1}$) | MeHg from multi-year sea-ice loss | 0.042 | [Beattie et al., 2014] |

Table S4. River and erosion fluxes

| | | | |
|-------------------------------------|---------------------------------------------------|---------------------------------------------------------|--------------------------------------------------|
| $M_{river-HgII}$ ($Mg\ a^{-1}$) | River Hg ^{II} discharge | 50 | [Dastoor and Durnford, 2014; Zhang et al., 2015] |
| $M_{river-MeHg}$ ($Mg\ a^{-1}$) | River MeHg discharge | $M_{river-HgII} \times F_{river-MeHg}$ | |
| $M_{erosion-MeHg}$ ($Mg\ a^{-1}$) | Erosion MeHg input | $M_{erosion-HgII} \times F_{MeHg-sediment}$ | |
| $M_{erosion-HgII}$ ($Mg\ a^{-1}$) | Erosion Hg ^{II} input | $TSS_{erosion} \times C_{erosion-HgII} \times 10^{-12}$ | |
| $F_{river-MeHg}$ | MeHg:Hg ^{II} fraction in river discharge | 0.05 (0.01-0.10) | (Table 4) |
| $TSS_{erosion}$ ($g\ a^{-1}$) | TSS from erosion | 430×10^{12} | (Figure 1) |
| $C_{erosion-HgII}$ ($ng\ g^{-1}$) | [Hg ^{II}] in eroded TSS | 81 (61-114) | [Leitch, 2006] |
| $F_{MeHg-sediment}$ | Fraction of eroded Hg ^{II} found as MeHg | 0.01 | [Hollweg et al., 2010] |

Table S5. Chemical transformation in the water column.

| | | | |
|-------------------------------------------|----------------------------------------------------------------------------|-----------------------------------------------------------------------------------|-----------------------------------------------------------|
| M_{ox} ($Mg\ a^{-1}$) | Oxidation flux | $M_{w-Hg0} \times (k_{phot-ox} + k_{dark-ox}) \times 365$ | |
| M_{red} ($Mg\ a^{-1}$) | Reduction flux | $M_{w-HgII} \times F_{red} \times (k_{bio-red} + k_{phot-red}) \times 365$ | |
| M_{met} ($Mg\ a^{-1}$) | Methylation flux | $M_{w-HgII} \times k_{met} \times 365$ | |
| M_{dem} ($Mg\ a^{-1}$) | Demethylation flux | $M_{w-MeHg} \times (k_{phot-dem} + (k_{bio-dem} \times F_{rad-area})) \times 365$ | |
| M_{met2} ($Mg\ a^{-1}$) | MeHg to Me ₂ Hg flux | $M_{w-MeHg} \times k_{met2} \times 365$ | |
| M_{2dec} ($Mg\ a^{-1}$) | Me ₂ Hg decomposition flux | $M_{w-Me2Hg} \times (k_{2dec} + k_{phot-2dec}) \times 365$ | |
| M_w (Mg) | Mass of Hg species in specified reservoir | | |
| $k_{phot-ox}$ (d^{-1}) | Photo-oxidation rate constant | $0.57 \times RAD \times (F_{rad-area})$ | |
| $k_{phot-red}$ (d^{-1}) | Photo-reduction rate constant | $0.16 \times RAD \times (F_{rad-area})$ | |
| $k_{dark-ox}$ (d^{-1}) | Dark oxidation rate constant | 0.0086 | [Soerensen <i>et al.</i> , 2010] |
| $k_{bio-red}$ (d^{-1}) | Biotic reduction rate constant | $0.0450 \times NPP$ | [Soerensen <i>et al.</i> , 2010] (NPP: Table 3) |
| k_{met} (d^{-1}) | Methylation rate constant | $0.0134 \times NPP + k_{met-base}$ | [Lehnher <i>et al.</i> , 2011] (NPP: Table 3) |
| $k_{met-base}$ (d^{-1}) | Methylation base rate | 0.0056 | [Monperrus <i>et al.</i> , 2007] |
| $k_{phot-dem}$ ($m^2\ E^{-1}\ d^{-1}$) | Photo-demethylation rate constant | $PAR \times 3 \times 10^{-3} \times F_{summer}$ | (see Table RATES) |
| $k_{bio-dem}$ (d^{-1}) | Biotic demethylation rate constant | 0.07-0.18 | |
| $k_{bio\ dem-base}$ (d^{-1}) | Demethylation base rate | 0.05 | [Monperrus <i>et al.</i> , 2007] |
| k_{met2} (d^{-1}) | MeHg to Me ₂ Hg rate constant | 0.0016 | (see Table RATES) |
| k_{2dec} (d^{-1}) | Me ₂ Hg dark decomposition rate constant | 0.0002 | (see Table RATES) |
| $k_{phot-2dec}$ ($m^2\ E^{-1}\ d^{-1}$) | Me ₂ Hg photodecomposition rate constant | $PAR \times 5 \times 10^{-4} \times F_{summer}$ | 50% of detection limit from Black <i>et al.</i> [2009] |
| F_{red} | Fraction of reducible Hg ^{II} | 0.4 | [Guentzel <i>et al.</i> , 1996] |
| $F_{rad-area}$ | Fraction of ocean where light can penetrate during summer (Apr-Sep) months | $F_{open-ocean} + F_{meltp}$ | (F _{open-ocean} : Table 3) |
| F_{meltp} | Area of ocean with summer melt ponds | 0.1 | [Boetius <i>et al.</i> , 2013] |
| F_{summer}/F_{winter} | Fraction of the year considered summer/winter | 0.5 | |
| PAR ($E\ m^{-2}$) | Shortwave intensity as a function of time | $RAD \times 0.432$ | |
| RAD ($W\ m^{-2}$) | Shortwave intensity at specified depth intervals | $\frac{1}{x_2 - x_1} \cdot \frac{R_i}{\eta} [e^{\eta x_1} - e^{-\eta x_2}]$ | (RAD _i : Table 3) |
| x_1 (m) | Surface depth | | |
| x_2 (m) | Bottom depth | | |
| η (m^{-1}) | Extinction coefficient for radiation | $\eta_w + \eta_{Chl} C_{Chl} + \eta_{DOC} C_{DOC}$ | |
| η_w (m^{-1}) | Extinction coefficient for water | 450 nm (vis) = 0.0145 | |
| η_{Chla} (m^{-1}) | Extinction coefficient for pigments | 450 nm (vis) = 31 | |
| C_{Chla} ($mg\ L^{-1}$) | Average concentration of Chl a in mixed layer | 0.6×10^{-3} | [Galand <i>et al.</i> , 2008; Jin <i>et al.</i> , 2012] |
| η_{DOC} ($mg\ L^{-1}$) | Extinction coefficient for DOC | 450 nm (vis) = 0.654 | |
| C_{DOC} ($mg\ L^{-1}$) | Concentration of DOC in water column | 0.8 | [Dittmar and Kattner, 2003; Hansell <i>et al.</i> , 2012] |

Table S6. Chemical transformation in sediment

| | | | |
|--------------------------------|--------------------------------------|---------------------------------------------------------------|--------------------------------------------------------------------------------|
| $M_{sed-met}$ ($Mg\ a^{-1}$) | Sediment methylation flux | $C_{psed-HgII} \times k_{sed-met} \times 365$ ($d\ y^{-1}$) | |
| $M_{sed-dem}$ ($Mg\ a^{-1}$) | Sediment demethylation flux | $C_{psed-MeHg} \times k_{sed-dem} \times 365$ ($d\ y^{-1}$) | |
| $C_{psed-HgII}$ (pM) | Porewater Hg^{II} concentration | 74.0 (14-258) | $C_{sed-HgII} \times 10^3 / kd_{HgII}$ (C_{sed} : Table 2) |
| $C_{psed-MeHg}$ (pM) | Porewater MeHg concentration | 2.4 (0.3-9.9) | $C_{sed-MeHg} \times 10^3 / kd_{MeHg}$ (C_{sed} : Table 2) |
| kd_{HgII} | Hg^{II} partition coefficient | $10^{4.0}$ ($10^{3.1}$ - $10^{4.2}$) | [Hollweg et al., 2010; Schartup et al., 2015; Sunderland et al., 2010] |
| kd_{MeHg} | MeHg partition coefficient | $10^{2.7}$ ($10^{2.2}$ - $10^{3.1}$) | [Hollweg et al., 2010; Schartup et al., 2015; Sunderland et al., 2010] |
| $k_{sed-met}$ (d^{-1}) | Sediment methylation rate constant | 0.03 | [Hammerschmidt and Fitzgerald, 2008; Heyes et al., 2006; Hollweg et al., 2010] |
| $k_{sed-dem}$ (d^{-1}) | Sediment demethylation rate constant | 0.58 | Derived based on the sediment MeHg: Hg^{II} ratio |

Table S7. Sediment burial

| | | | |
|-------------------------------|----------------------------------------------|------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------|
| M_{burial} ($Mg\ a^{-1}$) | Burial flux of Hg^{II} and MeHg | $M_{sed} \times Burial / D_{sed}$ | Burial rate found in Table 3 |
| M_{sed} (Mg) | Hg species conc. in sediments | $V_{sed} \times Sedsolid \times C_{sed} \times 10^{-15}$ ($Mg\ ng^{-1}$) $\times 10^6$ ($g\ kg^{-1}\ m^3\ l^{-1}$) | (C_{sed} : Table 2) |
| $Sedsolid$ ($kg\ l^{-1}$) | Concentration of solids in benthic sediments | $(1-POR) \times DSS$ | |
| DSS ($kg\ l^{-1}$) | Density of sediment solids | 1.5 | |
| V_{sed} (m^3) | Sediment volume | $A_{sed} \times D_{sed}$ | (A_{sed} : Table 3) |
| $D_{sed-deep}$ (m) | Depth of active deep ocean sediment | 0.01 | [Clough et al., 1997] |
| $D_{sed-shelf}$ (m) | Depth of active shelf sediment | 0.02 | [Clough et al., 1997; Kuzyk et al., 2013; Trefry et al., 2014] |

Table S8. Diffusive transfer

| <i>Sediment-water interphase</i> | | | |
|-----------------------------------------|-----------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------|
| $M_{sdiff-HgX}$ ($Mg\ y^{-1}$) | Sediment-water net diffusion of Hg | $F_{sw-diff} \times A_{sed} \times 10^4\ cm^2\ m^{-2} \times (60 \times 60 \times 24 \times 365\ s\ y^{-1})$ | |
| $F_{sw-diff}$ ($ng\ cm^{-2}\ s^{-1}$) | Diffusive flux based on Fick's law | $\frac{POR \times D_{sw} \times (C_{psed-Hg} - C_{w-Hg}) \times 1000}{TOR \times D_{sed}}$ | The diffusivity gradient is assumed to be in the top 1 cm of the sediment [Sunderland et al., 2010] |
| TOR | Tortuosity of sediments | $1 - \ln(POR^2)$ | [Gill et al., 1999] |
| POR | Porosity of sediments | 0.68±0.10 | [Hamden et al., 2013; Kuzyk et al., 2013] |
| D_{sed} ($cm^2\ s^{-1}$) | Sediment-water diffusion coefficient at temperature T_{sed} | $D_{sw25} / (1 + 0.048 \times (25 - T_{sed}))$ | [Gill et al., 1999] |
| D_{sw25} ($cm^2\ s^{-1}$) | Sediment-water diffusion coefficient at 25 °C | 6×10^{-6} (2×10^{-6} - 12×10^{-6}) | [Hollweg et al., 2010] |
| T_{sed} ($^{\circ}C$) | Temperature of sediments | 1 | Approximation, a 5 °C change in average temperature results in 12-15% change in diffusion flux |
| A_{sed} | Sediment area | | Table 3 |
| <i>Water column diffusion</i> | | | |
| $M_{wdif-HgX}$ ($Mg\ y^{-1}$) | In water column eddy diffusion of X species (Hg ^{II} , Hg ⁰ , MeHg, and Me ₂ Hg) | $D_w \times \left(\frac{C_{w-HgX}}{DiffDepth_{z2} - DiffDepth_{z1}} \right) \times 60 \times 60 \times 24 \times 365$ | |
| D_w ($m^2\ s^{-1}$) | Eddy diffusion coefficient | 0.8×10^{-4} ($< 0.05 \times 10^{-4}$ - 2.5×10^{-4}) | [Padman and Dillon, 1991; Shaw and Stanton, 2014; Wallace et al., 1987] |
| $DiffDepth_{so}$ (m) | Depth of diffusion gradient | 20 | |
| $DiffDepth_{ss}$ (m) | Depth of diffusion gradient | 140 | |
| $DiffDepth_{de}$ (m) | Depth of diffusion gradient | 300 | |
| $z1$ (m) | Upper diffusion depth | | |
| $z2$ (m) | Lower diffusion depth | | |

Table S9. Gas exchange parameterization for Me₂Hg

| | | | |
|-----------------------------------|---------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------|
| M_{eva-Me_2Hg} ($Mg\ y^{-1}$) | Total Me ₂ Hg evasion flux (A (areas); F: in Table 3) | $F_v \times (A_{shelf} + A_{deep}) \times \left(\frac{F_{open-sum} + F_{open-win}}{2} \right) \times 10^{-15} \times 24 \times 365$ | |
| F_v ($ng\ m^{-2}\ h^{-1}$) | Me ₂ Hg air-sea exchange flux | $K_w (C_w - C_a / H'_X(T))$ | |
| C_a ($ng\ m^{-3}$) | Concentration of Me ₂ Hg in air | 0.0038±0.0031 | [Baya et al., 2015] |
| C_w ($ng\ m^{-3}$) | Concentrations of Me ₂ Hg in PML | | Table 1 |
| $H'_{Me_2Hg}(T)$ | Temperature dependent dimensionless Henry's law constant for Me ₂ Hg | $\ln H' = \left(\frac{-2512.43}{T} + 7.27 \right)$ | [Lindqvist and Rodhe, 1985] |
| K_w ($m\ hr^{-1}$) | Water-side mass transfer coefficient | $A \times u_{10}^2 (Sc_{HgX} / Sc_{CO_2})^{-0.5}$ | [Wanninkhof, 1992] |
| A (unitless) | Constant based on the Weibull distribution of wind speeds over oceans | 0.25 | [Nightingale et al., 2000] |
| u_{10} ($m\ s^{-1}$) | Average wind speed at 10 m above sea surface | 5.6 (4-6) | [Nummelin et al., 2015; Serreze and Barry, 2005] |
| Sc_{CO_2} | Schmidt number for CO ₂ | $0.11T'^2 - 6.16T' + 644.7$ | [Poissant et al., 2000] |
| T' (°C) | Water temperature | 0 | Approximation, a 5 °C change in temperature only lead to 4% change in evasion |
| TK (K) | Water temperature | $T'+273.15$ | |
| Sc_{Hg} | Schmidt number for Me ₂ Hg | ν / D | |
| ν ($cm^2\ s^{-1}$) | Kinematic viscosity | $N/\rho = 0.017e^{(-0.025T')}$ | [Poissant et al., 2000] |
| ρ ($mg\ cm^{-3}$) | Seawater density | 1025 | |
| D ($cm^2\ s^{-1}$) | Me ₂ Hg diffusivity based on Wilke-Chang | $\frac{7.4 \times 10^{-8} (\phi_w M_w)^{1/2} T}{NV_B^{0.6}}$ | [Wilke and Chang, 1955] |
| N (cP) | Viscosity of water | See Soerensen et al. (2010) | [Soerensen et al., 2010] |
| M_w ($g\ mol^{-1}$) | Molecular weight of water | 18.0 | |
| V_B ($cm^3\ mol^{-1}$) | Molal volume of Me ₂ Hg at its normal boiling temperature | 72.11 | |
| ϕ_w | Solvent association factor | 2.26 | [Hayduk and Laudie, 1974] |

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