

Supporting Information to:

Cycling of PCBs and HCB in the Surface Ocean-Lower Atmosphere of the Open Pacific

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Methods

Sample Analysis Method

PCBs and HCB were analyzed using an Agilent GC 6890N with a DB-5MS capillary column (J&W Scientific) equipped with a Quattro microGC tandem MS (Waters). High purity Helium gas was used as the carrier gas with a flow rate of 1 mL/min. Injection port, GC-MS-MS interface, and ion trap temperature were set to 280°C, 240°C, 180°C, respectively. The oven temperature was programmed as follows: initial temperature 60°C, hold 3.5min, 11°C/min to 180 °C, 3 °C/min to 260 °C, 20°C/min to 300 °C, and hold for 6 min. The ion trap was operated in electron ionization-MS-MS mode. The filament emission current was 150 μ A.

Quality Assurance/Quality Control

PUFs were pre-extracted twice in dichloromethane (DCM) for 24 h and vacuum-dried. GFFs were baked at 450°C overnight. PUFs and GFFs were handled with nitrile gloves and rinsed pairs of tongs only and stored in solvent-rinsed tin cans and aluminum foils, respectively. They were firmly sealed and stored in freezer until extraction. Samples were extracted in a clean trace laboratory, which received HEPA-filtered air under positive pressure.

There were no major differences in the trace amounts of PCBs and HCB detected between field and laboratory blanks. Samples were therefore corrected using the mean of these two kinds of blanks and method detection limits were derived from the sum of mean blank concentrations and three times the standard deviation of mean blank concentrations.

Major concerns with trace sampling of organic contaminants on-board ship are linked to the contamination of samples from the ship's air during atmospheric sampling and handling (1). Results from our atmospheric sampling show that the R/V Revelle is essentially a clean ship (see results in main text), suggesting that our air and water samples were not affected by on-board contamination.

Extraction and analysis recoveries were routinely monitored using surrogates. Average and standard deviation of recoveries for air and water samples were as follows: $^{13}\text{C}_{12}$ -PCB 8, 80%±18%, 68%±14%; $^{13}\text{C}_{12}$ -PCB 28, 74%±16%, 65%±12%; $^{13}\text{C}_{12}$ -PCB 52, 71%±15%, 64%±11%; $^{13}\text{C}_{12}$ -PCB 118, 69%±16%, 63%±11%; $^{13}\text{C}_{12}$ -PCB 138, 72%±18%, 65%±12%; $^{13}\text{C}_{12}$ -PCB 180, 74%±18%, 63%±11%; $^{13}\text{C}_{12}$ -PCB 209, 63%±15%, 53%±10%; $^{13}\text{C}_{12}$ -p, p'-DDT, 67%±14%, 55%±11%.

Breakthrough of PCBs and HCB trapped on two/three PUFs in series was assessed by analyzing the PUFs separately and comparing the concentrations in each PUF.

Breakthrough was not of concern for all analytes except HCB in the air samples. The octanol-air partitioning coefficient (K_{oa}) for HCB (2) with temperature correction was used to infer air phase HCB concentrations by assuming equilibrium partitioning of HCB between PUF and air.

- Correction for DOC interference

PCBs and HCB partition to dissolved organic carbon (DOC) and this could lower truly dissolved concentrations (C_{sw_dis}) if the DOC concentration (C_{sw_DOC}) is high. The total concentration collected on PUFs (C_T) is the sum of C_{sw_dis} and C_{sw_DOC} . Freely dissolved PCB and HCB seawater concentrations ($[PCBs]_{sw_dis}$, $[HCB]_{sw_dis}$) were calculated by correcting for DOC interference. Most of the PCBs (>90% even for the very hydrophobic hexa-PCBs) and HCB were in the truly dissolved form because of very low C_{sw_DOC} .

The truly dissolved concentrations of PCBs and HCB were calculated by correcting the DOC interferences on the total concentrations collected on PUFs. (see SI for more details).

$$f_{sw_dis} = 1 / (1 + [DOC]K_{DOC}) \quad (1)$$

$$C_{sw_dis} = C_T * f_{sw_dis} \quad (2)$$

Where f_{sw_dis} is truly dissolved fraction of POPs.

K_{DOC} was estimated from the geometric mean regression equations derived by Burkhard for PCBs and HCB (3):

$$\log K_{DOC} = 0.71 * \log K_{OW} - 0.50 \quad (3)$$

where K_{OW} is the compound's octanol-water partitioning coefficient.

Surface [DOC] were calculated from regression models using sea surface temperature (SST) (4):

$$[\text{DOC}] = 0.993 * \text{SST} + 52.05 \quad (4)$$

Back Trajectories and Origins of Air Masses

The *R/V Revelle* sailed in the northern hemisphere (NH) trade winds after it left San Diego (air sample No. 2~8), until it reached the intertropical convergence zone (ITCZ). Origins of air sample No. 2~4 were from the west coast of US and Mexico. However, samples No. 5~8 were more likely originated from the coastal seas close to Middle America and had spent more than 10 days over the ocean. South of ITCZ, wind field switched to southern hemisphere (SH) trade winds. Air mass origins of sample No. 9~20 were traced back toward South America, but were above free troposphere (~3000m) 10-day back and descended toward the surface gradually. A more significant switch occurred after sample No. 20, sample No. 21~23, 100~122 were influenced by winds coming from east which were originally from the westerlies south of Australia and New Zealand. For the last few samples (No.123~125), polar easterlies dominated first and then changed to westerlies prior to be sampled.

Air-Water Gas Exchange Fluxes ($F_{a/w}$, $\text{ng m}^{-2} \text{d}^{-1}$) Calculation:

Air-water gas exchange fluxes ($F_{a/w}$, $\text{ng m}^{-2} \text{d}^{-1}$) were calculated using a modified version of the Deacon boundary layer model as described elsewhere (5). A positive $F_{a/w}$ value indicates a net flux from the water to the atmosphere:

$$F_{a/w} = v_{POP,a/w} \cdot (C_{sw_dis} - \frac{C_{gas}}{K_{aw}(T, sal)}) = v_{POP,a/w} \cdot (C_{sw_dis} - C_{gas}^*) \quad (5)$$

, where $v_{POP,a/w}$ is the air-water gas exchange mass transfer coefficient (MTC). If water is chosen as the reference phase, $v_{POP,a/w}$ is always of the form:

$$\frac{1}{v_{POP,a/w}} = \frac{1}{v_{POP,w}} + \frac{1}{v_{POP,a} K_{aw}(T, sal)} \quad (6)$$

The air-side MTC ($v_{POP,a}$) is determined by correlation with the MTC of water vapor ($v_{H_2O,a}$) across the air-side boundary layer (5, 6).

$$v_{POP,a} = \left(\frac{D_{POP,a}}{D_{H_2O,a}} \right)^{0.67} v_{H_2O,a} \quad (7)$$

where $D_{POP,a}$ and $D_{H_2O,a}$ are the molecular diffusivities of POPs and H_2O vapor in air, respectively. $v_{H_2O,a}$ is determined by the following empirical relationship:

$$v_{H_2O,a} = 0.2 \cdot u_{10} + 0.3 \quad (8)$$

where u_{10} is the wind speed ($m s^{-1}$) at 10m above the water surface. $v_{POP,w}$ was obtained using the MTC of CO_2 ($v_{CO_2,w}$) across the water-side boundary layer as reference.

$$v_{POP,w} = \left(\frac{Sc_{POP,w}}{600} \right)^{-a_{Sc}} v_{CO_2,w} \quad (9)$$

where $Sc_{POP,w}$ is the Schmidt numbers (kinematic viscosity/ molecular diffusivity) for POPs in the water boundary layer, which are strongly sensitive to water temperature because kinematic viscosity and molecular diffusivity are inversely related to temperature. The value of a_{sc} is 0.67 ($u_{10} \leq 4.2 \text{ m s}^{-1}$) or 0.5 ($u_{10} \geq 4.2 \text{ m s}^{-1}$), and $v_{CO_2, w}$ is calculated based on the model by Livingstone and Imboden (1993) (7). Schmidt numbers were corrected for each sample's water temperature and $v_{a/w}$ were calculated for each PCB congener and HCB.

Comparing Σ PCB fluxes with previous studies on different water bodies should be done cautiously (SI Table 4). Different K_{aw} and different wind speed dependency were used to derive MTC and subsequently quantified different PCB congeners.

Mass Balance Model:

For a better understanding of the PCBs cycling in the SPG, a mass balance model was used to discern what is driving the temporal changes in [PCB] in the atmosphere.

According to previous air mass origins discussions, advective transport is neglected in the mass balance model. Volatilization from surface seawater to air is considered as the only input/ source term of PCBs, while gaseous deposition and OH-radical depletion reaction are the loss terms. The change in $[PCB]_{air}$ with time can be described as:

$$\begin{aligned} \frac{dC_{gas}}{dt} &= \text{volatilization} - \text{deposition} - \text{OH radical depletion} \\ &= \frac{v_{POP,a/w} \cdot C_{sw_dis}}{ABL} - \frac{v_{POP,a/w} \cdot C_{gas}}{ABL \cdot K_{aw}} - k_{OH}[OH]C_{gas} \end{aligned} \quad (10)$$

where $v_{\text{POP}, a/w}$ is about $1\sim 2 \times 10^{-5} \text{ m s}^{-1}$, ABL is the ABL height which is about 1000m for these samples, $K_{a/w}$ is about 10^{-2} , k_{OH} is the lab-derived OH-radical depletion reaction constants for PCBs on the order of a few $10^{-12} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ (8), and the average OH radical concentration in the SPG is about $10^6 \text{ molecules cm}^{-3}$.

Results and Discussion

Interpretation of day-night variations

Temperature: several previous land-based studies identified temperature as the dominant factor driving short-term variations in ambient concentrations (26-28). During stable atmospheric conditions, a strong periodic correlation between air concentrations of several different POPs (PCBs, PAHs, and pesticides) and surface temperature were observed, with highest concentration in the early afternoon and lowest at approximately 6am. A sinusoidal wave was fit onto these time dependent variations of atmospheric concentrations (26, 27). Since daily surface temperature fluctuations also follow a sinusoidal wave, this was interpreted that temperature overruled other processes such as OH radical depletion reaction and dominated the diurnal variations of atmospheric concentrations. Seawater temperatures did not fluctuate by more than 2 °C (Fig 4). Thus, temperature alone cannot account for the diurnal fluctuations observed in the SPG.

Air mass origins: If the air masses were from different regions during day and night, this could cause higher nighttime $[\text{PCB}]_{\text{gas}}$ than the daytime concentrations. Back trajectories

showed the air masses for these four diurnal samples were from the same general location and had moved over the S Pacific for at least 10 days before arriving at the ship.

Air Boundary Layer (ABL) height: NOAA's HYSPLIT model did not indicate any variations in the ABL heights. As HCB is more resistant to OH radical attack, it was used as an internal reference compound to help distinguish air mass dilution effects from OH reactions. PCBs and HCB had different short-term variations in ambient concentrations, whereas any air mass dilution effect (due to changes in the ABL height) would affect all molecules similarly.

Tables

SI Table 1 Summary of Sampling and Meteorology Information a) Air; b) Water

a) Air No	Start Time	Stop Time	Lat	Long	Vol (L)	Temp(°C)		R.H (%)	W.S (m/s)	W.D (°)	S (PSU)	Fluo (µg/L)
2	2006/12/5 17:45	2006/12/6 17:30	18.8	-137.3	876	23.3	24.4	90.3	10.0	75	34.4	0.2
3	2006/12/6 17:45	2006/12/7 6:30	16.3	-140.3	450	24.7	25.6	92.1	9.0	73	34.0	0.3
4	2006/12/7 6:45	2006/12/7 17:45	14.6	-142.2	421	26.5	25.9	86.3	9.5	54	34.0	0.2
5	2006/12/7 18:00	2006/12/8 5:45	12.9	-144.0	449	25.4	26.1	90.6	11.3	59	33.9	0.3
6	2006/12/8 6:00	2006/12/8 15:30	11.4	-145.7	386	26.2	26.7	87.6	11.2	56	34.0	0.2
7	2006/12/8 17:45	2006/12/9 5:50	9.7	-147.5	482	26.4	27.1	87.2	12.6	53	34.1	0.3
8	2006/12/9 6:00	2006/12/9 18:00	7.8	-149.5	375	25.1	28.4	85.5	9.2	93	33.8	0.3
9	2006/12/9 18:15	2006/12/10 6:00	6.6	-150.7	458	26.8	28.8	84.5	8.6	69	34.0	0.7
10	2006/12/10 6:15	2006/12/10 17:45	4.7	-152.7	485	26.8	28.9	88.9	9.4	81	34.6	0.6
11	2006/12/10 18:00	2006/12/11 5:50	3.2	-154.2	471	27.0	28.7	88.2	8.4	80	35.0	1.9
12	2006/12/11 6:00	2006/12/11 17:45	1.3	-156.1	420	27.4	28.6	84.2	6.2	82	35.1	0.9
14	2006/12/11 18:15	2006/12/12 5:50	-0.5	-157.9	442	27.3	28.5	86.0	2.0	109	35.3	1.8
15	2006/12/12 6:10	2006/12/12 17:45	-2.2	-159.3	395	27.9	28.8	80.3	10.4	97	35.4	0.8
16	2006/12/12 18:00	2006/12/13 6:10	-3.5	-160.0	512	28.1	29.1	77.1	11.0	101	35.5	1.8
17	2006/12/13 6:25	2006/12/13 17:30	-5.6	-163.1	429	29.8	N/A	N/A	7.4	96	N/A	N/A
18	2006/12/13 17:45	2006/12/14 5:40	-7.2	-164.7	462	28.7	N/A	N/A	7.1	88	N/A	N/A
19	2006/12/14 6:00	2006/12/14 18:00	-8.8	-166.4	408	28.9	N/A	N/A	6.7	87	N/A	N/A
20	2006/12/14 18:20	2006/12/15 5:50	-10.3	-168.0	420	27.9	N/A	N/A	4.3	87	N/A	N/A
21	2006/12/15 6:00	2006/12/15 17:30	-11.8	-169.6	454	30.0	N/A	N/A	3.8	52	N/A	N/A
23	2006/12/15 18:00	2006/12/16 5:45	-13.1	-171.1	401	27.6	N/A	N/A	5.1	121	N/A	N/A
100	2006/12/18 6:30	2006/12/19 6:30	-17.5	-169.3	804	26.4	N/A	N/A	12.0	102	N/A	N/A
101	2006/12/19 6:40	2006/12/19 18:45	-20.7	-167.5	447	23.8	N/A	N/A	9.8	101	N/A	N/A
102	2006/12/19 18:55	2006/12/20 6:08	-23.5	-166.1	412	22.6	24.4	72.2	8.2	74	35.5	0.1
103	2006/12/20 6:29	2006/12/20 18:44	-23.9	-165.7	464	22.7	23.4	72.3	7.8	93	35.6	0.1
104	2006/12/20 18:49	2006/12/21 18:44	-23.9	-165.6	848	22.7	23.8	76.5	7.7	93	35.6	0.1
105	2006/12/21 19:02	2006/12/22 7:05	-24.3	-164.1	515	22.6	24.0	83.0	8.3	82	35.5	0.1
106	2006/12/22 7:20	2006/12/22 18:27	-24.8	-161.7	403	22.6	23.7	84.2	10.0	89	35.5	0.0
107	2006/12/22 18:35	2006/12/24 18:12	-25.9	-157.6	1695	21.6	22.7	79.9	8.3	83	35.5	0.0
108	2006/12/24 18:19	2006/12/25 18:00	-26.7	-154.2	863	21.2	23.1	74.1	6.4	47	35.5	0.0
109	2006/12/25 18:07	2006/12/26 15:50	-27.7	-149.8	737	21.7	22.9	78.6	8.8	52	35.4	0.0
110	2006/12/28 18:10	2006/12/29 8:40	-26.9	-140.7	464	25.2	24.1	N/A	4.8	31	35.5	0.0
111	2006/12/29 8:50	2006/12/29 18:11	-26.5	-138.3	161	25.3	24.3	N/A	5.9	76	35.3	0.0
112	2006/12/30 18:05	2006/12/31 17:46	-27.3	-134.9	755	26.3	24.1	N/A	5.8	18	35.4	0.0
113	2006/12/31 17:53	2007/1/2 17:50	-28.4	-130.4	1662	22.8	24.3	96.4	-3.6	188	35.4	0.0
114	2007/1/2 17:58	2007/1/3 9:07	-28.1	-126.1	548	23.7	24.7	85.2	1.5	86	35.7	0.0
115	2007/1/3 9:23	2007/1/3 17:05	-27.9	-123.4	290	24.2	24.9	82.2	2.5	47	35.7	0.0
116	2007/1/5 6:15	2007/1/5 16:44	-27.9	-120.0	367	23.7	25.1	80.4	5.8	110	35.9	0.0
117	2007/1/5 16:50	2007/1/7 3:29	-27.8	-117.7	1186	23.1	25.0	77.9	6.2	127	36.1	0.0
118	2007/1/7 3:29	2007/1/7 16:56	-28.9	-119.2	461	22.2	24.5	72.9	6.9	121	35.8	0.1
119	2007/1/7 17:03	2007/1/8 3:38	-30.4	-121.4	205	21.6	23.9	88.0	8.1	112	35.5	0.1
120	2007/1/8 3:52	2007/1/8 17:37	-31.9	-123.6	468	21.2	23.1	79.4	10.6	107	35.3	0.1
121	2007/1/8 17:46	2007/1/9 3:50	-33.5	-126.0	392	20.7	22.3	81.9	9.0	80	34.8	0.1
122	2007/1/9 4:05	2007/1/9 17:42	-35.0	-128.4	442	20.9	21.5	78.0	4.5	65	34.6	0.2
123	2007/1/10 3:55	2007/1/10 17:50	-37.8	-132.7	489	18.1	20.5	78.4	6.3	196	34.3	0.5
124	2007/1/12 4:00	2007/1/13 7:38	-38.9	-138.0	1044	16.4	18.8	81.3	8.7	259	34.2	1.0
125	2007/1/14 18:47	2007/1/15 6:28	-40.0	-143.0	436	13.2	17.1	52.6	8.2	160	34.3	1.9

R.H:Relative Humidity; W.S: Wind Speed; W.D: Wind Direction; Fluo: Fluorescence; N/A: not available

b) Water No	Start Time	Stop Time	Lat	Long	Vol (L)	Temp(°C)		R.H (%)	W.S (m/s)	W.D (°)	S (PSU)	Fluo (μ g/L)
						air	water					
2	2006/12/3 18:30	2006/12/4 8:30	25.6	-128.5	669	17.3	20.5	70.2	6.8	52	34.7	0.2
3	2006/12/4 8:30	2006/12/4 20:00	24.0	-130.7	525	19.0	21.1	67.4	6.7	65	34.7	0.2
4	2006/12/4 20:00	2006/12/5 8:30	22.4	-132.8	706	20.0	21.8	72.7	8.6	72	34.8	0.2
5	2006/12/5 8:45	2006/12/5 23:00	20.6	-135.2	477	21.7	23.1	85.3	9.3	73	34.8	0.1
6	2006/12/5 23:00	2006/12/6 8:20	19.0	-137.1	382	23.1	24.5	91.3	10.5	75	34.4	0.3
7	2006/12/6 8:30	2006/12/6 21:30	17.5	-138.9	336	24.3	25.2	90.4	10.0	82	34.3	0.2
8	2006/12/6 21:45	2006/12/7 9:15	15.8	-140.8	362	25.0	25.7	91.4	8.5	65	34.0	0.4
9	2006/12/7 9:30	2006/12/7 21:30	14.1	-142.9	492	26.3	26.0	87.5	10.2	54	33.9	0.3
10	2006/12/7 21:45	2006/12/8 9:10	12.5	-144.5	722	25.6	26.2	89.1	11.4	59	33.9	0.3
12	2006/12/8 9:30	2006/12/8 21:15	10.7	-146.4	398	26.2	27.0	88.8	12.2	57	34.0	0.2
13	2006/12/8 21:20	2006/12/9 10:00	9.0	-148.3	608	26.1	27.4	88.1	12.5	59	34.0	0.3
14	2006/12/9 10:00	2006/12/9 21:45	7.3	-150.0	336	25.7	28.6	82.6	7.2	90	33.9	0.4
15	2006/12/9 22:00	2006/12/10 9:30	5.9	-151.5	414	27.0	28.9	87.0	10.1	73	34.2	0.8
16	2006/12/10 9:45	2006/12/10 21:00	4.4	-153.0	260	26.7	28.8	89.9	8.8	78	34.7	0.9
17	2006/12/10 21:15	2006/12/11 8:15	2.7	-154.7	243	27.2	28.6	86.3	8.2	85	35.0	1.9
18	2006/12/11 8:20	2006/12/11 21:30	0.9	-156.5	603	27.4	28.6	84.7	6.1	88	35.2	1.0
19	2006/12/11 21:45	2006/12/12 8:20	-0.9	-158.4	172	27.3	28.6	85.7	8.0	109	35.4	1.7
20	2006/12/12 8:45	2006/12/12 20:50	-2.6	-159.6	384	28.0	28.9	77.9	10.8	95	35.5	0.9
21	2006/12/12 21:00	2006/12/13 8:35	-3.7	-160.0	154	28.1	29.2	79.4	10.7	104	35.6	1.9
23	2006/12/13 9:30	2006/12/13 21:15	-5.9	-163.4	512	30.0	N/A	N/A	7.1	96	N/A	N/A
24	2006/12/13 21:35	2006/12/14 9:00	-7.2	-164.7	317	28.8	N/A	N/A	7.6	83	N/A	N/A
25	2006/12/14 9:15	2006/12/14 22:35	-9.2	-166.9	621	29.9	N/A	N/A	5.9	89	N/A	N/A
26	2006/12/14 22:55	2006/12/15 10:15	-10.3	-168.4	574	28.4	N/A	N/A	3.8	76	N/A	N/A
27	2006/12/15 10:30	2006/12/15 20:45	-12.2	-170.1	449	30.4	N/A	N/A	3.2	54	N/A	N/A
28	2006/12/15 21:00	2006/12/16 7:40	-13.1	-171.1	429	27.3	N/A	N/A	5.7	121	N/A	N/A
100	2006/12/18 6:56	2006/12/19 6:50	-17.5	-169.3	860	26.4	N/A	N/A	12.0	102	N/A	N/A
101	2006/12/19 7:00	2006/12/19 19:05	-20.7	-167.5	623	23.8	N/A	N/A	9.8	101	N/A	N/A
102	2006/12/19 19:15	2006/12/20 6:38	-23.6	-166.0	561	22.5	24.3	72.6	8.1	75	35.5	0.1
103	2006/12/20 6:56	2006/12/20 18:58	-23.9	-165.7	621	22.7	23.4	72.2	7.8	93	35.6	0.1
104	2006/12/20 19:16	2006/12/21 19:12	-23.9	-165.6	327	22.7	23.8	76.7	7.7	92	35.6	0.1
105	2006/12/21 19:25	2006/12/22 9:00	-24.3	-163.8	534	22.6	23.9	83.1	8.4	84	35.5	0.1
106	2006/12/22 9:10	2006/12/22 18:44	-24.9	-161.5	513	22.6	23.8	84.1	10.3	87	35.5	0.0
107	2006/12/22 18:55	2006/12/24 19:01	-19.9	-135.3	1017	21.6	22.5	73.1	10.2	77	33.8	2.7
108	2006/12/24 18:40	2006/12/25 18:21	-26.7	-154.1	914	21.1	23.1	74.0	6.4	47	35.5	0.0
109	2006/12/25 18:31	2006/12/26 9:05	-27.6	-150.4	888	21.3	22.7	73.6	8.0	49	35.4	0.0
110	2006/12/28 19:31	2006/12/29 9:05	-26.9	-140.6	560	25.2	24.1	N/A	5.0	31	35.4	0.0
111	2006/12/29 9:20	2006/12/29 18:22	-26.5	-138.2	560	25.3	24.3	N/A	6.1	76	35.3	0.0
112	2006/12/30 18:24	2006/12/31 18:05	-27.3	-134.9	945	26.3	N/A	N/A	5.8	18	N/A	N/A
113	2006/12/31 18:14	2007/1/1 18:03	-28.4	-131.5	1027	25.5	23.4	N/A	5.0	344	34.7	-0.5
114	2007/1/2 19:22	2007/1/3 9:43	-28.0	-125.9	385	23.6	24.8	85.1	1.7	75	35.7	0.0
115	2007/1/3 9:50	2007/1/3 18:18	-27.9	-123.4	199	24.2	24.9	82.1	2.4	46	35.7	0.0
117	2007/1/5 18:10	2007/1/7 3:53	-27.8	-117.7	711	23.1	25.0	77.7	6.3	128	36.1	0.0
118	2007/1/7 4:02	2007/1/7 17:45	-28.9	-119.3	400	22.2	24.5	73.0	6.7	121	35.8	0.1
119	2007/1/7 18:18	2007/1/8 4:09	-30.5	-121.6	405	21.5	23.8	88.9	8.7	114	35.5	0.1
120	2007/1/8 4:09	2007/1/8 18:18	-32.0	-123.8	405	21.3	23.0	79.2	10.6	106	35.3	0.1
121	2007/1/8 18:04	2007/1/9 4:08	-33.5	-126.1	241	20.7	22.3	82.0	8.9	79	34.8	0.1
122	2007/1/9 4:18	2007/1/9 18:17	-35.1	-128.4	484	20.9	21.5	77.9	4.5	74	34.6	0.2
123	2007/1/9 18:28	2007/1/10 4:05	-36.6	-130.7	439	20.2	21.4	85.9	5.1	311	34.5	0.5
124	2007/1/10 4:10	2007/1/10 17:55	-37.8	-132.7	401	18.0	20.5	78.0	6.3	194	34.3	0.5
126	2007/1/12 18:56	2007/1/13 7:15	-39.2	-139.5	362	15.7	18.2	87.8	10.9	254	34.2	1.5

SI Table 2 Summary of Concentrations for Different PCB Congeners and HCB a) Air Phase (pg/m³); b) Water Phase (pg/L). PCB congeners not detected were left blank in the table.

a)Air	PCB 3	PCB 8	PCB 18	PCB 28	PCB 52	PCB 44	PCB 66	PCB 101	PCB 118	PCB 153	PCB 138	PCB 180	PCB 189	PCB 206	PCB 209	HCB
2	2.0	20.0	26.5	41.2	13.0	9.4	5.8	2.3	1.4	1.3		0.2				102.8
3	2.6	27.6	4.8	69.1	23.9	17.2	1.8	3.2	2.5	2.5	1.8			0.9		106.7
4	2.5	21.7	34.9	57.2	19.3	14.9	8.6	2.8	2.5	2.7	2.0					93.0
5	1.3	12.0	15.7	29.7				1.2		1.2						60.7
6	1.3	11.9	19.8	33.6	11.3	7.4	6.9	6.2	1.7	1.6	1.2					69.2
7	0.8	9.0	12.3	20.0	7.2	5.8	4.3	3.6	1.9	0.9						79.7
8	1.1	9.9	11.3	16.7	7.0	5.1	3.5	3.0	1.6							58.0
9	0.7	8.9	15.9	28.7	1.1	7.7	5.4	4.7	1.7	1.4	1.2					52.7
10	0.7	7.4	12.3	19.2	8.6	5.2	4.5	4.9	1.1	1.9	1.1				1.0	50.2
11	0.7	7.9	12.3	19.1	7.9	5.5	3.6	1.7		0.9						59.4
12	1.6	14.9	2.6	35.4	13.8	9.8	6.6	1.9		1.8						73.4
14	0.5	5.8	8.2	11.3	3.6	2.7	1.5	2.8								62.9
15	0.4	6.7	9.5	14.4	5.8	4.8	3.4	2.6								53.9
16	0.4	3.7	5.6	8.7	3.5	2.9	1.8									63.8
17	0.4	5.0	7.5	11.9	5.2	4.4	3.2	2.4								63.2
18	0.5	5.8	7.4	13.3	8.4	6.8	8.4	2.5	3.7	4.8	3.4			1.7	0.2	53.1
19	0.6	1.4	12.2	1.3	2.9		0.6									26.1
20	0.7	7.8	1.2	21.7	1.6	7.5	9.8	2.3	2.0	2.8	1.5					47.4
21	1.6	11.5	15.7	3.3	11.7	9.2	6.4	1.8	2.2	2.2	1.3					73.4
23	0.4	4.7	6.4	9.2	4.7	3.3	2.4	2.3	0.8							34.0
100	0.2	1.3	2.0	3.4	1.3	0.9	0.3	0.7				0.1				41.3
101	0.5	2.8	3.8	4.3	1.7	1.1										31.0
102	0.3	2.4	2.8	2.8	0.9	0.7										27.4
103	0.4	4.0	5.2	6.2	2.2	1.6	1.8	0.9						0.2		23.8
104	0.2	2.0	2.3	3.7	1.5	1.2	0.5	1.0	0.4		0.5	0.3		0.8	0.2	24.1
105	0.8	1.5	1.8	1.6	0.5	0.3										43.7
106		2.1	2.3	2.7	0.8	0.7	0.7			0.9				0.1		26.1
107	0.1	1.0	1.4	2.8	1.0	0.8	0.9	0.3				0.9		0.2	1.0	28.9
108	0.3	2.3	3.4	5.5	1.6	1.6	0.7	0.6				0.2			0.7	30.4
109	0.2	1.7	2.1	3.3	1.3	1.0	0.7	0.6								34.7
110	0.2	2.5	3.0	4.5	2.7	1.5	1.4	0.9								31.8
111	0.7	6.7	6.6	7.4	3.5	2.2										45.0
112	0.4	2.7	4.6	7.2	2.2	1.6	1.0	0.9				0.3				40.1
113	0.8	1.2	2.7	6.8	2.8	2.6	1.4	0.5	0.4	0.5	0.3	0.1				21.2
114	0.1	2.1	2.7	3.4	1.3	0.7	0.7									33.9
115	0.4	6.7	7.6	9.5	3.8	2.6	0.9									
116	0.2	2.3	2.6	2.4	0.9	0.9										
117	0.4	1.4	1.8	2.7	1.3	0.9	0.3	0.6	0.3	0.4		0.3			0.2	
118	0.6	4.1	4.4	6.4	2.6	2.0	1.7	0.9								35.5
119	0.6	7.9	9.4	13.4	4.3	5.0	2.9	2.6								17.3
120	0.2	2.5	2.9	4.4	2.6	1.9	0.8	2.9	2.2	1.9	2.9			1.3	0.3	31.1
121	0.5	5.6	6.2	7.4	2.6	1.9	2.1	1.3								36.3
122	0.6	8.1	1.1	17.8	7.6	5.1	3.9	3.1	1.4	1.1						
123	0.2		3.1	12.0	11.2	9.2	8.3	18.5	7.2	18.7	7.8	2.6		3.7	0.7	27.9
124	0.5	0.5	0.6	0.7		0.2						0.8	0.6			28.7
125	0.1	1.2	1.2	1.5		0.4										22.0

b) Water	PCB 3	PCB 8	PCB 18	PCB 28	PCB 52	PCB 44	PCB 66	PCB 101	PCB 118	PCB 153	PCB 138	PCB 180	PCB 189	PCB 206	PCB 209	HCB
2	0.16	2.24	1.49	1.49	3.23	1.57	0.78	4.05	1.53	2.76	1.56			0.83	0.15	1.43
3	0.10	2.49	1.56	1.15	2.24	1.01	0.42	2.37	1.14	1.89	1.22					1.94
4	0.08	1.37	0.59	0.79	1.58	0.60	0.37	2.12	0.48	1.34	0.95					
8		1.51	0.72		0.52	0.23		0.89	0.24		0.21					0.80
10	0.15	2.17	1.32	0.73	0.80	0.34	0.20	1.39	0.76	1.18	0.72					0.81
13		1.96	1.26	0.65		0.20	0.22	1.40	0.72	1.19	0.58					0.78
18	0.15	1.85	0.75						0.24							0.49
20		1.90	0.90			0.09			0.30		0.08					0.61
23		2.42	1.86	1.56	8.38	3.37	3.15	6.35	2.48	2.36	2.21					0.50
25	0.18	2.92	2.01	1.36	2.56	1.20	0.20	1.68	0.71	0.87	0.67				0.14	0.50
26	0.13	1.93	0.96		0.41	0.18			0.56		0.32					
100	0.10	1.65	0.79	0.46	0.23	0.12		0.26	0.30	0.38	0.35					
101		2.95	2.79	2.30	1.17	0.37	0.34		0.15		0.17					0.51
103		3.24	2.74	2.51	1.30	0.50		0.55	0.20							0.58
105		1.66	0.90			0.01										
106		1.34	0.85	0.61												
107		1.18	0.54	0.43												0.60
109	0.08	0.88	0.36													0.59
110		1.65	1.00	0.69	0.30	0.12										
111	0.09	1.38	1.11	0.72	0.45	0.20			0.41		0.44		0.32			
113	0.06	1.04	0.59	0.28									0.38			0.73
118		0.87	0.52								0.02					0.92
119		0.94	0.63		0.37											0.64
120		1.67	1.64	1.12	0.41	0.12					0.03					0.67
121		0.99	0.60													1.00
122		0.63														
123		0.99	0.44													

SI Table 3 Summary of Fugacity Ratios (FR) and Air-Water Gas Exchange Fluxes (ng m⁻² d⁻¹) for Different PCB Congeners and HCB

Air Sample No	Lat	Lon	PCB3		PCB6		PCB10		PCB20		PCB52		PCB44		PCB66		PCB101		PCB110		PCB153		PCB138		HCB	
			FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux	FR	Flux
3	16.3	-140.3			1.41	-1.04	0.30	0.75			2.02	-1.49	4.62	-1.30			0.29	0.92	1.61	-0.10			0.51	0.15	2.09	-2.60
5	12.9	-144.2	0.65	0.15	0.41	3.31	0.66	1.17	2.24	-2.36							0.07	2.92			0.08	2.29			1.58	-1.31
7	9.7	-147.5			0.31	4.09	0.50	1.90	1.57	-1.13			1.54	-0.32	1.08	-0.05	0.18	3.06	0.34	1.05	0.06	2.80			1.99	-2.51
12	1.3	-156.8	0.65	0.06	0.50	0.93	0.17	0.61																	2.69	-0.83
15	-2.2	-159.3			0.21	3.91	0.49	1.21					2.59	-0.36											1.55	-0.92
17	-5.6	-163.2			0.12	2.89	0.10	2.03	0.34	1.37	0.03	10.30	0.06	4.00	0.05	3.00	0.02	7.37							2.13	-0.70
19	-8.8	-166.4	0.20	0.14	0.03	2.63	0.27	1.32	0.04	1.17	0.05	2.00			0.14	0.15									0.89	0.05
20	-10.3	-168.0	0.35	0.03	0.26	0.44	0.06	0.27			0.20	0.09	2.12	-0.06					0.43	0.00			0.24	0.06		
100	-17.6	-169.3	0.13	0.27	0.06	4.41	0.13	1.94	0.39	0.80	0.32	0.42	0.44	0.19			0.20	0.51								
101	-20.8	-167.5			0.08	5.41	0.09	5.12	0.12	4.08	0.09	2.01	0.20	0.56											1.43	-0.47
103	23.9	165.7			0.11	3.04	0.13	3.10	0.17	2.70	0.12	1.44	0.23	0.40			0.16	0.54							1.04	0.04
105	-24.3	-164.6			0.08	2.38	0.13	1.22					2.39	-0.02												
106	-24.8	-161.7			0.14	2.39	0.18	1.46	0.29	0.91																
107	-25.9	-157.6			0.00	2.13	0.10	0.87	0.47	0.45															1.32	-0.42
109	-27.7	-149.0	0.30	0.00	0.10	0.99	0.42	0.29																	1.50	-0.51
110	-26.9	-140.7			0.13	0.72	0.19	0.39	0.42	0.19	0.64	0.05	0.02	0.01												
111	-26.5	-130.3	0.67	0.03	0.41	0.70	0.38	0.59	0.65	0.22	0.53	0.17	0.75	0.04												
113	-28.4	-130.4	1.18	-0.01	0.11	0.46	0.32	0.19	1.64	-0.09																
118	-28.9	-119.2			0.40	0.56	0.52	0.27																	0.91	0.09
119	-30.4	-121.4			0.75	0.39	0.97	0.03			0.81	0.11													0.67	0.36
120	-31.9	-123.6			0.14	3.01	0.12	3.04	0.27	1.73	0.40	0.42	1.10	-0.04									9.05	-0.30	1.22	-0.34
121	33.5	126.3			0.56	0.68	0.76	0.29																	1.00	0.01
122	-35.4	-128.4			1.35	-0.07																				

SI Table 4 Net air-water gas exchange fluxes of PCBs and HCB (ng m⁻² d⁻¹)

Area	Time	Fluxes ^a	Reference
PCBs			
		Freshwater	
Lake Superior	1986, Aug	19~141(Σ_{35} PCB)	9
	1988,Jul; 1990, Aug	35~110(Σ_{80} PCB)	10
	1992, May	-16~-40 (Σ_{80} PCB)	
Lake Michigan	1991, Sep	23.8~223 (Σ_{77} PCB)	11
	1993, Jun~Oct	13~1300 (Σ_{85} PCB)	12
	1994, May	-20~31 (Σ_{25} PCB)	13
	1994, Jul	-13~30 (Σ_{25} PCB)	
	1995, Jan	15~59 (Σ_{25} PCB)	
		Coastal Area	
New York /New Jersey Harbor Esturay	1999, Jul	400~2100 (Σ_{15} PCB)	14
Chesapeake Bay	1993,Annual	-63~800 (Σ_{46} PCB)	15
		Sea	
Arabian Sea and Bay of Bengal	1989~1990, Nov~ Mar	-20~20 (Σ_{36} PCB)	16 ^b
Baltic Sea	1999, Mar, May,Jun	Equilibrium	17
Mediterranean	1989, Nov~1990, Mar	-40~3 (Σ_{36} PCB)	16 ^b
	1997, 2002	2.6 (Σ_{18} PCB)	18
Bering Sea	1990, Jun~Aug	-120~0 (Σ_{36} PCB)	16 ^b
Chukchi Sea	1990, Jun~Aug	-60~-40 (Σ_{36} PCB)	16 ^b
Gulf of Alaska	1990, Jun~Aug	-60~0 (Σ_{36} PCB)	16 ^b
		Open Ocean	
E. Indian Ocean	1989, Apr~Jun	5~40 (Σ_{36} PCB)	16 ^b
Southern Ocean	1989, Apr~Jun	-20~0 (Σ_{36} PCB)	16 ^b
N. Atlantic	1989~1990, Nov~ Mar	-60~10(Σ_{36} PCB)	16 ^b
Atlantic	2005, Oct~Nov	-7~0.02 (Σ_5 PCB)	19
Northern North Atlantic and Arctic	2004,Jun~Aug	-4~0.08 (Σ_4 PCB)	20
Northern N. Pacific Ocean	1989, Jun~Jul	-100~10 (Σ_{36} PCB)	16 ^b
N. Pacific	1989, Nov~1990, Mar	-40~40 (Σ_{36} PCB)	16 ^b
Southern N. Pacific and S. Pacific	2006, Dec~2007, Jan	-0.4~30 (Σ_{15} PCB) ^c	this study
HCB			
Lake Baikal	1991, Jun	32	21
Arctic	1993, Annual	Equilibrium	22
Northern North Atlantic and Arctic	2004, Oct	Equilibrium	23
Southern N. Pacific and S. Pacific	2006, Dec~2007, Jan	Equilibrium	this study

^aPositive number means volatilization, negative number means deposition.

^bAll flux numbers in Iwata et al 1993 were case 1 from Fig 11.

^cFluxes only include numbers with ascertained air-water gas exchange directions (Fugacity Ratios >3.1

SI Table 5 Internally Consistent Air-Water (K_{aw}), Octanol-Water(K_{ow}), and Octanol-Air (K_{oa}) Partitioning Constants as well as Corresponding Internal Energy (U_{aw} , U_{ow} , U_{oa}) at 25°C for selected PCB Congeners and HCB

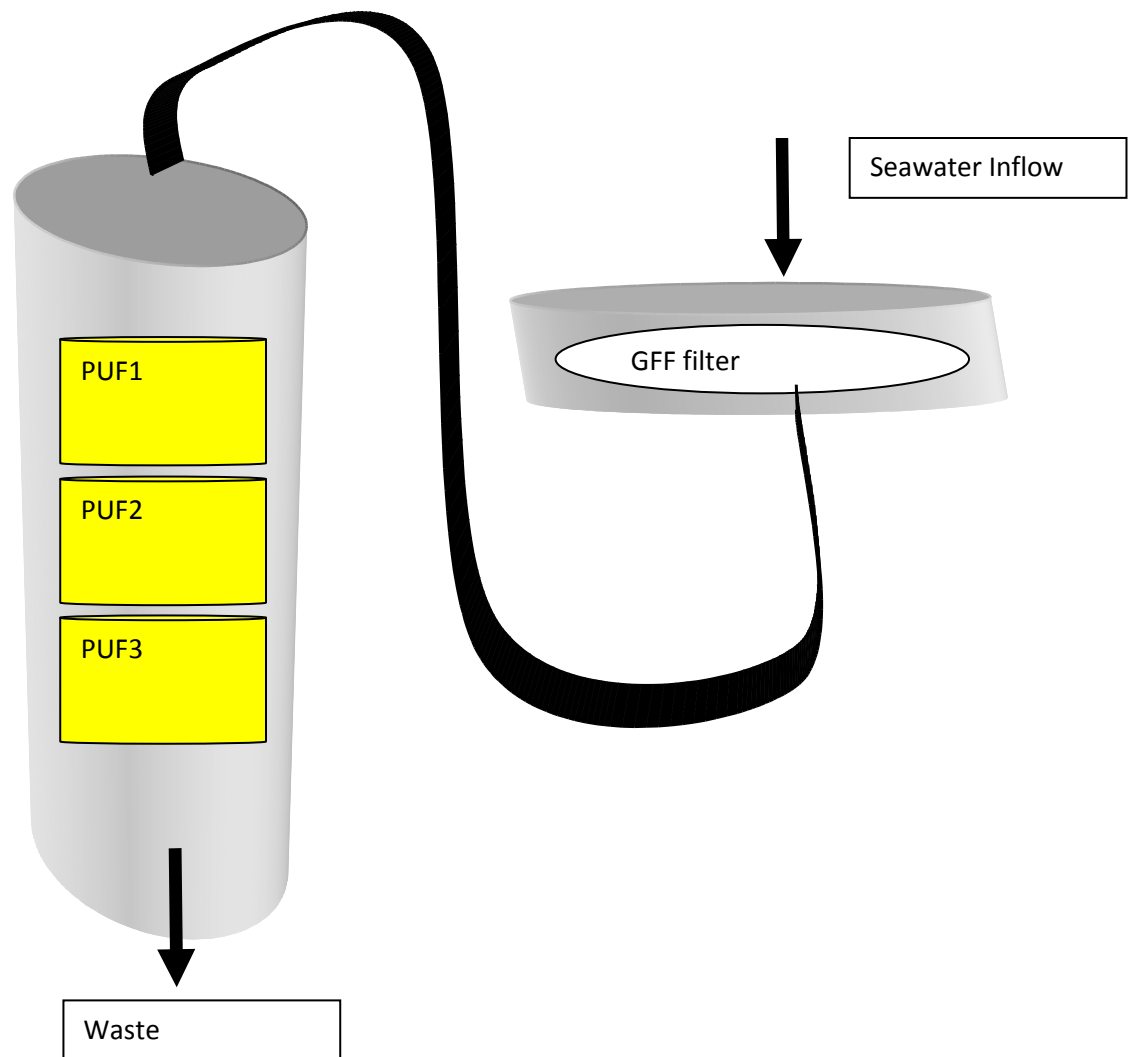
	$\log K_{ow}$	$\log K_{aw}$	ΔU_{aw} (kJ/mol)
PCB3	4.71	-2.06	52.9
PCB8	5.29	-2.06	54.9
PCB18	5.47 ^a	-1.93 ^b	51.8 ^b
PCB28	5.92	-1.93	51.8
PCB52	6.26	-1.96 ^c	53.8 ^c
PCB44	6.06 ^a	-1.96 ^c	53.8 ^c
PCB66	6.58 ^a	-1.96 ^c	53.8 ^c
PCB101	6.76	-2.08	65.2
PCB118	7.08	-2.36	65.2
PCB153	7.31	-2.13	68.2
PCB138	7.70	-1.97	64.7
PCB180	7.66	-2.51	69
PCB189	8.33 ^a		
PCB206	8.77 ^a		
PCB209	8.87 ^a		
Reference		(24)	

^a values from Schenker et al (24), missing values derived from a linear relationship with Hawker and Connell (25),

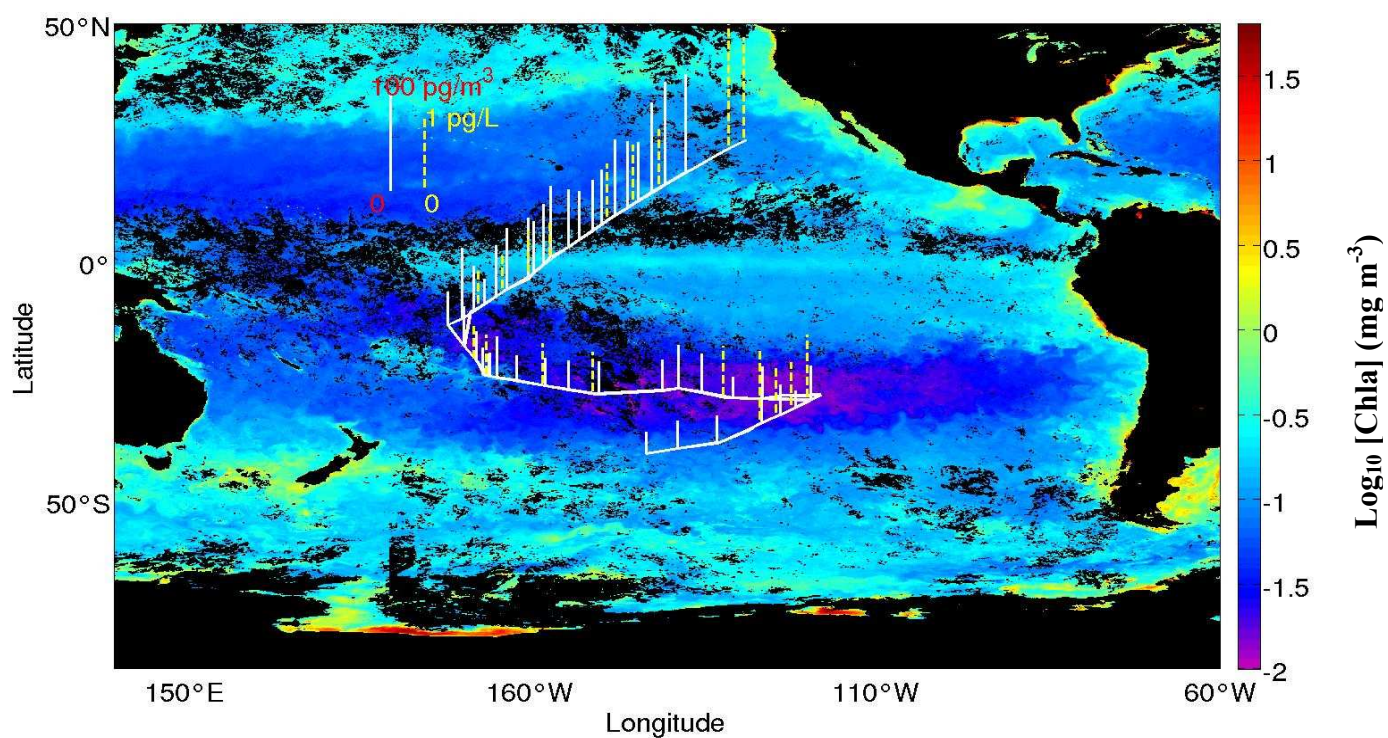
^b PCB-28 values used; ^c PCB-52 values used

	$\log K_{ow}$	$\log K_{aw}$	ΔU_{aw} (kJ/mol)	$\log K_{oa}$	ΔU_{oa} (kJ/mol)
HCB	5.61	-1.51	51.9	7.12	-75.9
Reference			(2)		

Figures



SI Figure 1 Sampling scheme for water samples



SI Figure 2 Air HCB Concentrations (White Bar), Water HCB Concentrations (Yellow Dash Lines), plotted on Aqua MODIS Jan-2007 Chlorophyll Concentration Map in the Pacific

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