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Supporting Information for

Spatial and Temporal Pattern of Pesticides in the Global Atmosphere

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Table S1 Sampling site information, site classification (AG = Agricultural, BA = Background, PO = Polar, RU = Rural and UR = Urban), latitude, longitude and length of sampling period in days for each sampling site.

					Sampling length			
Country	**Location[1]**		**Latitude**	**Longitude**	**1st year**	**2nd year**	**3rd year**	**4th year**
North America								
Canada	Bratt's Lake, SK	AG	50º 12' N	104º 42' W	371	359	344	371
Canada	Toronto, ON	RU	43º 46' N	79º 28' W	393	380	350	362
Canada	Whistler, BC	BA	50º 03' N	122º 57' W		315	344	375
Canada	Lasqueti Island, BC	BA	49º 29' N	124º 21' W		397	323	355
Canada	Little Fox Lake, YT	PO	61º 21' N	135º 38' W			227	387
Canada	Sable Island, NS	BA	43º N	60º W			368	377
Canada	Fraserdale, ON	BA	49º 53' N	81º 34' W			347	357
Canada	Alert, NU	PO	82º 27' N	63º 30' W			365	
Canada	Ucluelet, BC	BA	48º 54' N	125º 32' W			292	375
USA	Barrow, AK	PO	71º 18' N	156° 44' W	399	367	359	
USA	St. Lawrence Island, AK	PO	63º 42' N	170º 29' W	365			
USA	Athens, GA	AG	33º 22' N	83º 28' W	359			
USA	Dyea, AK	BA	59º 31' N	135º 21' W		372	98	391
USA	Point Reyes, CA	BA	33º 14' N	122º 19' W			341	366
USA	Mauna, Hilo	BA	19.54º N	155.58º W			325	362
USA	Tula, AS	BA	14.24º S	170.57º W			299	341
USA	Sydney, FL	UR	27º 57' N	82º 12' W			296	370
Bermuda	Tudor Hill	BA	32º 22' N	64º 39' W	386	327	343	364
Mexico and Central America								
Costa Rica	Tapanti National Park	BA	9 º 46' N	83º 47' W	365	351	356	392
Mexico	Veracruz	UR	19 º 25' N	96º 10' W	398			
Mexico	Tlahuac	UR	19°14' N	99° 00' W		183	355	
South America								
Colombia	Arauca	RU	7º 00' N	70º 44' W	391	368	290	624
Bolvia	Huayna Potosi La Paz	BA	16º 16' S	68º 08' W	327	380		
Chile	Chungura Lake	BA	18°13'S	69°10' W	452			
Chile	Coyhaique	BA	45°35' S	72° 02' W	419		365	
Brazil	Indaiatuba	RU	23º 09' S	47º 10' W	277	366	295	316
Brazil	St. Peter & St. Paul Rocks	BA	17º 37' S	47º 47' W				369
Argentina	Bahia Blanca	AG	38º 45' S	62º 15' W		326		

Table S1 continued

Europe								
Finland	Hollola	RU	61º 3' N	25º 39' E	365			
Finland	Pallas	BA	68º 00' N	24º 14' E		365	351	362
Czech Republic	Košetice	BA	49° 35' N	15° 05' E	370	354	347	369
Norway	Ny-Ålesund	PO	78º 54' N	11º 53' E	371		272	
Poland	Pomlewo	RU	54º 12' N	18º 22' E	349	365	348	
Turkey	Izmir	UR	38° 25' N	27° 08' E	365	372		
Iceland	Stórhöfði	BA	63º 24' N	20º 17' W	405	361	366	359
Ireland	Malin Head	BA	55° 23' N	7° 22' W	414	363	343	365
Spain	Barcelona	UR	41º 23' N	2º 11' E	365			
France	Paris	UR	48º 51' N	2º 21' E	365	321	363	362
Italy	Isola Marettimo	RU	37º 58' N	12º 04' E	365			
Russia	Danki	RU	54º 54' N	37º 48' E		343	355	367
Australia								
Australia	Cape Grim	BA	40º 41' S	144º 41' E	366	295	342	391
Australia	Darwin	RU	12º 22' S	130º 51' E	370	343	358	362
Africa								
Botswana	Kalahari	BA	25º'52' S	22º 54' E	345	482	250	366
South Africa	DeAar	BA	30º 40' S	24º 00' E	401	364	346	364
Ghana	Accra	RU	8º 00' N	2º 00' W	365	365		
Canary Islands	Telde, Las Palmas	RU	28º 59' S	15º 22' W	369	370	275	
Egypt	Cairo	RU	30º 08' N	31º 37' E		398		
Asia								
India	Delhi-C	AG	28° 40' N	77° 14' E	263	317	319	
India	Delhi-D	AG	28° 40'N	77° 14' E	263	317	319	
India	Coimbatore	BA	11° N	77° E			353	
Nepal	Dhulikhel	BA	27° 37'N	75 ° 32' E			145	
Malaysia	Danum Valley	BA	4º 95' N	117º 85' E	372	363	329	364
Kuwait	Abdaly	BA	29º 58' N	47º 42' E			344	382
Kuwait	Kuwait City	UR	29º 34' N	47º 90' E	365	358		
The Philippines	Manila	UR	14º 39'	121º 04' E	347	351		
The Philippines	Tagaytay City	BA	14º 08'	121º 00' E			352	368
Indonesia	Bukit Kototabang	BG	0.20º S	100.32º E		365	281	345
Korea	Pohang	RU	36° 0'N	129° 19' E			352	
Korea	Seoul	UR	37° 35' N	127° 10' E			346	
Korea	Gosan, Jeju Island	BA	33° 24' N	126° 00' E			205	
China	Nam Co. Tibet	BA	30° 46' N	90° 57' E			180	366
Antarctica								
	Mario Zucchelli Station, Italy	PO	74º 41' S	164º 07' E	365			

Detailed Information on Analytical Method and QA/QC

Sample Preparation. Soxhlet extracted XAD-2 resin was filled into mesh cylinders in a clean room at the National Water Research Institute in Burlington, Ontario, Canada, as described previously (Wania et al., 2003). During the third and fourth year, pre-cleaned XAD (Supelpak 2, Supelco, Bellefonte, PA) was used instead.

Extraction and Clean-up of PAS Samples. The XAD-2 resin from the samplers was transferred into an extraction thimble (cellulose) which had been pre-cleaned with dichloromethane (DCM) in a Soxhlet for 4 hours. After adding recovery standards, the resin was Soxhlet extracted with 350 ml of DCM for 20 hours. Extracts were volume reduced using a rotary evaporator and concentrated to around 1 ml using a gentle stream of nitrogen. The extracts from the first year samples were cleaned on a column with 1 g of 6% deactivated alumina (baked at 450 °C overnight) and 0.5 cm of sodium sulfate. The samples were eluted with 20 ml of DCM: petroleum ether (5:95; v/v). After reduction to 3 ml using a rotary evaporator, the extracts from the second, third and fourth year samples were passed through sodium sulfate (baked at 450 °C overnight) columns to remove water residues, but they were not cleaned using alumina. Extracts were again concentrated to 1 ml using a stream of nitrogen and then solvent-exchanged into iso-octane. The final volume of the extracts was 1 ml, and 100 ng of mirex was added to the samples for volume correction.

Analysis of Extracts. The extracts were analyzed for α- and γ-HCH, HCB, cis- (CC) and trans-chlordane (TC), trans-nonachlor (TN), heptachlor (HEPT), heptachlor exo-epoxide (HEPX), aldrin, dieldrin, endrin, p,p'-DDT, o,p'-DDT, p,p'-DDD, o,p'-DDD, p,p'-DDE, o,p'-DDE, α- and β-endosulfan (α-ES, β-ES), endosulfan sulphate (ESS), trifluralin (TF), chlorothalonil (CT), dacthal (DT), and pendimethalin (PM) using an Agilent 6890 gas chromatograph (GC) coupled to a 5973 mass selective detector with a negative chemical ionization source in selected ion mode. A DB-5 column (60 m, 0.25 mm i.d., 0.3 μm film thickness) was used for separation. Helium was used as the carrier gas at a flow rate of 1.2 ml min^{-1}. The GC oven temperature program was: 70 °C held for 0.5 min then increased at 15 °C·min^{-1} to 160 °C, then increased at 2 °C·min^{-1} to 250 °C and finally increased at 20 °C·min^{-1} to 270 °C and held for 5 min.

Quality Control and Assurance measures were used to monitor all analytical procedures. Field blanks were collected each year as additional XAD-PAS were shipped to several sampling sites. During the one-year deployment, mesh cylinders with resin were kept in the sealed sample holders, which were mounted on the post holding the sampler housing. These field blanks thus underwent

the same shipping, handling, and storage as the exposed XAD-PAS, except that the blanks were never exposed to air. Laboratory and field blanks were processed in the same way as the samples to determine the contamination introduced during extraction and clean-up, and by handling, shipping and storage, respectively. Very low blank concentrations indicated that no contamination had occurred during transport, storage or analysis. All data are blank-corrected using the average of 6 to 12 lab and 4 to 10 field blanks. Method detection limits (MDLs) were calculated as three times the standard deviation of the blanks. For those pesticides that were not detected in the blanks, MDLs were calculated by using three times the signal-to-noise ratio of the lowest calibration standard. The MDLs are included in the tables reporting the concentration data (Tables S2 and S3).

Recoveries of surrogates (d_6-γ-HCH, $^{13}C_{10}$-HEPX, $^{13}C_{10}$-TN, ^{13}C-Dieldrin, ^{13}C-*p,p'*-DDT) were between 72 and 105 %. The XAD had not been spiked with surrogates for the CUPs. To test for the loss of these compounds during the extraction and clean-up procedure, six replicates of 20 g of XAD-2 were spiked with five pesticides (CT, DT, PM, TF, chlorpyrifos) and then extracted and cleaned up in the same way as the samples. Recovery was between 76 to 111 % and thus sufficiently high to report concentrations. The recovery of the pesticides before clean-up with an alumina column ranged from 85 to 103 %. The reported data are not corrected for recovery.

Less persistent pesticides, such as chlorpyrifos, PM and TF, may be lost from the resin during the one year deployment as a result of degradation. In the case of chlorpyrifos we judged that possibility to be so high as to compromise the interpretation of its concentration in GAPS samples (Gouin et al., 2008). Therefore no chlorpyrifos data are being reported here. Although we can not rule out the possibility of degradation loss for PM and TF, extensive calibration studies suggested that their levels measured in XAD-PAS exposed for one year are sufficiently reliable to be interpreted with confidence (Hayward et al., 2010).

Table S2 Concentrations and method detection limits (MDL) in ng/PAS of selected current use pesticides (TF: trifluralin, CT: chlorothalonil, DT: dacthal, PM: pendimethalin, ES: Endosulfan, ESS: Endosulfan sulfate) in globally deployed passive air samplers. (ND: not detected, BD: below method detection limit).

Part 1 First sampling year (late 2004 to late 2005)

Location	TF	CT	DT	PM	α-ES	β-ES	ESS
North America							
Bratt's Lake, Canada	16.5	66.0	0.6	ND	5.2	0.4	ND
Toronto, Canada	6.8	78.2	8.4	2.8	42.9	4.3	0.5
Barrow, USA	BD	0.2	BD	ND	2.5	ND	ND
St. Lawrence Island, USA	BD	ND	ND	ND	0.1	ND	ND
Georgia, USA	1.9	22.1	2.3	4.2	11.6	2.1	0.7
Tudor Hill, Bermuda	0.2	2.3	0.5	55.2	3.5	0.1	ND
Mexico and Central America							
Tapanti, Costa Rica	ND	ND	ND	ND	1.9	ND	ND
Veracruz, Mexico	3.0	170	1.8	ND	216	48.2	5.8
South America							
Arauca, Columbia	0.1	0.6	0.5	ND	48.4	4.5	0.9
Huayna Potosi, Bolivia	ND	0.2	BD	ND	34.8	5.1	0.4
Chungura, Chile	ND	0.7	BD	ND	61.9	0.9	0.9
Coyhaique, Chile	ND	BD	BD	ND	1.0	ND	ND
Indaiatuba, Brazil	3.4	6.2	BD	ND	53.8	14.7	2.0
Europe							
Hollola, Finland	2.9	2.0	0.2	ND	2.8	0.1	ND
Košetice, Czech Rep.	89.1	8.2	0.2	103	10.3	1.5	ND
Ny-Ålesund, Norway	ND	0.2	0.1	ND	2.7	ND	ND
Pomlewo, Poland	19.0	0.3	BD	6.7	3.1	1.1	0.3
Izmir, Turkey	8.2	6.1	0.3	ND	26.6	8.9	0.4
Stórhöfði, Iceland	BD	3.2	0.2	ND	3.2	ND	0.1
Malin Head, Ireland	17.5	4.0	0.8	3.9	3.9	0.1	0.1
Barcelona, Spain	1.8	23.8	5.9	3.8	138	34.1	3.8
Paris , France	188	168	3.1	28.9	213	61.5	1.9
Isola Marettimo, Italy	0.5	15.1	3.0	ND	55.8	8.0	1.6
Australia							
Cape Grim, Australia	9.6	1.0	0.8	ND	9.0	0.6	0.1
Darwin, Australia	BD	0.9	0.1	ND	3.5	0.1	ND
Africa							
Kalahari, Botswana	0.3	6.8	0.3	ND	10.4	0.3	1.0
DeAar, South Africa	0.1	5.0	0.2	ND	14.3	1.9	1.1
Accra, Ghana	ND	BD	BD	ND	330	113	24.5
Telde, Las Palmas, Spain	0.5	3.4	0.1	0.7	60.2	12.7	0.2

Asia							
Delhi- C, India	1.4	ND	ND	2.5	755	56.1	6.7
Delhi-D, India	1.4	1.0	0.2	7.1	391	23.6	3.8
Danum Valley, Malaysia	ND	0.7	0.1	ND	6.9	0.4	0.1
Kuwait City, Kuwait	0.2	49.6	0.4	ND	21.7	7.7	1.5
Manila, The Philippines	0.1	0.3	0.1	0.7	2.3	ND	ND
Antarctica							
Mario Zucchelli Station, Italy	ND	ND	ND	ND	ND	ND	ND
MDL	0.08	0.13	0.04	0.09	0.09	0.03	0.02

Part 2 Second sampling year (late 2005 to late 2006)

Location	**TF**	**CT**	**DT**	**PM**	**α-E**	**B-E**	**ESS**
North America							
Bratt's Lake, Canada	6.5	232	2.3	ND	6.2	0.5	0.2
Toronto, Canada	7.0	135	4.5	3.0	23.4	4.5	0.5
Barrow, USA	ND	BD	BD	ND	1.9	ND	ND
Dyea, USA	BD	BD	BD	ND	BD	ND	ND
Lasqueti Island, Canada	1.0	12.2	0.5	ND	4.8	1.1	0.2
Tudor Hill, Bermuda	0.2	5.9	0.2	ND	3.2	BD	0.1
Whistler, Canada	ND	9.4	0.7	ND	8.3	0.8	0.3
Mexico & Central America							
Tapanti, Costa Rica	ND	1.7	ND	ND	5.6	0.5	0.2
Tlahuac, Mexico	6.0	278	0.5	ND	170	34.9	4.2
South America							
Arauca, Columbia	ND	BD	BD	ND	24.4	5.3	1.3
Huayna Potosi La Paz, Bolivia	0.2	2.3	BD	ND	34.4	0.3	0.7
Indaiatuba, Brazil	3.8	54.0	BD	ND	175	34.9	6.6
Bahia Blanca, Argentina	18.5	1.3	BD	ND	149	28.0	9.1
Europe							
Pallas, Finland	ND	17.7	0.2	ND	2.8	BD	0.1
Košetice, Czech Rep.	172	233	0.9	106	17.4	2.1	0.7
Danki, Russia	BD	3.2	BD	2.2	2.4	BD	0.1
Pomlewo, Poland	21.7	84.4	0.6	15.2	9.9	1.1	0.3
Izmir, Turkey	15.7	27.7	0.6	ND	42.2	20.2	2.1
Stórhöfði, Iceland	ND	12.0	0.4	ND	3.2	BD	0.1
Malin Head, Ireland	13.4	49.0	1.2	3.2	4.2	0.3	0.1
Paris , France	247	466	3.8	29.2	166	49.2	3.8
Australia							
Cape Grim, Australia	1.9	4.2	0.9	ND	4.8	0.4	0.2
Darwin, Australia	BD	5.1	BD	2.1	2.9	0.3	0.1

Africa							
Kalahari, Botswana	ND	5.0	0.3	ND	14.6	2.2	1.0
DeAar, South Africa	BD	6.8	0.6	ND	19	2.2	1.1
Accra, Ghana	BD	BD	BD	ND	353	119	23.3
Telde, Las Palmas, Spain	ND	47.3	0.6	ND	70	14.4	1.7
Cairo, Egypt	ND	13.0	0.6	ND	35	3.8	1.1
Asia							
Delhi-C, India	1.3	11.4	ND	ND	384	160	22.0
Delhi-D, India	0.9	6.8	ND	ND	430	159	20.1
Danum Valley, Malaysia	ND	2.3	BD	ND	4.9	0.4	0.3
Kuwait City, Kuwait	ND	56.7	0.2	ND	16.4	5.6	1.5
Manila, The Philippines	ND	4.7	ND	ND	4.5	0.5	0.1
Bukit Kototabang, Indonesia	ND	BD	BD	ND	6.9	0.8	0.4
MDL	0.05	1.16	0.19	0.28	0.54	0.05	0.03

Part 3 Third sampling year (late 2006 to late 2007)

Location	**TF**	**CT**	**DT**	**PM**	**A-E**	**β-E**	**ESS**
North America							
Bratt's Lake, Canada	10.0	111	2.1	ND	5.8	0.7	0.2
Toronto, Canada	2.7	185	7.7	4.0	33.5	7.9	0.9
Whistler, Canada	ND	6.7	1.7	ND	8.3	1.2	0.4
Lasqueti Island, Canada	0.2	4.7	ND	ND	1.8	0.4	0.1
Little Fox Lake, Canada	ND	ND	ND	ND	1.6	BD	ND
Sable Island, Canada	ND	12.9	1.0	ND	4.2	0.5	0.2
Fraserdale, Canada	ND	13.3	1.0	ND	4.2	0.4	0.3
Ucluelet, Canada	ND	0.7	ND	ND	1.1	BD	BD
Alert, Canada	ND	ND	ND	ND	1.6	ND	ND
Barrow, USA	ND	ND	BD	ND	1.1	ND	ND
Dyea, USA	ND	0.9	ND	ND	0.3	ND	ND
St. Lawrence Island, USA	ND	ND	ND	ND	ND	ND	ND
Point Reyes in California, USA	ND	3.5	0.9	ND	1.5	BD	0.1
Hilo, USA	ND	0.9	ND	ND	4.9	0.2	0.6
Tula in American Samo, USA	ND	ND	ND	ND	0.6	ND	ND
Sydney in Florida, USA	14.9	272	ND	54.3	369	117	14.8
Tudor Hill, Bermuda	ND	4.1	0.3	ND	2.9	0.1	0.1
Mexico and Central America							
Tapanti, Costa Rica	ND	1.7	ND	ND	6.3	0.8	0.4
Tlahuac, Mexico	2.3	109	0.7	ND	112	20.0	2.8
South America							
Arauca, Columbia	ND	ND	ND	ND	5.7	1.2	0.3
Chungura, Chile	ND	ND	0.2	ND	4.0	ND	0.1
Indaiatuba, Brazil	0.9	33.0	ND	ND	52.2	11.1	2.5

Europe							
Pallas, Finland	ND	3.2	0.2	ND	1.7	BD	BD
Košetice, Czech Rep.	104	161	1.1	259	14.7	2.2	0.8
Ny-Ålesund, Norway	ND	0.9	0.1	ND	1.1	ND	ND
Pomlewo, Poland	8.9	46.6	0.6	11.0	7.1	1.0	0.3
Stórhöfði, Iceland	ND	10.6	0.5	ND	3.1	0.1	0.1
Malin Head, Ireland	1.9	13.3	0.5	1.5	1.9	ND	BD
Paris , France	111	312	3.2	34.1	52.3	13.4	1.5
Danki, Russia	0.2	5.0	ND	ND	2.0	0.2	0.1
Australia							
Cape Grim, Australia	7.9	2.6	0.6	ND	7.1	0.6	0.3
Darwin, Australia	ND	2.5	ND	ND	4.6	0.9	0.2
Africa							
Kalahari, Botswana	0.1	2.0	0.3	ND	5.2	0.7	0.4
DeAar, South Africa	0.1	3.9	1.1	ND	21.0	2.0	1.3
Telde, Las Palmas, Spain	ND	15.6	0.4	ND	27.6	0.2	0.7
Asia							
Delhi- C, India	ND	39.8	ND	ND	846	230	22.7
Delhi-D, India	0.3	ND	ND	4.4	312	103	10.7
Danum Valley, Malaysia	ND	1.1	ND	ND	1.7	0.2	0.1
Kuwait City, Kuwait	ND	7.7	ND	ND	11.8	4.5	2.4
Manila, The Philippines	ND	1.9	ND	ND	7.6	0.4	0.3
Bukit Kototabang, Indonesia	ND	ND	ND	ND	4.3	0.3	0.2
Coimbatore, India	0.3	5.5	ND	9.0	676	207	19.5
Kuntabesi, Nepal	ND	ND	ND	ND	337	86	11.5
Pohang, Korea	0.2	92.7	ND	ND	263	59.8	8.0
Seoul, Korea	0.4	99.6	ND	12.3	225	53.9	5.8
Jeju Island, Korea	0.7	110	ND	504.6	483	150	24.8
Tibet, China	ND	0.6	ND	ND	26.8	0.1	0.7
MDL	0.09	0.17	0.08	0.07	0.12	0.06	0.05

Part 4 Fourth sampling year (late 2007 to late 2008)

Location	**TF**	**CT**	**DT**	**PM**	**α-E**	**β-E**	**ESS**
North America							
Bratt's Lake, Canada	6.0	54.9	1.61	ND	6.7	0.7	0.9
Toronto, Canada	1.1	74.6	3.2	1.6	24.1	5.1	3.0
Whistler, Canada	ND	3.5	0.6	ND	5.6	0.7	0.9
Lasqueti Island, Canada	0.1	3.4	BD	ND	1.8	0.2	ND
Little Fox Lake, Canada	ND	0.4	BD	ND	2.2	ND	ND
Sable Island, Canada	ND	7.3	0.6	ND	4.6	0.3	0.6
Fraserdale, Canada	ND	11.0	0.7	ND	3.2	0.2	0.8
Ucluelet, Canada	ND	0.8	BD	ND	1.9	ND	ND

Dyea, USA	ND	0.5	BD	ND	0.4	ND	ND
Point Reyes in California, USA	ND	2.3	1.3	ND	1.7	ND	ND
Hilo, USA	ND	0.5	BD	ND	2.0	ND	1.4
Tula American Samo, USA	0.1	0.5	BD	ND	2.1	ND	ND
Sydney in Florida, USA	2.9	262	1.7	12.0	438	139	73.7
Tudor Hill, Bermuda	ND	1.8	0.2	ND	2.4	ND	0.4
Tapanti, Costa Rica	BD	1.7	ND	ND	7.7	0.9	1.8
South America							
Arauca, Columbia	ND	ND	ND	ND	3.7	0.6	0.9
St. Peter and St. Paul Rocks, Brazil	ND	0.4	ND	ND	206	34.7	32.3
Indaiatuba, Brazil	0.3	70.8	ND	ND	7.4	0.3	0.7
Europe							
Pallas, Finland	ND	1.3	BD	ND	1.2	ND	ND
Košetice, Czech Rep.	47.5	120	0.8	92.3	5.9	0.6	1.5
Stórhöfði, Iceland	ND	8.3	0.4	ND	3.2	ND	ND
Malin Head, Ireland	0.2	30.6	0.6	ND	2.2	ND	ND
Paris , France	18.0	135	1.9	9.9	11.2	2.2	1.7
Danki, Russia	ND	1.6	BD	ND	1.1	ND	ND
Australia							
Cape Grim, Australia	1.3	1.5	0.5	ND	6.9	0.5	1.3
Darwin, Australia	ND	1.0	0.2	ND	3.6	0.3	0.9
Africa							
Kalahari, Botswana	ND	1.9	0.4	ND	15.3	2.5	5.6
DeAar, South Africa	BD	2.1	0.8	ND	19.6	1.9	5.6
Asia							
Danum Valley, Malaysia	ND	0.8	BD	ND	3.5	0.2	0.8
Kuwait City, Kuwait	ND	4.0	BD	ND	9.7	2.3	7.9
Manila, The Philippines	ND	1.0	BD	ND	5.5	0.2	1.6
Bukit Kototabang, Indonesia	ND	ND	ND	ND	5.4	ND	1.0
Tibet, China	ND	ND	ND	ND	39.1	0.2	5.9
MDL	0.08	0.12	0.09	0.1	0.1	0.05	0.05

Table S3 Concentrations and method detection limits (MDL) in ng/PAS of selected pesticides in globally deployed passive air samplers (ND: not detected, BD: below method detection limit.).

Part 1 First sampling year (late 2004 to late 2005).

Location	*α*-HCH	*γ*-HCH	HCB	CC	TC	TN	HEPT	HEPX	Dieldrin	*o,p'*-DDE	*p,p'*-DDE	*o,p'*-DDT
North America												
Bratt's Lake, Canada	8.6	7.0	32.8	0.5	0.6	0.6	ND	2.8	ND	ND	ND	ND
Toronto, Canada	7.0	4.5	28.1	2.1	2.0	2.0	0.5	1.8	4.4	ND	7.2	ND
Barrow, USA	13.5	1.7	34.9	0.4	0.2	0.4	ND	0.2	ND	ND	ND	ND
St. Lawrence Island, USA	7.6	0.9	19.2	0.2	0.1	0.2	ND	ND	ND	ND	ND	ND
Georgia, USA	6.2	8.4	240	3.1	4.8	1.6	2.3	2.5	1.7	ND	ND	ND
Tudor Hill, Bermuda	3.4	1.3	74.2	1.2	1.3	1.1	ND	0.6	6.7	ND	ND	ND
Mexico & Central America												
Tapanti, Costa Rica	0.3	2.3	7.2	0.1	0.1	0.1	ND	ND	ND	ND	ND	ND
Veracruz, Mexico	14.1	47.5	164	1.6	2.0	1.2	ND	2.5	BD	ND	ND	ND
South America												
Arauca, Columbia	1.6	3.7	111	0.3	0.2	0.1	ND	ND	ND	ND	8.1	ND
Huayna Potosi La Paz, Bolivia	0.9	0.5	8.9	ND	BD	BD	ND	ND	ND	ND	ND	ND
Chungura, Chile	1.2	1.4	16.3	ND	BD	BD	ND	ND	ND	ND	ND	ND
Coyhaique, Chile	0.3	0.3	15.7	BD	BD	ND	ND	ND	ND	ND	ND	ND
Indaiatuba, Brazil	3.5	5.3	9.4	0.3	0.8	0.2	1.0	ND	BD	ND	8.1	ND
Europe												
Hollola, Finland	5.7	2.6	28.0	0.3	0.2	0.3	ND	0.1	ND	ND	ND	ND
Košetice, Czech Rep.	9.5	13.2	78.1	0.3	0.3	0.4	ND	ND	ND	0.4	8.2	1.3
Ny-Ålesund, Norway	6.2	1.1	27.0	0.3	0.2	0.4	ND	0.3	ND	ND	ND	ND
Pomlewo, Poland	7.2	5.8	32.9	0.3	0.2	0.3	ND	0.4	ND	ND	4.2	ND
Izmir, Turkey	17.7	8.7	36.5	0.3	0.3	0.4	ND	0.7	ND	ND	7.5	ND
Stórhöfði, Iceland	9.6	2.0	36.3	0.6	0.3	0.5	ND	0.5	ND	0.5	ND	ND
Malin Head, Ireland	7.6	4.1	39.9	0.5	0.3	0.4	ND	0.6	1.5	ND	ND	ND
Barcelona, Spain	8.1	18.8	42.0	1.4	3.3	0.8	ND	4.0	3.9	0.8	8.0	ND
Paris , France	16.5	118	57.1	1.7	1.9	1.9	1.7	7.4	7.2	ND	2.9	1.7
Isola Marettimo, Italy	8.6	8.2	54.5	0.7	0.5	0.6	ND	1.6	1.5	0.7	5.7	8.8
Australia												
Cape Grim, Australia	0.4	0.2	31	0.2	0.2	0.1	ND	ND	ND	ND	ND	ND
Darwin, Australia	0.5	1.1	54.7	1.5	6.1	3.3	4.0	1.9	7.6	ND	ND	ND
Africa												
Kalahari, Botswana	2.0	1.8	80.7	0.3	0.3	0.1	ND	ND	ND	ND	ND	ND
DeAar, South Africa	44.2	16.4	33.0	0.2	0.2	0.2	ND	ND	ND	ND	ND	ND
Accra, Ghana	0.6	24.5	12.2	ND	0.1	ND	ND	ND	ND	ND	ND	ND
Telde, Las Palmas	4.9	22.9	33.6	0.8	0.9	0.6	0.9	0.6	4.2	2.0	23	2.6

Asia												
Delhi- C, India	306	731	184	2.5	8.0	0.8	54.0	ND	ND	10	66	27
Delhi-D, India	221	321	254	1.4	7.4	0.6	ND	ND	ND	9.0	42	22
Danum Valley, Mal.	1.0	0.6	8.7	0.1	0.2	0.2	ND	ND	2.0	ND	ND	ND
Kuwait City, Kuwait	14.4	17.6	110	1.3	1.5	1.1	ND	ND	2.0	1.9	5.4	ND
Manila, Philippines	1.6	3.6	21	23.9	35.8	17.8	13.2	1.6	6.9	1.0	5.9	1.7
Antarctica												
Mario Zucchelli Station, Italy	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
MDL	0.14	0.1	0.95	0.06	0.04	0.09	0.20	0.12	0.46	0.34	0.34	1.2

Part 2 Second sampling year (late 2005 to late 2006).

Location	***α*-HCH**	***γ*-HCH**	**HCB**	**CC**	**TC**	**TN**	**HEPT**	**HEPX**	**Dieldrin**	***o,p'*-DDE**	***p,p'*-DDE**	***o,p'*-DDT**
North America												
Bratt's Lake, Canada	5.1	3.4	33.6	0.2	0.3	0.3	ND	2.2	ND	ND	ND	ND
Toronto, Canada	2.5	1.5	22.6	0.7	0.8	0.8	ND	1.1	ND	ND	5.0	ND
Barrow, USA	5.3	0.8	26.2	0.1	ND	0.2	ND	ND	ND	ND	ND	ND
Dyea, USA	2.0	0.2	12.5	ND	ND	ND	ND	ND	ND	ND	ND	ND
Lasqueti Island, Canada	5.1	0.7	25.7	0.2	0.2	0.2	ND	ND	ND	ND	ND	ND
Tudor Hill, Bermuda	1.1	0.5	19.7	0.5	ND	0.6	ND	ND	9.4	ND	ND	ND
Whistler, Canada	5.0	0.9	24.5	0.2	ND	0.1	ND	ND	ND	ND	ND	ND
Mexico and Central America												
Tapanti, Costa Rica	BD	BD	7.2	ND	ND	ND	ND	ND	ND	ND	ND	ND
Tlahuac, Mexico	4.1	19.2	37.4	0.9	1.3	0.9	ND	ND	ND	ND	ND	ND
South America												
Arauca, Columbia	0.3	0.5	11.0	ND	0.5	0.1	ND	ND	ND	ND	6.3	ND
Huayna Potosi, Bolivia	0.7	0.7	16.0	ND	0.2	ND	5.8	ND	ND	ND	ND	ND
Indaiatuba, Brazil	2.7	3.9	11.4	0.2	0.5	0.1	1.3	ND	ND	ND	ND	ND
Bahia Blanca, Argentina	0.5	0.7	21.4	ND	0.2	ND	ND	ND	ND	ND	5.2	ND
Europe												
Pallas, Finland	2.9	1.0	26.3	ND	0.1	0.1	ND	ND	ND	ND	ND	ND
Košetice, Czech Rep.	4.9	7.2	57.6	0.2	0.1	0.2	ND	0.9	ND	0.4	10.2	ND
Danki, Russia	5.9	2.5	25.4	ND	0.1	0.1	ND	ND	ND	ND	ND	ND
Pomlewo, Poland	4.7	4.9	37.5	0.2	0.1	0.2	ND	ND	ND	ND	ND	ND
Izmir, Turkey	10.0	4.7	45.0	0.2	0.1	0.2	ND	ND	ND	ND	6.2	ND
Stórhöfði, Iceland	6.8	1.4	53.9	0.3	0.2	0.3	ND	ND	ND	0.5	ND	ND
Malin Head, Ireland	4.1	3.3	37.3	0.3	0.2	0.2	ND	0.7	ND	ND	ND	ND
Paris , France	8.3	71.9	30.6	0.7	0.9	1.1	1.3	10.3	9.0	ND	7.3	ND

Australia												
Cape Grim, Australia	BD	ND	22.8	ND	0.1	ND	ND	1.4	ND	ND	ND	ND
Darwin, Australia	BD	0.3	13.0	0.8	3.0	0.9	3.2	ND	9.4	ND	ND	ND
Africa												
Kalahari, Botswana	1.0	1.7	10.9	0.1	0.1	0.1	ND	ND	ND	ND	ND	ND
DeAar, South Africa	41.1	18.7	19.4	0.1	0.2	0.1	ND	ND	ND	ND	ND	ND
Accra, Ghana	0.4	14.2	11.0	ND	0.1	ND	ND	ND	ND	ND	ND	ND
Cairo, Egypt	2.6	12.7	58.6	0.4	0.3	0.3	ND	ND	ND	ND	ND	ND
Telde, Las Palmas, Spain	14.0	5.9	32.4	0.3	0.4	0.2	ND	0.8	ND	ND	15.1	ND
Asia												
Delhi-C, India	204	491	158	1.0	3.7	0.5	ND	ND	ND	ND	80.0	ND
Delhi-D, India	127	219	172	0.8	4.5	0.3	30.7	ND	ND	ND	45.2	ND
Danum Valley, Malaysia	0.4	0.4	14.3	ND	ND	ND	ND	ND	ND	ND	ND	ND
Kuwait City, Kuwait	4.3	4.6	46.1	0.6	0.6	ND	ND	ND	ND	ND	ND	ND
Manila, The Philippines	0.7	4.4	25.0	9.9	16.2	11.0	8.8	1.5	10.9	ND	ND	ND
Bukit Kototabang, Indonesia	0.4	2.0	13.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
MDL	0.17	0.19	0.52	0.07	0.03	0.04	0.4	0.4	0.9	0.50	0.55	1.5

Part 3 Third sampling year (late 2006 to late 2007).

Location	***α*-HCH**	***γ*-HCH**	**HCB**	**CC**	**TC**	**TN**	**HEPT**	**HEPX**	**Diel-drin**	***o,p'*-DDE**	***p,p'*-DDE**	***o,p'*-DDT**
North America												
Bratt's Lake, Canada	3.3	2.8	20.3	0.2	0.3	0.3	ND	2.1	2.2	ND	ND	ND
Toronto, Canada	3.5	2.3	21.7	1.2	1.3	1.1	ND	ND	ND	ND	ND	ND
Whistler, Canada	5.1	0.7	22.8	0.1	ND	0.1	ND	ND	ND	ND	ND	ND
BC, Canada	3.3	0.3	9.6	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Lasqueti Island, Canada	2.4	0.4	9.8	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Little Fox Lake, Canada	4.6	0.5	17.5	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Sable Island, Canada	15.3	1.5	34.0	0.4	0.4	0.4	ND	ND	1.2	ND	ND	ND
Fraserdale, Canada	4.8	0.8	18.2	0.3	ND	0.3	ND	20.7	ND	ND	40.6	ND
Ucluelet, Canada	3.3	0.3	9.6	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Alert, Canada	3.0	0.3	15.8	0.1	ND	0.1	ND	ND	ND	ND	ND	ND
Barrow, USA	5.0	0.5	13.7	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Dyea, USA	1.1	0.4	4.4	ND	ND	ND	ND	ND	ND	ND	ND	ND
Reyes, California, USA	4.3	0.4	12.9	0.3	0.3	0.2	ND	ND	ND	ND	ND	ND
Hilo, USA	2.3	0.4	22.2	0.1	ND	0.1	ND	ND	ND	ND	ND	ND
Tula, American Samo	ND	ND	7.4	BD	ND	0.1	ND	ND	ND	ND	ND	ND
Sydney, Florida, USA	1.1	2.2	16.2	4.3	6.9	4.7	ND	ND	4.3	ND	ND	ND
Tudor Hill, Bermuda	1.3	0.7	13.5	0.4	1.2	0.5	ND	ND	6.5	ND	ND	ND

Mexico and Central America												
Tapanti, Costa Rica	ND	16.9	6.4	ND	ND	ND	ND	ND	ND	ND	ND	ND
Tlahuac, Mexico	2.6	8.9	15.9	0.5	0.8	0.5	ND	ND	ND	ND	ND	ND
South America												
Arauca, Columbia	ND	ND	4.4	ND	ND	ND	ND	ND	ND	ND	ND	ND
Chungura, Chile	ND	ND	8.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
Indaiatuba, Brazil	1.2	2.3	5.5	0.1	0.3	BD	ND	ND	ND	ND	ND	ND
Europe												
Pallas, Finland	2.5	0.6	16.9	0.1	BD	0.1	ND	ND	ND	ND	ND	ND
Košetice, Czech Rep.	5.6	7.2	49.0	0.3	BD	0.2	ND	ND	ND	ND	15.9	ND
Ny-Ålesund, Norway	2.1	0.3	12.6	ND	ND	0.1	ND	ND	ND	ND	ND	ND
Pomlewo, Poland	4.3	3.2	23.2	0.1	BD	0.2	ND	ND	ND	ND	ND	ND
Stórhöfði, Iceland	5.8	6.2	30.7	0.4	BD	0.3	ND	ND	ND	ND	ND	ND
Malin Head, Ireland	2.4	1.9	18.8	0.2	BD	0.1	ND	ND	ND	ND	ND	ND
Paris , France	7.8	52.4	23.3	0.6	0.9	1.1	1.2	15.8	8.8	ND	ND	ND
Danki, Russia	ND	ND	20.9	0.1	ND	BD	ND	ND	ND	ND	ND	ND
Australia												
Cape Grim, Australia	ND	ND	13.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
Darwin, Australia	ND	ND	7.4	0.6	3.1	0.7	3.2	1.0	7.1	ND	ND	ND
Africa												
Kalahari, Botswana	0.7	1.5	9.0	BD	ND	BD	ND	ND	ND	ND	ND	ND
DeAar, South Africa	18.4	8.9	13.4	0.1	ND	0.1	ND	ND	ND	ND	ND	ND
Telde, Las Palmas	1.4	6.3	14.5	0.2	0.4	0.1	ND	6.6	3.3	ND	ND	ND
Asia												
Delhi- C, India	212	797	384	0.9	3.5	0.4	ND	45.5	ND	ND	78.7	ND
Delhi-D, India	58	143	151	0.6	3.3	0.2	19.2	29.2	ND	ND	43.4	ND
Danum Valley, Malaysia	ND	ND	4.6	BD	ND	ND	ND	ND	ND	ND	ND	ND
Kuwait City, Kuwait	4.1	5.1	40.7	0.3	ND	0.3	ND	ND	ND	ND	ND	ND
Manila, Philippines	1.0	0.8	20.5	1.2	2.0	1.1	ND	ND	ND	ND	ND	ND
Bukit Kototabang, Indonesia	0.3	9.9	5.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
Coimbatore, India	31.7	51.1	23.3	ND	0.8	0.1	ND	17.2	ND	ND	28.5	ND
Kuntabesi, Nepal	50.8	18.3	8.7	ND	ND	ND	ND	19.3	ND	ND	31.7	36.4
Pohang, Korea	8.8	4.0	45.3	0.4	ND	0.3	ND	ND	ND	ND	ND	ND
Seoul, Korea	12.0	19.1	63.9	0.3	0.7	0.3	ND	ND	ND	ND	ND	ND
Jeju Island, Korea	5.6	1.8	27.4	0.4	ND	0.3	ND	ND	ND	ND	ND	ND
Nam Co., Tibet, China	5.9	1.7	11.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
MDL	0.12	0.17	0.35	0.06	0.08	0.09	0.25	0.28	0.42	0.38	0.40	1.4

Part 4 Fourth sampling year (late 2007 to late 2008).

Location	***α*-HCH**	***γ*-HCH**	**HCB**	**CC**	**TC**	**TN**	**HEPT**	**HEPX**	**Diel-drin**	***o,p'*-DDE**	***p,p'*-DDE**	***o,p'*-DDT**
North America												

Bratt's Lake, Canada	4.2	2.1	29.5	0.2	0.3	0.4	ND	3.0	2.0	ND	ND	ND
Toronto, Canada	1.8	1.3	15.4	0.9	0.9	1.0	ND	ND	ND	ND	ND	ND
Whister, Canada	6.0	0.7	9.5	0.1	0.2	ND	ND	ND	ND	ND	ND	ND
Lasqueti Island, Canada	2.2	0.4	7.4	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Little Fox Lake, Canada	5.2	0.4	23.6	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Sable Island, Canada	13.5	1.6	37.5	0.5	0.5	0.6	ND	ND	ND	ND	ND	ND
Fraserdale, Canada	4.1	0.8	20.2	0.2	ND	0.2	ND	19.3	0.8	ND	38.1	ND
Ucluelet, Canada	5.3	0.3	21.0	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Dyea, USA	1.5	0.3	7.3	ND	ND	ND	ND	ND	ND	ND	ND	ND
Reyes, California, USA	4.0	0.4	17.8	0.2	0.3	0.2	ND	ND	ND	ND	ND	ND
Hilo, USA	3.0	0.4	25.0	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Tula, American Samo	0.2	BD	15.9	ND	ND	ND	ND	ND	ND	ND	ND	ND
Sydney, Florida, USA	1.0	2.0	18.0	4.0	5.4	4.3	ND	ND	3.9	ND	ND	ND
Tudor Hill, Bermuda	1.1	0.8	18.7	0.4	0.7	0.8	ND	ND	6.0	ND	ND	ND
Mexico and Central America												
Tapanti, Costa Rica	0.8	92.4	12.6	ND	0.2	ND	ND	ND	ND	ND	ND	ND
South America												
Arauca, Columbia	0.2	ND	8.6	ND	ND	ND	ND	ND	ND	ND	ND	ND
St. Peter & St. Paul Rocks, Brazil	1.5	2.8	17.8	ND	0.3	ND	ND	ND	ND	ND	ND	ND
Indaiatuba, Brazil	1.2	2.9	9.8	0.1	0.4	ND	ND	ND	ND	ND	ND	ND
Europe												
Pallas, Finland	2.2	0.5	14.9	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Košetice, Czech Rep.	3.5	5.7	45.8	0.3	ND	ND	ND	ND	ND	ND	14.6	ND
Stórhöfði, Iceland	5.0	1.6	45.3	ND	ND	ND	ND	ND	ND	ND	ND	ND
Malin Head, Ireland	2.3	1.7	23.0	0.2	ND	ND	ND	ND	ND	ND	ND	ND
Paris , France	6.8	39.0	20.1	0.5	0.7	0.8	1.0	7.8	8.4	ND	ND	ND
Danki, Russia	4.7	2.3	17.0	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Australia												
Cape Grim, Australia	BD	BD	13.5	ND	ND	ND	ND	ND	ND	ND	ND	ND
Darwin, Australia	BD	BD	11.0	0.4	2.0	0.4	3.0	0.8	6.8	ND	ND	ND
Africa												
Kalahari, Botswana	0.8	1.4	11.6	ND	ND	ND	ND	ND	ND	ND	ND	ND
DeAar, South Africa	18.0	14.1	14.2	0.1	ND	ND	ND	ND	ND	ND	ND	ND
Asia												
Danum, Malaysia	BD	BD	8.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
Kuwait City, Kuwait	2.9	4.4	38.2	0.4	0.3	0.4	ND	ND	ND	ND	ND	ND
Manila, Philippines	1.1	0.7	22.0	0.4	0.9	0.5	ND	ND	ND	ND	ND	ND
Bukit Kototabang, Indonesia	0.3	BD	10.1	ND	ND	ND	ND	ND	ND	ND	ND	ND
Nam Co., Tibet, China	8.8	2.6	23.7	ND	ND	ND	ND	ND	ND	ND	ND	ND
MDL	0.11	0.14	0.40	0.06	0.07	0.08	0.22	0.25	0.40	0.36	0.38	1.1

Table S4 Results of randomized block design ANOVA test for the concentrations of HCHs, α–endosulfan, HCB and total chlordanes in North America, South and Europe for the year 2005 to 2008. $p < 0.05$ indicates that the levels of OCPs significantly differ among the four years sampling (2005 to 2008) or between two years (e.g. 2005 and 2006).

Year		Location	p				
From	To		α-HCH	γ-HCH	HCB	α-endosulfan	Total chlordane
		North America					
All four years			**0.03**	**0.05**	0.25	0.72	**0.02**
2005	2006		0.056	0.086	0.47	0.73	**0.02**
	2007		**0.04**	0.10	0.24	0.98	0.063
	2008		**0.04**	**0.05**	0.38	0.82	**0.04**
2006	2007		0.99	0.99	0.94	0.89	0.74
	2008		0.99	0.95	0.99	0.99	0.94
2007	2008		1.00	0.90	0.98	0.95	0.95
		South					
All four years			0.26	**0.03**	**0.004**	0.49	0.21
2005	2006		0.98	0.79	**0.02**	0.82	0.35
	2007		0.39	0.06	**0.006**	0.98	0.27
	2008		0.38	0.05	**0.01**	0.77	0.24
2006	2007		0.59	0.27	0.95	0.62	0.99
	2008		0.58	0.24	0.99	0.99	0.99
2007	2008		1.00	1.00	0.99	0.55	1.00
		Europe					
All four years			**0.00**	0.33	**0.03**	0.40	**0.04**
2005	2006		**0.002**	0.68	0.65	0.99	0.15
	2007		**0.001**	0.49	**0.04**	0.64	0.075
	2008		**0.00**	0.29	0.08	0.44	**0.03**
2006	2007		0.89	0.98	0.21	0.87	0.96
	2008		0.30	0.87	0.39	0.60	0.75
2007	2008		0.65	0.97	0.96	0.98	0.95

Table S5 Results of the regression analysis between the levels of OCPs and time (2005 to 2008) for fifteen sites. $p < 0.05$ and negative correlation (bold font) indicate that the levels of OCPs are significantly different from years 2005 to 2008 and decreasing during the sampling period.

Sites	α-HCH		γ-HCH		Total Chlordane		HCB		α-endosulfan	
	slope	p	slope	p	slope	p	slope	p	slope	p
Bratt's Lake, Canada	**negative**	**0.006**	**negative**	**0.02**	negative	0.55	positive	0.79	negative	0.53
Toronto, Canada	**negative**	**0.01**	**negative**	**0.01**	**negative**	**0.04**	negative	0.61	negative	0.34
Tudor Hill, Bermuda	**negative**	0.23	negative	0.47	negative	0.34	negative	0.22	**negative**	**0.007**
Tapanti, Costa Rica							positive	0.30	***positive***	***0.03***
Arauca, Columbia							negative	0.19	**negative**	**0.05**
Indaiatuba, Brazil	**negative**	**0.05**	negative	0.14	negative	0.14	negative	0.76	positive	0.47
Košetice, Czech Rep.	**negative**	0.13	negative	0.13	**negative**	**0.05**	negative	0.06	negative	0.59
Stórhöfði, Iceland	**negative**	0.08	**negative**	**0.04**	**negative**	**0.02**	negative	0.15	negative	0.18
Malin Head, Ireland	**negative**	**0.05**	negative	0.79	**negative**	**0.05**	positive	0.95	negative	0.74
Paris , France	**negative**	0.15	**negative**	**0.04**	negative	0.13	negative	0.09	**negative**	**0.01**
Cape Grim, Australia							**negative**	**0.05**	negative	0.70
Darwin, Australia					negative	0.11	negative	0.20	positive	0.64
Kalahari, Botswana	**negative**	**0.01**	**negative**	**0.01**			negative	0.23	positive	0.85
DeAar, South Africa	negative	0.08	negative	0.31	**negative**	**0.05**	negative	0.11	positive	0.24
Danum Valley, Malaysia							negative	0.69	negative	0.20

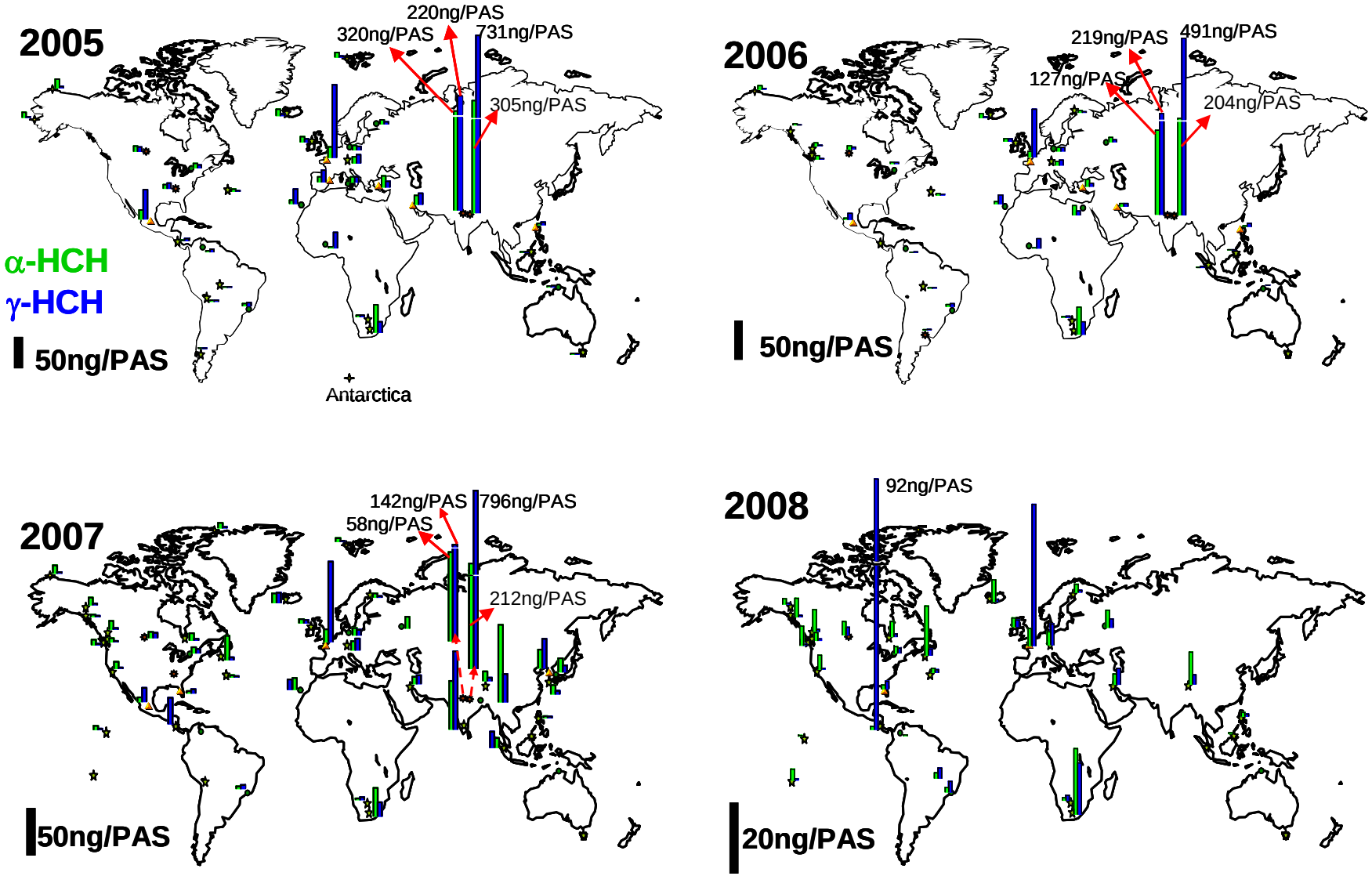

Figure S1-1 The distribution of hexachlorocyclohexanes in the global atmosphere during the first four years of GAPS.

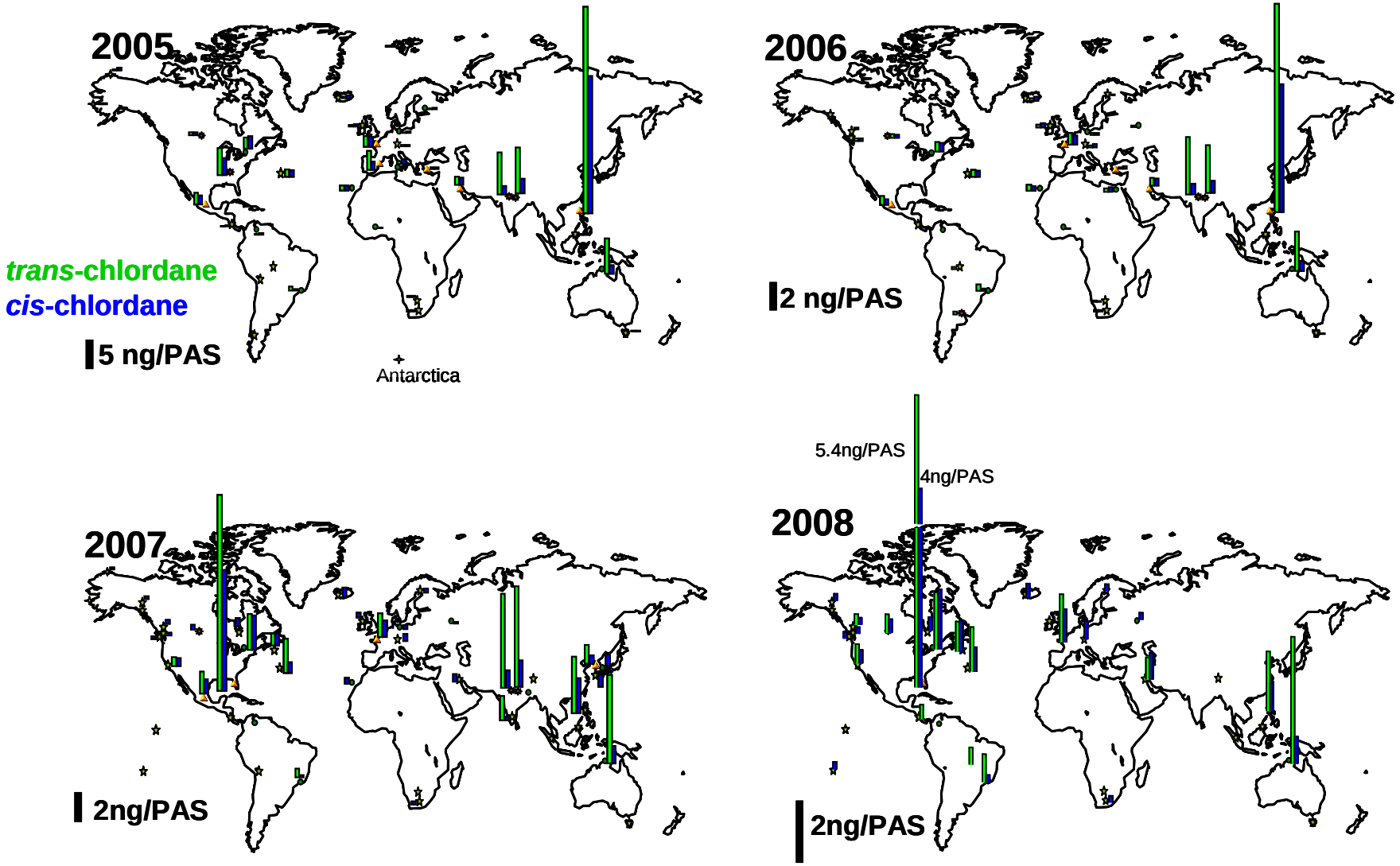

Figure S1-2 The distribution of *trans*- and *cis*-chlordane in the global atmosphere during the first four years of GAPS.

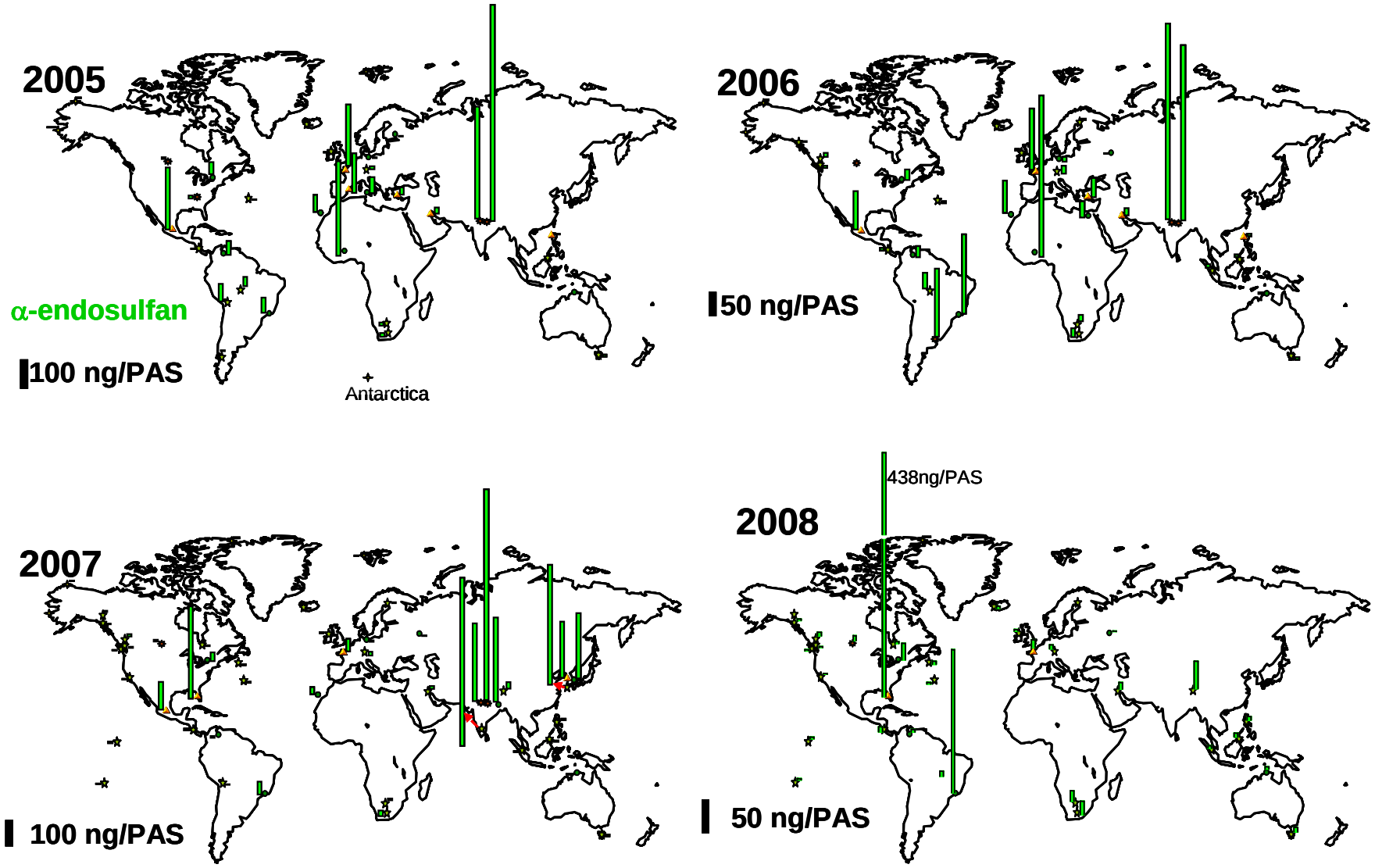

Figure S1-3 The distribution of α-endosulfan in the global atmosphere during the first four years of GAPS.

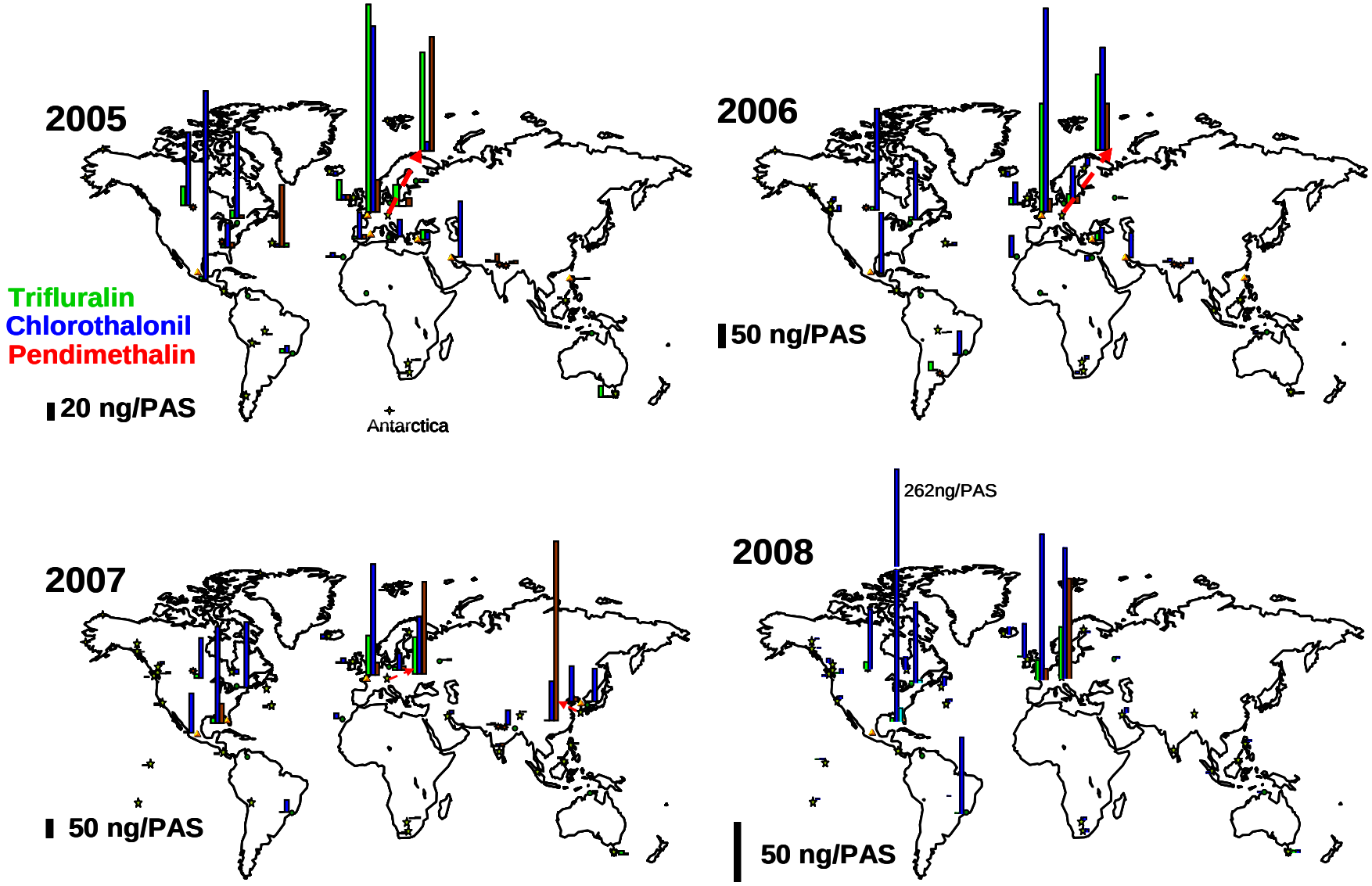

Figure S1-4 The distribution of trifluralin, chlorothalonil, and pendimethalin in the global atmosphere during the first four years of GAPS.

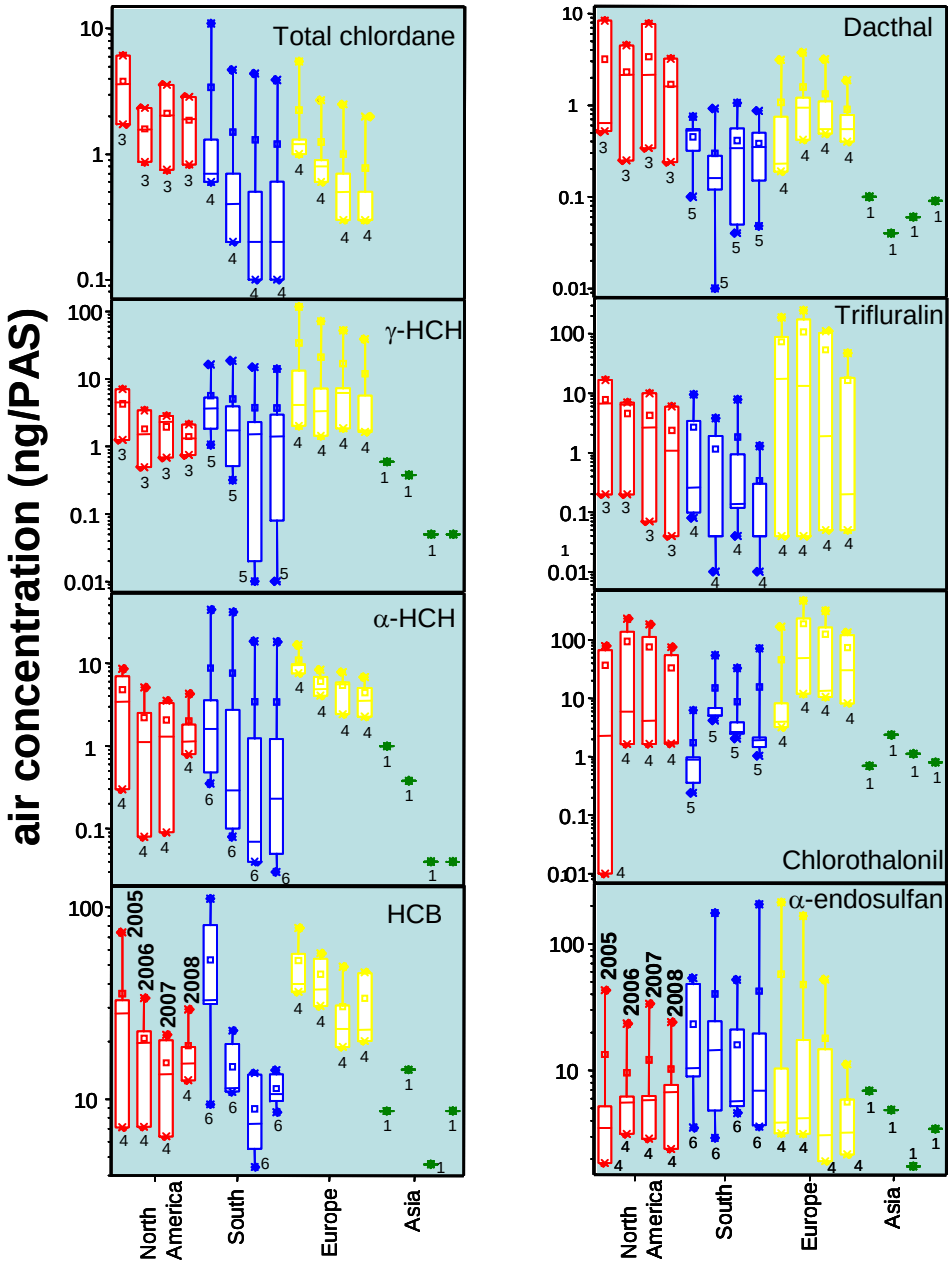

Figure S2 Box-and-whisker plot of the pesticide concentrations in passive air samplers (ng/PAS) deployed at the 15 GAPS sites that had PAS installed during all four sampling years. The number next to each box indicates the number of sampling sites. If that number decreases from year to year, it is because the levels at some sites dropped below the limit of detection. (Sites with non-detected level was assigned with random value of method of detection limit).

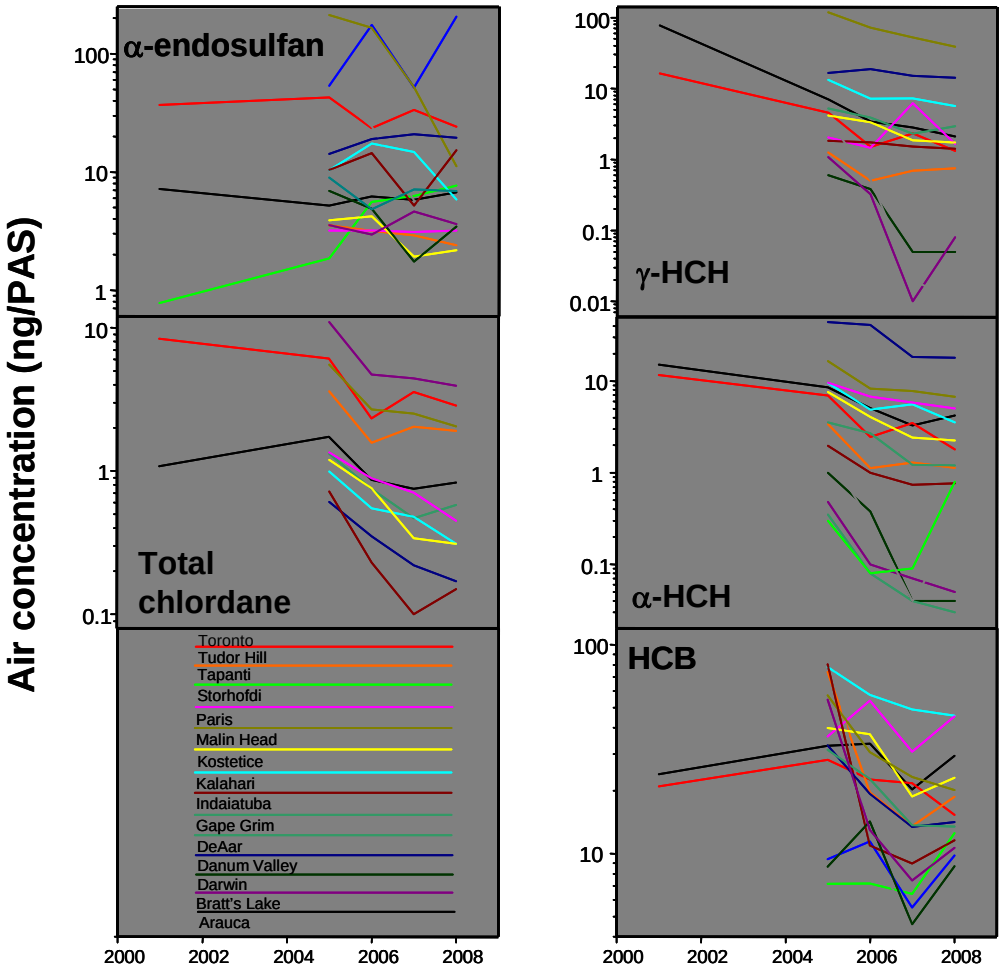

Figure S3 Time trends in pesticide concentrations at the 15 GAPS sites that had PAS installed during all four sampling years. Toronto and Bratt's Lake also had samplers deployed in 2000/1.[28,29] (Sites with non-detected level was assigned with random value of method of detection limit).

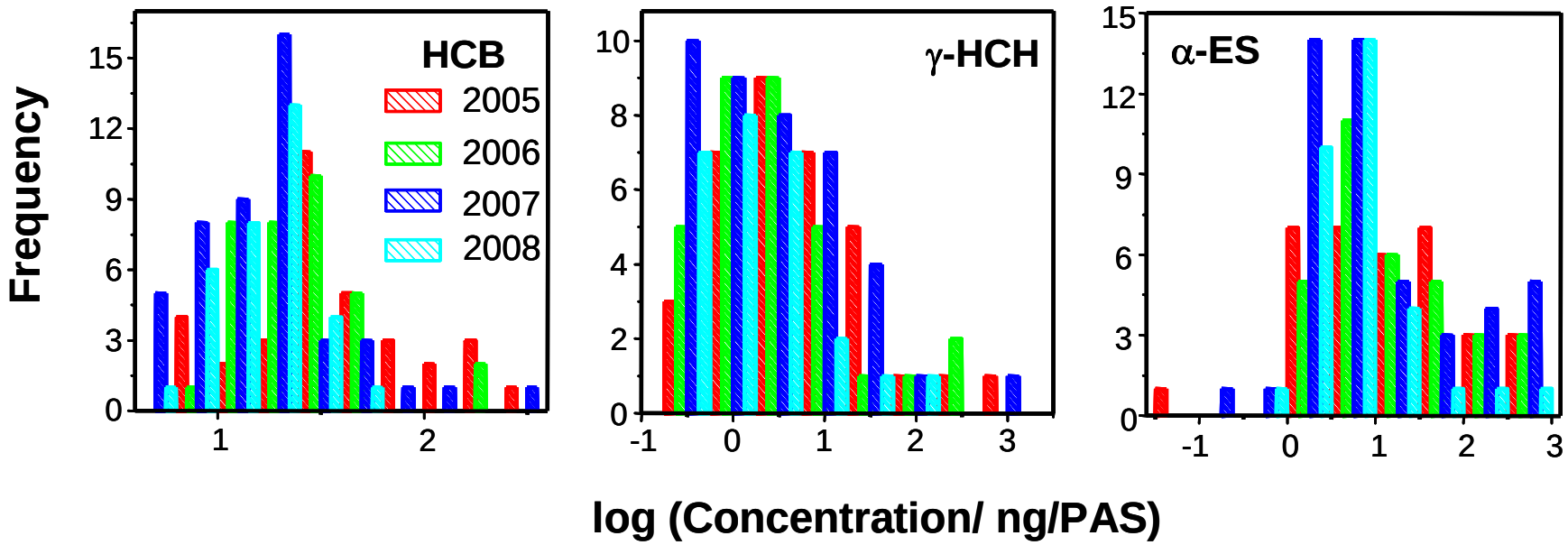

Figure S4 Histogram of logarithm of the PAS concentration for hexachlorobenzene, γ-hexachlorocyclohexane, and α-endosulfan.

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