Electronic Supplementary Information for

Cluster Analysis of Passive Air Sampling Data Based on the Relative Composition of Persistent Organic Pollutants

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SECTION 1: Chengdu-Wolong Natural Reserve, Southwestern China[6]

Duplicate XAD-PAS samples were collected at seven sites in Wolong Natural Reserve (WNR), a mountainous area at the eastern edge of the Tibetan Plateau in Western Sichuan Province, China, for six-month intervals for two and half years.[6] From April 2007 to April 2008, duplicates samples were also deployed for two consecutive six month periods (one winter and one summer period) at a site in Chengdu, a city of 11 million inhabitants located in the Sichuan basin about 80-105 km to the East of the WNR. Therefore, samples taken during this year were used for the cluster analysis. With the WNR, the sites are along a valley and up the windward slope of Balang Mountain reaching the Pass at 4485 m a.s.l. The lowest and highest sites within WNR were approximately 80 kilometers apart. The sampling duration was 182 days for all samples. There were 30 PAS samples retrieved, i.e. 15 for winter and summer periods, respectively. (see Figure S1)

The data are notated according to sampling period and site. The first capital letter (S or W) stands for summer or winter sampling period. This is followed by one or two capital letter(s) for different sites (CD for Chengdu; in the WNR (N) in the order of increasing altitudes: N1 for Gengda; N2 for Panda Center; N3 for Sandaoqiao; N4 for Dengsheng; N5 for Beimuping; N6 for 95 kilometer milestone; N7 for Pass. The last lower-case letter (a or b) indicates one of the duplicates. For example, S-N1a and S-N1b stand for summer duplicates at Gengda; W-N1a and W-N1b for winter duplicates at the same site. No duplicate was available for the highest site (S-N7 and W-N7).

Eleven POPs compounds were measured, including HCB, α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, o,*p'*-DDT, PCB28, and PCB52.

The result of the cluster analysis is displayed in Figures 1 and 2 in the main paper. As listed in Table 2 in the main paper there are five groups identified by cluster analysis based on compositional data. The samples were classified principally by season, as a winter group and three summer groups are formed. Thirteen of the fifteen winter samples are in the 'winter' group with highest HCB fraction. Nine of the fifteen summer samples are in the 'summer' group. 'Summer-urban' group consists of summer duplicates from Chengdu (S-CDa, S-CDb) with a composition high in γ-HCH, *p,p'*-DDT, *p,p'*-DDE , *o,p'*-DDT, PCB28, and PCB52. The 'summer-local contamination' group consists of summer duplicates from Sandaoqiao (S-N3a, S-N3b) with a composition high in α-HCH and β-HCH. In previous studies it was reported that the soil at Sandaoqiao was contaminated by HCHs in the past. This was also the case for the soil at the Panda Center site.[7]

There is also a 'mixed' group with four samples, two summer samples from Panda Center (S-N2a, S-N2b) and two winter samples from Pass (W-N7) and 95 kilometer milestone (W-N6a). This mixed group disappears when HCB was not included in the cluster analysis (Fig. 3 in the main text).

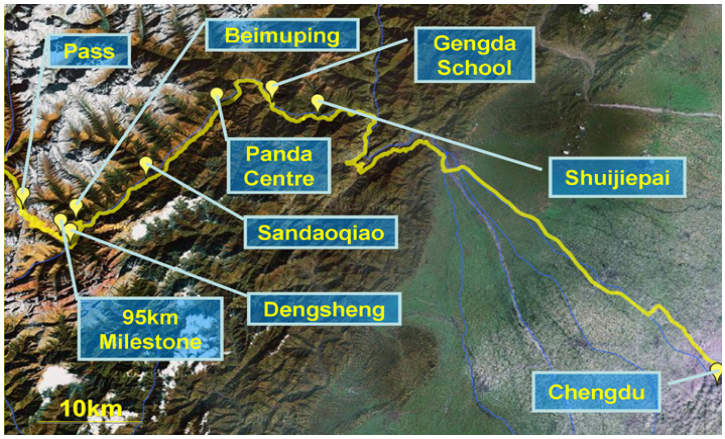

Figure S1 ***Location of sampling sites in Chengdu and Wolong Nature Reserve (WNR) (modified from ref.[6])***

SECTION 2: Botswana[5]

In this study XAD-PAS samples were collected at 15 sites, an inland area of Botswana (Figure S2). All sites have similar altitudes ranging 295 m-371 m a.s.l. Five sites (E1 through E5) were along the Eastern border of Botswana where the majority of the population resides and where pesticides are used on vegetables and crops. Ten sites (from O1 through O10) were in the Okavango Delta area (only three of those sites are shown in Figure S2), which was repeatedly sprayed with DDT and endosulfan in the past.[5] The 13 compounds measured in this work included HCB, α-HCH, γ-HCH, o,*p'*-DDE, TC, CC, TN, dieldrin, dacthal, Endo-I, Endo-II, Endo. Sulfate, and CT (chlorothalonil).

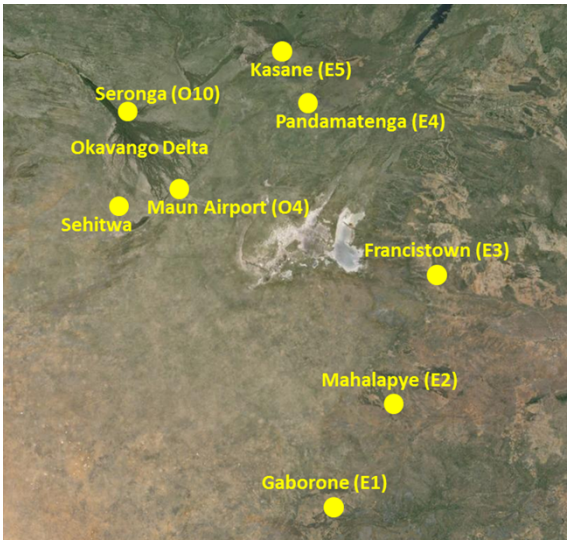

Figure S2 ***Location of selected sampling sites in a nation-wide PAS network in Botswana***

For clustering results, please see Figure 4 in the main text and Table S1. Five groups were formed. Since HCB was found very much evenly distributed nation-wide in this study, the highest HCB fraction value of 0.374 indicates the 'O-Delta' group is the cleanest. Also according to the sum of POPs, the O-Delta group had the lowest levels (Table S1). In

chemical compositions, this group was dominated mainly by HCB and endosulfan, a pesticide used in the past. This group contains 6 sites being scattered over the Okavango Delta. It implies that the Delta area shares a very uniform POPs composition and thus belongs to the same airshed, having similar contributions from both regional and long-range atmospheric transport.

There were another four sites in the Okavango Delta area that fell into another three groups due to contributions from local emission sources. Nokaneng, a village, was unique in chemical composition, having very high o,p'-DDE and chlordane contributions, and formed a group of its own ('O7-DDE' group in Table S1).

O1 and O4 had high lindane contributions and constituted the 'lindane' group, together with sites E1 and E3 (Table S1). Sites in this group were from rather different geographical locations. It is the similar POPs source profiles and, in turn, the similar POPs compositions that make them a group. O1 is near a tourist camp and O4 is at the airport. It is quite possible that lindane was used in these locales.

Table S1 Normalized compositions of the groups in the cluster analysis of data from Botswana

Group	no. of sites	Sum of POPs	HCB	α-Endo	trans-chlordane	dieldrin	lindane	o,p-DDE
E4-dieldrin	1	**79** ng/PAS	0.085	**0.519**	0.013	**0.236**	0.018	0.003
O-Delta	6	17 ng/PAS	**0.374**	0.411	0.014	0.015	0.051	0.012
EEO	3	31 ng/PAS	0.241	0.319	0.095	0.008	0.096	0.007
O7-DDE	1	51 ng/PAS	0.116	0.136	**0.181**	0.005	0.073	**0.305**
Lindane	4	73 ng/PAS	0.124	0.286	0.038	0.031	**0.354**	0.004

The same argument holds for site in the village of Seronga (O10) which formed a group with Mayalapye (E2) and Kasane (E5), two towns in the eastern part of Botswana. This group ('EEO' in Table S1) was featured by a relatively high portion of trans-chlordane. What Seronga and Kasane also have in common is that the sampling site was on the property of the police station. Maybe the police had used chlordane as termiticide.

Site E4, Pandamatenga, again in a police station, is also a single site group, having high portion of dieldrin and α-Endosulfan ('E4-dieldrin' in Table S1).

In this study it is clearly demonstrated that cluster analysis is able to separate sites that are heavily influenced by local sources from those that are not.

SECTION 3: Chile[3]

20 PAS were deployed in 2006-2007 for one year along three altitudinal transects in Chile.[3] The three transects were very far apart; the distance between the northern and southern transect was ca. 3000 kilometers. The transect in Northern Chile (18˚S) had 6 sites from Arica (N1, 48 m a.s.l.) to Lago Chungara (N6, 4400 m). The transect in Central Chile (38˚S) had 7 sites, from Lago Lleu Lleu (C1, 10 m a.s.l.) to Paso Pino Hachado (C7, 1874 m); plus a site on the campus of the University of Concepcion (C8, 33 m). The transect in Southern

Chile (44˚S) had 5 sites, from Puerto Cisnes (S1, 50 m a.s.l.) to Alto Rio Cisnes (S5, 700 m); plus a site close to the town of Coyhaique (S6, 427 m) (see Figure 6 in the main text). The 15 compounds measured in this work included HCB, α-HCH, γ-HCH, heptachlor, Hepx, TC, CC, TN, dieldrin, E-I, E-II, E-S, CT, trifluralin, and pendimethalin.

Clustering yielded four groups (Figure 5 in the main text): the first group ("polluted-1") contains only one site (C4, 75 m a.s.l.) with a composition dominated by pendimethalin and trifluralin. This is a 'hot-spot' site with an elevated total contamination level (Table S2). This site C4, Fundo El Vergel in Angol, is on a farm where pyrethroid type pesticides are produced. Located in a valley with intense agricultural activity, its exceptional composition is reasonable. The second group ("polluted-2") also has elevated contamination (Table S2) and comprises the three most "urban" sites within the network: N1 and N2 (48 m and 255 m a.s.l.) are within or close to the town of Arica, whereas C8 is in the city of Concepcion. Their composition was dominated by chlorothalonil and dieldrin.

The remaining two groups comprise many sites and are called regional groups. "Regional-1" contains 8 sites from the southern and central transects, having low overall POP levels (Table S2) and very similar compositions dominated by HCB (64 %). The sites from the central transect within this group (C1, C2, C3) were located at relatively low altitudes (10 m, 355 m, 1189 m a.s.l.) and to the west of the agricultural valley in this region of Chile. Most likely they are more influenced by "clean" air masses coming in from the South Pacific. In contrast, sites C5 to C6 are east of this valley and may thus be influenced by pesticide use there.

"Regional-2" also contains 8 sites (N3 to N6; S6, C5 to C7) with a composition dominated by HCB, ES-1 and CT. These sites were from all three transects and were located at relatively high altitudes. S6 site is close to the town of Coyhaique and therefore is not representative of the southern transect. "Regional-2" mainly reflects the northern transect, as all sites from that transect, except the polluted sites, N1 and N2, fell into this group. The other three sites, C5, C6 and C7 were from high altitudes of the Central transect (954 m, 1165 m, 1874 m a.s.l.). "Regional-2" has relatively higher levels of E-S and CT and relatively lower levels of HCB when compared to "regional-1". This could be because they are downwind of local/regional sources of ES and CT in Chile. In the North, they are downwind of Arica. In the Central part, they are downwind of the central agricultural valley. The site in Coyhaique (S6) may be influenced by some local use. Sites in "regional-2" thus have in common that they were exposed to both regional and LRAT contributions;[3] while sites in "regional-1" were less exposed to regional sources and mostly under the influence of clean Pacific Ocean air. "Regional-1" was a factor of 3 cleaner than "regional-2", when measured by the sum of the contaminants (see Table S2).

Importantly, several of the clusters included sites from more than one transect, even though they were very far apart. Sites fall into different groups not merely based on latitude, but also based on their location downwind from regional sources, which in Chile to some extent may be related to longitude and altitude, those being further East/inland/higher having potentially more regional source influence.

Table S2 Normalized compositions of the groups in the cluster analysis of data from Chile

Group	no. of sites	Sum of POPs (ng/PAS)	HCB	E-I	CT	dieldrin	pendimethalin
Polluted-1	1	100	0.068	0.016	0.201	0.003	**0.520**
Polluted-2	3	**140**	0.055	0.049	**0.675**	**0.136**	0.002
Regional-1	8	9	**0.638**	0.137	0.058	0.029	0.034
Regional-2	8	30	0.320	**0.367**	0.223	0.009	0.010

SECTION 4: North America[2,4]

Shen et al.[2,4] deployed XAD-PAS at 40 sites across North America, a continental scale PAS network (Figure S4), which included sites in Arctic Canada (3, 24, 25, 26, 27, 28, 29), the Maritime Provinces of Canada (4, 5, 6, 7, 8, 9), the Pacific Coast of Southern Canada (22, 23), the Great Lakes (1, 2, 10, 11, 12, 30, 31), the Eastern USA (32, 33, 34, 35, 36), the Canadian Prairies (13, 14, 15, 16), the Western Canadian Mountains (17, 18, 19, 20, 21), and Mexico and Central America (37, 38, 39, 40). The 16 compounds measured in this work included PeCB, HCB, α-HCH, γ-HCH, *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, Hept, Hepx, Oxy, TC, CC, TN, dieldrin, Endo-I, Endo-II.

Figure S3 Location of the sampling sites in the North American study

Eight groups are formed by clustering (Figure 7 in the main text), which are reduced to six groups by combining smaller groups into clusters called 'DDXs' and 'endosulfan' (Table S3). The first three groups in Table S3 have total POP levels two or three times higher than the other groups: The 'lindane' group contains only one site in the Canadian Prairies (15), where agricultural application of lindane led to a high relative abundance of this pesticide. The 'DDXs' group comprises three sites in Mexico and Central America, (37, 38, 39), where high DDT/DDE ratios implies recent input of DDT. The 'endosulfan' group contains 6 sites. This is a very diverse group (6, 10, 14, 21, 31, 40) and it is difficult to find any commonalities in character. It includes a place on the Atlantic island of PEI (6), a town in rural Quebec (10), a place in rural Manitoba (14), a place in rural central BC (21), a big city (31) and a relatively remote place in Costa Rica (40). The first four are rural places with agricultural activity indicating the influences of past or on-going endosulfan use in the vicinity of the sampling sites.

As shown in Figure 7 in the main text, the other three groups are relatively big, containing more sites, and their sum amount of POPs are much lower (Table S3). And Regional-1 was split into two sub-groups (Figure 7, Table S3). These groups are distinctive in POPs compositions and fairly different in POPs concentration levels.

Table S3 Normalized compositions of the groups in the cluster analysis of data from North America.

Group	No. of sites	Sum ng/PAS	HCB	HCH			DDXs	CC+TC	dieldrin	α-Endo
				α-	γ-	ratio				
lindane	1	**140**	0.172	0.108	**0.558**	0.2	0.002	0.007	0.002	0.054
DDXs	3	**174**	0.101	0.007	0.025	0.3	**0.582**	0.008	**0.106**	0.136
endosulfan	6	114	0.165	0.100	0.119	0.8	0.067	0.023	0.009	**0.418**
waterside	14	48	**0.374**	**0.296**	0.047	6.3	0.022	0.015	0.006	0.061
Regional-1-others	6	64	**0.311**	0.168	0.156	1.1	0.025	0.036	0.014	0.109
Regional-1-mountain	4	46	**0.312**	**0.213**	0.056	3.8	0.062	0.011	0.005	0.193
Regional-2	6	85	0.193	0.108	0.090	1.2	0.086	**0.100**	0.032	0.186

The sites in the 'waterside group' were located adjacent to, or not far from, the ocean or large lakes such as sites 3, 4, 5, 7, 8 in Atlantic Canada, sites 24 to 29 in Arctic Canada, sites 22 and 23 on the Pacific coast, and site 11 on Lake Superior. This group had the lowest total POP levels and a composition rich in α-HCH, probably due to re-evaporation from those water bodies.[4] Being evenly distributed continent-wide in North America, HCB accounted for the highest fractional value (37 %), indicating the low overall contamination at the sites of this group.[2] The same is true for 'regional-1' group, where HCB makes up 31 %. "Regional-1" could be split into two subgroups; one contains only Western Canadian Mountain sites (17 to 20); another contains six sites that could be further split into two small group; site 13 and 16 from the Canadian Prairies, and four other sites 2, 12, 32, 35, again waterside sites. 'Regional-2' group was featured by a higher fraction of CC and TC. It merges with 'regional-

1' group and both had similar POP compositions. The sites in 'regional-2' group were mixed with those in 'regional-1' geographically. They were all scattered in middle US and east coast areas.

The three regional groups are intriguing, especially the water-side group with so many far apart sites having highly uniform compositions. It implies that regional representative compositional fingerprints may exist.

SECTION 5: Global Atmospheric Passive Sampling (GAPS)[14]

Although four years of data for the XAD-PAS within the GAPS network have been published,[14] we used the data for years 2007 and 2008 when more sites were sampled. There were 47 sites in 2007, and 33 in 2008. The cluster analysis for 2007 only includes 32 sites that correspond with those sampled in 2008 (Table S4). In Table S4, the first two letters stand for the continent (NA for North America, CA for Central America, SA for South America, Eu for Europe, As for Asia, Af for Africa, and Au for Australia). The capital letter(s) after the hyphen indicates the nation, for example C for Canada, U for USA, CR for Costa Rica, Me for Mexico, and so forth. The number at the end is used to distinguish sites from the same nation. Site SA-Br2 had data for 2008 only.

Table S4 Site ID for Global Atmospheric Passive Sampling (GAPS)

No.	Site ID	location	No.	Site ID	Location
1	NA-C1	Bratt's Lake, Canada	18	Eu-Cz	Košetice, Czech Rep.
2	NA-C2	Toronto, Canada	19	Eu-Ic	Stórhöfði, Iceland
3	NA-C3	Lasqueti Island, Canada	20	Eu-Ir	Malin Head, Ireland
4	NA-C4	Whistler, Canada	21	Eu-Fr	Paris , France
5	NA-C5	Little Fox Lake, Canada	22	Eu-Fi	Pallas, Finland
6	NA-C6	Sable Island, Canada	23	Eu-Ru	Danki, Russia
7	NA-C7	Fraserdale, Canada	24	Au-1	Cape Grim, Australia
8	NA-C8	Ucluelet, Canada	25	Au-2	Darwin, Australia
9	NA-U1	Dyea, USA	26	Af-Bo	Kalahari, Botswana
10	NA-U2	Point Reyes, Calif., USA	27	Af-SA	DeAar, South Africa
11	NA-U3	Hilo, Hawaii, USA	28	As-Ma	Danum Valley, Malaysia
12	NA-U4	Sydney, Florida, USA	29	As-Ku	Kuwait City, Kuwait
13	NA-Be	Tudor Hill, Bermuda	30	As-Ph	Manila, The Philippines
14	NA-Sa	Tula, American Samoa	31	As-Ind	Bukit Kototabang, Indonesia
15	CA-CR	Tapanti, Costa Rica	32	As-Ch	Nam Co, Tibet, China
16	SA-Co	Arauca, Columbia	33	SA-Br2	Peter & Paul Rocks, Brazil
17	SA-Br1	Indaiatuba, Brazil			

There are 11 compounds quantified in both 2007 and 2008: chlorothalonil, dacthal, three endosulfans (α-, β-, and sulfate), α-HCH, γ-HCH, HCB, cis-chlordane, trans-chlordane, trans-nonachlor. Figures S4 and S5 show the results of the cluster analysis performed on the data from 2007 and 2008, respectively. The dendrogram for both GAPS year shows seven groups, three groups contain relatively clean samples, from a total of 21 (2007) or 20 (2008) sites.

The other four groups contain between 2 and 4 sites, some of them having high pollutants levels. In Tables S5 and S6 the sum of contaminants sequestered (ng/PAS) is listed along with the average group composition. This way, both absolute concentration levels and relative compositions can be evaluated at the same time. In the tables, bold numbers indicate those features that make a group different from others.

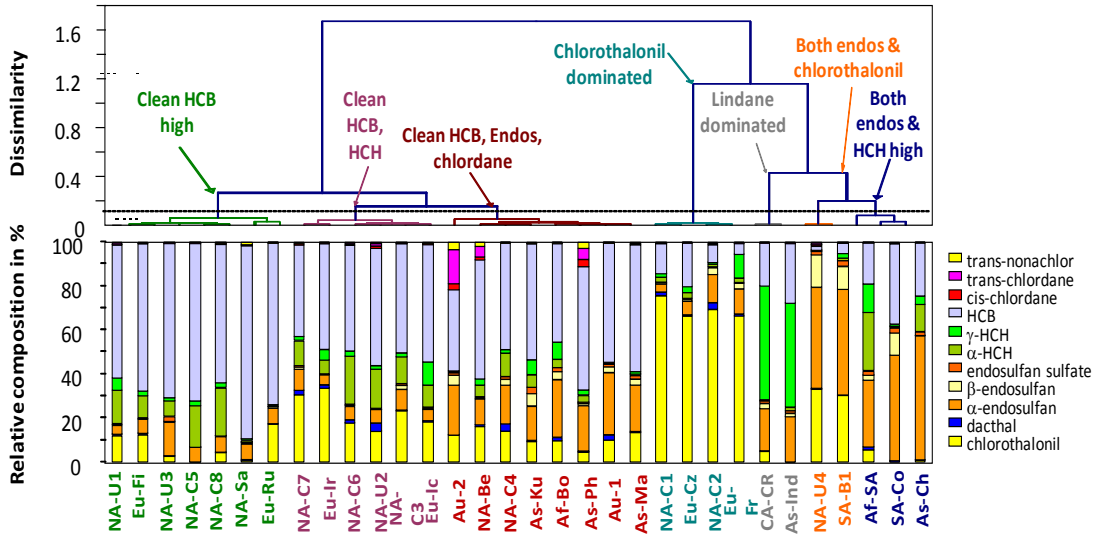

Figure S4 Dendrogram and composition plot for GAPS data from 2007.

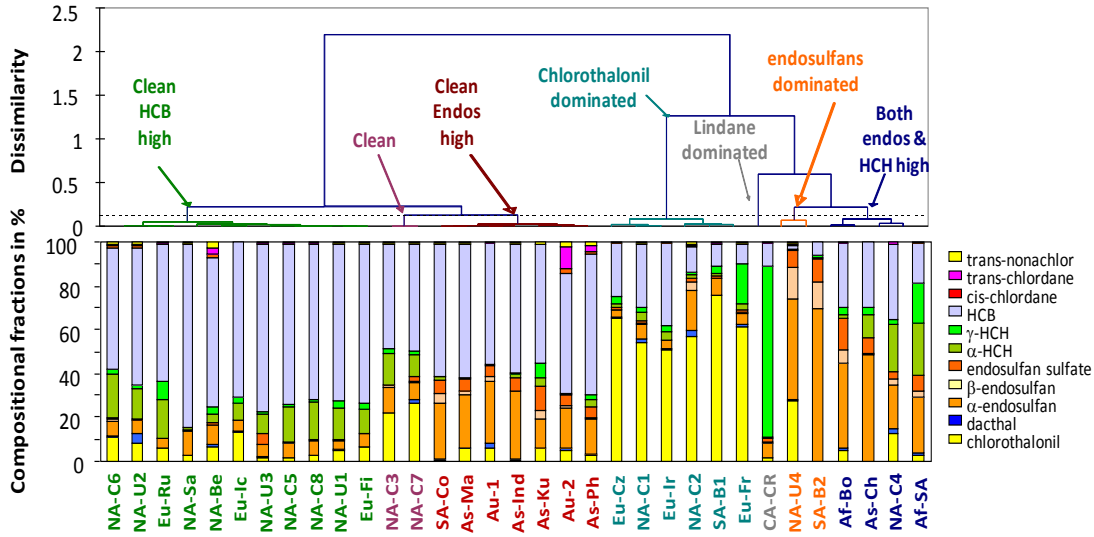

Figure S5 Dendrogram and composition plot for GAPS data of 2008

In previous studies HCB was often evenly distributed.[14] As a result, HCB accounted for high fraction values at clean sites, and vice versa. This was also the case globally, with three clean groups having low POP levels and high HCB fractions (Tables S5, S6). Those three clean groups contained 21 or 20 sites. The last two groups in Table S5 and the last group in Table S6 contained sites that also had low levels. There were thus only a small number of sites with high POP levels. Clearly, the majority of GAPS sites were indeed in clean background

regions.

Table S5 Group averages for cluster analysis based on GAPS-2007 normalized compositional data

Group	number of sites	sum of contaminants	chlorothalonil	α-endosulfan	β- endosulfan	endosulfan-SO_4	α-HCH	γ-HCH	HCB
CT-dominated	4	281 ng/PAS	**0.695**	0.084	0.018	0.003	0.019	0.042	0.118
Clean-HCB, Endos, CT	**8**	**32** ng/PAS	**0.115**	**0.204**	**0.028**	**0.013**	**0.036**	**0.028**	**0.506**
Clean-HCB high	**7**	**20** ng/PAS	**0.073**	**0.077**	**0.004**	**0.005**	**0.105**	**0.021**	**0.701**
Clean-HCB, α-HCH	**6**	**43** ng/PAS	**0.232**	**0.068**	**0.007**	**0.004**	**0.131**	**0.039**	**0.488**
Both Endos & CT	2	458 ng/PAS	**0.321**	**0.469**	0.124	0.021	0.006	0.012	0.035
Lindane-dominated	2	27 ng/PAS	0.028	0.199	0.019	0.011	0.008	**0.495**	0.234
Both endos & HCH	3	43 ng/PAS	0.025	**0.446**	0.044	0.019	**0.130**	0.056	0.268

Table S6 Group averages for cluster analysis based on GAPS-2008 normalized compositional data

Group	number of sites	sum of contaminants	chlorothalonil	α-endosulfan	β- endosulfan	endosulfan-SO_4	α-HCH	γ-HCH	HCB
Chlorothalonil-dominated	6	132 ng/PAS	0.606	0.075	0.010	0.009	0.026	0.050	0.205
Clean- HCB	**11**	**33** ng/PAS	**0.058**	**0.065**	**0.001**	**0.007**	**0.121**	**0.025**	**0.701**
Clean	**2**	**29** ng/PAS	**0.241**	**0.096**	**0.009**	**0.010**	**0.120**	**0.022**	**0.480**
Both α-HCH & endos high	4	56 ng/PAS	0.050	0.332	0.029	0.080	0.145	0.069	0.279
Endos-dominated	2	623 ng/PAS	0.139	0.579	0.132	0.093	0.003	0.006	0.040
Lindane-dominated	1	118 ng/PAS	0.014	0.065	0.008	0.015	0.007	0.782	0.107
Clean-endos	**7**	**28** ng/PAS	**0.037**	**0.226**	**0.019**	**0.062**	**0.016**	**0.015**	**0.586**

There was no clear separation of sites from different continents. No group exists that contained sites from only one continent. However, North American sites were more likely to be in the same groups as European sites, for example, in the groups 'Clean-HCB', 'Clean-HCB, HCH', and 'CT-dominated'. These groups contained no sites from other continents. The three groups containing only North American and European sites were characterized by high HCB, α-HCH, and chlorothalonil, whereas Asian sites had higher fractions for endosulfans. This is consistent with conclusions in previous work.[14] North America and Europe are in the Northern Hemisphere and the synoptic air mass connects them. It seems reasonable that they are compositionally similar.

SECTION 6: Western Canadian Mountains[1]

The 17 PAS monitoring sites were set along three altitudinal transects within relatively close proximity: Two transects west of the continental divide are situated in Mount Revelstoke and Yoho National Park, while the Observation Peak transect in Banff National Park is east of the divide (Figure S6 and also Figure 9 in the main text).

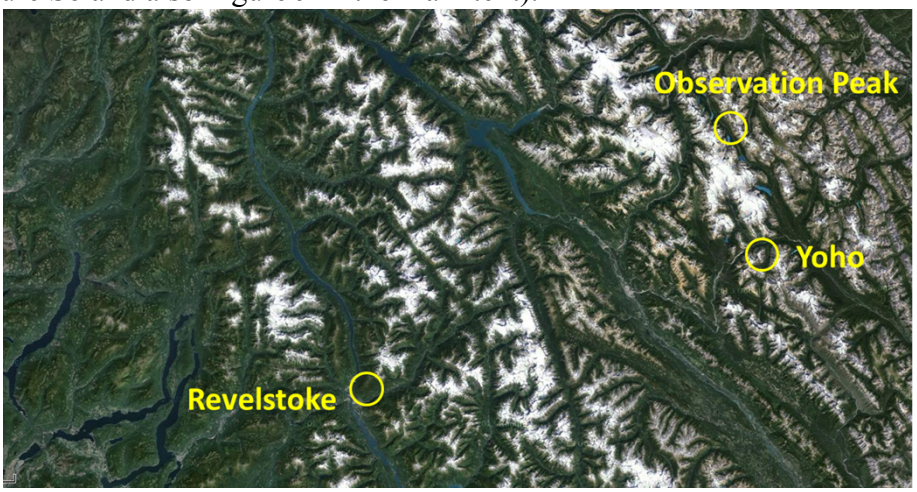

Figure S6 Location of the transects Revelstoke, Yoho and Observation Peak in Western Canada

The Revelstoke transect had six sites, R1 through R6, in order of increasing altitudes, from 570 m to 1550 m above sea level (a.s.l.). And the Yoho transect had five sites, Y1 through Y5, from 1109 m to 2280 m a.s.l. Finally, the Observation Peak transect again had six sites, O1 through O6, from 1402 m to 2611 m a.s.l..

The 22 POPs compounds measured in this work included α- and γ-hexchlorocylochexane (HCH), dieldrin, endrin, heptachlor, Hepx (heptachlor epoxide), TC (trans-chlordane), CC (cis-chlordane), TN (trans-nonchlor), E-I (endosulfan-I), E-I (endosulfan-II), E-S (endosulfan sulfate), hexachlorobenzene (HCB), CT (chlorothalonil), Dacthal, o,p-DDT, p,p'-DDT, o,p'-DDE, p,p'-DDE, o,p'-DDD, p,p'-DDD, and PCNB (pentachloronitrobenzene).

The result of the cluster analysis on the compositional data is shown in Figure 8 in the main text. The R and Y group each are made up of three sites at relatively low altitude on the Revelstoke (R1, R3, R4) and Yoho (Y1, Y2, Y3) transect, respectively. However, the O-plus group comprises 11 sites, including all six sites of the Observation Peak transect and four high-altitude sites from the Revelstoke and Yoho transects (R5, R6, Y4, Y5). R2 site with low altitude is also in this group and regarded as an outlier. Clearly, site elevation plays a role: the R- and Y groups comprise low-altitude sites, while those in the O-plus group are almost

all at high altitude. The three groups had more or less the same absolute level of POPs, as the average sum of POPs sequestered were 33.6, 36.7, 29.4 ng/PAS for the O-plus, Y- and R-group, respectively. The overall average was 34.0 ng/PAS. In terms of relative composition, the R-group had more DDT-related compounds, TC, and CT; the Y-group had more dieldrin and DDTs and their degradation products; while the O-plus group had more HCB and α-HCH and few DDT-related contaminants (see Table S7). HCB and α-HCH are both POPs with a high potential for long-range atmospheric transport, whereas DDT and its degradation products have a much lower propensity for transport in the atmosphere.[2]

Table S7 Average normalized composition of the groups in the cluster analysis of data from the Western Canadian Mountains

group	HCB	α-HCH	γ-HCH	α/γ ratio	Chlorothalonil	dieldrin	DDT	DDE+DDD
R	0.174	0.086	0.024	3.6	**0.101**	0.009	**0.203**	**0.117**
Y	0.268	0.111	0.030	3.7	0.015	**0.044**	**0.096**	**0.195**
O plus	**0.406**	**0.133**	0.029	**4.6**	0.025	0.026	0.023	0.079

In summary, the importance of site elevations was clearly revealed and two kinds of atmospheric transport, regional (valley) and synoptic flow (long-range), could be distinguished.

SECTION 7: Tianjin and Changdao Islands, Northern China[8,9,10]

Tianjin used to be a major area of organochlorine pesticides (OCPs) manufacture in China. Large scale OCPs factories were located in industrial districts of Tanggu and Hangu. Recent studies reported high concentrations of HCHs, DDTs, and HCB.[11,12,13] Under strong influence of the Asian monsoon, Tianjin could be a major source of OCPs for the Asian-Pacific region. Changdao consists of several small islands in the Bohai Bay, about 400 kilometers to the east of Tianjin, right on the pathway of the monsoon from continental Asia towards North Pacific Ocean. With low economic activities Changdao has been an ideal location to study regional phenomena. In this study in Tianjin and Changdao Islands, PAS samples were retrieved at three months interval from March 2007 to May 2008. It is therefore possible to observe seasonal variations and site differences[8,9,10].

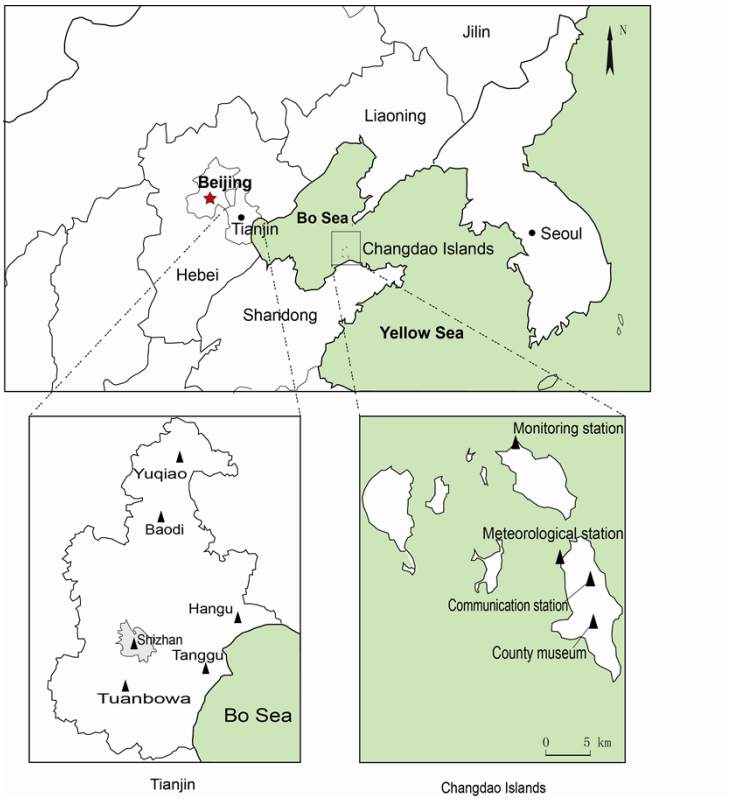

Figure S7 ***Location of six sampling sites in Tianjin and four sampling sites on the Changdao Islands***

PAS samplers were deployed synchronously in six sites in Tianjin and four sites in Changdao (Figure S7). In Tianjin the six sampling sites included the industrial sites Tanggu (T1) and Hangu (T2), the urban site Shizhan (T3), the suburban site Tuanbowa (T4), and the rural sites Baodi (T5) and Yuqiao (T6). On the Changdao Islands the four sites included the monitoring station (C1), the meteorological station (C2), the communication station (C3), and the county museum (C4). The capital letters in parenthesis will be used for site (sample) notation. T stands for Tianjin, and C for Changdao. The number starts from industrial sites through urban, suburb and rural sites for Tianjin area and from northwestern corner to southeastern end for the Changdao Island. The sampling campaign lasted for 15 months, so seasonal samples were collected for five periods. The 50 sets of compositional data are notated according to sampling period and site. The first capital letter (S, U, A, or W) stands for spring, summer, autumn and winter seasons, respectively. Since two spring periods were involved, S1 and S2 had to be used for spring 2007 and 2008, respectively. The following capital letter and number indicating different sites; the first letter stands for Tianjin (T) and Changdao (C); the number designates each sampling site. For example, S1-T1 and S2-T1 stand for two spring samples from Tanggu in Tianjin, and U-C1 and A-C1 are summer and autumn samples from the monitoring station site on the Changdao Islands.

Eleven POPs compounds were measured, including HCB, α-HCH, β-HCH, γ-HCH, δ-HCH, *p,p'*-DDT, *p,p'*-DDE, *p,p'*-DDD, o,*p'*-DDT, PCB28, and PCB52. The result of the cluster analysis is displayed in Figure 4 and Table 3 in the main text. Associated discussions are also in the main paper.

SECTION 8: Synthetic data set for three regional studies[1,3,5]

The synthetic data set combines data from three regional PAS network studies (Western Canadian Mountains, Chile, Botswana).[1,3,5] For details we refer to the first, second, and fourth section of the ESI. Since many PAS samples were involved, the sample notation needs some modification. Samples from the Observation Peak transect in Canada are now referred to with the letter P, from P1 to P6. The letter O is therefore exclusively used for samples from the Okavango Delta in Botswana. The 10 compounds commonly measured in the three studies were used in this cluster analysis. They were HCB, α-HCH, γ-HCH, TC, TN, dieldrin, Endo-I, Endo-II, Endo. Sulfate, and CT (chlorothalonil).

The dendrogram shows six groups (Fig. 11 in the main text). The 'Canadian mountains' group contains sixteen of the seventeen samples from Western Canada. The only non-Canadian site in this group is S5 from southern Chile. This group has the highest α-HCH fraction (Table S8). The Canadian site R1 seems to be an outlier, as it appears in a group together with four sites from Chile.

Table S8 Group averages for cluster analysis based on synthetic dataset from three regional studies based on 10 compounds

Group	N	HCB	α-HCH	γ-HCH	Die	TC	TN	α-end	β-end	end-S	CT
Botswana – polluted	4	0.129	0.012	**0.364**	0.031	0.040	0.016	0.294	**0.047**	0.008	0.059
Botswana – clean	15	0.309	0.015	0.058	0.027	0.047	0.015	**0.410**	0.028	0.011	0.079
Chile - polluted	4	0.097	0.005	0.050	**0.105**	0.010	0.001	0.050	0.008	0.001	**0.674**
Chile	5	0.301	0.023	0.040	0.009	0.016	0.002	0.276	0.017	0.007	0.309
Chile – clean	7	**0.684**	0.009	0.056	0.032	0.003	0.006	0.133	0.004	0.003	0.069
Canadian mountains	17	0.450	**0.151**	0.038	0.035	0.038	0.002	0.223	0.018	0.010	0.034

N: number of sites; Die: dieldrin; TC: trans-chlordane; TN: trans-nonachlor; α-end: α-endosulfan; β-end: β-endosulfan; end-S: endosulfan-SO_4; CT: chlorothalonil

The fifteen samples from Botswana fall into two groups. The 'Botswana-clean' group is big, containing 11 sites from Botswana as well as 4 from Chile. It is relatively clean, with a composition dominated by endosulfans; while the 'Botswana-polluted' group contains 4 sites from Botswana, having the highest γ-HCH fractions, likely due to the recent input of lindane.

Fifteen of the twenty samples from Chilean mountains can be found in three groups ('Chile-polluted', 'Chile-clean', 'Chile') with the remaining five samples falling into other groups. Since the sites in Chile are from three transects in northern, central, and southern Chile, that are very far away from each other, this more complicated situation is not unexpected. The 'Chile-clean' group only comprises Chilean sites (7 sites from the southern and central transects) and is the cleanest group with a composition with the highest HCB fraction. The 'Chile-polluted' group, comprised of four sites from Chile, is a polluted group with a composition with high fractions of dieldrin and chlorothanonil. In the groups 'Botswana-clean', 'Chile', and 'Canadian mountains', Chilean sites are mixed with sites from other studies.

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