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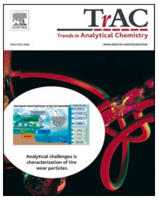

Persistent organic pollutants in air across the globe using a comparative passive air sampling method

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ABSTRACT

For the global monitoring plan (GMP) established under the Stockholm Convention on Persistent Organic Pollutants (POPs), passive air samplers (PAS) equipped with polyurethane foam disks (PUFs) were recommended for generating comparative results on the presence of POPs in countries. One representative sampling location was designed in each of the 42 countries and the PAS/PUFs exposed for three months to collect POPs during 2017–2019. The PUFs were analyzed in three laboratories according to their chemical nature and concentrations reported *per* PUF and 90 ± 3 days of exposure. In total, 381 PUFs were analyzed for organochlorine pesticides (OCPs), industrial chlorinated POPs, dioxin-like POPs (dl-POPs), brominated flame retardants (BFRs), and perfluoroalkane substances (PFAS). Concentrations typically varied by a factor of up to three for a POP and often within the same geographic region or other denominator such as similar latitudes or altitudes. Maximum values were found for DDT (895 ng/PUF) in Africa, but the highest medium value was in the Latin American countries. Also, polychlorinated biphenyls (PCB) and hexachlorobenzene (HCB) were most abundant across all parameters assessed. Noteworthy were high values in single countries, such as DDT in the Solomon Islands, toxaphene in Mexico, β-HCH and lindane in Tanzania, hexachlorobutadiene and hexabromocyclododecone in Mongolia, decaBDE and PFOS in Zambia. Although we had two sampling locations at very high altitudes, >2000 m, we could determine increasing POPs concentrations with increasing altitudes only for HCB but not for other POPs. A general dependency on latitudes with higher concentrations towards the north and south pole could not be found, although the most northern station in Mongolia had two of the maxima and elevated values for some legacy POPs.

1. Introduction

The adoption of the Stockholm Convention on Persistent Organic Pollutants (POPs) in 2001 has triggered governments, researchers, and industry to have a closer look into the sources, occurrence, fate but also toxicology and other impacts of these chemicals [1–3]. The Stockholm Convention, in 2023 has 186 Parties, *i.e.*, countries legally bound to implement the provisions of the Convention [4]. The Stockholm Convention initially started with twelve POPs but since 2009, new POPs have been listed so that today, the Annexes of the Convention include 34 (groups of) chemicals [5–29]. It must be noted that not all Parties, but most of them, have ratified these additions. Further, the provisions for the latest three POPs listed, methoxychlor, Dechlorane plus, and UV-328 [30–32], will not enter into force before August 2024. In its Articles 11 on 'research, development and monitoring', 12 on 'technical assistance', and specifically Article 16 on 'effectiveness evaluation', a global monitoring plan (GMP), was established to provide the Conference of the Parties of the Convention 'with comparable monitoring data on the presence of the chemicals listed in Annexes A, B and C as well as their regional and global environmental transport' [33]. In a section on modelling, the global monitoring reports provide context on important processes, including long-range or sources of POPs to assist interpretation of levels and trends of POPs in air and how they may impact the effectiveness of control measures. One of the important environmental processes is 'global fractionation' or 'cold condensation' as postulated by Wania and Mackay [34]. They explain often surprisingly high concentrations in Arctic regions by the temperature-dependent partitioning of certain organic chemicals with relatively low volatility become

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latitudinally fractionated, *i.e,* they are ‘condensing’ at different ambient temperatures dependent on their volatility. The approach is used for trend analysis and risk assessment [34–37] in many programmes such as the Arctic Monitoring and Assessment Programme (AMAP) [38,39].

The Convention also provides financial and technical support to developing countries and countries with economies in transition to implement the Convention [3]. For the GMP, a guidance document was first developed in 2007 and updated as new POPs were listed, core matrices to be sampled and assessed were added or regional and global arrangements have changed. The latest version is of 2021 [40].

Passive air samplers are widely used to monitor POPs concentrations in air. The guidance document of the GMP under the Stockholm Convention recommends various sorbents to capture the pollutants: (i) polyurethane foam disks (PUFs) were primarily recommended since they collect both, gas-phase and particle-phase POPs (section 4.1.2 in Ref. [40]), (ii) XAD and polyethylene (PE) films mainly target gas-phase chemicals. A paper by Wania and Shunthirasingham describe the strengths and limitations as to the applicability and reliability of the various types of passive air sampling [41].

Regional projects to support the GMP component under the Stockholm Convention in developing country regions are largely financed by the interim financial mechanism of the Convention, the Global Environment Facility (GEF; www-thegef.org) and were coordinated by the United Nations Environment Programme (UNEP) since 2005. The latest round, including 42 countries in three regions of the United Nations (UN) started in 2016 and ended in 2021 [42–45]. One of the core matrices under the GMP is ambient air to study presence, fate, and transport but also changes with time and geographies of POPs. The GMP recommends the use of passive air samplers (PAS) equipped with polyurethane foam (PUF) disks as a simple and cost-effective tool to measure and assess atmospheric concentrations of POPs [40]. In order to obtain comparable data, 42 participating countries were provided with up to 12 passive air samplers, cleaned and pre-conditioned polyurethane foam disk (PUFs) to allow for quarterly sampling at one site in the respective country. The PUFs were exposed for two years and changed every three months between 2017 and 2019. The projects addressed 26 of the 34 POPs listed in either Annex A, B, or C of the Convention, whereby polychlorinated biphenyls (PCB) were included twice, namely as the sum of six indicator PCB (PCB$_6$; *i.e.*, congeners PCB 28, PCB 52, PCB 101, PCB 153, PCB 180) and as dioxin-like PCB (12 dl-PCB). Not included were chlordecone [7], polychlorinated naphthalenes [16], pentachlorophenol (PCP) [18], short-chain chlorinated paraffins (SCCP) [21], and dicofol [23] as well as methoxychlor [30], Dechlorane plus [31], and UV-328 [32].

Results of the projects have been published for organochlorine pesticides (OCPs), industrial chlorinated POPs (indPOPs), and brominated flame retardants (BFRs) [46], dioxin-like POPs (dl-POPs) [47], and perfluoroalkyl substances (PFAS) [48]. At the Dioxin2022 symposium in New Orleans, U.S.A., we presented a spatial assessment for dl-POPs-based toxic equivalents [49]. In addition, the temporal and spatial changes of POPs in PAS/PUFs were published [50].

Here we assess the amounts of 28 POPs measured in 381 ambient air samples on a comparative basis for national and regional occurrence, *i.e.,* within a UN region but also for geographic location such as latitude or altitude. The objective is to test the hypothesis that through long-range transport POPs concentrations increase towards the poles, with increasing altitudes or accordingly with declining temperatures. Further, it is attempted to identify common pattern as to the global distribution of POPs, and if possible, give recommendations for national action or at least future monitoring programmes.

2. Materials and methods

2.1. Sampling locations and identity of samples

The geographical location of the 42 sampling sites is provided in Fig. 1. In the text, tables and figures the country names are referred to as ISO3 alpha code and used to identify the samples. The physical address as well as geographic latitude and altitude are provided in the Supplementary information as Table S1 The sampling locations were maintained throughout the project and were identical with the sites from a previous GMP1 project that was implemented in 2010/2011 [51,52]. There were 15 countries and sampling locations in Africa, 7 in Asia, 9 in the Pacific Islands countries (PAC), and 11 in GRULAC (Group of Latin America and the Caribbean). Most sampling sites, 24 or 57 %, were located on the northern hemisphere contributing 222 samples. Further characteristics include that 16 (or 38 %) of the sampling locations were around the equator at ±10° N or S latitude; thus, had tropical climate conditions. The most northern location was in Mongolia at 47° 55′ N and the most southern in Chile at 36° 39′ S. 62 % (26) of the sampling locations were at low altitudes (<200 m). With Ethiopia in Africa and Ecuador in GRULAC, there were also two locations at very high altitudes (>2000 m).

The preparation of the PUFs and the setup of the PAS/PUFs followed established procedures [53]. Most importantly, PUFs were pre-cleaned with water and acetone and then, preconditioned with dichloromethane to capture OCPs and BFRs, toluene to capture dl-POPs (and BFR in GRULAC), and methanol for PFAS. The flow of the PUFs (samples) is as follows and summarized in Table 1. Six PAS were equipped with solvent pretreated PUFs, which were exchanged every three months. After exposure, PUFs were sent to the analytical laboratory for analysis of the POPs. The number of scheduled samples *per* year was four and one

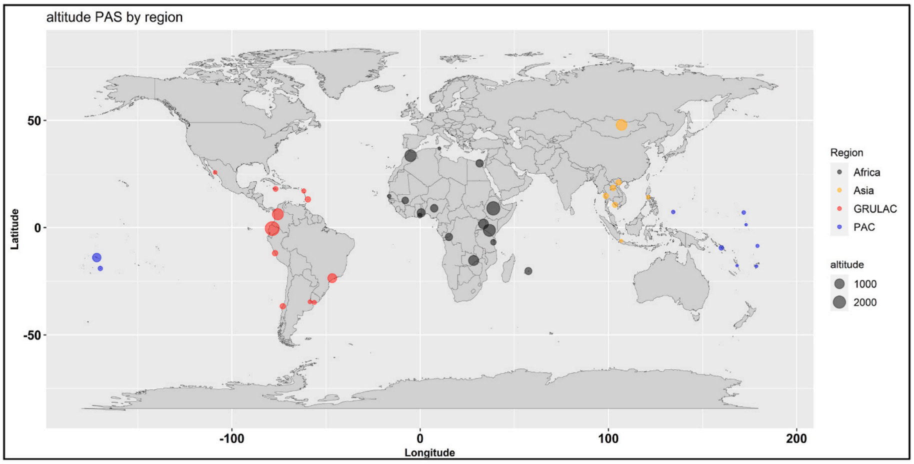


Fig. 1. Location of the 42 sampling sites, colored according to the region. The size of the dots indicates the altitude of the site.

Table 1
Flow of PUFs from exposure to chemical analysis.

PAS	PUF and pre-treatment	POP group and POP analytes	No. analyses per year and lab
PAS 1	I, II, III, IV Dichloromethane	**Initial OCPs**: aldrin, endrin, dieldrin, chlordane, DDT, heptachlor, mirex, endosulfan, toxaphene **Newly listed OCPs**: endosulfan, α-HCH, β-HCH, lindane, **indPOPs**: HCB, pentachlorobenzene, hexachlorobutadiene	4 (VU University or CSIC) 1 (toxaphene in GRULAC by CSIC)
PAS 3	I, II, III, IV Dichloromethane	**indPOPs**: 6 indicator PCB	4 (VU University or CSIC)
PAS 5	I, II, III, IV Toluene	**dl-POPs** – annual sample: 17 PCDD/PCDF, 12 dl-PCB	1 (CSIC)
PAS 7	I, II, III, IV Toluene	**dl-POPs** quarterly samples: 17 PCDD/PCDF, 12 dl-PCB	4 (CSIC)
PAS 9	I, II, III, IV Dichloromethane (Africa, Asia, PAC) Toluene (GRULAC)	**BFRs**: 8 PBDE, HBCD, PBB 153	4 (VU University or CSIC)
PAS 11	I, II, III, IV Methanol	**PFAS**: PFOS, PFOA, PFHxS, FOSA, NMeFOSA, NEtFOSA, NMeFOSE, NEtFOSE	4 + 1 annual sample (Örebro University)

for the dl-POPs. In addition, in the PFAS laboratory, all PUFs from the same year were combined and analyzed in one (annual) sample for PFAS.

Table 2 summarizes the samples obtained and assessed: N indicates northern and S southern hemisphere; the numbers thereafter indicate the latitude in degree; the number after letter A indicate a range of altitudes in meter (m). In total, 381 samples were assessed with the following regional distribution: 149 samples were collected in Africa, 104 in GRULAC and 64 each in Asia and the Pacific Islands countries. In the year 2018, 194 samples were collected, 139 in 2017, and only 48 in 2019. The vast majority of the samples (229) were collected at sampling locations with lower altitudes, between sea level and 249 m; on the other hand, there were 20 samples from altitudes >2000 m (Ethiopia and Ecuador).

2.2. Chemical analyses

After exposure, PUFs were shipped with express mail to the laboratories abroad at VU University for analysis of OCPs, indPOPs, BFRs, at CSIC for dl-POPs and OCPs, indPOPs, BFRs, or at Örebro University for PFAS. Table S2 provides an overview on the POPs, the reference for their listing, the recommended substances to be analyzed according to chapter 2 of the GMP guidance documents [40], and the abbreviation used in this paper (referred to as designation). Chemical analysis including instrumentation, procedures, and quality control/quality assurance has been described elsewhere [46–48].

The procedure for the chemical analysis of OCPs and indPOPs (PCB_6, HCB, PeCBz, HCBD) has been published [46,50] and followed the standard operational procedure as established under the UNEP/GEF projects [54]. Samples from Africa, Asia, and PAC were analyzed by the Vrije Universiteit Amsterdam, the Netherlands and samples from GRULAC by CSIC, Barcelona, Spain. In brief chemical analysis used gas chromatography with capillary columns coupled to mass selective detectors (GC/MS or GC/MSMS) or – in rare cases – to electron capture detector (ECD).

Brominated flame retardants (BFRs) were analyzed by the validated in-house methods at VU and CSIC. Instrumentation for PBDE, PBDE 209 and PBB 153 were GC-MS/MS or GC/HRMS but HBCD isomers were separated on a LC column and then determined using LC-MS/MS (all samples analyzed at VU). Details are contained in Ref. [46].

Dioxin-like POPs were analyzed in the laboratory of CSIC in Barcelona, Spain. Dl-POPs comprised 7 congeners of PCDD, 10 PCDF, and 12 dioxin-like PCB (dl-PCB). In this paper, the amounts are reported as mass concentrations *per* PUF in ng/PUF since we consider fate parameters, which cannot be determined for toxic equivalents. Extraction, clean-up, separation, identification and quantification of the dl-POPs was achieved using high resolution gas chromatographic columns (HRGC) coupled to sector-field high-resolution mass spectrometers (HRMS). The

Table 2
Number of samples analyzed by region, latitude, altitude or hemisphere Latitude in degrees north or south, Eq = equator including ±10° Altitude as A_ followed by elevation in meter.

Region	Africa	Asia	PAC	GRULAC	Overall
No of samples	149	64	64	104	381
			Sampling year		
Y2017	63	10	17	49	139
Y2018	70	36	35	53	194
Y2019	16	18	12	2	48
			Latitude		
N_23+	29	10	0	10	49
N_10-23	21	44	0	29	94
Eq	79	10	35	20	144
S_10-23	20	0	29	5	54
S_23+	0	0	0	40	40
			Altitude		
A_0-49	21	17	42	39	119
A_50-249	18	37	20	35	110
A_250-999	62	0	2	10	74
A_1000-1999	38	10	0	10	58
A_2000+	10	0	0	10	20
			Hemisphere		
Northern	99	54	20	49	222
Southern	50	10	44	55	159

analytical procedures followed general principles [55–57] and specifically [47].

PFAS were analyzed by MTM Örebro University. The method has been described in detail by Camoiras Gonzalez et al. [48]. In brief, PUFs were Soxhlet-extracted using a mixture of methyl *tert*-butyl ether (MTBE) and methanol (60:40 v/v), the extracts loaded to SPE-WAX cartridges and if necessary, ENVI-Carb SPE tubes (250 mg for the individual and 1 g for the combined), which were stacked up under the SPE-WAX cartridge for elution (new ENVI-Carb SPE tube for each fraction). Elution was done first using methanol (for neutral PFAS), and second by 0.1 % ammonium hydroxide in methanol (for ionic PFAS). Fractions were kept separately and concentrated by using RapidVap to 250 μL and 100 μL, respectively. To the second fraction, 150 μL of 2 mM ammonium acetate in water and 5 μL of injection standard solution (200 pg/μL) were added. All sample extracts were analyzed with a liquid chromatograph, separated on a BEH (ethylene bridged hybrid) C$_{18}$-column (1.7 μm, 2.1 mm × 100 mm; Waters Corporation Milford, USA) coupled to a tandem mass spectrometer (LC–MS/MS) with electrospray ionization (ESI) operating in negative mode.). Mobile phases used were methanol:water 70:30 (v/v) (A) and 100 % methanol (B) with 2 mM ammonium acetate in both phases [58]. Analysis was performed by a multiple reaction monitoring (MRM) method that monitored two mass transitions (parent ion/product ion) for each analyte except for MeFOSA, EtFOSA, MeFOSE, and EtFOSE for which only one product ion was monitored.

The chemical laboratories at MTM Örebro University, CSIC Barcelona, and Vrije Universiteit, Amsterdam have successfully performed in the UNEP-coordinated interlaboratory assessments for POPs in the air extract samples [59–66].

The sample IDs were set-up as follows: ISO3 (YYYY-season) whereby YYYY refers to the sampling year. The seasons covered a period of three months and were defined one to four with Roman numbers indicating the seasons for the PAS/PUFs, whereby the seasons I to IV indicate sampling dates at the end of March (I), end of June (II), end of September (III) and end of December (IV). The amounts of all POPs, including metabolites or defined congeners as well as sum parameters, are given in ng/PUF for comparative reasons and illustration. All PUF values refer to one PUF and an exposure period of three months (90 days ± 3 days).

2.3. Data handling

All data were maintained in Microsoft Office 365 Excel®; visualization, statistical evaluations, and principal component analysis (PCA) were made using R (version 4.2.2) and R packages with R-Studio (2022.12.0 Build 353, 2009–2022 Posit Software, PBC).

Following a normality test using histogram and density tests, the samples did not show normal distribution. Non-parametric testing was performed using the Kruskal–Wallis H test to determine if there are statistically significant differences between the independent variables and dependent variables. Post-hoc analysis was performed using the pairwise Wilcoxon test. Adjustment of the p-value was made using the Benjamini–Hochberg method. Significance level was set to $p = 0.05$. Principal component analysis (PCA) was used to extract important information and express this information as a set of few new variables, the principal components. These new variables correspond to a linear combination of the originals. The variation along the principal component is maximal.

In the text and tables, the values for the limit of detection (LOD) or limit of quantification (LOQ) are indicated by "<". For statistical operations and since the chemical analysis was very sensitive and selective, values below the LOQ or LOD were set zero. In box whisker plots, outliers were defined as values above (or below) the interquartile range multiplied by 1.5. The interquartile range is defined as the length of the middle 50 % of data points, *i.e.*, the difference between the third or upper quartile (75 % of data points) and the first or lower quartile (25 % of data points).

3. Results and discussion

3.1. Descriptive statistics according to region, latitude, and altitude

Since not all countries achieved to have eight exposures and not all POPs were analyzed in all regions, *e.g.*, PBDE 209 and HBCD were not analyzed in GRULAC samples, the results are based on 381 samples (see Table 2) with the number of analytes shown in Table 3.

The quantitative results are summarized in the Appendix by region in Table S3, latitude in Table S4 or altitude in Table S5. Therein, cells containing the maximum value are highlighted in yellow color. It can be seen that the values ranged across several orders of magnitude, often for the same POP.

The highest overall mean and median values were found for DDT (30.7 ng/PUF; 11.8 ng/PUF, resp.), which is a factor of almost 3 between the values. Next within the median values were HCB and PCB$_6$ having 3.59 ng/PUF and 3.49 ng/PUF, resp. With respect to mean values, PCB$_6$ had 10.3 ng/PUF followed by PBDE 209 with 6.64 ng/PUF and dieldrin (6.16 ng/PUF). The mean value for HCB was 4.89 ng/PUF, corresponding only to the 8th highest value. From the above, depending on the evaluation parameter the ranking of the POPs differs. The standard deviations (SD) of the mean values, see Table S3-Table S5, often were larger than the mean value. For many POPs, the measured values were below the limit of detection or quantification; highlighted by the fact that the median value for several POPs within their categories – region, range of latitudes or altitudes – were below the LOD/LOQ, depicted as "0" in the tables. These include aldrin, endrin, toxaphene, β-HCH, PBB 153, PFHxS, and FOSA.

Sixteen of the 27 maximum values were found in African countries (Table S3), namely DDT (895 ng/PUF), heptachlor (8.2 ng/PUF), a-HCH (18.0 ng/PUF), β-HCH (104 ng/PUF), lindane (61 ng/PUF), endosulfan (140 ng/PUF), PCB$_6$ (290 ng/PUF), PBDE (16 ng/PUF), PBDE209 (130 ng/PUF), PBB153 (0.87 ng/PUF), PCDF (0.6 ng/PUF), dl-PCB (20.7 ng/PUF), PFOS (36.0 ng/PUF), PFOA (3.2 ng/PUF), PFHxS (7.9 ng/PUF), and FOSA (1.9 ng/PUF). Asian countries had four maximum values as follows: chlordane (66 ng/PUF), HCB (27 ng/PUF), HCBD (334 ng/PUF), and HBCD (76 ng/PUF) as well as had GRULAC: aldrin (2.6 ng/PUF), endrin (3.1 ng/PUF), toxaphene (4.8 ng/PUF), and PeCBz (55 ng/PUF). PAC had only two maximum values: mirex (1.9 ng/PUF) and PCDD (1.9 ng/PUF).

Table 3
Number of results for each group of POPs and by region (*HCBD and PBDE 209 not measured in GRULAC).

	Initial OCPs		New OCPs	Ind.POPs	BFRs	dl-POPs	PFAS
	Aldrin, dieldrin, endrin, chlordane, DDT, heptachlor, mirex	toxaphene	α-HCH, β-HCH, lindane, endosulfan	PCB$_6$, HCB, PeCBz, HCBD	PBDE, PBDE209, HBCD, PBB153	PCDD, PCDF, dl-PCB	PFOS, PFOA, PFHxS, FOSA
Africa	113	113	113	113	113, 111, 113, 113	89	127
Asia	49	49	49	49	49, 48, 49,49	30	46
GRULAC	83	21	83	83*	82, 0*, 79, 82	53	101
PAC	49	49	49	49	49, 48, 49, 49	23	43
Total	294	232	294	294, 211*	293, 207, 290, 293	195	317

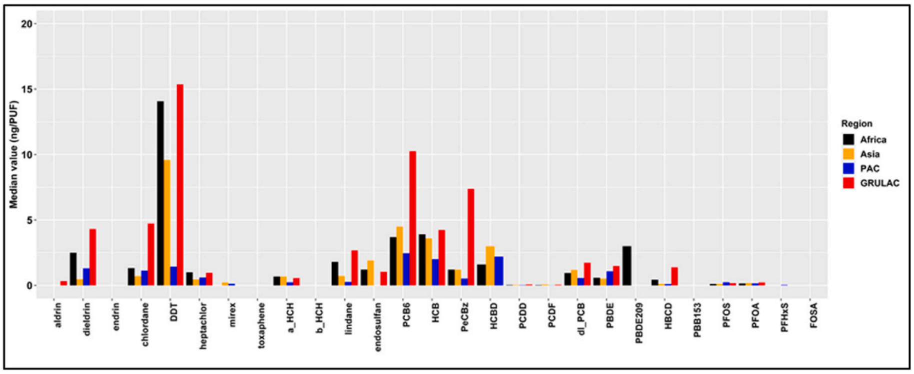


Fig. 2. Barplot displaying the median values of the POPs according to the region.

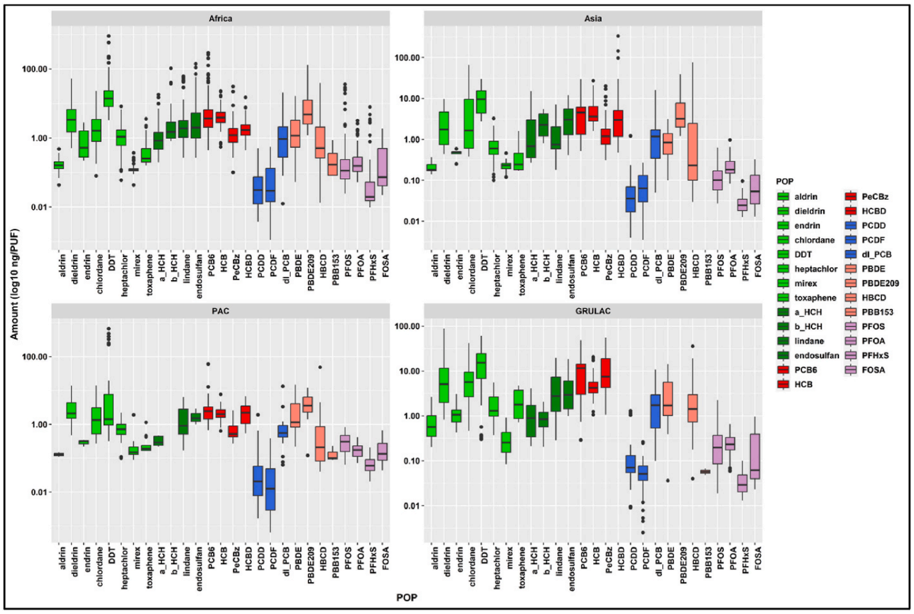


Fig. 3. Box whisker plots of POPs concentrations by region and colored as POP group (green = initial OCPs, dark green =newly listed OCPs, pink = BFRs, blue = dl-POPs, and purple = PFAS).

Because of the large SDs, we present the regional, latitudinal, and altitudinal results as median values. Overall, the highest median values are associated with the initial POPs (DDT, HCB, PCB_6, dieldrin). In Fig. 2, it can be seen that the highest median values were found in GRULAC, surprisingly including dieldrin (4.32 ng/PUF), DDT (15.4 ng/PUF), PCB_6 (10.3 ng/PUF), HCB (4.24 ng/PUF), and PeCBz (7.38 ng/PUF). In general, PAC had low median values for all POPs in general but also in comparison to the other three regions. Low in scale but higher median values than in other regions were found for PBDE (1.08 ng/PUF; note: GRULAC value higher), PFOS (0.23 ng/PUF), and PFHxS (0.03 ng/PUF).

Fig. 3 summarizes the statistical data as box whisker plots on a logarithmic scale. The dominance of the various POPs in the different regions can be seen.

From the literature it is known that semi-volatile chemicals, such as POPs, travel towards the poles or higher latitudes and accumulate. This phenomenon of “cold condensation” is heavily influenced by changes in temperature, among others by largescale climate patterns, which have the potential to increase volatilization of chemicals, enhance their transformation/degradation, and alter long-range transport pathways towards higher latitudes or altitudes. Although modelling studies found so far that the effects of climate change on contaminant transport are small compared to the expected effects of global regulatory efforts on reducing contaminants emissions such as the Stockholm Convention on POPs [39], we find such assessment necessary. The graphical representation of our results in Fig. 4, above, depicts the median values according to the latitude; accordingly, lowest values should occur at the equator (Eq) or in the tropical zones (N_10–23 and S_10–23) and the highest towards the poles. Often, the yellow bars, corresponding to the equator showed high values but the above hypothesis seems to apply for PCB_6, HCB, PeCBz, and dl-PCB.

In Fig. 4, bottom, the median values are plotted according to increasing altitudes. Following common understanding of POPs fate, highest concentrations should be found at higher altitudes. For DDT, HCB, lindane, PeCBz, PBDE 209, endosulfan, and α-HCH the largest median values were associated with the highest altitude (A_2000+). Especially for DDT, an increasing trend from low to high altitudes could be seen. An assessment using trend analysis is discussed in section 3.2.3.

3.2. Trends

3.2.1. Regions and years

The 381 samples were assessed for metadata. With only three years to compare, no formal trend analysis could be done but some graphical presentation is included in the Supplementary information for illustration (Figure S1). Narrow grey shaded areas indicate that the values do

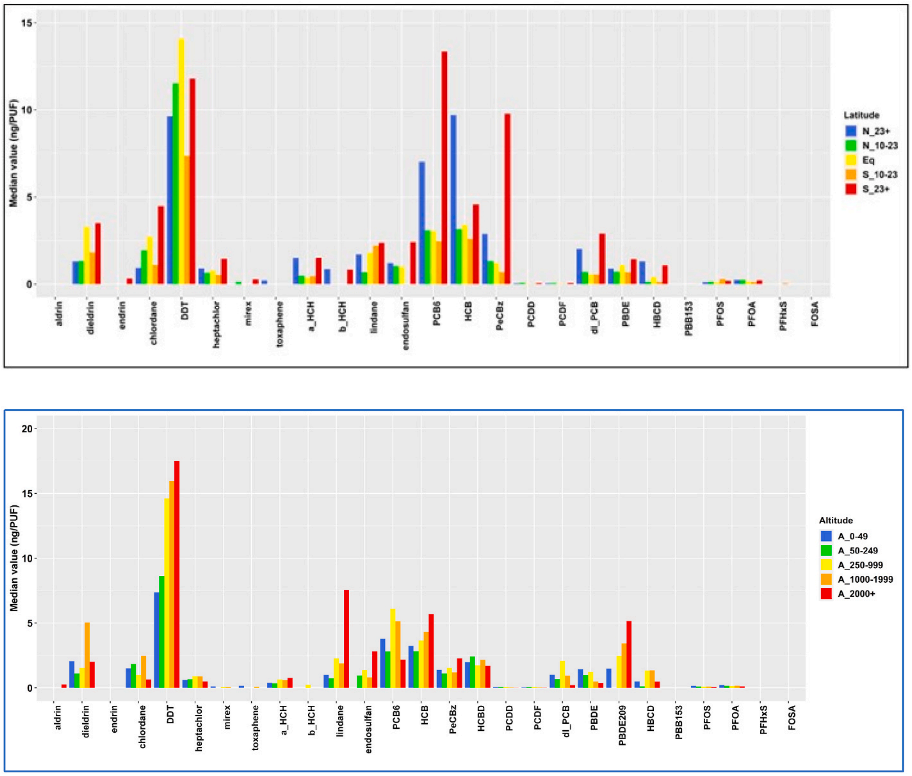


Fig. 4. Barplots displaying the median values of POPs according to latitude (above) and altitude (below).

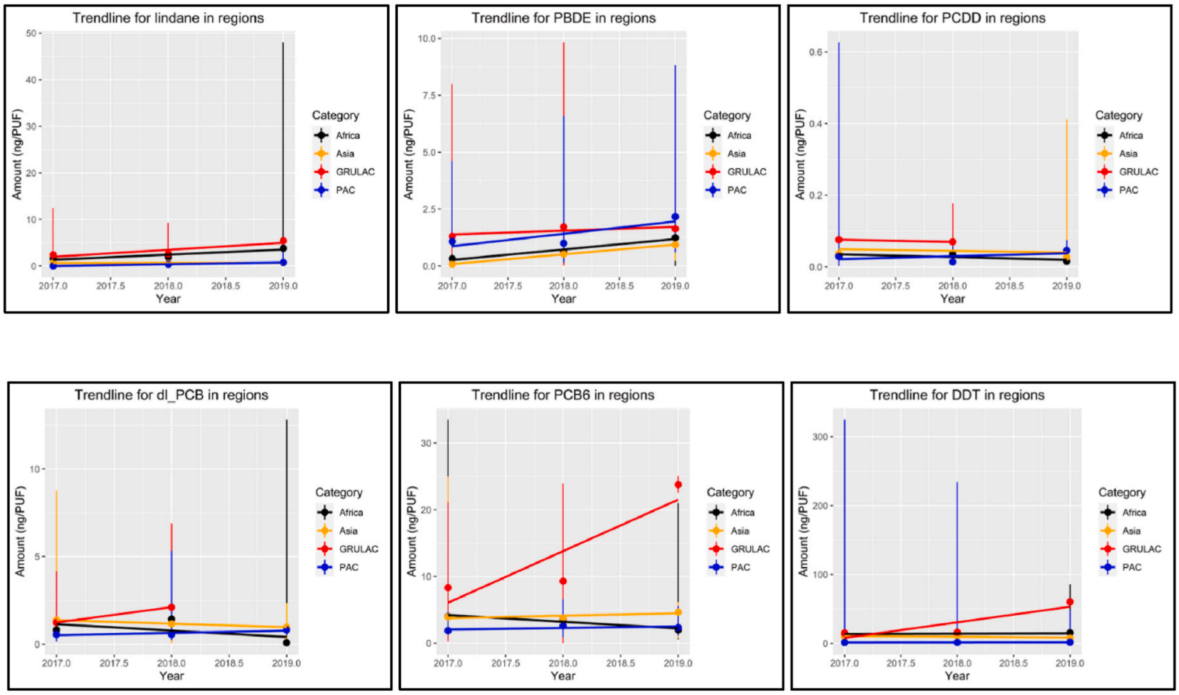


Fig. 5. Regional trendlines (linear model) by year and POP across all samples (N = 381).

not spread widely but it is obvious that in 2018, when most data were available, median values (represented by the black dot) were close to the trendline but that the 90th percentile values were highest for HBCD and dl-PCB.

The global trendlines, as depicted in Fig. S 1 were further disaggregated into the regions to assess if the tendencies were the same; Fig. 5 gives examples were the slopes were either in parallel or diverging. The trendlines in the first row of Fig. 5 are very similar in all four regions and do not have much slope: For lindane and PBDE the slopes are slightly positive whereas for PCDD, at least three are negative. For dl-PCB and PCB$_6$, opposite slopes in the regions were seen for the same POP; whereas dl-PCB increase in GRULAC, they concentrations

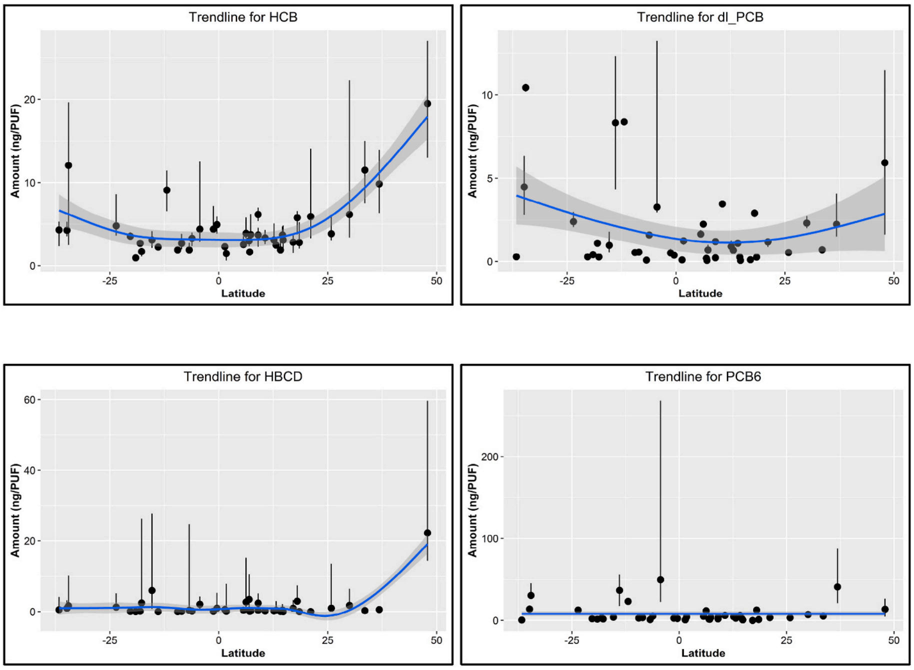


Fig. 6. Latitudinal trendlines (gam model) by latitude (from south to north) and POP across all samples (N = 381).

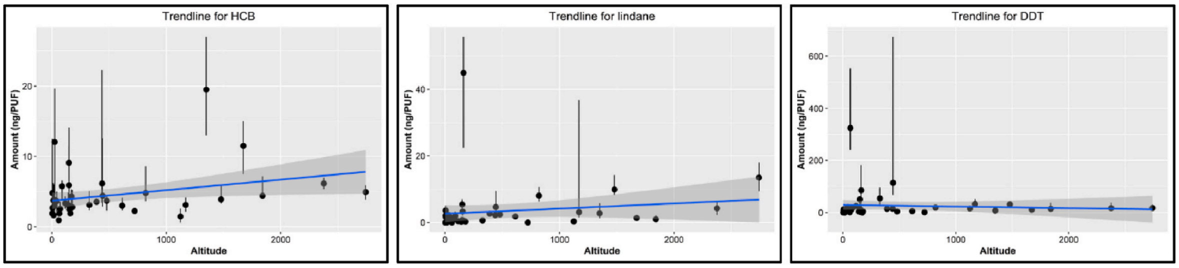


Fig. 7. Altitudinal trendlines (linear model) by altitude (in m) and POP across all samples (N = 381).

decrease in the other three regions. For PCB_6, a strong increase was seen from 2017 to 2019 whereas in the other regions there was almost no variation. Lastly, DDT showed only small variation as to the median values with an increase mainly in GRULAC but had large values for the 90th percentiles, especially for PAC (the upper and lower end of the lines indicate 90th and 10th percentiles of the measured values). In general, the trendlines are just a different model and confirm the findings from section 3.1 and Fig. 2.

3.2.2. *Latitude*

From Fig. 4, it was implied that for some POPs, greater amounts would be found at the locations closer to the poles. The trend analysis applied used the ‘gam’ model since the concentrations at the equator (in the center of the x-axis) should be lowest. The findings from the optical inspection were confirmed for HCB and dl-PCB (Fig. 6, upper row) with the highest median values at the extreme latitudes and the lowest at the equator. For PCB_6, such trendline could not be established (Fig. 6, lower row). For HBCD, the very high values in Mongolia are seen at the highest northern latitude (close to 50°).

3.2.3. *Altitude*

The trend analysis, using the linear model and assuming increasing values with increasing altitude, shows that an increasing trend was seen only for HCB and to a lesser extent for lindane, but not for DDT. Although bar graphs of median values of Fig. 4 seem to indicate a clear increasing tendency, the very high values in Africa (895 ng/PUF) at A_250–999 or the Pacific Islands (667 ng/PUF) at A_50–249 drive the 90th percentile towards higher values for the lower altitudes (Fig. 7). Some examples are included in the Supplementary information (Fig. S 2), showing that Pearson correlation coefficients for HCB and lindane were small but significant.

3.3. *Multivariate analysis*

The principal component analysis (PCA) has only limited power since the first two dimensions explain only 25 % of the variation. The biplot in Fig. 8 (upper row) shows that for the first dimension (Dim1), the major driving forces were from the four PFAS (PFOS and PFHxS, both >20 %, PFOA 19 %, FOSA 14 %), followed by PBDE 209 with 11 %: all other POPs with less than 5 %. For Dim2, endrin and β-HCH had 16 %, followed by α-HCH and lindane with 12 % and 10 %, resp.; all other POPs had less than 10 % contribution of the variables to the PCA. The outliers for Dim1 were the samples from Zambia and for Dim2 from Tanzania, both with black dots indicating Africa. The PCAs show that the African samples spread much wider than the other regions as shown by the wide concentration ellipse.

In the biplots shown in Fig. 8 (lower row) ellipses could be generated for latitude and altitude. The ellipses referring to latitudes were quite narrow for all except for the red ellipse referring to the southern latitudes between 10° and 23° to include the PFAS-driven samples from Zambia in the 4th quartile. All other ellipses point into the 1st quartile with the higher weight of the OCPs. The Mongolia samples, dark green

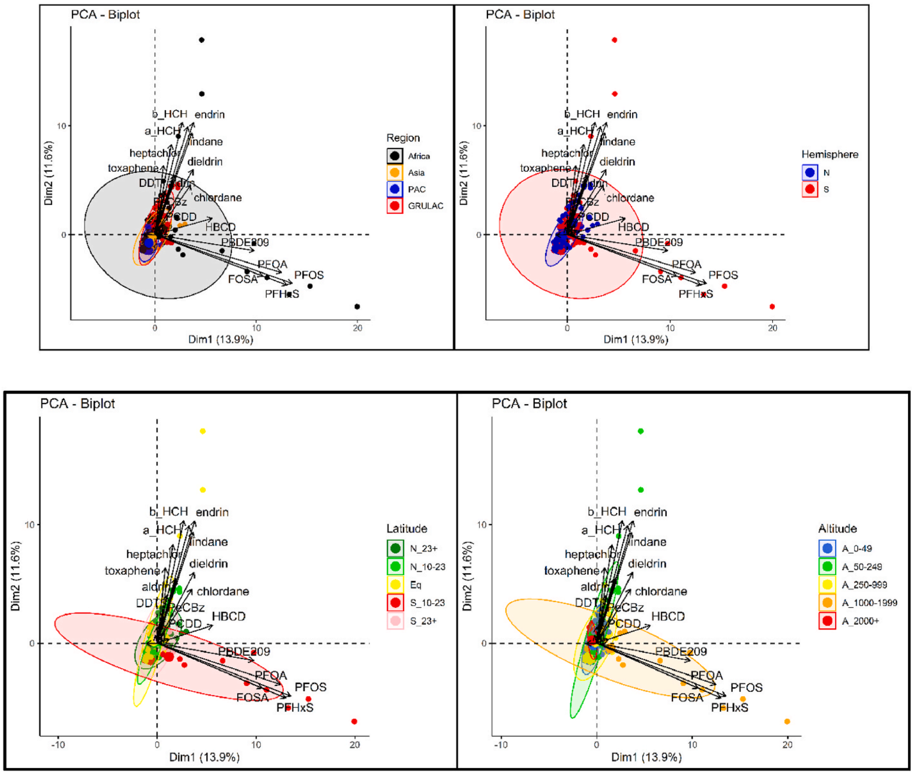
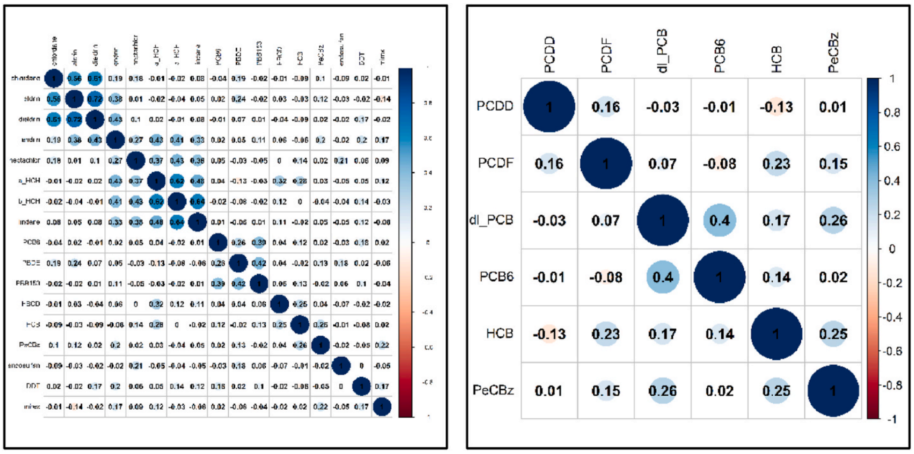


Fig. 8. PCA biplots for all POPs with concentration ellipses around the regions and the hemisphere (upper row) and ellipses around latitude and altitude (lower row).



	chlordane	aldrin	dieldrin	endrin	heptachlor	a_HCH	b_HCH	lindane	PCB6	PBDE	PBB153	HBCD	HCB	PeCBz	endosulfan	DDT	mirex
chlordane	1	0.56	0.61	0.19	0.18	-0.01	-0.02	0.08	-0.04	0.19	-0.02	-0.01	-0.09	0.1	-0.09	0.02	-0.01
aldrin	0.56	1	0.72	0.38	0.01	-0.02	-0.04	0.05	0.02	0.24	-0.02	0.03	-0.03	0.12	-0.03	-0.02	-0.14
dieldrin	0.61	0.72	1	0.43	0.1	0.02	-0.01	0.08	-0.01	0.07	0.01	-0.04	-0.09	0.02	-0.02	0.17	-0.02
endrin	0.19	0.38	0.43	1	0.27	0.43	0.41	0.33	0.02	0.05	0.11	0.06	-0.06	0.2	-0.02	0.2	0.17
heptachlor	0.18	0.01	0.1	0.27	1	0.37	0.43	0.38	0.05	-0.03	-0.05	0	0.14	0.02	0.21	0.06	0.09
a_HCH	-0.01	-0.02	0.02	0.43	0.37	1	0.62	0.48	0.04	-0.13	-0.03	0.32	0.28	0.03	-0.05	0.05	0.12
b_HCH	-0.02	-0.04	-0.01	0.41	0.43	0.62	1	0.64	-0.02	-0.08	-0.02	0.12	0	-0.04	-0.04	0.14	-0.03
lindane	0.08	0.05	0.08	0.33	0.38	0.48	0.64	1	0.01	-0.06	0.01	0.11	-0.02	0.05	-0.05	0.12	-0.06
PCB6	-0.04	0.02	-0.01	0.02	0.05	0.04	-0.02	0.01	1	0.26	0.39	0.04	0.12	0.02	-0.03	0.18	0.02
PBDE	0.19	0.24	0.07	0.05	-0.03	-0.13	-0.08	-0.06	0.26	1	0.42	0.04	-0.02	0.13	0.18	0.02	-0.06
PBB153	-0.02	-0.02	0.01	0.11	-0.05	-0.03	-0.02	0.01	0.39	0.42	1	0.06	0.13	-0.02	0.06	0.1	-0.04
HBCD	-0.01	0.03	-0.04	0.06	0	0.32	0.12	0.11	0.04	0.04	0.06	1	0.25	0.04	-0.07	-0.02	-0.02
HCB	-0.09	-0.03	-0.09	-0.06	0.14	0.28	0	-0.02	0.12	-0.02	0.13	0.25	1	0.26	-0.01	-0.08	0.02
PeCBz	0.1	0.12	0.02	0.2	0.02	0.03	-0.04	0.05	0.02	0.13	-0.02	0.04	0.26	1	-0.02	-0.05	0.22
endosulfan	-0.09	-0.03	-0.02	-0.02	0.21	-0.05	-0.04	-0.05	-0.03	0.18	0.06	-0.07	-0.01	-0.02	1	0	-0.05
DDT	0.02	-0.02	0.17	0.2	0.06	0.05	0.14	0.12	0.18	0.02	0.1	-0.02	-0.08	-0.05	0	1	0.17
mirex	-0.01	-0.14	-0.02	0.17	0.09	0.12	-0.03	-0.06	0.02	-0.06	-0.04	-0.02	0.02	0.22	-0.05	0.17	1

	PCDD	PCDF	dl_PCB	PCB6	HCB	PeCBz
PCDD	1	0.16	-0.03	-0.01	-0.13	0.01
PCDF	0.16	1	0.07	-0.08	0.23	0.15
dl_PCB	-0.03	0.07	1	0.4	0.17	0.26
PCB6	-0.01	-0.08	0.4	1	0.14	0.02
HCB	-0.13	0.23	0.17	0.14	1	0.25
PeCBz	0.01	0.15	0.26	0.02	0.25	1

Fig. 9. Correlation between POPs groups (Pearson correlation coefficients).

color, are all located close to the origin of the biplot but along Dim2. The same picture obtained for the ellipses colored according to altitudes, where the orange color warps the Zambia sample at 1170 m.

3.4. Correlations

A correlation assessment between POPs was done using Pearson correlations for samples that had (i) all the OCPs and BFRs and (ii) industrial and dl-POPs quantified (Fig. 9). There is not a single strong or even moderate correlation coefficients for the POPs. Except β-HCH with lindane (across all samples), which had r = 0.64.

The results across all samples and all POPs did not give statically significant results for the northern and the southern hemisphere (p-value = 0.37). With respect to the altitude, the p-value was 4.8×10^{-8}, thus a significant difference across all altitudes. The pairwise correlation using the Benjamini Hochberg adjusted p-value, then did not result in significant differences between the values at the higher altitudes: A_2000+ had p-values >0.10 with all other altitudes (with A_0–49 p = 0.78, with A_50–249 p = 0.097, with A_250–999 p = 0.45, and with A_1000–1999 p = 0.15), in addition, for A_1000–1999, the p-value to A_250–999 was 0.29; but there were significant differences to A_0–49 (p = 0.004) and A_50–249 (p = 4.2×10^{-7}).

4. Conclusion

The project has generated an abundance of data which first informs the countries about the chemical identity and the scale of POPs present in their country or region. Further, the data are useful in determining temporal and spatial trends for the Stockholm Convention POPs.

Despite some drawbacks and simplifications in the sampling and measurement approaches, including the inclusion of new POPs that do not necessarily follow the original sources, occurrence and definition of the legacy OCPs and PCB, and omission of data transformation from PUF into volume, we can make some useful recommendations:

1. The application of PAS/PUFs with defined pre-cleaning to optimize capture of groups of POPs and selective and sensitive targeted analysis has proven robust in implementation onsite, transport, and analytical laboratory.
2. Sampling periods of three months provide enough disaggregation to capture weather events. However, since some POPs level off or due to slow uptake kinetics, it is recommended to combine quarterly PUFs into annual samples for analysis. This approach has proven to be successful especially for dl-POPs and for PFAS.
3. It is recommended to cover full calendar years; however, it does not seem to be necessary to have monitoring programmes every year. Sampling for two consecutive years within one GMP reporting period under the Stockholm Convention seems sufficient (six years).
4. Sampling locations should remain the same for several monitoring programmes since it seems that local sources have a larger impact than long-range transport.
5. Geographic metadata including regional approaches are not a good predictor for POPs in air rather the results are site-specific on a relative scale but the same for a given POP.

The correlation coefficients for POPs with either latitude or altitude were all low; some positive, some negative. Many of these were significant so that more data would not change the picture and cannot be used for optimizing the global monitoring strategy.

With two years of sampling and selective and sensitive POPs analysis, a large number of data has been generated. In the first place, the data have been generated to close geographic gaps in monitoring data from developing country regions as was found in the first global monitoring reports of the first effectiveness evaluation under the Stockholm Convention [67]. The data then contributed to the development of the second and third global and regional monitoring reports as mandated by the Conference of the Parties. Thus, they were used for global interpretation on a comparative basis. In the future, they may contribute to new global assessments when climate change, and especially raising temperatures, may have a measurable impact on transport or transformation pattern. The advantage of the approach is that at national level, these results were obtained through application of a simple sampling equipment but sophisticated selective and sensitive chemical analysis. For countries, the data will serve to prioritize sampling and analytes in future monitoring programs. They may be used for establishing local/national models and serve as a starting point for transfer and accumulation schemes. If consistently and evenly applied, some so far overlooked countries or locations may be discovered.

The data would also be a model when considering establishing POPs laboratories for air monitoring as to prioritizing POPs and develop analytical methods as well as setting sensitivity requirements Countries and laboratories must be aware that many of the POPs substances are levelling off and may no longer be quantified even by sophisticated targeted analysis. Definitively, these low concentrations cannot be captured by screening methods.

It must be noted that ambient air monitoring using PAS/PUFs may be used to test efficiencies of release reduction measures at hotspots; however, these ambient air measurements do not replace a national source and release inventory.

Author credit statement

Heidelore Fiedler: Funding acquisition Örebro University, coordination and supervision of analysis of PFAS and dioxin-like POPs, Data curation, Validation, Visualization, Writing – original draft, review and editing. Esteban Abad: Funding acquisition CSIC, coordination and supervision chemical analysis of POPs in GRULAC and all dl-POPs, PCB6 in GRULAC), Validation, Writing – review & editing. Jacob de Boer: Funding acquisition Vrije Universiteit Amsterdam, coordination and supervision chemical analysis of OCPs, industrial chlorinated POPs, and brominated flame retardants, Validation, Writing – review & editing

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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