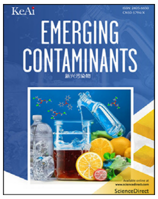
Emerging Contaminants 9 (2023) 100251

Contents lists available at ScienceDirect

Emerging Contaminants

journal homepage: www.keaipublishing.com/en/journals/emerging-contaminants



POPs in the Mongolian environment

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A R T I C L E I N F O

Article history:
Received 5 May 2023
Received in revised form
26 June 2023
Accepted 26 June 2023
Available online 28 June 2023

Keywords:
Developing country
Stockholm Convention on Persistent Organic Pollutants
Cross-matrix assessment
Multi-pollutants
Chemical analysis

A B S T R A C T

An environmental and human monitoring project analyzing the persistent organic pollutants (POPs) listed in the Annexes of the Stockholm Convention on POPs was implemented in Mongolia with sampling activities from 2017 until 2019. In support of the global monitoring plan implementation, a total of 51 samples were analyzed for POPs whereby 23 POPs were detected frequently. Two POPs, endrin and hexabromobiphenyl (PBB 153), were not quantified in any of the samples; toxaphene was found only twice in air samples; and dicofol, pentachlorophenol, and short-chain chlorinated paraffins were analyzed only in one human milk sample. Most results were for perfluorooctanoic acid (PFOA, 32 of 43 samples) and perfluorooctanesulfonic acid (PFOS, 31 of 43 samples). With respect to detection frequency, dioxin-like polychlorinated biphenyls (dl-PCB) were detected in 100% of the samples analyzed ($N = 28$). On scale, highest amounts were found in soil samples, almost 30 000 ng/g for lindane, more than 13 000 ng/g for α-hexachlorocyclohexane (α-HCH), and almost 10 000 ng/g for dichlorochlorodiphenyltrichloroethane (DDT) and its transformation products. The soil and ambient air samples were the drivers towards highest values in the abiotic samples, due to the presence of legacy POPs such as DDT, indicator PCB, HCHs but also hexachlorobenzene (HCB), and pentachlorobenzene (PeCBz). Concentrations of dioxin-like POPs were low in general as were the perfluorinated substances (PFAS). With respect to brominated flame retardants, the presence of hexabromocyclododecane in air samples was surprising. At present, the analytical capacity to analyze POPs in various matrices under the global monitoring plan for POPs is being set-up to strengthen future environmental monitoring activities in Mongolia.

1. Introduction

Mongolia is a Party to the Stockholm Convention on Persistent Organic Pollutants (POPs) since 2004 and has actively contributed to the Convention implementation. Mongolia has ratified all amendments to the Convention, *i.e.*, should address all listed POPs [32]. A global monitoring plan (GMP) was established under the Stockholm Convention to monitor changes of POPs in the environment and humans over time and with spatial resolution. Guidance documents on practical aspects of implementation had been developed and the most recent version was concluded in 2021 [31]. Further, the GMP contributes to the effectiveness evaluation of the Convention as stipulated in Article 16 of the Convention [23]. So far, Mongolia had not undertaken systematic analysis of POPs in the environment but had participated in the POPs monitoring project in East Asia (POPsEA), which is coordinated by the Japan Environmental Sanitation Center (JESC) and supported by the Ministry of Environment Japan. This project provided first data on their presence of POPs in ambient air through measurements using active air samplers (AAS) [28]. Since 2005, Parties to the Convention had been supported by capacity building and data generation projects coordinated by regional projects coordinated by the United Nations Environment Programme (UNEP) and with financial assistance from the Global Environment Facility (GEF) or others. From 2015, Mongolia participated in a monitoring project to support the implementation of the GMP under the Convention in seven Asian countries [27]. Mongolia participated in all subprojects and recruited teams to participate in the air, human milk, and water sampling networks as well as in interlaboratory assessments, and

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Peer review under responsibility of KeAi Communications Co., Ltd.

https://doi.org/10.1016/j.emcon.2023.100251
2405-6650/

capacity building activities. The GMP had defined ambient air and water as core matrices for environmental occurrence and transport and human milk or human blood for human exposure. Results for POPs in these core matrices from Mongolia have been published recently [22].

On the other hand, many countries are interested in determining POPs concentrations in foodstuffs, including for regulatory purposes, or sediment and soil. Within the UNEP/GEF GMP2 projects, these additional samples were named ‘national samples’ since most countries were interested in matrices other than the core matrices; *e.g.*, soil or sediment and especially food samples present in their daily consumption. A specific standard operational procedure (SOP) was prepared addressing soil, sediment, meat, fish, dairy products or eggs and applied for these projects [30]. These so-called national samples were analyzed in the project's central laboratories for the POPs and their transformation products as recommended in Table 2.2 of the GMP guidance document [31].

A first inventory of POPs uses in Mongolia found that from the POPs pesticides, technical mixtures of hexachlorocyclohexanes (HCH) have been used widely under the common name of "Dust" containing α-HCH (12%) or an HCH emulsion (16%) that had been used from 1958 to 1985 as an insecticide in livestock ectoparasites (to fight mites, scabs, ticks, bloodsuckers, etc.), as well as for disinfecting animal shelters and ordure. HCH in powder form was mainly spread over small animals by hand, whereas big animals and cattle were driven into a cubicle where they were fumigated with HCH. In addition, hexachlorobenzene (HCB), chlordane, aldrin, dieldrin, and heptachlor was reported to have been used from 1969 to 2003 [21].

The occurrences, relative abundances, and distribution of 26 POPs listed in either Annex A, B, or C of the Stockholm Convention were analyzed in a limited number, but broader spectrum of abiotic and biota samples collected in Mongolia and analyzed abroad. Five of the listed POPs were not included in this project: chlordecone, polychlorinated naphthalenes, short-chained chlorinated paraffins, pentachlorophenol, and dicofol. Here, we present the quantitative results and make an initial assessment for prioritization of POPs in the Mongolian environment and future monitoring activities.

2. Materials and methods

2.1. Origin and characteristics of the samples

The Institute of Chemistry and Chemical Technology of the Mongolian Academy of Sciences (MAS) was responsible for the national implementation of the project including selection and collection of matrices. All the national samples were prepared following the protocol for the “Sampling and pre-treatment of national samples within the UNEP/GEF projects to support the global monitoring plan of POPs” [30]. When preparing the national samples, close contact was maintained with the expert laboratories in the Netherlands and Sweden. The national samples were chosen according to the most consumed foods for the everyday life of Mongolians. In addition, soil samples close to the animal wash basin of a veterinary clinic and water were collected and analyzed. The two river water samples are not included in this paper and have been reported already in a recent publication [22]. The matrices and the analytes are shown in Table 1.

2.2. Chemical analysis

Chlorinated POPs included aldrin, dieldrin, endrin, hexachlorobenzene (HCB), pentachlorobenzene (PeCBz), mirex, chlordecone, hexachlorobutadiene (HCBD), endosulfan (only α-endosulfan was quantified), alpha-hexachlorocyclohexane (α-HCH), beta-hexachlorocyclohexane (β-HCH), and lindane (γ-HCH) as single compounds, and five POPs that consist of more than one compound: Dichlorodiphenyltrichloroethane (DDT) included analysis of *o,p’*- and *p,p’* isomers of DDT, DDD, and DDE and are reported as DDT; PCB_6 includes PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, and PCB 180, toxaphene included three Parlar congeners, namely Parlar 26, Parlar 50, and Parlar 62; chlordane included *cis*- and *trans*-chlordane, *cis*- and *trans*-nonachlor, and oxychlordane; heptachlor included heptachlorepoxide. This group of chlorinated organochlorine pesticides (OPCs) and chlorinated industrial chemicals (ind.POPs) is further referred to as OCPs + ind.POPs. Since α-HCH, β-HCH, and lindane were listed as separate POPs [24–26], their results were not summed-up to HCHs as was done in the UNEP regional reports. Brominated flame retardants (BFR) included hexabromobiphenyl #153 (PBB153), three stereoisomers of hexabromocyclododecane, α-HBCD, β-HBCD, and γ-HBCD, referred to as HBCD. The commercial mixtures of tetra-, penta-, hexa- and octabromodiphenyl ethers were combined to PBDE and included the six recommended compounds PBDE 47, PBDE 99, PBDE 100, PBDE 153, PBDE 154, PBDE 175/183. The two congeners recommended for air samples, PBDE 17 and PBDE 28, were measured by the analytical laboratory and reported elsewhere [6,16] . For comparative reasons, PBDE 17 and PBDE 28 were not included in the sum parameter of this paper; thus, all PBDE refers to $PBDE_6$. Decabromodiphenyl ether (deca-BDE), since listed as an individual POP [29], was analyzed separately and referred to as PBDE 209. Dioxin-like POPs (dl-POPs) comprise three listings in Annex C of the Stockholm Convention, namely 7 congeners of 2,3,7,8-substituted PCDD, 10 congeners of 2,3,7,8-substituted PCDF, and 12 dioxin-like PCB (dl-PCB) using the 2005 WHO toxicity equivalency factors (TEFs) [40]. They are reported as toxic equivalents (TEQ), specifically as TEQ_PCDD, TEQ_PCDF, and TEQ_PCB. Perfluoroalkyl substances (PFAS) included perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), and perfluorohexanesulfonic acid (PFHxS), these were reported as the respective anion; thus, including acids and salts.

PUFs and national samples were analyzed by the Department Environment & Health Vrije Universiteit Amsterdam, the Netherlands, for brominated and chlorinated POPs (except dl-POPs). PCB and OCPs were analyzed by GC-MS/MS, toxaphene, $PBDE_6$ and PBB by GC/MS, and the HBCD diastereomers by LC-MS/MS [6,7]. All PFAS analysis was performed by MTM Research Centre at Örebro University, Sweden) using UPLC/MS-MS instrumentation [2,4,13–15,41]. dl-POPs were analyzed as follows: Air samples from active and passive samplers were analyzed by CSIC Barcelona [1] and national samples by an accredited commercial laboratory in Germany [11]. All laboratories followed generally accepted analytical principles [8,38,39] and applied their validated methods using high resolution gas chromatographic columns (HRGC) coupled to sector-field high-resolution mass spectrometers (HRMS).

The POPs laboratories at MTM Örebro University, Vrije Universiteit Amsterdam, and CSIC had successfully participated in the rounds of the UNEP-coordinated interlaboratory assessments for POPs with the majority of z-scores <|2| or within ±25% from assigned value (the individual reports are available at the UNEP WebPage [33–36] or published in the literature [5,17–19,41]. The commercial laboratory serves as a national reference laboratory for food and feed of an EU member state and successfully participated in all the required proficiency tests. Table 2 shows the types of samples and the analytes. It can be seen that not all samples were analyzed for the same spectrum of POPs. Dairy samples, one cow milk, one mare milk, and four butter samples were analyzed for dl-POPs and PFAS only, the butter sample was also analyzed for brominated flame retardants. The soil samples were not analyzed for dl-POPs; water samples only for PFAS. For data presentation, the

Table 1
Identity of samples and analytes.

Matrix	Description	Location, GIS coordinates, amounts available for analysis
PUF	PAS/PUFs, exposed for 3 months from April 2017 until end of March 2019	Bayanzurkh district in Ulaanbaatar city
AAS	An active air sampler was installed, and two samples collected during three consecutive days	GIS: 47°55′06″ N, 106°58′15″ E
Soil	Soil1 and Soil2 were background samples; Soil3 and Soil4 from locations close to veterinary hospital with animal washing basin	50 g each OCPs/PCB and BFR analysis; 25 g each for dl-POPs and PFAS analysis
Water	Eight GMP2 samples from March 2017 until Dec 2018; at 30 cm–50 cm depth from referred to as GMP (201x-1/2/3/4.	Collected in 1 L in HDPE bottles, each From the middle of the Tuul river GIS: 47.889, 106.910
	Two samples from Bayanzurkh district in Ulaanbaatar referred to as WR1 and WR2.	WR1 from a well at 47° 54′ 28″ N, 106° 59′ 39″ E WR2 was collected at the Uliastai river at 47° 54′ 13″ N and 107° 1′ 51″ E
Fish	Four fish samples referred to as Fish1, Fish2, Fish3, and Fish4	Commercial product from Ulaanbaatar markets/supermarket
Meat	Mutton, horse meat, pork Fat from sheep tail, mutton/lamb	Commercial product from Ulaanbaatar markets/supermarket
Butter	Four samples of different brands of butter, referred to as Butter1, Butter2, Butter3, Butter4	300 g each for OPC/PCB and BFR analysis; 300 g for dl-POPs/PFAS analysis
Milk	Cow milk and one mare milk sample	Mare milk: 300 g 250 mL for cow milk
Chicken	Chicken 1 – imported chicken 2 – locally raised	
Egg	Two samples from locally raised hens referred to as Egg "Tumen shuvuut" and Bayan (Egg1, Egg2) Imported eggs – NBTs (Egg3) local eggs with Se (Egg4)	Four eggs per sample for PCB analysis, 3 eggs for BFR, up to 500 g for dl-POPs and PFAS analysis
Plant	Sea buckthorn and flax for OCPs/PCB Sea buckthorn oil for dl-POPs/PFAS analysis	500 g for OCPs/PCDF; 300 g for dl-POPs and PFAS
Oil	Sheep tail fat oil	
Human milk	1 sample as pool of 50 mL each from 50 *primiparae* living in 7 districts in the Ulaanbaatar area, collected Feb–May 2018; sampled by hand expression; stored in refrigerator	Ethics committee approval took another 2 months, samples sent to laboratory in August 2018

Table 2
Number of samples and analytes.

Sample	Matrix	OCPs	endo-sulfan	PCB$_6$+HCB +PeCBz	HCBD	PBDE$_6$ +HBCD	PBDE209	dl-POPs	PFAS
PAS/PUF	PUF	8	8	8		8		6	9
AAS	AAS	2		2		2	2	2	
Soil	Soil	4	4	4	4	4	4		4
Water	Water								10
Fish	Fish	3	3	3	3	2	2	3	4
Meat (beef, horse, mutton, pork, sheep tail)	MeDaEg	4	3	4	4	2	2	3	3
Butter						4	4	4	4
Milk (cow, mare)								2	1
Chicken meat		2	1	2	2	2	2	2	2
Egg		4	4	4	4	4	4	4	4
Fruit (buckthorn)	Plant	1	1	1	1				
Oil (buckthorn, flax)[a]		1	1	1	1			1	1
Human milk	HM	1	1	1	1	1	1	1	1
Total		**30**	**26**	**30**	**20**	**29**	**29**	**28**	**43**

[a] The sea buckthorn oil was analyzed for dl-POPs and PFAS, flax oil was analyzed for OCPs + ind.POPs and BFR.

terrestrial food samples of meat, dairy, and eggs were designated as ‘MeDaEg’. In total, about 700 quantitative results from 51 samples were generated and assessed.

2.3. Data handling and assessment

All data were maintained in Microsoft 365 Excel®, version 2211 (Microsoft Corporation). Statistical evaluations and visualization were made using R version 4.2.2 and R packages with R-Studio version 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, where necessary 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).

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). For water samplings, these were punctual collections occurring once in three months at the end of the season. Water samples were referred to by year and season with Arabic numbers 1–4 used.

Amounts of OCPs + ind.POPs and BFR were reported in nanogram per PUF (ng/PUF), ng/g or ng/m^3; amounts of dl-POPs were reported in picogram *per* PUF (pg/PUF), pg/g, or pg/m^3. Amounts of PFAS were reported as pg/PUF and pg/g fresh weight (f.w.). The basis for the units were air dried samples for soil and sediment, fresh weight (f.w.) for fish and plants, and fat for terrestrial foodstuffs and human milk (except for PFAS). All PUF values refer to one PUF and an exposure period of three months (one quarterly season or 90 days ± 3 days).

3. Results

3.1. Occurrence and scale of POPs in the Mongolian environment

Aldrin, endrin, chlordane (including isomers and transformation products), heptachlor (including isomers and transformation products), and PBB 153 were not quantified in any of the samples. Toxaphene (sum of three as Parlar #26, #50, #62) was found in only one fish sample (Fish1 at 0.04 ng/g). These POPs are not included in the graphics. PFHxS was quantified in the water samples only; thus, water samples have data only for PFAS, and this matrix is not reflected when visualizing other POPs. The cow milk sample had PFOA at 229 pg/g f.w. and all other PFAS were below LOQ (PFOS and PFHxS: LOQ <6.2 pg/g f.w.); this sample was excluded from further assessment [14].

3.1.1. Organochlorine pesticides and chlorinated industrial chemicals

There were 30 samples analyzed for OCPs and chlorinated industrial POPs, namely PCB_6, HCB, PeCBz, and HCBD (OCPs + ind.POPs), refer to Table 2. The descriptive statistics are contained in the supplementary information as Table S 1. The scale of the OCPs + ind.POPs as stacked bars of the quantified POPs is shown in Fig. 1, upper row, with concentrations at left and the pattern at right. An illustration by POP in the different matrices is shown in the supplementary information, Fig. S 1. The cow milk and the mare milk samples were not analyzed for Br/Cl-POPs and the butter samples and Fish1 not for the chlorinated POPs. It can be seen that in fish and chicken, PCB_6 was most prominent whereas could not be quantified in eggs (N = 4) or plants (N = 2). In addition, chlorinated industrial chemicals such as HCB, PeCBz, and HCBD were found in meat and eggs; to a lesser extend in fish. The soil samples contained small amounts of PCB_6 (0.1 ng/g-4.5 ng/g), HCB (0.1 ng/g-8.2 ng/g), PeCBz (4.7 ng/g-74 ng/g), and HCBD (<LOQ-0.41 ng/g) but very high concentrations of certain organochlorine pesticides.

Soil1 was highly contaminated with DDT (9 880 ng/g) and had also the highest value for PeCBz. Soil2 had quantifiable levels of most chlorinated POPs with 419 ng/g of α-HCH. Soil3 had more β-HCH (3 406 ng/g) and lindane (2 321 ng/g) than α-HCH (573 ng/g). Soil4 had extreme values for the three HCHs with 13 062 ng/g for α-HCH, 2 600 ng/g for β-HCH, and 29 300 ng/g for lindane. These results confirmed the earlier use of pesticides in the veterinary sector. The PUFs had high amounts of HCBD but no comparison could be made with the high-volume samples, since due to analytical interference, HCBD could not be quantified in the two AAS samples.

The box whisker plots in the lower row of Fig. 1 illustrate, that certain POPs are dominating in selected matrices. PCB_6, HCB, and PeCBz were dominating the POP pattern in the air samples (PUFs and AAS) but were not found in soil samples. Soil samples had by far the highest absolute contamination: mainly α-HCH, β-HCH, and especially lindane present. The fish samples had quite high amounts of PCB_6, with a wide range between measurements (mean = 4.12 ng/g f.w. ±5.64 ng/g f.w.; min = 0.71 ng/g f.w., max = 10.6 ng/g f.w.) and DDT to a lesser extent (mean = 1.04 ng/g f.w.). The food samples from terrestrial animals had highest concentrations for HCB (mean = 0.79 ng/g lipid ±0.70 ng/g lipid), β-HCH, and dieldrin (for numeric results, see Table S 1).

Statistical assessment reveals that there were significant differences between the categories and the matrices (p-values $<2.2 \times 10^{-16}$) (Table S 2). The pairwise assessment showed that there were significant differences between the PUFs and most other matrices but not with soil and human milk (p = 0.306 and 0.066), but also between soil and AAS (p = 0.035), fish (p = 0.021), MeDaEg ($p = 3.8 \times 10^{-6}$), or plant (p = 0.0007). For human milk, no statistical significance could be determined; all pairwise p-values were >0.07.

3.1.2. Brominated flame retardants

There were 29 samples analyzed for BFR (for details, see Table 2). The descriptive statistics are contained in the supplementary information as Table S 3. The scale of the three quantified BFR, namely, $PBDE_6$, PBDE 209, and HBCD, as stacked bars is shown in Fig. 2, left and the pattern in Fig. 2, right. The large amounts of PBDE 209 and HBCD in air samples was striking. PBB 153 could not be quantified in any sample and is not contained in the visualization. An illustration by POP in the different matrices is shown in the supplementary information, Fig. S 2. The fish samples had quite high amounts of $PBDE_6$.

Statistical assessment reveals that there were significant differences between the categories ($p = 3.0 \times 10^{-5}$) and the matrices ($p = 8.2 \times 10^{-7}$) (Table S 4). The pairwise assessment showed that there were significant differences only for PUFs with all matrices except human milk (p = 0.161).

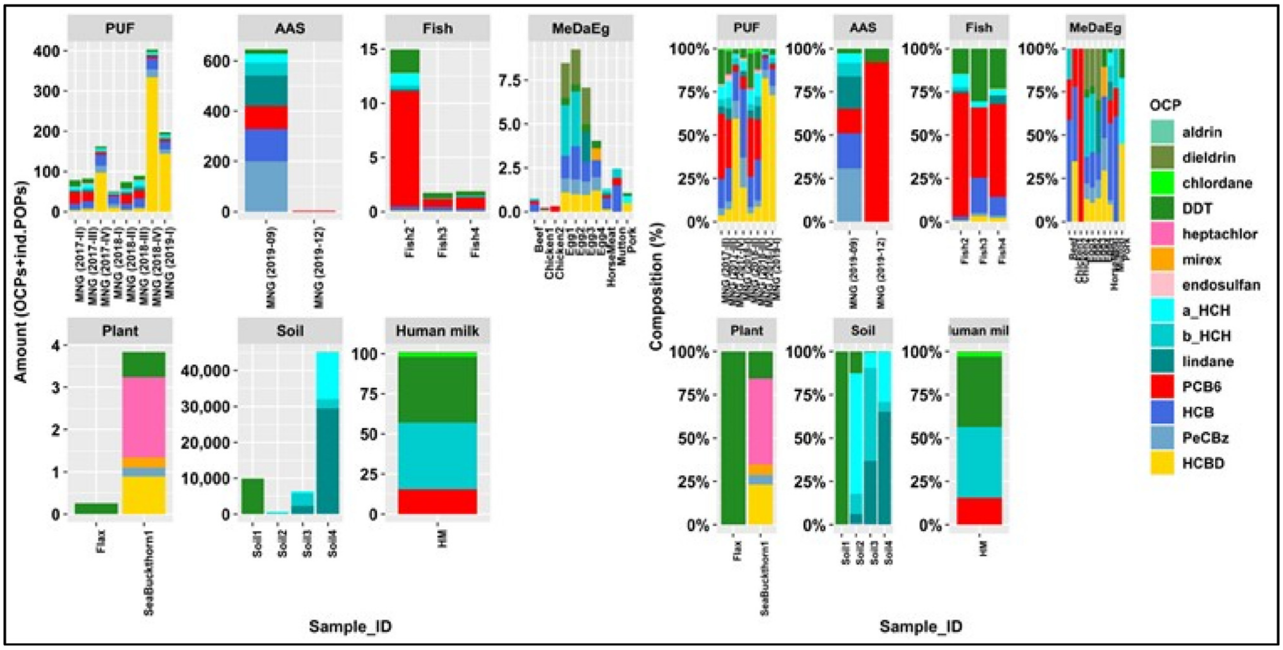
3.1.3. Dioxin-like POPs

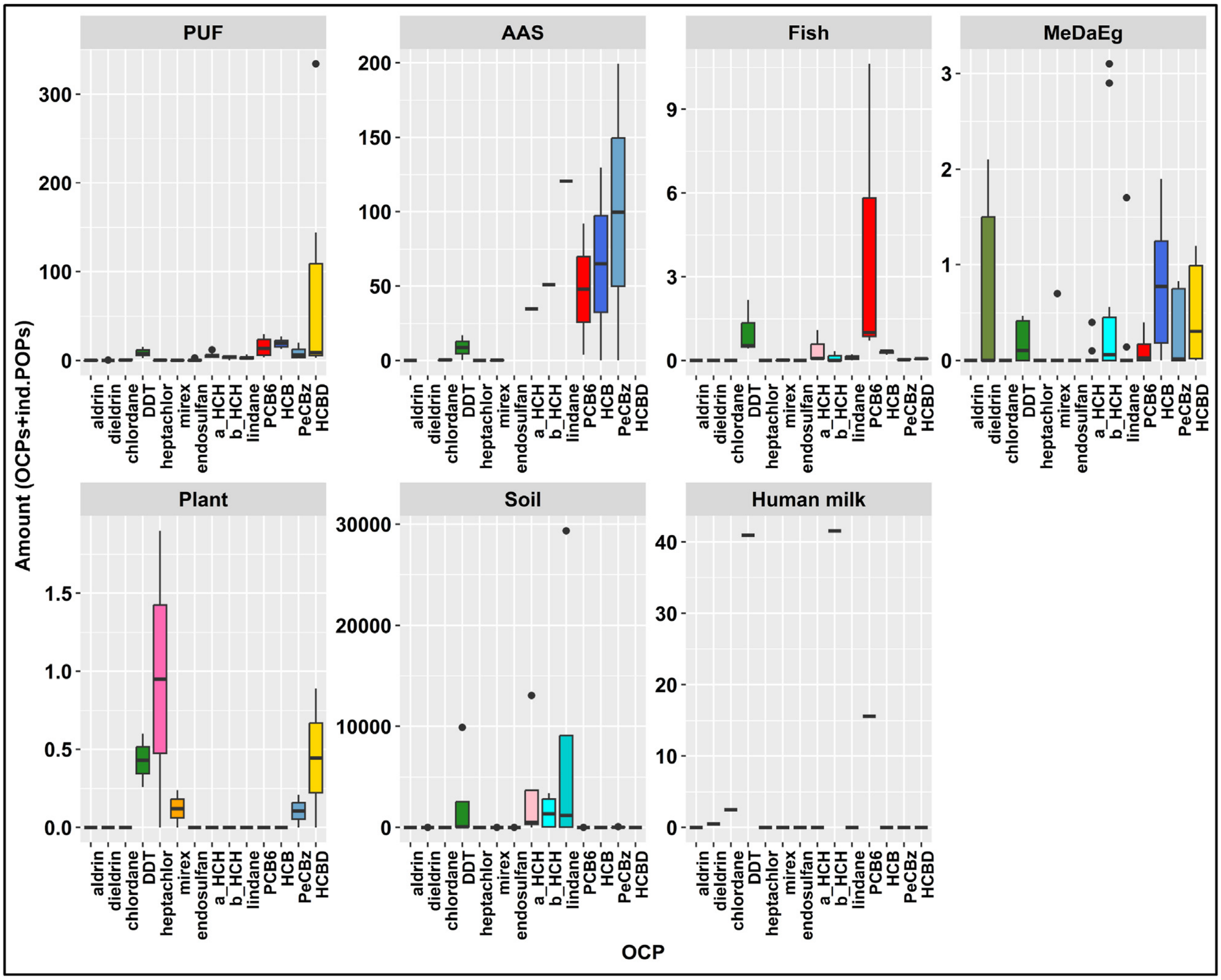
There were 28 samples analyzed for dl-POPs (for details, see Table 2). The descriptive statistics are contained in the supplementary information as Table S 5. The scale of the three dl-POPs as stacked bars is shown in the upper row of Fig. 3, left and the pattern in Fig. 3, right. An illustration by POP in the abiotic and biota matrices is shown in the lower row of Fig. 3. It can be seen that the Mongolian biota samples showed very low amounts of dl-POPs; and if quantified, PCDD were found in terrestrial foodstuffs like meat, dairy and egg but not in fish, whereas PCDF and especially dl-PCB were also found in fish. Soil samples were not analyzed for dl-POPs (see Table 2). dl-PCB were higher than PCDD or PCDF (as TEQ). Detailed analysis shows that PCDD was not quantified in the fish samples but in meats and dairy products. The human milk sample had the highest amounts for PCDD and PCDF, and the only one fish sample had more dl-PCB (see supplementary information, Fig. S 3). The plant matrix refers to the sea buckthorn sample that contained 0.006 pg TEQ/g; PCDD and PCDF were below the LOQ; as well as the three PFAS.

Statistical assessment reveals that there were significant differences between the categories ($p = 1.0 \times 10^{-4}$) and the matrices ($p = 3.96 \times 10^{-7}$) (Table S 6). The pairwise assessment showed that there were significant differences between almost all pairs of matrices with the exception of human milk with PUFs (p = 0.475) and fish (p = 0.083) as well as for the plant samples (p = 0.104).

3.1.4. Perfluorinated acids

There were 43 samples analyzed for PFAS (for details, see Table 2). The descriptive statistics are contained in the supplementary information as Table S 7. The amounts of the three PFAS as stacked bars is shown in the upper row of Fig. 4, left and the pattern


PUF
AAS
Fish
MeDaEg
Plant
Soil
Human milk
Amount (OCPs+ind.POPs)
Composition (%)
Sample_ID
Sample_ID
0
100
200
300
400
600
15
10
5
7.5
5.0
2.5
0.0
40,000
30,000
20,000
10,000
75
50
25
4
3
2
1
0%
25%
50%
75%
100%
MNG (2017-II)
MNG (2017-III)
MNG (2017-IV)
MNG (2018-I)
MNG (2018-II)
MNG (2018-III)
MNG (2018-IV)
MNG (2019-I)
MNG (2019-09)
MNG (2019-12)
Fish2
Fish3
Fish4
Beef
Chicken1
Chicken2
Egg1
Egg2
Egg3
Egg4
Horsemeat
Mutton
Pork
Flax
SeaBuckthorn1
Soil1
Soil2
Soil3
Soil4
HM
OCP
aldrin
dieldrin
chlordane
DDT
heptachlor
mirex
endosulfan
a_HCH
b_HCH
lindane
PCB6
HCB
PeCBz
HCBD


PUF
AAS
Fish
MeDaEg
Plant
Soil
Human milk
Amount (OCPs+ind.POPs)
OCP
0
100
200
300
50
150
3
6
9
1
2
1.5
1.0
0.5
0.0
10000
20000
30000
10
20
30
40
aldrin
dieldrin
chlordane
DDT
heptachlor
mirex
endosulfan
a_HCH
b_HCH
lindane
PCB6
HCB
PeCBz
HCBD

in Fig. 4, right. An illustration by POP in the different matrices is shown in the lower row of Fig. 4. PFOS was higher in fish than PFOA, but PFOA had higher detection frequencies. Fish2 had the highest PFOS value (309 ng/kg f.w.). The sea buckthorn oil sample is not included in the graphical representation since none of the three PFAS could be quantified. PFHxS was analyzed in all 43 samples but could be quantified in four water samples only (range from 0.027 ng/L to 0.076 ng/L). The water samples were collected at three different locations with eight samples at the “national” location Tuul river and two samples, referred to as “WR1” and WR2 at the Uliastai river, a contributary to the Tuul river. PFHxS was quantified in three water samples from the Tuul river and one from the Uliastai river. The visualization graph by sample is contained in the supplementary information as Fig. S 4. With respect to amounts, the concentration ellipse in Fig. 6, upper row right and lower row, shows that all water samples were located in the 2nd and 3rd quartile, so that they were very similar although the pattern varied but this was due to the low concentrations.

Statistical assessment reveals that differences were not significant for categories ($p = 0.20$) but for matrices ($p = 0.0009$) (Table S 8). The pairwise assessment showed that only the PUFs differed significantly from water ($p = 0.022$)), terrestrial foods (MeDaEg, $p = 0.0012$), and soil ($p = 0.019$). The p-values between water, fish, MeDaEg, and soil were all greater than 0.16.

3.2. Multivariate analysis

The graphs from the principal component analysis below show the complex pictures as to the chemical composition and scale of the POPs quantified in the Mongolian samples. All PCA graphs show concentration ellipses around the category, either abiotic or biota, and the matrix. All graphics show that the biota samples have much narrower ellipses than the abiotic samples. With respect to matrices, only a few ellipses could be drawn, which means that the scales and composition of the POPs group within a matrix is not necessarily similar.

The PCA graphs in Fig. 5 for OCPs + ind.POPs visualize the location and contribution of these POPs to the first two components (Dim1 and Dim2). The first two dimensions explain 48% of the data. The location of each sample is displayed in the first row at left of the graphic. The extreme dot in the 1st quartile represents Soil4, having almost 30 000 ng/kg of lindane but also 13 000 ng/kg of α-HCH and 74 ng/kg of PeCBz. The soil samples are very different and are dominated – in scale – by the HCHs. The air samples, with sample_IDs containing the Roman numbers for the seasons representing the PAS/PUF results formed the extreme dots associated with high values. For both, PCB_6, and HCB were associated with the highest values. Soil4 had the highest contribution to Dim1 (~80%) but hardly to Dim2 (5%); the overall contribution from Soil4 to the first two dimensions was almost 45%. Dim2 had the largest contributions from the seasonal PAS/PUF samples (see Fig. 5, row 1, left). The vectors of the variables (Figure 5, 2nd row) show that lindane, α-HCH, and PeCBz have most influence on Dim1 (24.4%, 24.0%, and 22%) whereas HCB, aldrin, and PCB_6 had the largest contributions to Dim2 (25%, 16%, and 15%). The soil and PUF samples were therefore responsible for the wide ellipse around the abiotic samples. The ellipse around the biota samples is very small, indicating little variation between the samples; further, the location close to the origin indicates small values.

The PCA for BFR in Fig. 5, top row right, for the location of major contributors and lower row as biplot with the ellipses show that $PBDE_6$ and PBDE 209 are located in different quartiles. The first two dimensions represent almost 100% of the variance. Most of the abiotic samples have strong contributions, corresponding to high concentrations, along Dim1 whereas the biota samples had larger contributions to Dim2, corresponding to $PBDE_6$ contamination. The highest values in the 1st and 4th quartile are PAS/PUF air samples. The soil samples are close to the biota samples and form a very small ellipse with low amounts (3rd quartile); for the BFR, the soil samples did not have the extreme values as for the chlorinated POPs. Among the biota samples, the highest amount was found in one fish sample (Fish2; 1.28 pg/g f.w. for $PBDE_6$) as shown by the highest dot along Dim2. Since most analytical determinations were below the LOQ for the three BFR, these conclusions must be taken with care.

The PCAs for the dl-POPs and PFAS are displayed in Fig. 6. The location of the samples in the first two dimensions for the dl-POPs are shown in the upper row at left; the first two components explain 98% of the data. The extreme dots are associated with the annual PAS/PUF samples and one sample from the 2nd season in 2017 (MNG (2017-II), each contributing between 21% and 32% to Dim1. They also had the highest contributions to Dim2; in addition, the human milk sample had 9.5% towards Dim2. In general, most dots and all food samples were located close to the origin, indicating low concentrations. The vectors of the variables (Figure 6, 2nd row) show that PCDD are located in the 1st quartile whereas PCDF and dl-PCB are closer together in the 4th quartile. The vectors nicely support findings that PCDF and dl-PCB correlate more than with PCDD. For matrices, one very wide ellipse could be created for the PUFs, and one very narrow ellipse for the food samples (MeDaEg).

The PCA for PFAS in Fig. 6, top row right for the location of major contributors and lower row as biplot with the ellipses show that PFHxS is located in the 2nd quartile whereas PFOS and PFOA are closely together in the 1st quartile. This means that PFOS and PFOA have a positive contribution and PFHxS a negative contribution to the PCA. Overall, the first two dimensions explain 92% of the data. The water samples are almost all located in the 2nd quartile since they are the only ones with PFHxS quantified (see Fig. 6, lower row, at right). The PUFs formed an extremely narrow ellipse and also the fish samples are grouped together. The sample Fish2 does not show up as outlier in the PCA despite the highest value for PFOS (309 pg/g f.w.).

4. Discussion

In this paper, we have combined the POPs results from various abiotic and biota samples, total of 51 samples, collected in Mongolia from 2017 to 2019. Other POPs results from the Mongolia environment are scarce and referred to below. The results presented here are for one of 42 countries within four regional projects and summarizes the work over six years. The number of samples collected and analyzed is higher than the average assigned to the project countries (planned: 25). The Mongolian results should be compared with the actual air data on the global level published in three papers [1,4,6] and put in perspective with global but limited trend data [10]. Mongolia participated for the first time in the human milk survey and its results should be compared with the values obtained at global level for PFAS [13,15] and within a trend assessment for all POPs [12]. Comparative data for PFAS in surface waters from the UNEP/GEF projects can be found in the paper by Baabish *et al.* [2]. Finally, all the results from the national samples

Fig. 1. OCPs and industrial POPs in each matrix (N = 30). Upper row: Stacked barplots for each sample (left); units in ng/g, ng/PUF, ng/g fat for human milk and MeDaEg, or ng/m^3 (left) and pattern at 100% (right). Lower row: Scaled box whisker plots by matrix); units in ng/g, ng/PUF, ng/g fat for human milk and MeDaEg, or ng/m^3 (AAS).

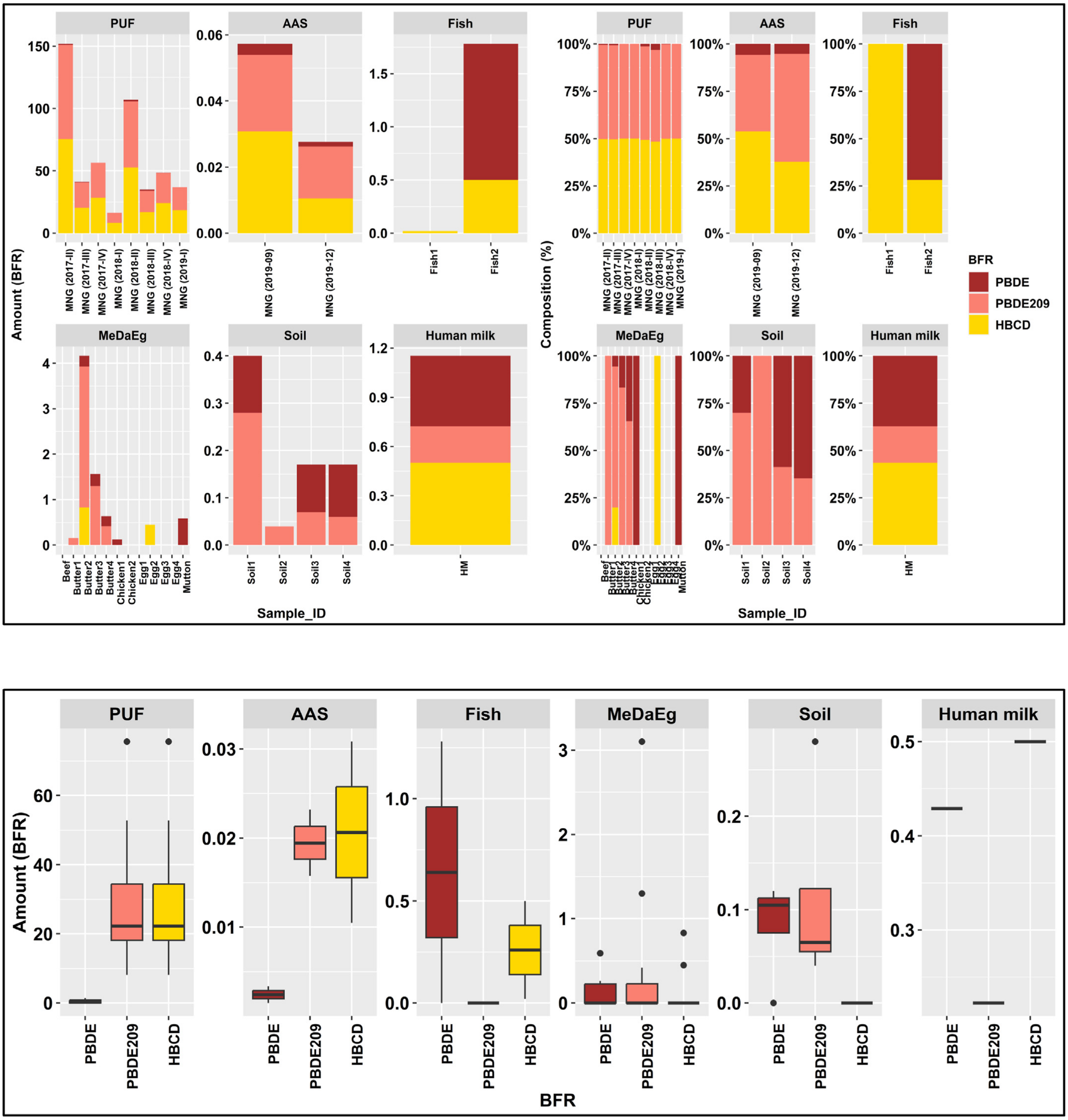


Fig. 2. BFR in each matrix (N = 29). Upper row: Stacked barplots for each sample by matrix; units in ng/g, ng/PUF, ng/g fat for human milk and MeDaEg, or ng/m^3 (left) and pattern at 100% (right). Lower row: Scaled box whisker plots by matrix; units in ng/g, ng/PUF, ng/g fat for human milk and MeDaEg, or ng/m^3 (AAS).

sent by almost 40 countries for PFAS and dl-POPs were presented and assessed [11,14].

Our present results complement a previous publication describing the scale of POPs in the core matrices of the Stockholm Convention in Mongolia [22]. In general, it was found that for Mongolia, POPs concentrations tend to be low, especially in biota samples. The legacy industrial chemicals, PCB$_6$, HCB, PeCBz, and HCBD, could be found in some of the foodstuffs. Dieldrin was present in egg samples. The soil samples demonstrated that locally, very high POPs contamination could be found; especially DDT, α-HCH, β-HCH, and lindane indicating past applications.

A study published in the Mongolian Journal of Chemistry [9] analyzed air and soil samples and reported PCB, HCHs, DDT, and HCB; thereafter referred to as “Lit 2014”. Our findings of the high contamination with OCPs from the cattle dips are in agreement with information compiled by Vijgen *et al.* [42] on European HCH

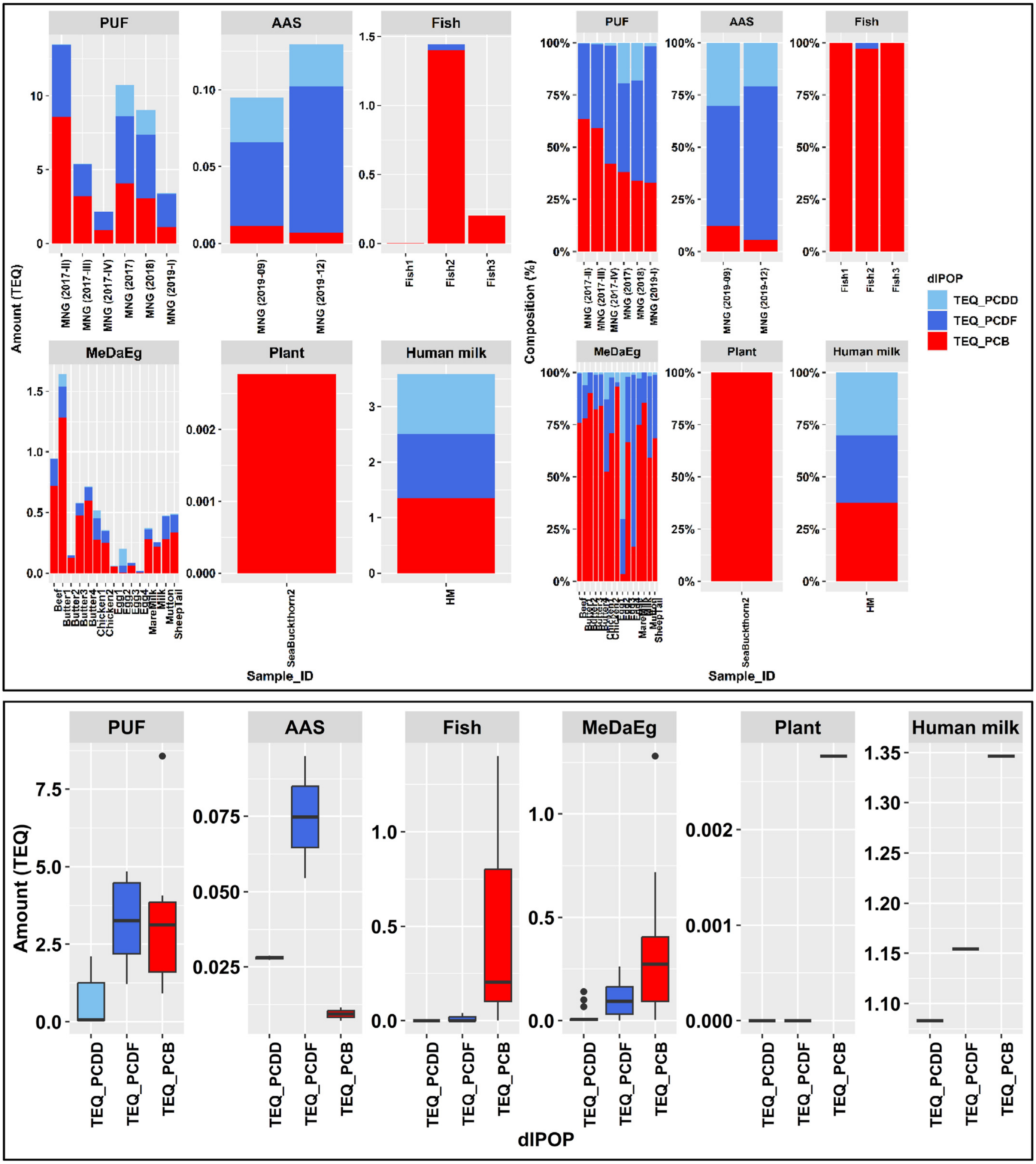


Fig. 3. Dioxin-like POPs (as TEQ) in each matrix (N = 28). Upper row: Stacked barplots for each sample; units in pg TEQ/g, pg TEQ/PUF, pg TEQ/g fat for human milk and MeDaEg, or pg/m^3 (left) and pattern at 100% (right). Lower row: Scaled box whisker plots by matrix; units in pg TEQ/g, pg TEQ/PUF, pg TEQ/g fat for human milk and MeDaEg, or pg/m^3 (AAS).

hotspots. Data for DDT, HCHs, and PCB from PAS/PUFs from the cities of Khovd and Ulaanbaatar were published together with those from other cities in East Asia [3]; thereafter referred to as “Lit 2013”. The data warehouse of the Stockholm Convention contained two samples from active air sampling obtained through the POPsEA project in 2006 and 2013 [37]; thereafter referred to as “GMP1”. The following Fig. 7 presents the pattern of the sums of DDT, PCB, HCHs and HCB, where available, from Mongolian air and soil samples, and compares them with our data; thereafter referred to as “GMP2”. The pattern within the DDT, HCH, and PCB$_6$ are contained in the

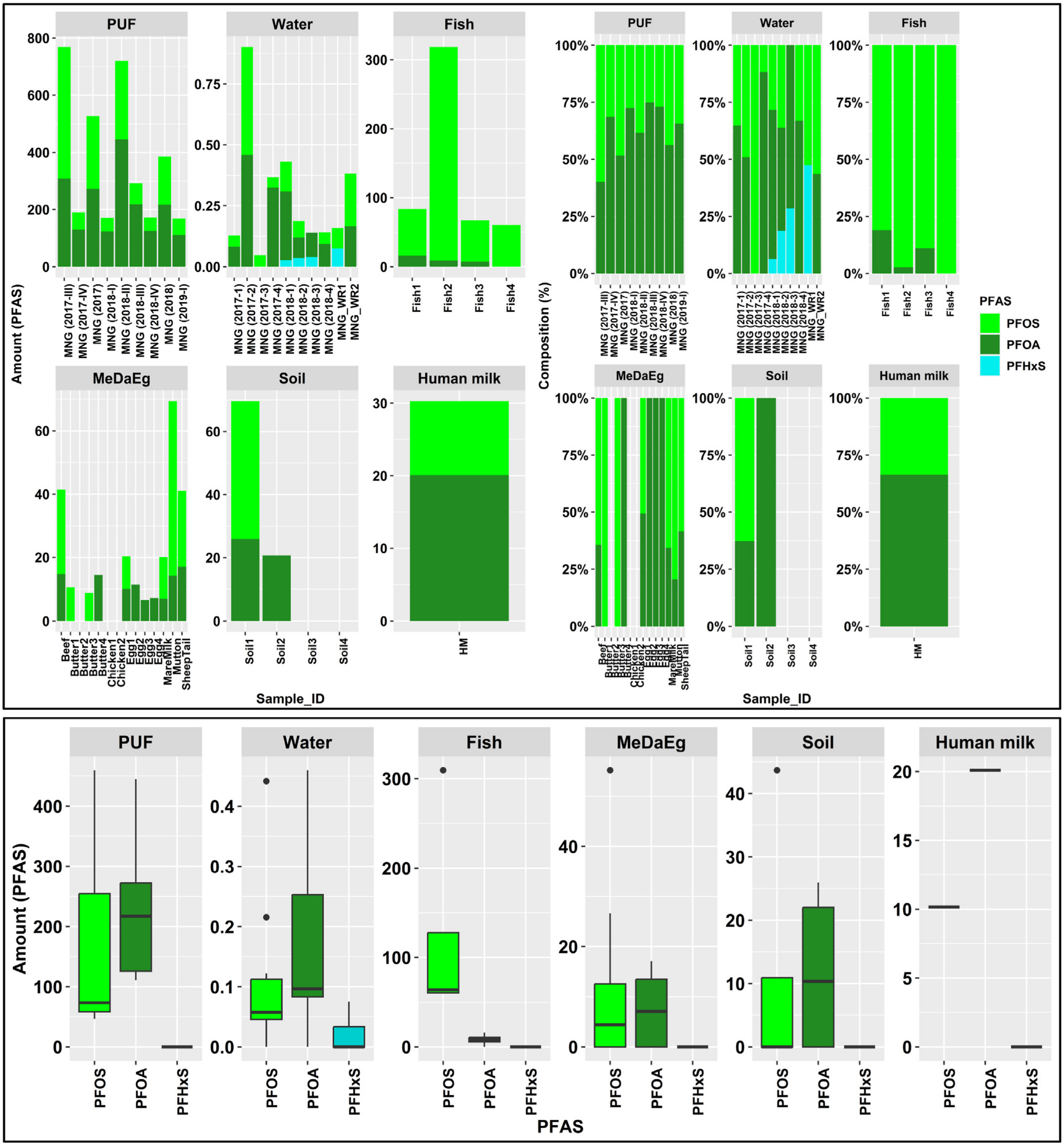


Fig. 4. PFAS in each matrix (N = 43). Upper row: Stacked barplots for each sample; units in pg/g f.w., pg/PUF or ng/L (for water) (left) and pattern at 100% (right). Lower row: Scaled box whisker plots by matrix; units in pg/g f.w., pg/PUF or ng/L (water) for POPs (lower row).

Supplementary information as Fig. S 5. Although not all the four POPs have been analyzed in all samples, some conclusion can be drawn: (i) Air samples, independently if were PAS/PUFs or AAS had more homogenous pattern than soil samples: Such finding is not surprising since soils most likely were impacted by nearby sources and are not as representative as the air samples. (ii) Air samples had a high contribution of *p,p*-DDT beside *p,p'*-DDE indicating relatively recent uses of DDT in Mongolia. (iii) Application of technical mixtures of HCHs with high α-HCH content was demonstrated in the soil samples from our project and was present at larger shares in the air samples. It shall be noted that HCHs in air samples had not been include since only α-HCH could be quantified [9]; however, the presence of α-HCH supports this conclusion. (iv) the high shares of PCB_6 in soil samples as was reported by Erdenebayasgalan *et al.*

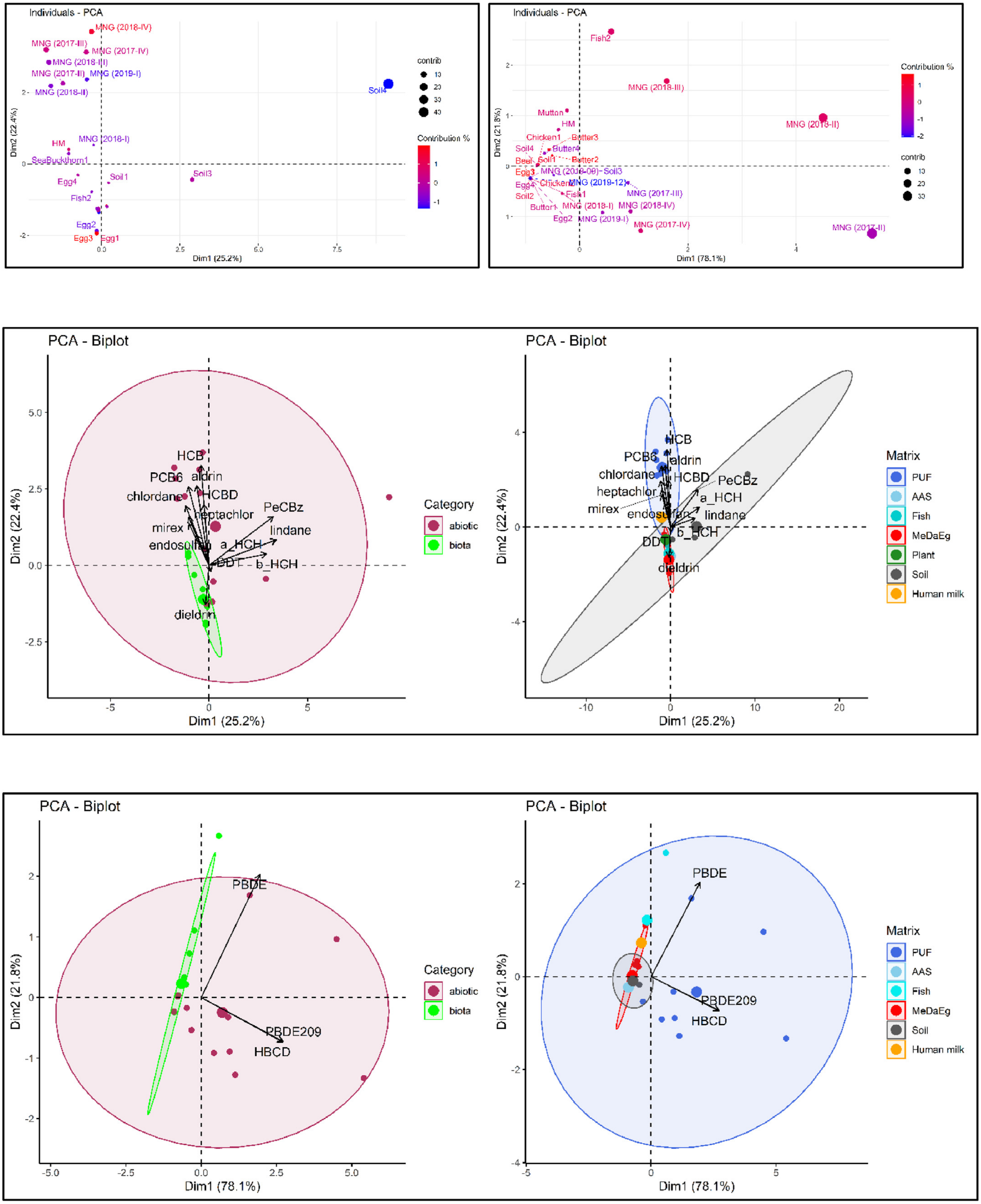


Fig. 5. PCA Row 1: Location of individuals for OCPs + ind POPs (left, N = 30) and for BFR (right, N = 29). Row 2: Biplot for OCPs + ind.POPs with ellipses around abiotic/biota samples and matrices. Row 3: Biplot for BFR with ellipses around abiotic/biota samples and matrices.

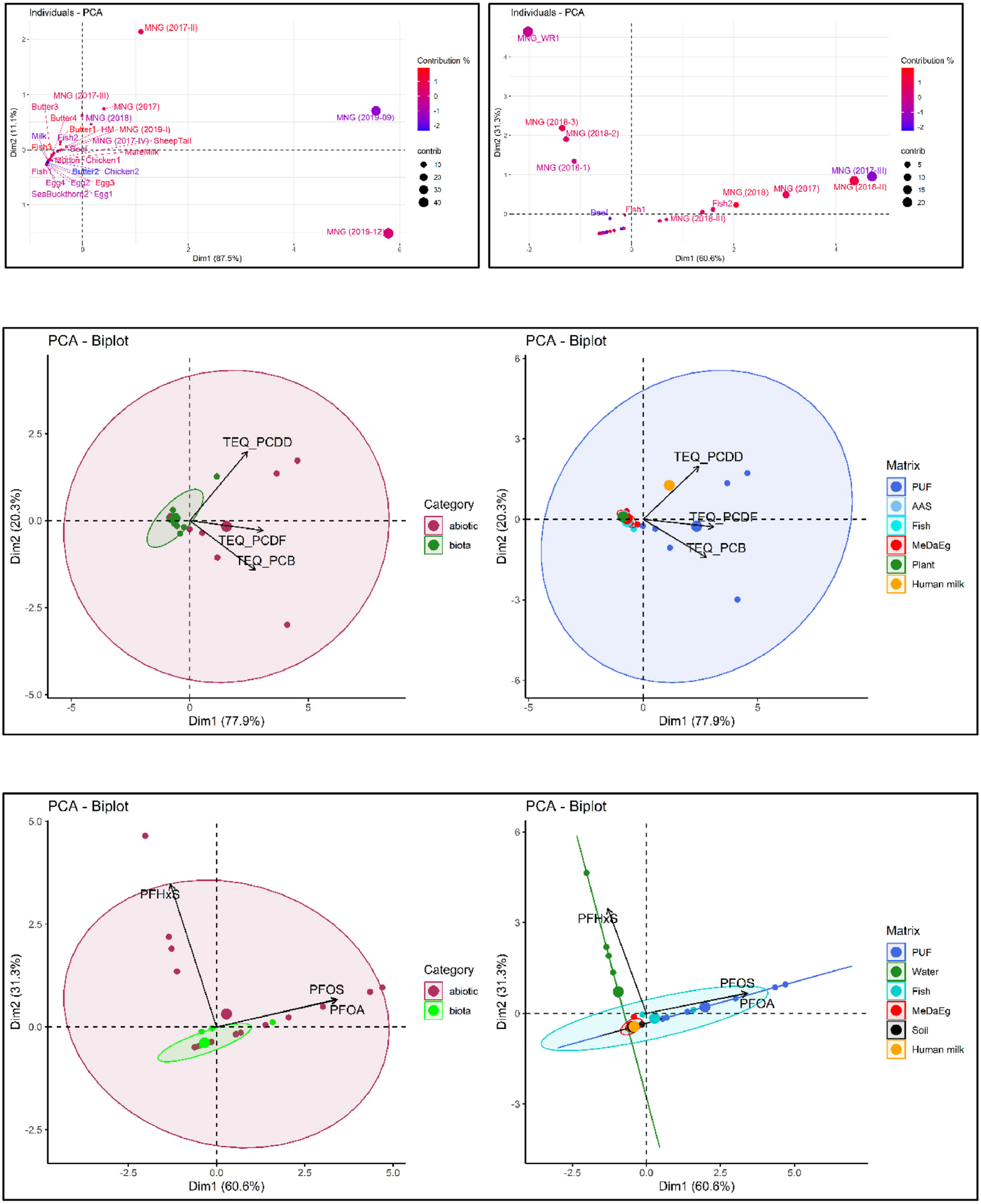


Fig. 6. PCA Row 1: Location of individuals for dl-POPs (left, N = 28) and for PFAS (right, N = 42). Row 2: Biplot for dl-POPs with ellipses around abiotic/biota samples and matrices. Row 3: Biplot for PFAS with ellipses around abiotic/biota samples and matrices.

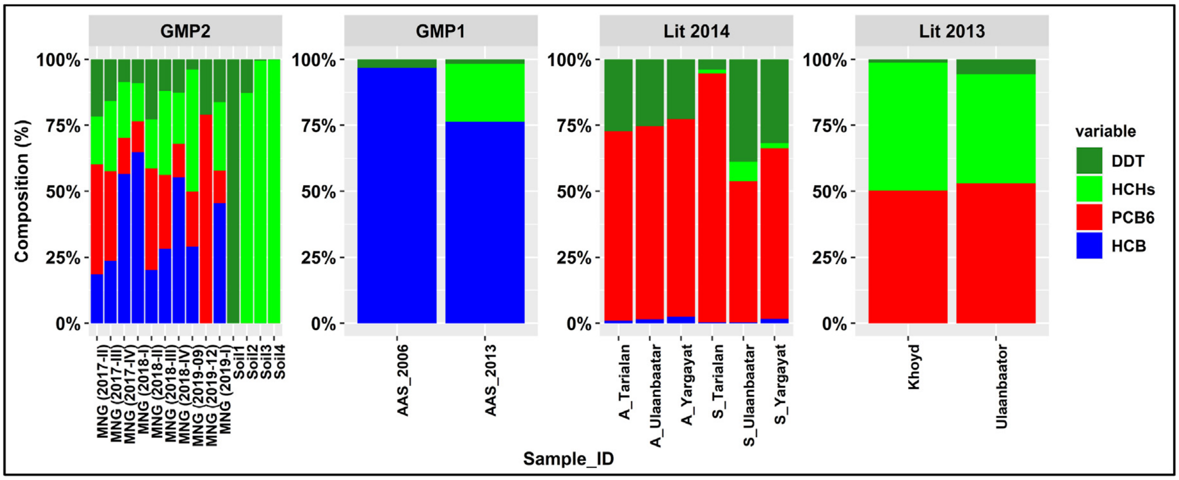


Fig. 7. Composition of major POPs in air and soil samples from Mongolia.

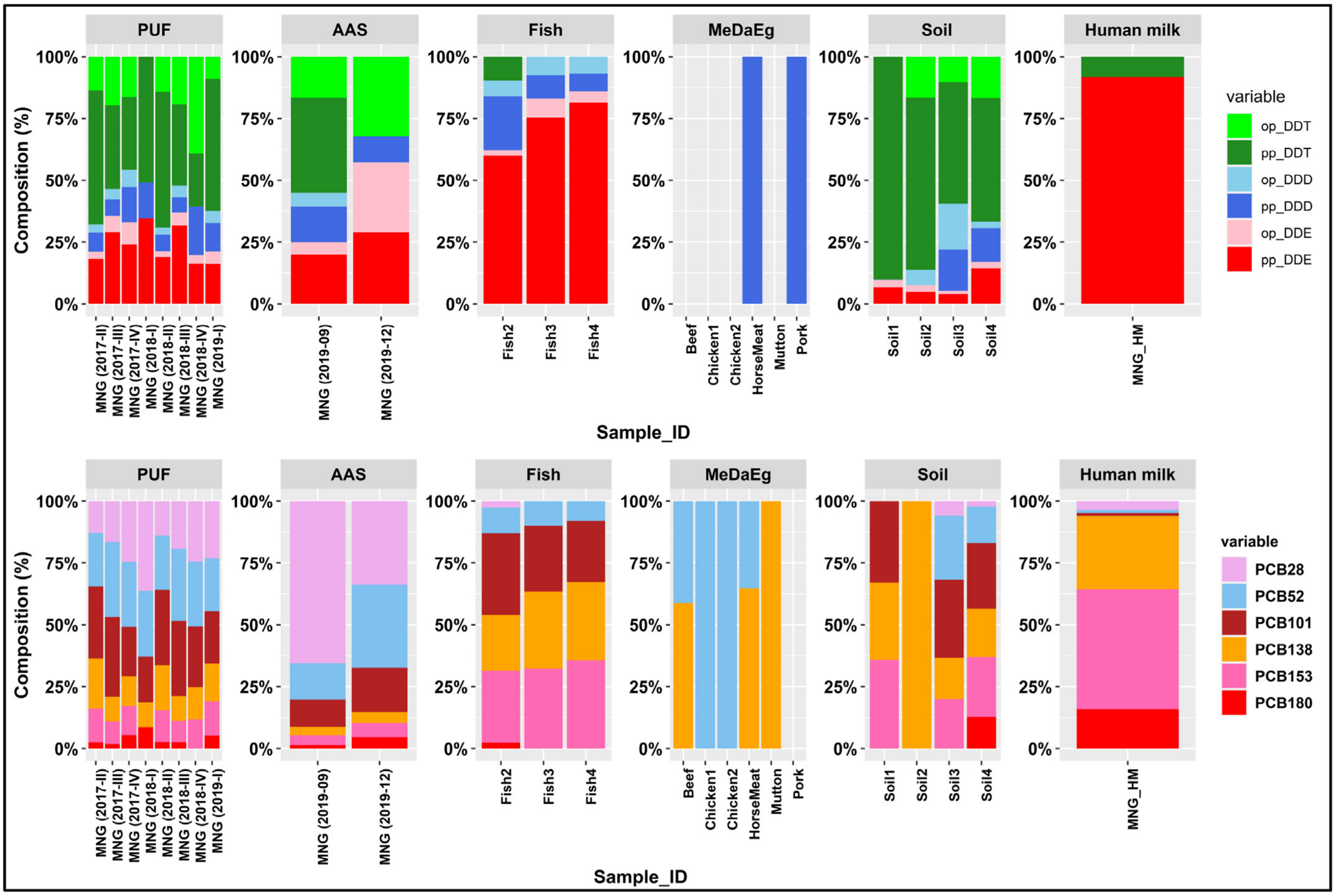


Fig. 8. Congener pattern for DDT and PCB_6 in Mongolian GMP2 samples.

and Baek *et al.* [3,9] was not found in our samples; however, it must be stated that our study targeted the OCPs uses and not PCB contamination. (v) HCB was found at high prevalence but was not often analyzed.

A pattern analysis of the POPs chemicals contributing to DDT and PCB_6 across all the GMP2 samples is shown in Fig. 8. The human milk sample is dominated by the more persistent and bioaccumulating compounds such as *p,p'*-DDE and the higher chlorinated PCB (PCB 138, PCB 153, and PCB 180). With respect to the DDT, the fish samples reflect closely the pattern of the human milk but not the PCB_6. The much higher shares of the lower chlorinated PCB (PCB 28, PCB 52, and PCB 101) in the fish samples indicate that whereas humans can metabolize especially the lower chlorinated PCB and thus, the pattern in humans is enriched by the higher chlorinated PCB, fish cannot. The abundance of the lower chlorinated PCB 52 and PCB-138 in the meats (group MeDaEg) is striking. Also, the soil samples were not dominated by the more persistent DDT and PCB as was expected. Only the three fish samples had ≫50% of ΣDDT from *p,p'*-DDE.

For comparison with other data reporting schemes, we converted our PAS/PUF results for dl-POPs to volume-based results using the equations as established by Harner [20]. There were three PAS/PUF samples corresponding to exposures of several months in 2017 (three samples, 3 months exposure each), 2018 (four samples, 3 months exposure each), and 2019 (one exposure from January through March 2019) and two samples using an AAS with three days of exposure in 2019. The characteristics of the samples are shown below.

Sample_ID	Year	Days	Temperature (°C)
MNG (2017-q234)	2017	271	9.3
MNG (2018-A)	2018	365	6.6
MNG (2019-I)	2019	93	−12.8

Fig. 9 shows very different scales of the data recalculated from the PAS/PUF measurements and the two AAS measurements; all values are in pg/m^3 for easy comparison. Whereas the amounts from the AAS measurements are not visible in the graphs. The pattern reveals for the 17 PCDD/PCDF congeners that they are very similar distributed for all samples, *i.e.*, the annual PUF samples (MNG (2017), MNG (2018), and MNG (2019) and the two AAS samples (MNG (2019–09) and MNG (2019–12)). The same was found for the dl-PCB, although especially one of the non-ortho substituted PCB, PCB 77, has higher shares in the AAS samples than the PUFs. The pattern for the TEQs shows clear differences between the 2017/2018 samples and the 2019 samples but very good distribution between the PUF and the volume composition. The pattern of the TEQ results is quite different with about 5 times lower contribution from the dl-PCB (red color).

5. Conclusions

The present study was the first systematic survey to monitoring POPs in the environment (air, soil, and water), in foodstuffs, and humans in Mongolia. The study results allowed for a prioritization of POPs, which will lead to follow-up studies and creation of national chemical analytical capacities to be used in future

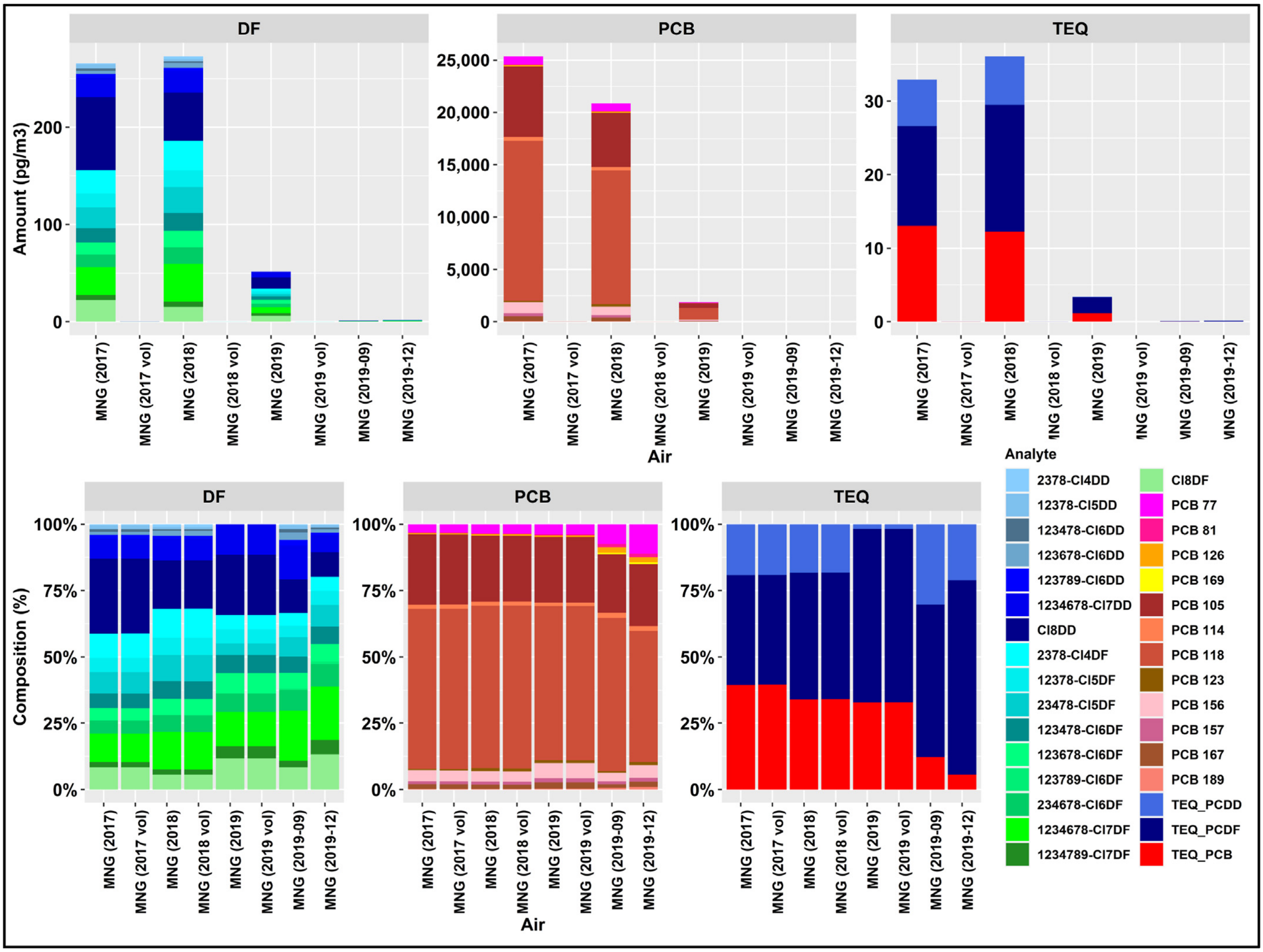


Fig. 9. Stacked barplots for sums of 17 PCDD/PCDF, 12 dl-PCB, and three TEQs for annual PUF samples for 2017, 2018, and 2019 (amounts in pg *per* PUF and amounts in pg/m^3 next to each other) and for two AAS measurements in 2019. At left: stacked bar plots; right: scaled to 100%.

programmes. Such analytical capacity should prioritize legacy POPs since these were more abundant in all matrices and especially DDT and HCH as there may be more contaminated sites from cattle dips; in addition, storage sites should be included. The presence of HBCD in air samples may need further investigation if Mongolia had used large amounts of EPS/XPS foams treated with HBCD in building insulation and despite low migration potential, HBCD was unintentionally released and are reflected in air and were quantifiable in human milk. On the other hand, brominated flame retardants, dioxin-like POPs, and PFAS were found at lower levels and may not warrant priority attention.

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.

Acknowledgments

The contribution of the UNEP/GEF project to support POPs monitoring in the Asian developing country regions financed by the Global Environment Facility (GEF) and implemented by UN Environment (GEF ID 4894, UNEP Code 4F32) is greatly acknowledged. The Ministry of Environment and Tourism, Mongolia, is acknowledged for their support during the implementation of GMP2 project.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.emcon.2023.100251.

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