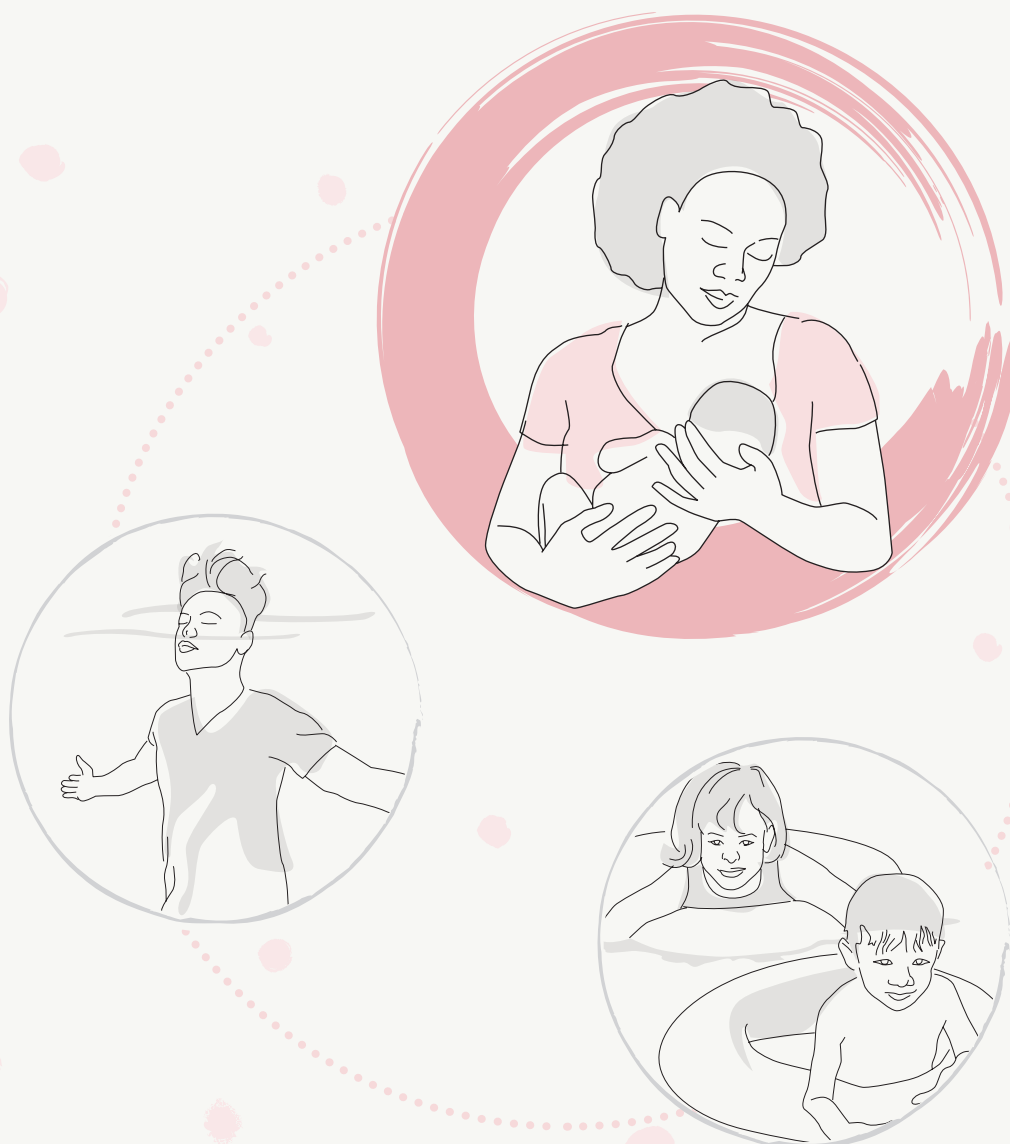


Sectoral Report

Results of the 2016-2019 WHO/UNEP

# Human Milk Survey

on Persistent Organic Pollutants



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# LIST OF ACRONYMS

BDE	brominated diphenyl ether
CPs	chlorinated paraffins
CV	coefficient of variation
DL-PCBs	dioxin-like PCBs
GEF	Global Environment Facility
GMP	Global Monitoring Plan
GRULAC	The Group of Latin America and the Caribbean
HBCD	hexabromocyclododecanes
MCCPs	medium-chain chlorinated paraffins
NDL-PCB	non-dioxin-like PCB
PBDEs	polybrominated diphenyl ethers
PCBs	polychlorinated biphenyls
PCDDs	polychlorinated dibenzo-p-dioxins
PCDFs	polychlorinated dibenzofurans
PCNs	polychlorinated naphthalenes
PFAS	perfluorinated alkane substances
PFHxS	perfluorohexane sulfonic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonic acid
POPs	persistent organic pollutants
PT	proficiency test
REP	relative effect potency
SC	Stockholm Convention
SCCPs	short-chain chlorinated paraffins
TEF	toxic equivalency factor
TEQ	toxic equivalent
UNEP	United Nations Environment Programme
WHO	World Health Organization
WHO-PCB-TEQ	sum of toxic equivalents of DL-PCBs
WHO-PCDD/PCDF-TEQ	sum of toxic equivalents of PCDD/PCDF
WHO-TEF	toxic equivalency factors assigned by WHO assessments (used here: factors of reevaluation in 2005)
WHO-TEQ	Total sum of toxic equivalents of mixtures of PCDD/PCDF and DL-PCBs (WHO-PCDD/PCDF-PCB-TEQ <sup>a</sup> ; "WHO <sub>2005</sub> -TEQ")

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## Executive summary

# EXECUTIVESUMMARY

This report presents the findings of the survey on Persistent Organic Pollutants (POPs) in human milk coordinated by the United Nations Environment Programme (UNEP) and the World Health Organization (WHO) from 2016-2019. This survey is part of the Global Environment Facility-funded UNEP POPs Global Monitoring Plan project in the Africa, Asia, The Pacific Islands and Group of Latin America and the Caribbean (GRULAC) regions. 36 project countries including 15 in Africa, 4 in Asia, 8 in the Pacific Islands and 9 in GRULAC submitted samples to this survey. Seven self-funded countries from other UN regions participated in this survey on a voluntary basis.

To ascertain comparability of results with previous WHO/UNEP-coordinated exposure studies carried out between 2000 and 2019, human milk samples were collected following WHO- and UNEP-designed protocols under the supervision of a national coordinators in each country. A large number of individual samples were collected. Subsequently, from equal amounts of these samples a pooled sample was prepared that was considered to be representative of the country.

In 2019 the POPs listed under the Stockholm Convention had increased to 30 chemicals or groups of chemicals (28 chlorinated or brominated, two perfluorinated). Since no multi-method exists that would have allowed the determination of all POPs of interest, various analytical methods with comprehensive quality control were applied.

The collection of human milk as a non-invasive sampling method, and preparation of pooled samples considered representative of a country, had a number of advantages. The most important are: i) this approach is highly cost-effective; ii) owing to the relatively high fat content of human milk and the large volume of the pooled (composite) samples, sufficient sample material was available to apply different methods in the determination of all 30 listed POPs as of 2019, as well as the two chemicals proposed at that time for listing under the convention.<sup>1</sup>

This project aimed at supporting the Convention's implementation by providing data for the effectiveness evaluation, as required under Article 16. Temporal tendencies in POPs concentrations are indicated for 24 of the 36 countries with repeated participation in WHO/

UNEP-coordinated exposure studies by comparing the 2016-2019 results with those for previous years. Moreover, these projects have contributed to the derivation of statistically significant time trends in the United Nations regional groups and globally on the basis of data from 82 countries for the period 2000-2019.

From the results of the human milk survey, there has been an observed decrease in the global general levels of legacy POPs such as DDT, PCB and dioxins. This indicates the difference made by restricting or banning the production and usage of legacy POPs and improving waste and emission management.

Concentrations of POPs in some areas remain high. Results of the 2016–2019 human milk survey showed that globally, DDT accounted for the largest proportion of POPs on average, followed by and chlorinated paraffins and PCB. Industrial POPs counted for about 60% of the total load of POPs in human milk in the Asia-Pacific Region and 40% in the Africa and GRULAC regions.

The high levels of new POPs listed under the Stockholm Convention such as chlorinated paraffins and PFASs raise concerns over their sources of exposure. This shows the need for further monitoring to address the expanding list of POPs and to support addressing gaps in policies and actions.

How the human milk survey was carried out, and its findings, are summarized in more detail in Part 4, summary and conclusions.

The WHO/UNEP human milk survey is both the largest and longest-running global study on human exposure to POPs. The continuation of this monitoring effort is vital in order to secure enough data for a proper time trend assessment. In total, 82 countries from all UN regions participated between 2000 and 2019. An ongoing effort with repeated country participation and an increased number of participating countries would further support the effectiveness evaluation of the Stockholm Convention.

Given the health impacts of many POPs, monitoring is crucial for risk prevention. Strengthening the collaboration between the environment and health sectors is also necessary to support cross-cutting and effective policy making.

<sup>1</sup> These chemicals were medium-chain chlorinated paraffins (MCCPs) and perfluorohexane sulfonic acid (PFHxS). In June 2022 the Conference of the Parties amended Annex A to the Convention as follows: "List perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds in Annex A without specific exemptions" (Decision SC-10/13) (Stockholm Convention 2022).

# SECTION 1

**Introduction: Concept and participating countries**



# 1. INTRODUCTION: CONCEPT AND PARTICIPATING COUNTRIES

## 1.1. Aim and concept of this report

Biomonitoring of persistent organic pollutants (POPs) can be used to assess integrated human exposure, occurring mainly through foods. It reflects the body burdens of POPs due to their long half-lives. Between 2000 and 2019 the World Health Organization (WHO) and the United Nations Environment Programme (UNEP) performed five global studies on concentrations of POPs in human milk. The last round was conducted in the period 2016-2019 through Global Environment Facility (GEF) funded POPs Global Monitoring Plan projects (UNEP/GEF POPs GMP-2). It supported implementation of the Stockholm Convention's POPs Global Monitoring Plan (GMP).

Human milk is a core matrix under the GMP. Human biomonitoring within the GMP includes WHO- and WHO/UNEP-coordinated human milk studies. The purpose is to evaluate the effectiveness of the Stockholm Convention by identifying temporal and, as appropriate, spatial trends in levels of POPs in humans.

The objective of this report is the compilation and interpretation of the results of the human milk survey conducted under the UNEP/GEF POPs GMP-2 projects in Africa, Asia, the Pacific Islands, and the Group of Latin America and the Caribbean including a summary of the outputs and outcomes. The project covered 42 invited countries. Samples were obtained from 36 of them. While the report could have limited the presentation and the discussion of the results to these countries and to the period 2016-2019, a widened view has been taken, including results for these 36 countries from earlier WHO/UNEP-coordinated human milk studies to indicate changes between the testing periods.

Furthermore, in the WHO/UNEP-coordinated human milk studies between 2000 and 2019, 82 countries from all United Nations regions (including the Eastern European Group [EEG] and the Western European and Others Group [WEOG]) participated, with 50 countries taking part in more than one study. They included seven self-funded countries in the period 2016-2019. With results publicly available through the

Stockholm Convention Global Monitoring Plan Data Warehouse (GMP DWH), a comprehensive review of this overall database is being prepared to support presentation of the data and outcomes of all WHO/UNEP-coordinated exposure studies on human milk to stakeholders by compiling them into a special issue with more than a dozen specific publications (Malisch, Fürst and Šebková 2023).

Therefore, two important aspects are covered in this report beyond a limited view of data only from this project:

Some of the 36 countries participated in earlier rounds of the studies. In these cases the results obtained between 2000 and 2015 are included for comparison with the most recent results and as an indication of temporal tendencies.

The overall ranges found in all 82 countries in the period 2000-2019 are provided as additional information for comparison. This approach allows the report to present and discuss the results obtained under the UNEP/GEF POPs GMP-2 projects as comprehensively as possible.

## 1.2. Guidelines

To ascertain comparability of results, human milk samples were collected under the supervision of a national coordinators in each country, following WHO- and UNEP-designed protocols, during the studies performed between 2000 and 2019. For the guidelines of the 2016-2018 survey, see *the Global Monitoring Plan: Protocol for POPs sampling - Human milk (UNEP 2017)*. Generally, the concept of the WHO/UNEP-coordinated exposure studies had four basic elements:

- Collection of a large number of individual samples from mothers based on the standardized WHO/UNEP protocol (since 2004, recruitment of 50 individual donors per pooled sample in countries with up to 50 million population);
- Preparation, from equal amounts of the individual samples, of pooled samples considered to represent the average levels of POPs in human milk for a country or subgroup/region of that country at the time of sampling;
- Performance of the analysis of the pooled samples at the reference laboratories for the WHO/UNEP-coordinated exposure studies in the period 2000-2019. For chlorinated and brominated POPs in the period 2000-2019 analyses were performed at the CVUA (State Institute for Chemical and Veterinary Analysis of Food; Chemisches und Veterinäruntersuchungsamt), Freiburg,

Germany, and for perfluoroalkane substances in the period 2009-2019 at Örebro University, Sweden. For the analytical results a high degree of reliability was achieved;

- Repeated participation of countries, permitting the assessment of temporal trends, which can be used for risk management purposes as well as for evaluating the effectiveness of the Stockholm Convention in eliminating or reducing emissions of POPs.

This approach is very cost-effective. The analysis of one or of a few pooled human milk samples, considered to be representative of a country or a subgroup/region, is far less expensive than the analysis of a high number of individual samples.

The option to have the individual samples analysed for old pesticide POPs and marker polychlorinated biphenyls (PCBs) by a competent national laboratory and the analysis of pooled samples by the reference laboratory is a contribution to capacity building, particularly in developing countries. Comparison of the mean of the individual samples for these analytes with the results of the reference laboratories serves as an internal check, as the average of the results of the individual samples should be comparable to the result of the pooled sample, which is prepared from equal aliquots of the individual samples.

### 1.3. Strategy for obtaining data on all chemicals listed under the Stockholm Convention and two additional POPs proposed for listing

Between the adoption of the Stockholm Convention in 2001 and 2019, the number of listed POPs in Annexes A (elimination), B (restriction) and C (reduction of releases from unintentional production) increased from the initial 12 to 30 chemicals or groups of chemicals. Many of these chemicals have numerous congeners, homologous groups, forms and transformation products, which significantly increases the number of recommended analytes (UNEP 2019). In addition to the 30 listed chemicals (28 chlorinated or brominated and two perfluorinated), two additional chemicals proposed for listing under the Convention were of interest (Table 1).<sup>2</sup>

As no multi-residue method exists that would allow the simultaneous determination of all analytes of interest, various analytical methods have to be applied (see Part 2).

<sup>2</sup> The two chemicals proposed for listing were medium-chained chlorinated paraffins (MCCPs) and perfluorohexane sulfonic acid (PFHxS). In June 2022 the Conference of the Parties amended Annex A to the Convention as follows: "List perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds in Annex A without specific exemptions" (Decision SC-10/13) (Stockholm Convention 2022).

The collection of human milk is a non-invasive sampling method. Thus it has many practical and procedural advantages over collection of other biological samples such as blood or adipose tissue. Furthermore, from an analytical point of view the relatively high fat content of human milk (about 4 per cent) makes the extraction of sufficient amounts of lipids easier. These aspects, in combination with the use of a large volume of pooled (composite) samples (often over 1 litre) considered to represent a country or a subgroup at the time of sampling, have significant advantages: i) considerably reduced costs; ii) simplified logistics; iii) lowered limits of quantification; iv) improved precision of measurements; and v) the possibility to apply various determination methods for all the POPs listed in the annexes to the Stockholm Convention as well as for the two chemicals proposed for listing as of 2019. With regard to the last point, large sample volumes were necessary for application of the various analytical methods.

Samples taken during the most recent survey (2016-2019) were therefore analysed for 32 POPs: for the first time data on all 30 listed POPs and the two chemicals proposed for listing became available for most samples. It should be noted that for small countries the collection of the desired number of individual samples for certain rounds was not always possible. In some cases collection of the recommended sample volume was not possible. Consequently, in a few cases a smaller subset of POPs was analysed.



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**Table 1:** Chemicals (30 listed POPs and two chemicals proposed for listing as of 2019) and recommended analytes covered by the UNEP/WHO human milk survey under the UNEP/GEF POPs GMP-2 project

Initial POPs (2001)		Recommended analytes for human milk
1	Aldrin	Aldrin
2	Chlordane	cis- and trans-chlordane; cis- and trans-nonachlor, oxychlordane
3	DDT	4,4'-DDT, 2,4'-DDT, 4,4'-DDE, 2,4'-DDE, 4,4'-DDD, 2,04'-DDD
4	Dieldrin	Dieldrin
5	Endrin	Endrin
6	HCB	HCB
7	Heptachlor	Heptachlor and heptachlorepoxide
8	Mirex	Mirex
9	PCB	ΣPCB6 (6 congeners): 28, 52, 101, 138, 153, and 180 PCB with TEFs* (12 congeners): 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189
10	PCDD	2,3,7,8-substituted PCDD (7 congeners)
11	PCDF	2,3,7,8-substituted PCDF (10 congeners)
12	Toxaphene	Congeneres P26, P50, P62
* PCB with TEFs (Toxic Equivalency Factors) assigned by WHO in 1998		
POPs listed at COP-4 (2009)		
13	Chlordecone	Chlordecone
14	Alpha-hexachlorocyclohexane	alpha-HCH
15	Beta-hexachlorocyclohexane	beta-HCH
16	Gamma-hexachlorocyclohexane	gamma-HCH
17	Hexabromobiphenyl	PBB 153
18	Pentachlorobenzene	PeCBz
19	Tetra- and pentabromodiphenyl ether	PBDE 47, 99; optional: PBDE 100
20	Hexa- and pentabromodiphenyl ether	PBDE 153, 154, 175/183 (co-eluting)
21	Perfluorooctane sulfonic acid	PFOS (linear and branched isomers)
POPs listed at COP-5 (2011)		
22	Endosulfan	alpha-, beta-endosulfan; endosulfan sulfate
POPs listed at COP-6 (2013)		
23	Hexabromocyclododecane	alpha-, beta-, gamma-HBCD
POPs listed at COP-7 (2015)		
24	Hexachlorobutadiene (in Annex A)	HCBD
25	Pentachlorophenol	[Pentachloranisole (PCA)]
26	Polychlorinated naphthalenes	[PCN (congeneres to be decided)]
POPs listed at COP-8 (2017)		
27	Decabromodiphenylether (DecaBDE)	PBDE-209
28	Short-chained chlorinated paraffins	[SCCP]
(24)	Hexachlorobutadiene (in Annex C)	HCBD
POPs listed at COP-9 (2019)		
29	Dicofol	[Dicofol]
30	Perfluorooctanoic acid	PFOA
Voluntary (POPs proposed for listing)		
31	Medium-chained chlorinated paraffins	[MCCP]
32	Perfluorohexane sulfonic acid	PFHxS [POP]: to be decided.

**Note:** The two chemicals proposed for listing were medium-chained chlorinated paraffins (MCCPs) and perfluorohexane sulfonic acid (PFHxS). In June 2022 the Conference of the Parties amended Annex A to the Convention as follows: "List perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds in Annex A without specific exemptions" (Decision SC-10/13) (Stockholm Convention 2022).

## 1.4. Participating countries

In early 2016, 42 countries from four regions (Africa, Asia, the Pacific Islands, and GRULAC) were invited to participate in this UNEP/GEF POPs GMP-2 project. For each country a national coordinator was identified. The national coordinator was to be responsible for overall planning and implementation of the survey, assisted by health, laboratory and administrative staff. Within the framework of this project CVUA Freiburg supplied glassware for sampling of individual breast milk samples and received shipment of the pooled breast milk sample back from the participating country for POPs analysis. CVUA Freiburg also handled the preparation of a sample aliquot to be shipped to Örebro University in Örebro, Sweden for subsequent perfluorinated alkane substances (PFAS) analysis.

After identification of the national coordinators, glassware was shipped to all countries in December 2016/January 2017. Pooled samples from 36 countries were received by CVUA Freiburg between November 2017 and October 2019. The six remaining countries did not send samples (*Table S1 in the Appendix*). In a few cases collection of the recommended sample volume was not possible and a smaller subset of POPs was analysed.

*Table 2* lists the 36 countries in Africa, Asia, the Pacific Islands and GRULAC that submitted samples in the 2016-2019 round. The years of their participation in the WHO/UNEP-coordinated human milk surveys between 2000 and 2015 are also shown. Data from the period 2000-2015 are available for 24 countries. In the case of repeated participation, results from 2016-2019 are also discussed in Part 3 of this report with regard to temporal trends.

## 2. ANALYTICAL METHODS AND QUALITY

Africa					
	2000-2003	2004-2007	2008-2011	2012-2015	2016-2019
Democratic Republic of the Congo			X		X
Egypt	X				X
Ethiopia				X	X
Ghana			X		X
Kenya			X		X
Mali			X		X
Mauritius			X		X
Morocco					X
Nigeria			X		X
Senegal			X		X
United Republic of Tanzania					X
Togo			X		X
Tunisia					X
Uganda			X		X
Zambia					X
Asia					
	2000-2003	2004-2007	2008-2011	2012-2015	2016-2019
Cambodia					X
Mongolia					X
Thailand					X
Viet Nam					X

The Pacific Islands					
	2000-2003	2004-2007	2008-2011	2012-2015	2016-2019
Fiji	X	X	X		X
Kiribati		X	X		X
Marshall Islands			X		X
Niue			X		X
Palau			X		X
Samoa			X		X
Solomon Islands			X		X
Vanuatu					X

GRULAC					
	2000-2003	2004-2007	2008-2011	2012-2015	2016-2019
Antigua and Barbuda			X		X
Argentina					X
Barbados			X		X
Colombia					X
Ecuador					X
Jamaica			X		X
Mexico			X		X
Peru			X		X
Uruguay			X		X

Table 2: Countries in Africa, Asia, the Pacific Islands, and GRULAC participating in the UNEP/GEF POPs GMP-2 (2016-2019) and their participation in the previous WHO/UNEP-coordinated human milk surveys (2000-2015)

# SECTION 2

## Analytical methods and quality control

# CONTROL

The analytical performance of a laboratory contributes to both the accuracy and precision of results. It can therefore enhance the interpretation of time trends. For all samples of the WHO/UNEP-coordinated exposure studies from the period 2000-2019, rigid quality control programmes were carried out by the Reference Laboratories to ensure high quality data and comparability of results.

## 2.1. Chlorinated and brominated POPs

Chlorinated and brominated POPs were determined by CVUA Freiburg as the reference laboratory for these analytes during the WHO/UNEP-coordinated exposure studies in 2000-2019. This laboratory was designated as the WHO reference laboratory for the 2000-2003 round of the exposure studies and for studies thereafter, as it met all performance criteria for analyses in a quality assessment study on levels of marker and dioxin-like PCBs, polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in human milk conducted by WHO (UNEP 2017).

To further ensure consistency in measurements of the subsequent exposure studies organized by WHO and UNEP, all samples were analysed by CVUA Freiburg for the chlorinated and brominated POPs listed under the Stockholm Convention using validated methods. Successful annual participation in international proficiency tests (PTs) has been part of the comprehensive quality control programme of CVUA Freiburg as an accredited laboratory since 1998. The analytical methodology used fulfils the requirements of the general criteria for the operation of testing laboratories as laid down in International Organization for Standardization (ISO) and International Electrotechnical Commission (IEC) standard ISO/IEC 17025 – Laboratory Competence.

In 2006 CVUA Freiburg was designated as the European Union reference laboratory (EURL) for PCDDs, PCDFs and PCBs in feed and food and as the EURL for pesticide residues in food of animal origin and commodities with high fat content. In 2018 the tasks of the EURL with regard to PCDDs, PCDFs and PCBs in feed and food were extended to all halogenated POPs. With respect to the analysis of human milk for WHO and UNEP-coordinated exposure studies, the overlapping responsibilities of CVUA Freiburg had significant synergistic effects, in particular regarding development of analytical methods and quality control.

### 2.1.1. Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs)

The analytical method used for the determination of PCDDs, PCDFs and PCBs in human milk comprised the extraction of lipids, the use of all relevant  $^{13}\text{C}_{12}$  labelled PCB and PCDD/PCDF internal standards, several chromatographic purification steps, and high-resolution gas chromatography coupled with high-resolution mass spectrometry (HRGC/HRMS). A comprehensive quality control programme was used to assure the reliability of the results of the human milk samples received for WHO/UNEP-coordinated exposure studies between 2000 and 2019. This included procedural blanks, the use of fortified vegetable oil and numerous quality control samples as in-house reference material, duplicate analyses, and successful participation in 32 proficiency tests (PTs) covering 81 samples of food of animal origin or human milk.

Trueness was estimated from the PT samples in the relevant range for human milk above 1 pg (picogram) WHO-TEQ2005/g lipid: the deviation was less than 10 per cent from the assigned values for WHO-PCDD/PCDF-PCB-TEQ (= WHO-TEQ2005) and WHO-PCDD/PCDF-TEQ and less than about 15 per cent for WHO-PCB-TEQ for about 90 per cent of the results. For the sum of six non-dioxin-like PCBs ("marker PCBs"; relevant occurrence range 1-1000 ng/g [nanograms per gram] lipid), approximately 90 per cent of the results differed by less than 15 per cent from the assigned values. A long-term precision of <15 per cent (coefficient of variation of within-laboratory reproducibility) was achieved, based on quality control samples analysed in 2000 and 2019.

The analytical methods fulfilled the requirements of the analytical criteria for PCDDs/PCDFs and PCBs in feed and food specified in European Union (EU) legislation and the criterion for monitoring information for Parties to the Stockholm Convention.

### 2.1.2. Organochlorine pesticides and industrial contaminants

The analytical method for the determination of non-polar organochlorine pesticides and industrial contaminants in human milk comprised the extraction of lipids, the use of internal standards, and two chromatographic separation steps for clean-up and gas chromatography on various columns of different polarity and different detectors (GC-ECD, GC-MS/MS). The more polar analytes were determined by applying the Quick Easy Cheap Effective Rugged Safe (QuEChERS) method and high-performance liquid chromatography tandem mass spectrometry (HPLC-MS/MS) measurements.

For many chemicals it is recommended not only to determine concentrations of the parent molecule, but also to include certain metabolites, degradation products, and/or by-products during manufacture. The sum of the parent POP and its other compounds of concern can be calculated based on the determined levels with two options: i) without correction for molecular weight, or ii) after correction for molecular weight. The *Guidance on the Global Monitoring Plan for Persistent Organic Pollutants* (UNEP 2019) does not provide guidance on this important analytical detail.

The sum parameters in this report (complexes) were calculated after correction of the detected analytes using correction factors for molecular weight and applying the “lower bound approach”, which uses only quantifiable results. This follows the principles for regulations for maximum levels in feed and food according to existing EU regulations (Malisch *et al.* 2008) and is harmonized internationally with those of the Codex Alimentarius Commission. As an example, for the DDT complex the sum of o,p'-DDT, p,p'-DDT, p,p'-DDE and p,p'-DDD was calculated using correction factors for molecular weight for p,p'-DDE and p,p'-DDD. Similarly, other complexes for chlordane, heptachlor, endrin and endosulfan were calculated using the correction factors as listed in *Table S2*.

A comprehensive quality control programme was applied to prove the long-time reliability of results between 2000 and 2019, comprising, for example, numerous quality control samples and participation in 49 proficiency tests covering test samples of food of animal origin, plant oils and solutions between 2000 and 2019. The assessment of long-term quality control charts with spiked samples for mean recoveries showed a range of 86 to 111 per cent (median 99 per cent) for 29 analytes with a median

coefficient of variation (CV) of 12 per cent. Furthermore, left-over samples of oil of plant and animal origin were spiked with several organochlorine pesticides from PTs and analysed as in-house quality control samples. For these samples the mean recovery was 95.0 per cent with a CV of 7.6 per cent. Then, between 2000 and 2019, CVUA Freiburg participated in 53 proficiency tests (PTs) to determine pesticide residues in test samples, mainly of food of animal origin, but also plant oils and, in two cases, human milk. The average deviation from the assigned value was 14 per cent; about 90 per cent of the results differed by less than 21 per cent from the assigned value.

The Stockholm Convention guidance criteria for monitoring of POPs were met, as well as the analytical criteria for analyses of organochlorine pesticides and industrial contaminants in feed and food set in EU legislation (“Analytical quality control and method validation procedures for pesticide residues analysis in food and feed”, Document No. SANTE/12682/2019).

### 2.1.3. Polybrominated diphenyl ethers (PBDEs)

Six prevalent analytes for the group from tetra-BDE to hepta-BDE (BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-175/183) were recommended as analytes for PBDE analysis. The sum of these six PBDEs was used as the most important summarizing parameter until 2017, when BDE-209 was added. The analytical method used for the determination comprised the extraction of lipids, use of <sup>13</sup>C<sup>12</sup> labelled PBDE as internal standards for the six prevalent BDE and BDE-209, several chromatographic purification steps, and HRGC/HRMS measurements.

Six different quality control samples were used as in-house reference material for monitoring the precision of PBDE analysis between 2004 and 2021. CVs for the sum parameters  $\sum$  PBDE<sub>6</sub> and  $\sum$  PBDE<sub>7</sub> were between 9 and 16 per cent and between 6 and 34 per cent, respectively. Between 2006 and 2021 CVUA Freiburg also participated in numerous rounds of interlaboratory studies and proficiency tests for PBDEs, generating results for individual congeners and sum parameters for up to 53 different food test samples. The mean absolute deviation of the reported results from the assigned values was 12 per cent for  $\sum$  PBDE<sub>6</sub> and 14 per cent for  $\sum$  PBDE<sub>7</sub>. Thus the criterion for monitoring information for the Parties to the Stockholm Convention was met.

#### 2.1.4. Hexabromocyclododecane (HBCD)

The analytical method for determination of alpha-HBCD, beta-HBCD and gamma-HBCD comprised the extraction of lipids, the use of internal standards, two chromatographic separation steps for clean-up, and HPLC-MS/MS for determination. The CV of 106 replicates of a quality control sample used as in-house reference material was between 15 and 17 per cent for these three stereoisomers. While gamma-HBCD is the main compound in technical HBCD, alpha-HBCD is more persistent in the environment and biota, including humans, and was the predominant stereoisomer in human milk. The mean deviation of alpha-HBCD based on 31 results from proficiency tests was 24 per cent, while that of the sum parameter for alpha-, beta- and gamma-HBCD was 27 per cent.

#### 2.1.5. Chlorinated paraffins

The analytical method for determination of chlorinated paraffins (CPs) consisted in the extraction of lipids, the use of internal standards, two chromatographic separation steps for clean-up and, until 2016, gas chromatography-electron ionization-triple quadrupole mass spectrometry (GC-EI-MS/MS) determination. This allowed for reliable and comparable quantification of the total CP amount in the sample, but not for distinguishing between short-chain CPs (SCCPs) and medium-chain CPs (MC-CPs) (Krätschmer and Schächtele 2019). In 2016 a new quantification method using gas chromatography, coupled with electron capture negative ion high-resolution Orbitrap mass spectrometry (GC-ECNI-Orbitrap-HRMS) technology, was established for CPs. The very high mass resolution of this instrument allowed for a differentiation between SCCPs and MCCPs, while operation in full scan mode reduced analysis time compared to other GC-ECNI-MS methods (Krätschmer 2021).

A comprehensive quality control programme was undertaken to prove the long-term reliability of results between 2012 and 2021. It included spiked samples containing different levels and different kinds of quality control samples. Possible systematic errors were checked by the analysis of reference materials or participation in numerous interlaboratory studies. Initial validation of the GC-ECNI-Orbitrap-HRMS method with two different kinds of matrices at two different levels showed recoveries in the range of 84 and 105 per cent with CVs between 2 and 5 per cent. Between 2017 and 2020 several different matrices were prepared as quality control samples to provide a good fit

with the sample matrix. In the case of human milk samples, raw cow's milk was analysed as a procedural blank and fortified with SCCP and MCCP standards. Besides such matrix-specific QC samples, fortified coconut fat or lard samples from the 2017 and 2018 interlaboratory studies on SCCPs and MCCPs in food were routinely added to each sample batch, providing a more accurate view of the long-term stability and repeatability of the method. The recoveries of the fortified raw cow's milk samples analysed in tandem with each human milk batch 2017-2020 for SCCPs, MCCPs and  $\Sigma$  CPs were between 80 and 120 per cent and thus within the warning levels for daily quality control.

Interlaboratory studies and PTs on chlorinated paraffins in biota are, to this day, very rare. The performance of the GC-EI-MS/MS method was compared to that of other laboratories in the interlaboratory testing scheme organized by Quality Assurance of Information in Marine Environmental Monitoring in Europe (QUASIMEME) in 2011-2017, which focused on SCCPs in environmental matrices and standard solutions. Additionally, the EURL for Halogenated Persistent Organic Pollutants in Feed and Food (EURL POPs) organized yearly interlaboratory studies and, later, PTs on SCCPs and MCCPs in food matrices starting in 2017.

Due to the very complex and specialized field of analysis of the GC-ECNI-Orbitrap-HRMS method used in the second study to determine SCCPs and MCCPs separately, the number of participants in each study was comparatively lower than for well-established analyte groups. This led to some evaluations being only provisional or completely impossible. All z-scores achieved in interlaboratory comparisons for the duration of the human milk studies discussed in the results for chlorinated paraffins were within  $\pm 2$  z and thus satisfactory.

#### 2.1.6. Polychlorinated naphthalenes (PCNs)

The analytical method for determination of 26 native PCN congeners comprised the extraction of lipids, use of eight  $^{13}\text{C}_{10}$  labelled standards, sample purification by DEXTech Plus on three chromatographic columns for clean-up, and HRGC/HRMS determination. Confirming measurements were carried out using GC-Orbitrap Q Exactive (ThermoFisher) at resolution 60,000. Quality control parameters were based on the sum of 26 congeners and 13 congeners showing toxicological relevance due to high REP factors. The mean recoveries for the quality control of milk fat and butter samples for each of the 26 PCN congeners were in the range of 86 per cent (PCN 42) and 131 per cent (PCN 31) with a CV between 0.7 and 11 per cent.

In order to control the analytical performance at different concentration levels of PCN congeners, two quality control samples (fish oil contaminated at different levels of single PCN congeners) were analysed together with human milk samples. With these samples, the linearity of the response for PCN (range between 0.2 pg/g lipid for PCN 70 and 130 pg/g lipid for PCN 52/60) was checked. The CV of 7 per cent for  $\Sigma\text{PCN}_{26}$  and 6 per cent  $\Sigma\text{PCN}_{13}$  obtained by repeated analysis as a quality control sample demonstrated the high precision of the analytical method. The two quality control fish oil samples that were analysed together with the human milk samples did not show exceedance of any warning level or control level for the individual 26 PCN congeners. In conclusion, based on this quality control sample, the methodology achieved a long-term precision of below 10 per cent over the period 2019-2021 for the  $\Sigma\text{PCN}_{13}$  and  $\Sigma\text{PCN}_{26}$ .

Due to the lack of available PTs at the time the human milk samples from the period 2016-2019 were measured, an external validation for the control of trueness was performed as an interlaboratory comparison with an independent laboratory. The maximal deviation of the  $\Sigma\text{PCN}_{13}$  in five samples between the external lab and CVUA Freiburg was 20 per cent.

At a later stage, CVUA Freiburg took part in an interlaboratory comparison study on cod liver oil for 26 polychlorinated naphthalene (PCN) congeners conducted by the EURL-POPs in the second half of 2021. The number of participants in this first interlaboratory study for PCNs was comparatively lower than for well-established analyte groups. Assigned values could be derived for four congeners. For these parameters the z-scores achieved by CVUA Freiburg were within  $\pm 2$  z and thus satisfactory. The results submitted by CVUA Freiburg for all 26 PCN congeners were in accordance with the median of the results of all participants.

## 2.2. Perfluorinated alkane substances (PFAS)

With the inclusion of perfluorooctane sulfonic acid (PFOS) and related compounds in 2009, additional expertise was needed and perfluorinated chemicals were analysed at the Man-Technology-Environment (MTM) Research Centre of Örebro University, Örebro, Sweden.

The target analytes – PFOS, perfluorooctanoic acid (PFOA) and perfluorohexane sulfonic acid (PFHxS) – were extracted using alkaline digestion and solid liquid extraction followed by weak anion exchange, solid-phase extraction and additional clean-up. The target PFAS were separated and quantified by using an ultra-performance liquid chromatograph electrospray interface (ESI) operating in the negative ion mode coupled to a triple-quadrupole mass spectrometer detector. The validation of the method applied to food samples resulted in recoveries >70 per cent for all three PFAS. The limits of quantification (LOQs) in all food matrices were 3.1 pg g<sup>-1</sup>, 3.4 pg g<sup>-1</sup>, and 4.9 pg g<sup>-1</sup> for PFHxS, PFOA and L-PFOS, respectively (Sadia, Yeung and Fiedler 2020). A procedural blank and one quality control sample (human milk sample obtained from the fourth round of the WHO/UNEP-coordinated Biennial Interlaboratory assessment) were run together with the human milk samples. The recoveries of mass-labelled standards were in the range of 95-97 per cent for the three targeted PFAS (Fiedler and Sadia 2021).

# SECTION 3

## Results and discussion





# 3. RESULTS AND DISCUSSION

The results of the pooled samples from the 36 countries participating in the UNEP/GEF POPs GMP-2 project in the period 2016-2019 are discussed below on a regional basis (Africa, Asia, the Pacific Islands and GRULAC), including results from previous periods. Data from the period 2000-2015 are available for 24 countries. Complementing this data and for comparison, the ranges found for 82 countries from all UN regions (including EEG and WEOG) participating in the WHO/UNEP-coordinated human milk studies between 2000 and 2019 are given.

Temporal trends for POPs in human milk can be assessed by considering only countries with repeated participation in WHO/UNEP-coordinated exposure studies. In contrast to a general estimation of time trends from all participating countries, this is a more precise approach because levels among countries are often highly variable. However, only a very few time points from these 36 countries are available, which prevents deriving statistically significant temporal trends in these cases. Yet the existing data can indicate decreasing or increasing tendencies in POP concentrations. Statistically significant time trends in the UN regional groups and globally can be derived on the basis of data from all 82 countries during the period 2000-2019.

## 3.1. Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs)

Of the theoretically possible congeners of PCDD and PCDF (75 PCDDs and 135 PCDFs), the 17 congeners that have at least four chlorine atoms with substitution in the 2,3,7,8-positions were shown to cause similar toxic responses and considered to be of particular relevance for human health. Similarly, from the 209 theoretically possible PCB congeners, 12 congeners (eight mono-ortho substituted and four non-ortho substituted) have dioxin-like properties. These congeners show different toxic potencies that are expressed as toxic equivalency factors (TEFs) compared to the most toxic congener, 2,3,7,8-tetrachloro dibenzo-p-dioxin (2,3,7,8-TCDD). With the TEF the toxicity of a mixture of PCDD/PCDF and dioxin-like PCB (DL-PCB) can be expressed in a single number, the toxic equivalent (TEQ). This is defined by the sum of the products of the

concentration of each compound (17 PCDD/PCDF congeners with 2,3,7,8-substitution and 12 DL-PCB congeners) multiplied by their corresponding TEF value. This is an estimate of the total 2,3,7,8-TCDD-like toxicity of the mixture. WHO2005-TEF values, as proposed at the WHO-IPCS expert meeting held in 2005, were applied (Van den Berg *et al.* 2006).

The acceptable difference between lower- and upper-bound values is of particular importance for the analysis of samples intended to be used as a control of time trends for the effectiveness evaluation of the Stockholm Convention. If the difference is too great, changes in WHO-TEQ levels might actually be caused by changes in the analytical sensitivity and not by changes of the real levels of POPs in samples. In particular, samples with limited amounts or those with low fat levels are at considerable risk of showing a high difference between lower- and upper-bound WHO-TEQ levels.

Therefore, regardless of whether human milk, human blood, air or other matrices are analysed, all studies intended to be used for an effectiveness evaluation of the Stockholm Convention should report lower- and upper-bound WHO-TEQ levels that are within the acceptable range. The 2019 version of the *Guidance on the Global Monitoring Plan for Persistent Organic Pollutants* (UNEP 2019) recommends that this difference between the lower-bound (LB) and upper-bound (UB) concentrations should be less than 20 per cent. *Tables S3-S5* present the results of all relevant congeners and the resulting sum parameters, including for TEQ levels. The LB and UB results are identical in nearly all cases and differ by about 0.1 per cent in other cases. Therefore, the criterion of the GMP guidance document is met.

The following sum parameters are used: i) the sum of six Indicator PCBs ( $\Sigma\text{PCB}_6$ ) for non-dioxin-like PCB (NDL-PCB), including congeners number 28, 52, 101, 138, 153 and 180; ii) the sum of toxic equivalents (TEQ) of PCDD/PCDF (WHO-PCDD/PCDF-TEQ); iii) the sum of toxic equivalents (TEQ) of dioxin-like PCB (WHO-PCB-TEQ); and iv) total sum of toxic equivalents ("total TEQ") of mixtures of PCDD/PCDF and dioxin-like PCB (WHO<sub>2005</sub>-TEQ).

Overall, in samples from the 82 countries during the period 2000-2019 the highest concentrations in the single pooled samples were 41.2 pg/g for TEQ from PCDD and PCDF, 13.6 pg/g for TEQ from dioxin-like PCB and 49.0 pg/g for total TEQ, all found during the 2000-2003 round. The highest concentration in single pooled samples was 1009 ng/g for the sum of six indicator PCBs in the period 2000-2003.

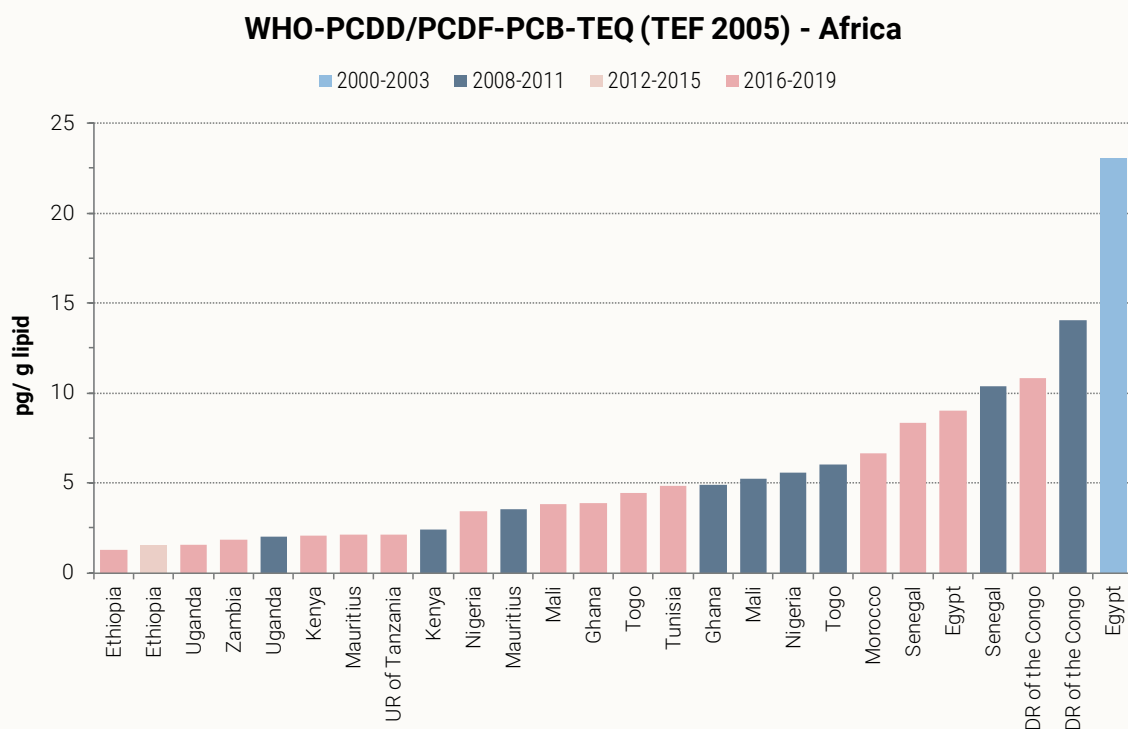
### 3.1.1. Africa

Over the whole period between 2000 and 2019 Africa had the widest variation in contamination of human milk with total TEQ observed in any group. *Figure 1* illustrates these results, with the five four-year studies between 2000 and 2019 shown in different colours as in some of the other figures in this report.

At one extreme was Ethiopia, with the lowest levels of total TEQ of any country in the 2000-2019 studies (1.54 pg WHO<sub>2005</sub> TEQ/g in 2012 and 1.29 pg/g in 2019). At the other was Egypt with a median of 23.0 pg WHO<sub>2005</sub>-TEQ/g for nine pooled samples collected in 2001 and 2002, which are comparable to levels found in Europe at that time. With 49.0 pg WHO<sub>2005</sub>-TEQ/g, one of the pooled samples from Egypt submitted in 2001 had a very high level of total TEQ and was probably from a contaminated area. The pooled 2019 sample from Egypt (9.0 pg WHO<sub>2005</sub>-TEQ/g) had considerably lower concentrations.

Uganda (2009 and 2018), Zambia (2019), Kenya (2009 and 2019), Mauritius (2018) and the United Republic of Tanzania (2019) were at the lower end of the frequency distribution among African countries (below 3 pg WHO<sub>2005</sub>-TEQ/g). Nigeria (2008 and 2019), Mali (2009 and 2019), Ghana (2009 and 2019), Tunisia (2019) and Morocco (2019) were in the middle (range 3-7 pg WHO<sub>2005</sub>-TEQ/g). Senegal (2009 and 2018), the Democratic Republic of the Congo (2009 and 2017) and, as mentioned, Egypt (2001 and 2019) were in the upper third of the frequency distribution in the African group (range 8-23 pg WHO<sub>2005</sub>-TEQ/g).

Temporal tendencies of WHO<sub>2005</sub>-TEQ concentrations can be discussed for 11 countries with repeated participation between 2000 and 2019. Most countries participated for the first time during the period 2008-2011; in these countries WHO<sub>2005</sub>-TEQ concentrations fell on average by about 22 per cent (range 14-40 per cent) in comparison to 2016-2019. The highest declining rate was observed in Egypt, where PCDD/PCDF showed very high levels in 2001-2002.



**Figure 1:** Results of the 2016-2019 survey for total TEQ in human milk (pg WHO-PCDD/PCDF-PCB-TEQ [TEF 2005]/g lipid) in countries in Africa in comparison to three previous surveys (with an indication of the period)

*Figure S1* illustrates the relative contributions of toxic equivalency resulting from PCDD (WHO-PCDD-TEQ), PCDF (WHO-PCDF-TEQ) and dioxin-like PCB (WHO-PCB-TEQ) to total TEQ (WHO-PCDD/PCDF-PCB-TEQ) in human milk in

countries in Africa. Interestingly, the two countries with the highest levels of total TEQ, the Democratic Republic of the Congo (2009) and Egypt (samples from 2001-2002) were among the countries with low contributions from dioxin-like

PCB (range 8-22 per cent). This observation indicates that elevated TEQ levels are not caused by PCB contamination. However, whereas Egypt (2001) had the highest contribution to total TEQ from PCDF (41 per cent from WHO-PCDF-TEQ), the Democratic Republic of the Congo (2009 and 2017) had the highest contribution to total TEQ from PCDD (range 65-85 per cent from WHO-PCDD-TEQ). Furthermore, Côte d'Ivoire, which did not participate in the UNEP/GEF POPs GMP-2 project in 2017-2019, showed high contributions to total TEQ from PCDD (2010 and 2015).

The PCDD-dominated patterns in human milk from the Democratic Republic of the Congo and Côte d'Ivoire mirror the patterns found in certain clays with high concentrations of PCDD/PCDF collected on the Dutch market which originate from African countries. These congener patterns would be expected after bioaccumulation and hint at consumption of such clays ("geophagy") by pregnant women in these countries as the likely source of remarkably high levels in human milk (Reeuwijk *et al.* 2013).

The mixture of a PCDF-dominated pattern with a particularly high contribution of 2,3,4,7,8-PeCDF and PCDD to the TEQ levels, as found in nine human milk samples from Egypt (2000-2002), could indicate combustion and drying processes as a source of this contamination (Hoogenboom *et al.* 2020). While Egypt covers an area of about 1,000,000 square kilometres, the great majority of its nearly 100 million people live along the banks of the Nile River

with about half the population living in urban settings. In this relatively small area possible emissions from industrial production, as well as open burning of waste, occur in close proximity to agricultural production areas. This might explain finding high concentrations of PCDD and PCDF in foods in the 1990s, particularly in butter (Malisch and Saad 1994; Malisch and Saad 1996), and elevated levels of these contaminants in human milk collected in 1997 (Malisch, Fouzy and Saad 2000). In 2019 these levels decreased to 9.04 pg total TEQ/g lipid.

Figure 2 illustrates the results of the sum of six Indicator PCBs ( $\Sigma\text{PCB}_6$ ), with the period of participation (between 2000 and 2019) indicated. Ethiopia also had the lowest levels of any country in the 2000-2019 studies for this parameter: 2.15 ng  $\Sigma\text{PCB}_6$ /g lipid in 2012 and 0.90 ng/g in 2019. The highest level in Africa was in Senegal, where the 2018 sample (90.3 ng/g lipid) showed an increasing trend in comparison to the sample of 2009 (65.8 ng/g lipid). In Egypt a reduction of approximately 85 per cent was observed between the median of nine pooled samples from various regions submitted in the period 2000-2003 and the one pooled sample submitted in the period 2016-2019, which is considered to represent the country at that time. Most countries participated for the first time during the period 2008-2011; in these countries  $\Sigma\text{PCB}_6$  concentrations fell by about 50 per cent on average until the period 2016 to 2019 (ranging from a 71 per cent decrease to a 37 per cent increase).

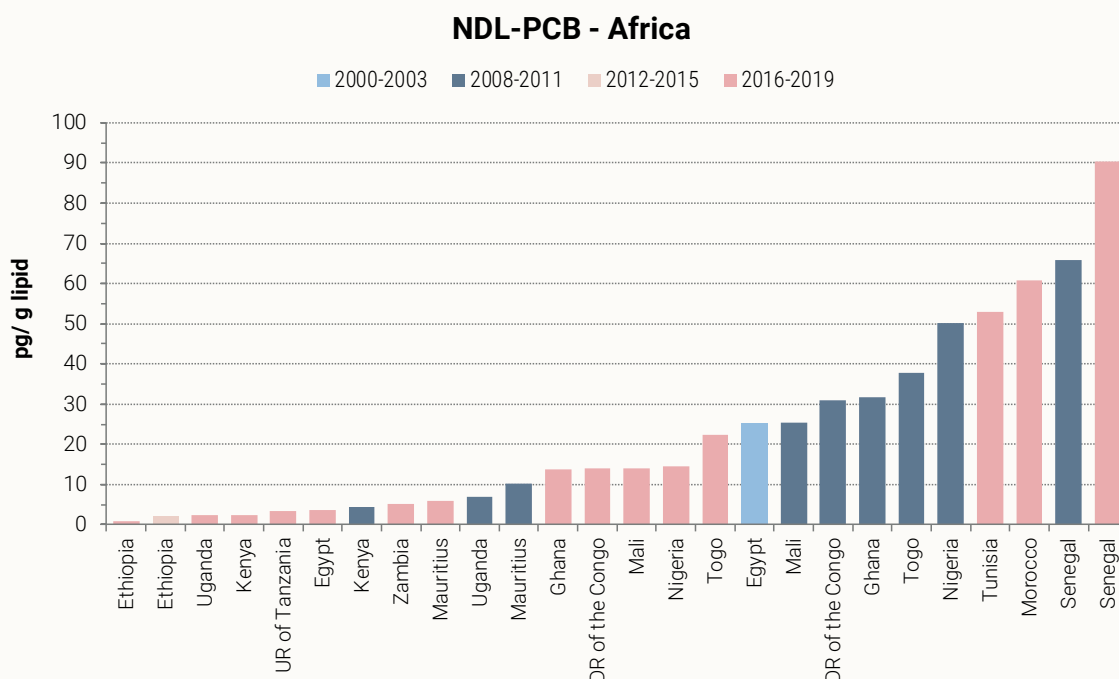


Figure 2: Results of the 2016-2019 survey for  $\Sigma\text{PCB}_6$  (ng/g lipid) in human milk in countries from Africa in comparison to three previous surveys (with an indication of their periods)

### 3.1.2. Asia

Figure 3 illustrates the results for total TEQ and Figure 4 those for NDL-PCB in the four Asian countries that participated in the 2016-2019 round. Concentrations below 5 pg WHO<sub>2005</sub>-TEQ/g lipid and 20 ng ΣPCB<sub>6</sub>/g lipid were found in these samples (there are no data for the period 2000-2015 for these countries).

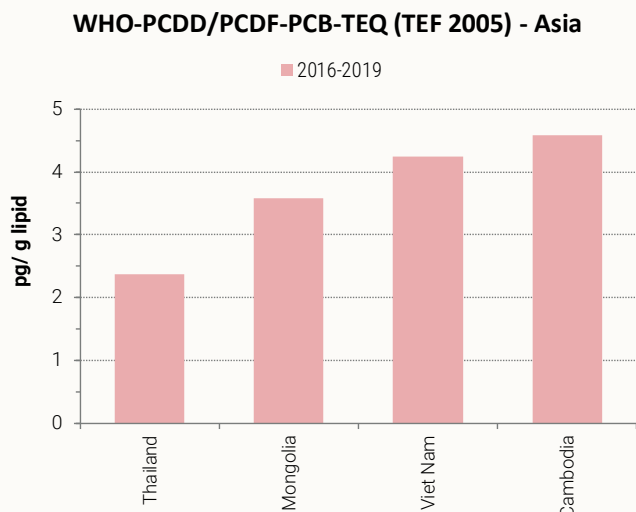


Figure 3: Results of the 2016-2019 survey for total TEQ in human milk (pg WHO-PCDD/PCDF-PCB-TEQ [TEF 2005]/g lipid) in four countries in Asia (no data available for previous surveys between 2000 and 2015)

### NDL-PCB - Asia

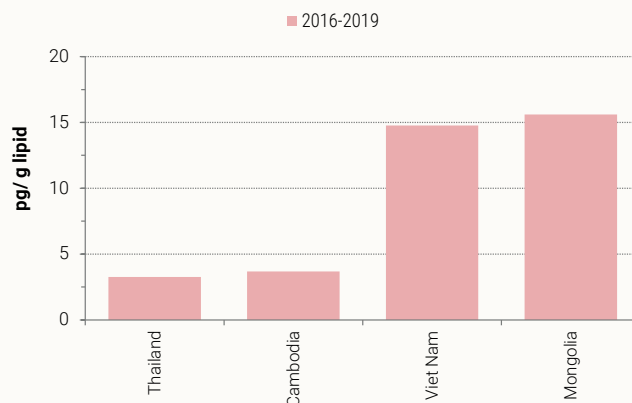


Figure 4: Results of the 2016-2019 survey for ΣPCB<sub>6</sub> (ng/g lipid) in human milk in four countries in Asia (no data available for previous surveys between 2000 and 2015)

### 3.1.3. The Pacific Islands

Figure 5 illustrates the results of total TEQ for samples from the Pacific Islands countries participating in the period 2016-2019 and the respective levels obtained in previous surveys, if applicable. All samples submitted between 2000 and 2015 were approximately in the range 3 to 6 pg of WHO<sub>2005</sub>-TEQ/g lipid. In nearly all samples from the period 2016-2019 concentrations were below 4 pg/g. Downward tendencies were observed in nearly all countries with repeated participation.

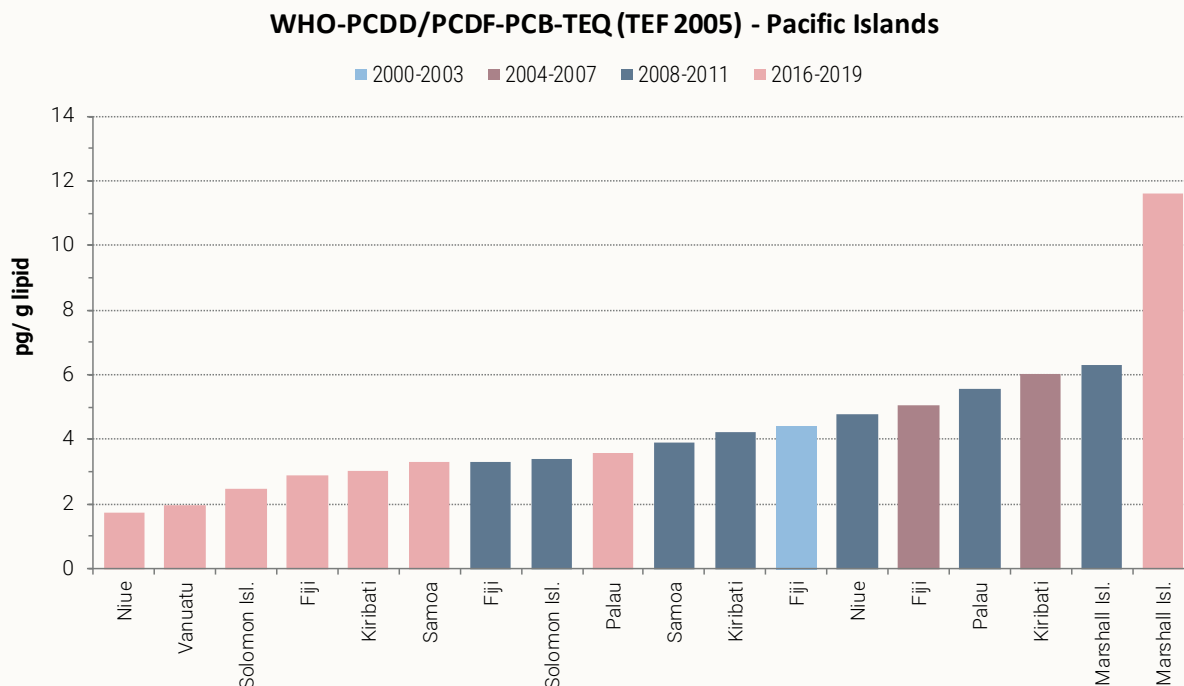


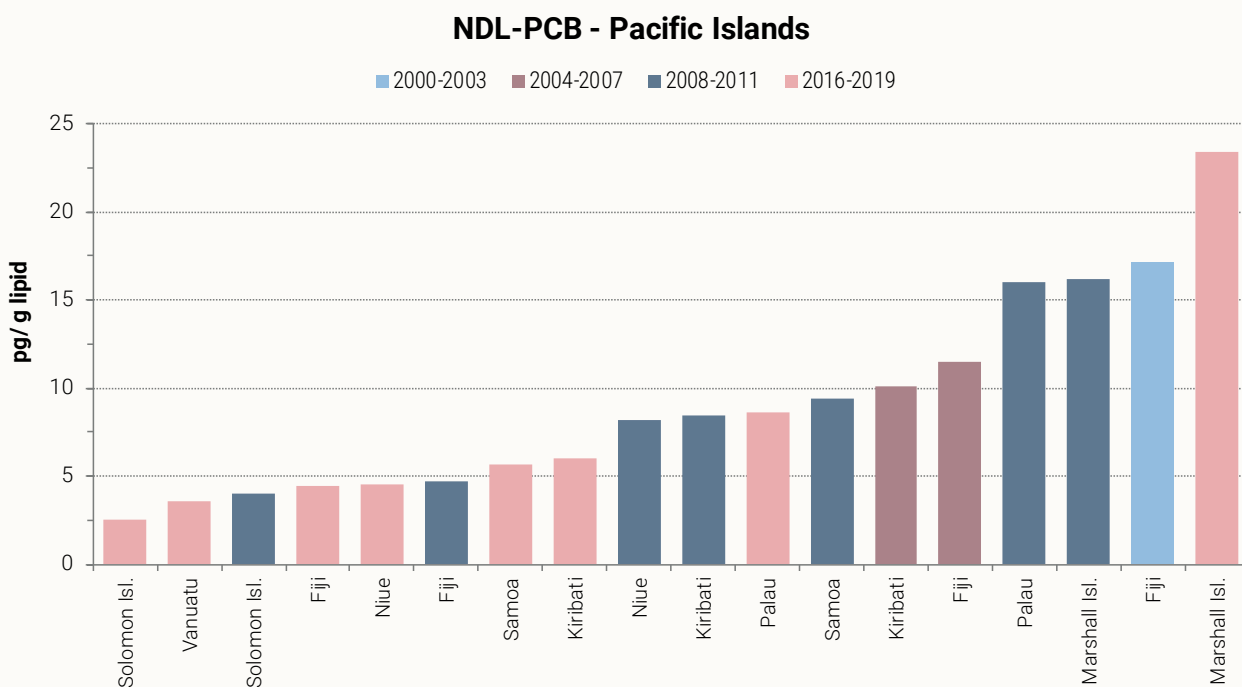
Figure 5: Results of the 2016-2019 survey for total TEQ in human milk (pg WHO-PCDD/PCDF-PCB-TEQ [TEF 2005]/g lipid) in Pacific Islands countries in comparison to three previous surveys (with an indication of their periods)

Only one sample from the Marshall Islands (2019) had a substantially higher concentration of 11.6 pg/g, which was nearly double the 6.32 pg total TEQ/g lipid found in the 2011 sample from that country and the highest concentration found in the Pacific Islands subgroup in the period 2000-2019.

With regard to the increasing concentration in the Marshall Islands from 2011 to 2019 and the overall elevated concentration range of its two samples in the Pacific Islands subgroup, the changes in the relative contribution (per cent) of PCDDs, PCDFs and dioxin-like PCBs to the total toxic equivalents are of interest (Figure S3). The 73 per cent contribution of PCDD to total TEQ in the sample from

2019 is the highest found in the Asia-Pacific Group. In this sample 7 per cent came from PCDF and 20 per cent from dioxin-like PCB. This is a considerable change in comparison to its 2011 sample, when 40 per cent of the total TEQ came from PCDD, 23 per cent from PCDF and 37 per cent from dioxin-like PCB (DL-PCB).

Regarding NDL-PCB concentrations, samples collected between 2000 and 2015 were in the range of approximately 4-17 ng/g for  $\Sigma$  PCB<sub>6</sub> whereas most samples for the period 2016-2019 were in the range of approximately 3-9 ng/g  $\Sigma$  PCB<sub>6</sub>. Only the Marshall Islands showed an increase, from 16 to 23 ng/g  $\Sigma$  PCB<sub>6</sub> between 2011 and 2019 (Figure 6).



**Figure 6:** Results of the 2016-2019 survey for  $\Sigma$ PCB<sub>6</sub> (ng/g lipid) in human milk in countries in Pacific Islands countries in comparison to three previous surveys (with an indication of their periods)



### 3.1.4. The Group of Latin America and the Caribbean (GRULAC)

Figure 7 illustrates the results for total TEQ in countries belonging GRULAC which participated during the period 2016-2019 and the respective levels found in previous

surveys, if applicable. Figure 8 shows the results for NDL-PCB levels. Most countries participated for the first time in 2008-2011. In these countries a decrease was observed in 2016-2019. In Mexico background levels of NDL-PCB remained unchanged.

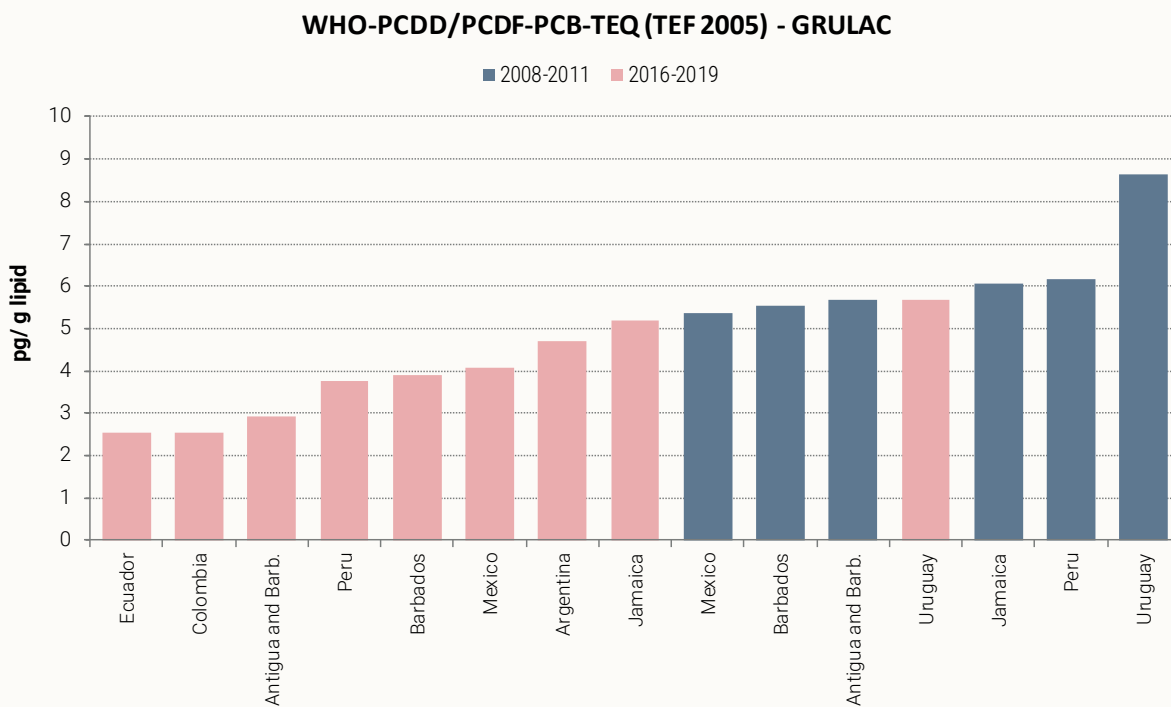


Figure 7: Results of the 2016-2019 survey for total TEQ in human milk (pg WHO-PCDD/PCDF-PCB-TEQ [TEF 2005]/g lipid) in GRULAC countries in comparison to the survey conducted in the 2008-2011 period

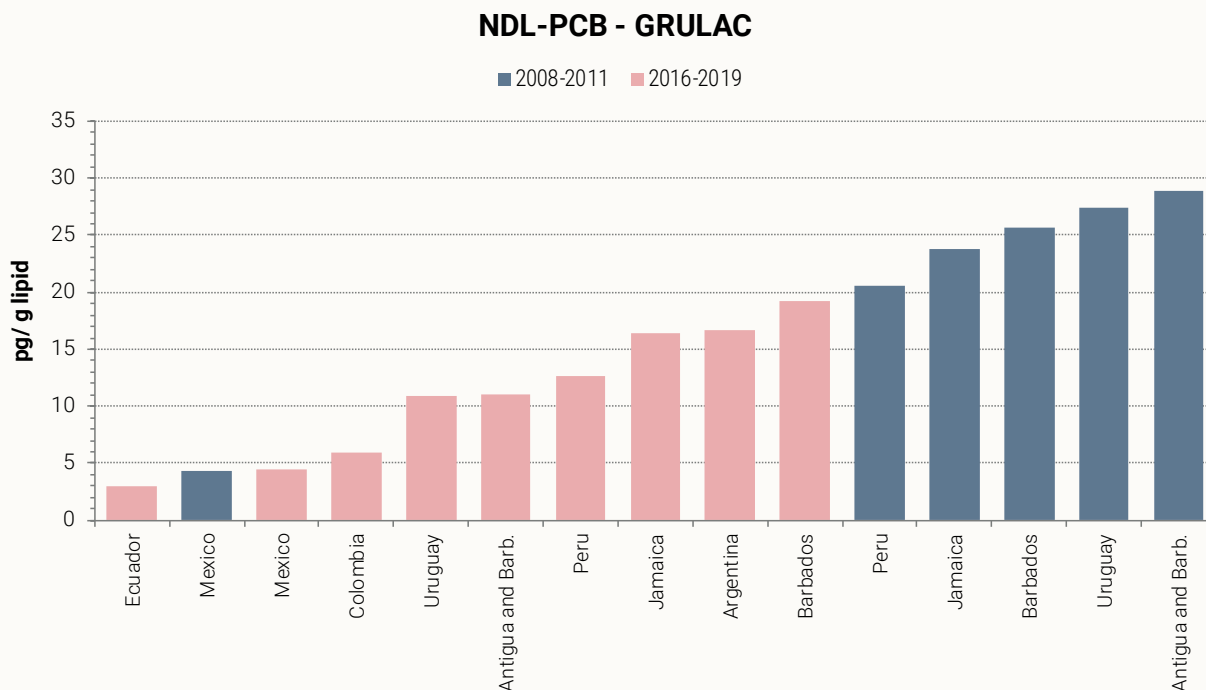


Figure 8: Results of the 2016-2019 survey for  $\Sigma$ PCB<sub>6</sub> (ng/g lipid) in human milk in GRULAC countries in comparison to the survey conducted in the 2008-2011 period

## 3.2. Organochlorine pesticides and organochlorine industrial chemicals

For organochlorine pesticides and organochlorine industrial chemicals, *Table S6* presents the results of all recommended analytes and resulting sum parameters.

### 3.2.1. DDT

Commercial DDT is a mixture mainly of the desired para-para' substituted isomer ( $p,p'$ -DDT = 4,4'-DDT) as major component and the ortho-para' substituted isomeric impurity ( $o,p'$ -DDT = 2,4'-DDT). Due to degradation and metabolism, the transformation products 4,4'-DDE (dichlorodiphenyltrichloroethylene) and 2,4'-DDE, respectively, and 4,4'-DDD (dichlorodiphenyldichloroethane) and 2,4'-DDD, respectively, are of interest. For calculation of the summarizing parameter "DDT complex" correction factors for molecular weight as shown in *Table S2* were applied.

The comparison of ranges for all 82 countries participating between 2000 and 2019 revealed large differences, with a minimum of 17  $\mu\text{g}$  (micrograms) DDT complex/kg lipid found in 2019 in one country in Africa and a maximum of 23,500  $\mu\text{g}$  DDT complex/kg lipid found in 2012 in another country in Africa.

#### 3.2.1.1. Africa

Of all the UN regions, Africa showed the widest variation in contamination of human milk with DDT complex. *Figure 9* illustrates these results for countries, with the five four-year

periods between 2000 and 2019 shown in different colours.

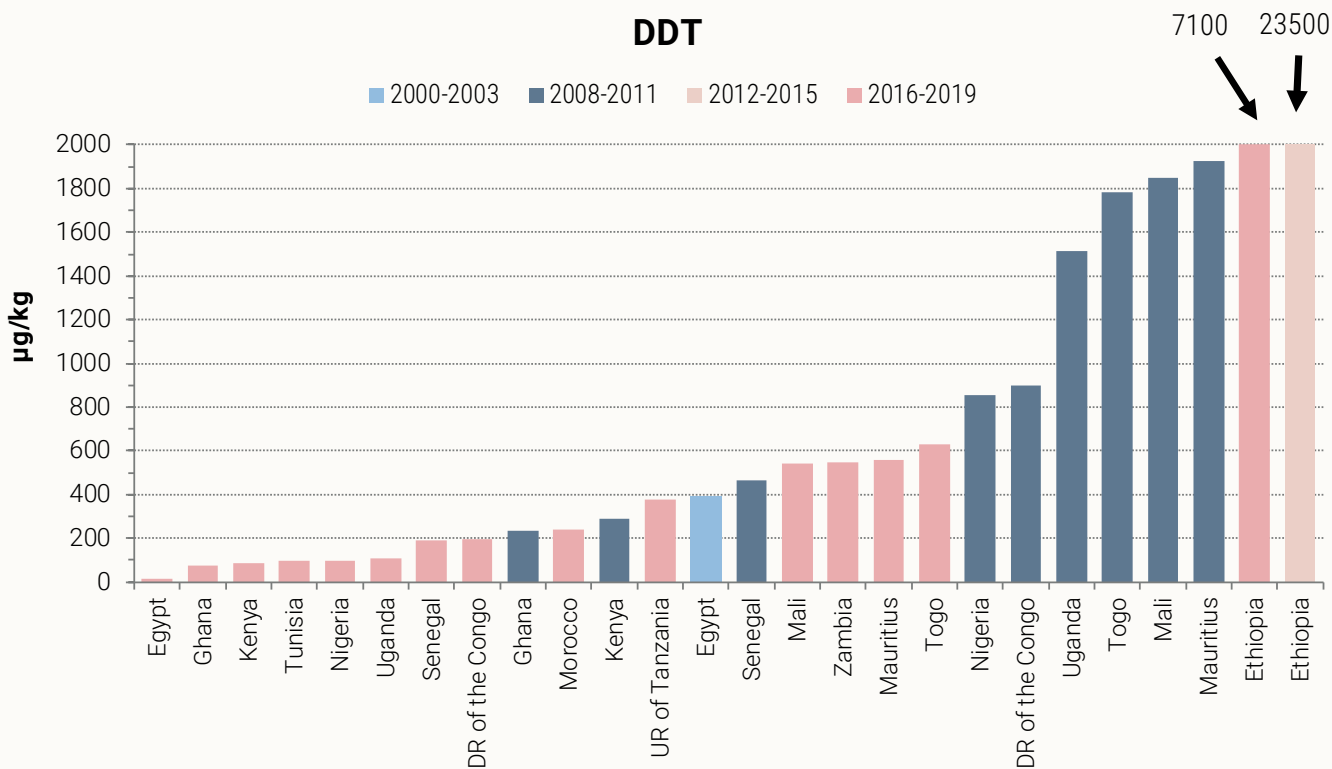
The lowest concentration (17  $\mu\text{g}$  DDT complex/kg lipid) was found in Egypt in 2019. The highest concentration, 23,500  $\mu\text{g}$  DDT complex/kg lipid (or 23.5 mg DDT complex/kg lipid), was found in Ethiopia in 2012. In this sample about 50 per cent of the DDT complex came from  $p,p'$ -DDE, 46 per cent from  $p,p'$ -DDT and 4 per cent from  $o,p'$ -DDT. This is indicative of more recent use and contamination, probably due to public health use of DDT to combat mosquitos for malaria control. DDT is currently listed in Annex B to the Stockholm Convention, with its production and/or use restricted to disease vector control purposes when no equally effective and efficient alternative is available, and in accordance with related WHO recommendations and guidelines (Stockholm Convention 2019).

These conclusions are in line with findings of high mean levels of DDT complex of 12,680  $\mu\text{g}/\text{kg}$  lipid (calculated without correction factors) in human milk from Ethiopia collected in 2010 in three cities in malarious areas where annual spraying for malaria control was common. Between 55 and 71 per cent of DDT complex was attributed to  $p,p'$ -DDT, revealing the continued use of DDT at that time. A number of measures were recommended to reduce the levels of DDT exposure (Gebremichael *et al.* 2013). As a result, the 2019 sample from Ethiopia showed a considerable downward trend with 7,100  $\mu\text{g}$  DDT complex/kg lipid. Importantly, in the 2019 sample  $p,p'$ -DDE contributed nearly 80 per cent and  $p,p'$ -DDT 20 per cent to DDT complex.

Most countries participated for the first time in the period 2008-2011; in these countries DDT concentrations fell by about 70 per cent on average between that period and 2016-2019 (a decrease of between 59 and 93 per cent).



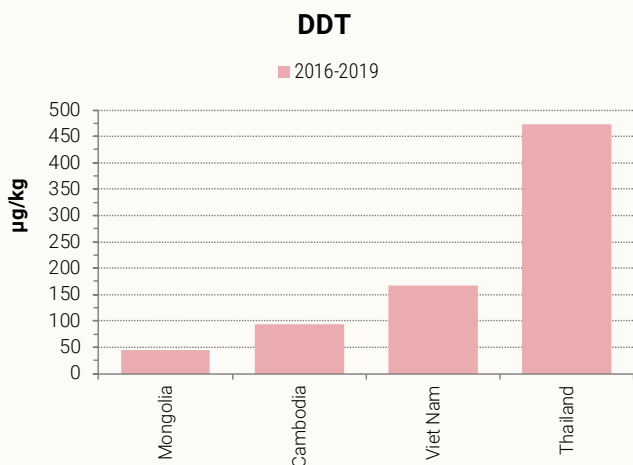
Caroline Obinju Ocholla performs Indoor Residual Spraying (IRS) of an insecticide to prevent malaria in her home in Kenya.



**Figure 9:** Concentration figures of DDT in human milk ( $\mu\text{g } \Sigma \text{ DDT complex/kg lipid}$ ) from African countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.2.1.2. Asia

Figure 10 illustrates the DDT complex results for the four Asian countries participating in the 2016-2019 round (there are no data for the period 2000-2015). A range of between 45 and 473  $\mu\text{g } \Sigma \text{ DDT complex/kg lipid}$  was found.



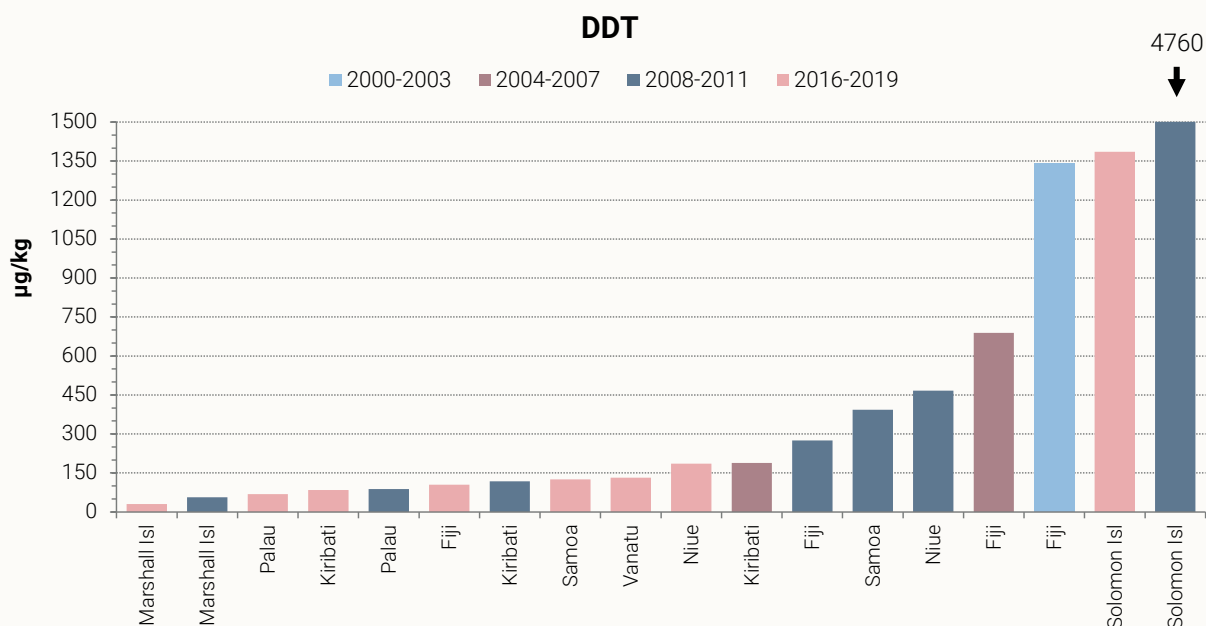
**Figure 10:** Concentrations of DDT in human milk ( $\mu\text{g } \Sigma \text{ DDT complex/kg lipid}$ ) from Asian countries in the period 2016-2019 (No data available for previous surveys between 2000 and 2015)

### 3.2.1.3. The Pacific Islands

Figure 11 illustrates the DDT complex results for the Pacific Islands countries that participated during the period 2016-2019 and levels found in previous surveys if applicable. A wide range of DDT concentrations was found, with the lowest concentration in the Marshall Islands in 2019 (31  $\mu\text{g DDT complex/kg lipid}$ ) and the highest in the Solomon Islands in 2011 (4,760  $\mu\text{g DDT complex/kg lipid}$ ). In this sample with (in other units) 4.8 mg DDT complex/kg lipid, 91 per cent of the contribution to the sum parameter “DDT complex” came from *p,p'*-DDE. With 1390  $\mu\text{g DDT complex/kg lipid}$ , the 2019 sample from the Solomon Islands had considerably lower DDT levels, with a contribution from *p,p'*-DDE of 95 per cent.

Fiji’s results from 2001, 2006, 2011 and 2019 show a considerable downward trend from 1340  $\mu\text{g DDT complex/kg lipid}$  to 105  $\mu\text{g/kg lipid}$ , with the contribution of *p,p'*-DDE to DDT complex increasing gradually from 82 per cent in 2001 to 96 per cent in 2019.



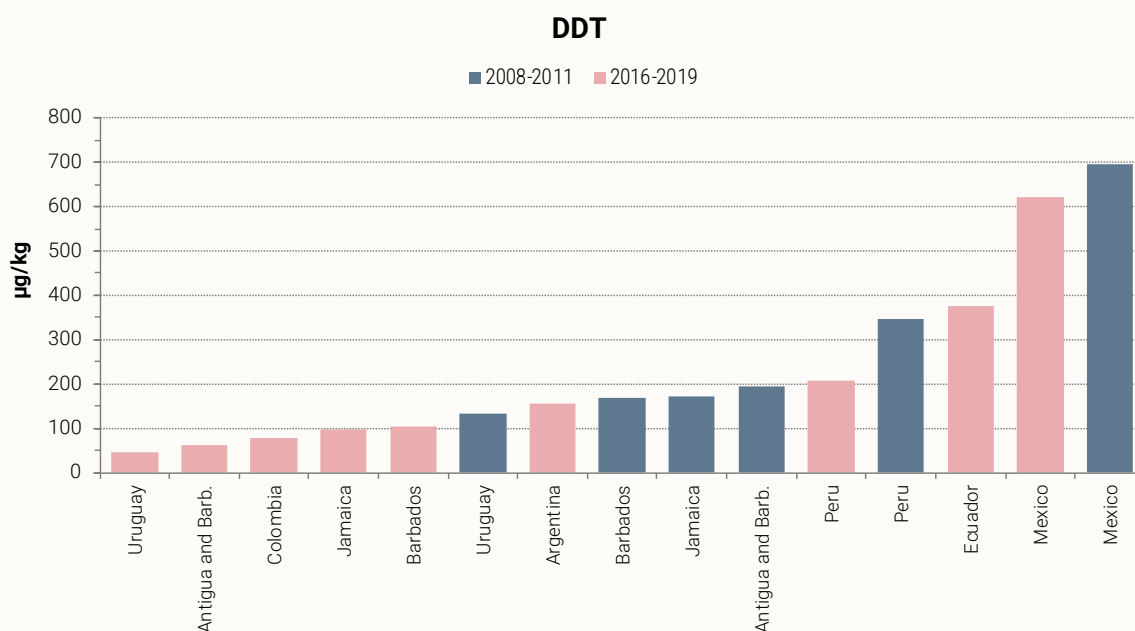


**Figure 11:** Concentrations of DDT in human milk ( $\mu\text{g} \Sigma \text{ DDT complex/kg lipid}$ ) from The Pacific Islands countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.2.1.4. The Group of Latin America and the Caribbean (GRULAC)

Figure 12 illustrates the DDT results for GRULAC countries participating during the period 2016-2019 and the levels found in previous surveys, if applicable. A range

of DDT concentrations was found, from 46  $\mu\text{g}$  DDT complex/kg lipid in Uruguay (2019) to 696  $\mu\text{g}$  DDT complex/kg lipid in Mexico (2011). For the countries that participated between 2008 and 2011 a decrease was observed in subsequent years (decrease rates: median 41 per cent; range 11-67 per cent).



**Figure 12:** Concentrations of DDT in human milk ( $\mu\text{g} \Sigma \text{ DDT complex/kg lipid}$ ) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

### 3.2.2. Hexachlorocyclohexane (alpha-HCH, beta-HCH, gamma-HCH)

Technical grade hexachlorocyclohexane (HCH) is a mixture mainly of three isomers comprising about 65-70 per cent alpha-HCH, 7-20 per cent beta-HCH and 14-15 per cent gamma-HCH. Note that only gamma-HCH (lindane) has insecticidal properties. Due to metabolism, mainly beta-HCH accumulates in humans. Alpha-HCH, beta-HCH and gamma-HCH are the most important isomers. In 2009 they were listed in the Stockholm Convention's Annex A (Stockholm Convention 2019). In this report HCH complex is defined as the sum of alpha-HCH, beta-HCH and gamma-HCH.

As result of the metabolism of hexachlorocyclohexane isomers in humans, concentrations of alpha-HCH and gamma-HCH in most human milk samples were below the limit of quantification (<0.5 µg/kg lipid) with a median of quantifiable residues of about 1 µg/kg lipid and maximum 10.5 µg/kg for alpha-HCH and 16 µg/kg for gamma-HCH. In most cases where HCH complex concentrations are above 10 µg/kg lipid, about 95-100 per cent of this sum parameter is attributed to beta-HCH. Therefore, only the ranges found for beta-HCH are discussed in more detail in the following subsection.

Comparison of the ranges of beta-HCH in all 82 countries participating between 2000 and 2019 revealed great differences, with a minimum of <0.5 µg beta-HCH/kg lipid (found in a few countries) and a maximum of 1020 µg beta-HCH /kg lipid.

#### 3.2.2.1. Africa

The range of beta-HCH in samples from African countries varied between <0.5 µg/kg lipid in three countries and 51.3 µg/kg lipid in Egypt in 2002 (Figure 13). In Egypt a considerable downward trend, with 19.7 µg/kg lipid measured in 2019. In both samples from Egypt 94 per cent of the HCH complex was from beta-HCH.

In contrast, 16 µg gamma-HCH/kg lipid was found in the 2009 sample from Senegal, corresponding to 25 per cent HCH complex with 48.3 µg beta-HCH/kg lipid. In Senegal the contribution of gamma-HCH to HCH complex fell to 13 per cent until 2018 while the beta-HCH concentration fell to 11.7 µg/kg.

Most countries participated for the first time in the period 2008-2011 (range 1.9 µg/kg lipid to 48.3 µg/kg lipid; median 8.3 µg/kg lipid). In nearly all countries beta-HCH concentrations fell by about 63 per cent on average until the period 2016-2019 (range of decrease rates between 43 and 92 per cent).

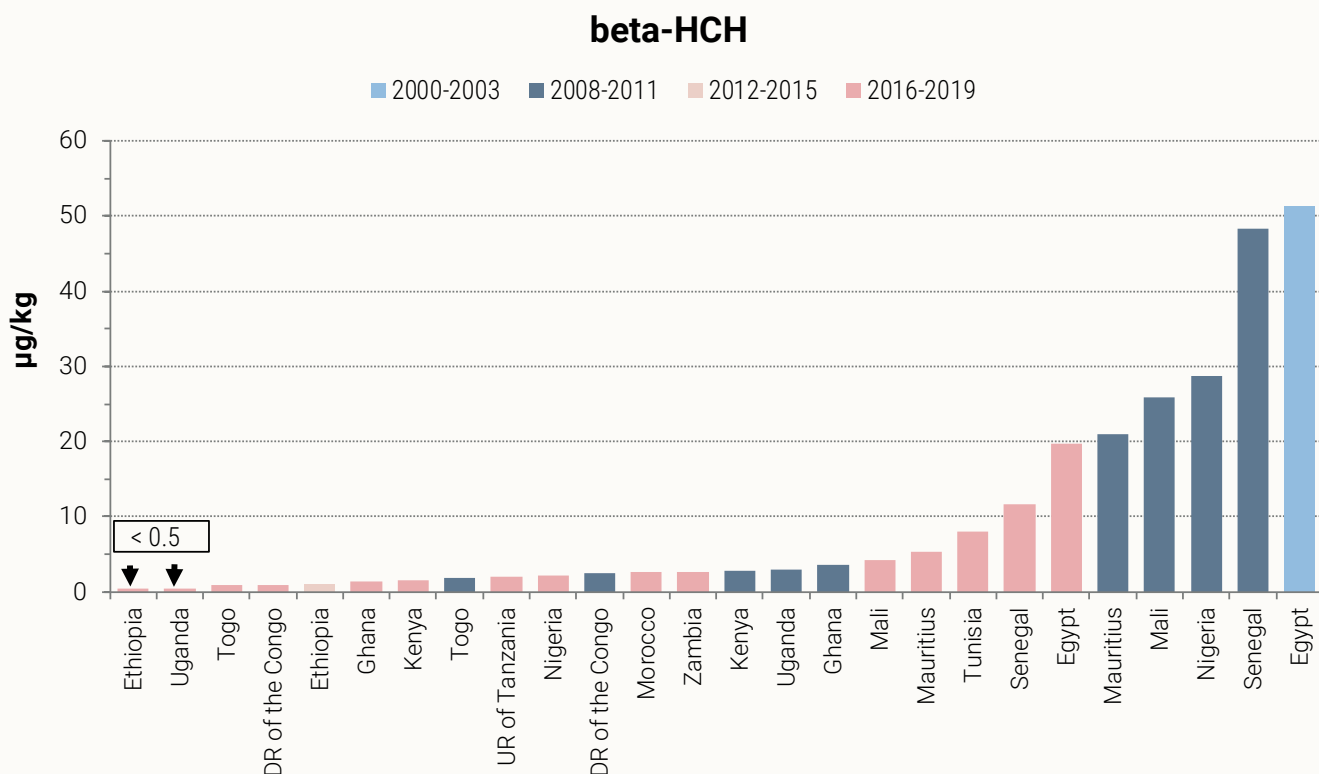
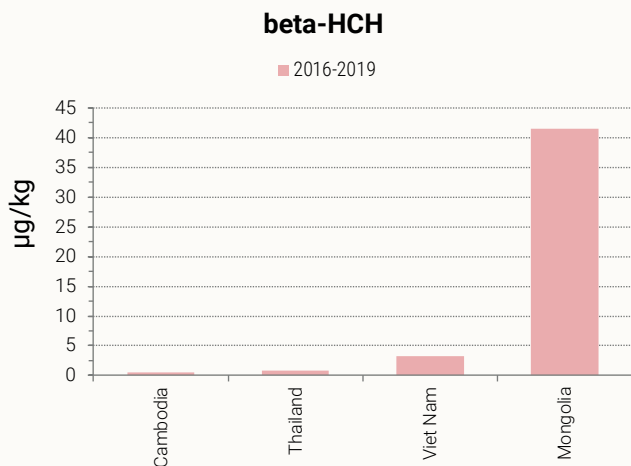


Figure 13: Concentrations of beta-HCH in human milk (µg beta-HCH/kg lipid) from African countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.2.2.2. Asia

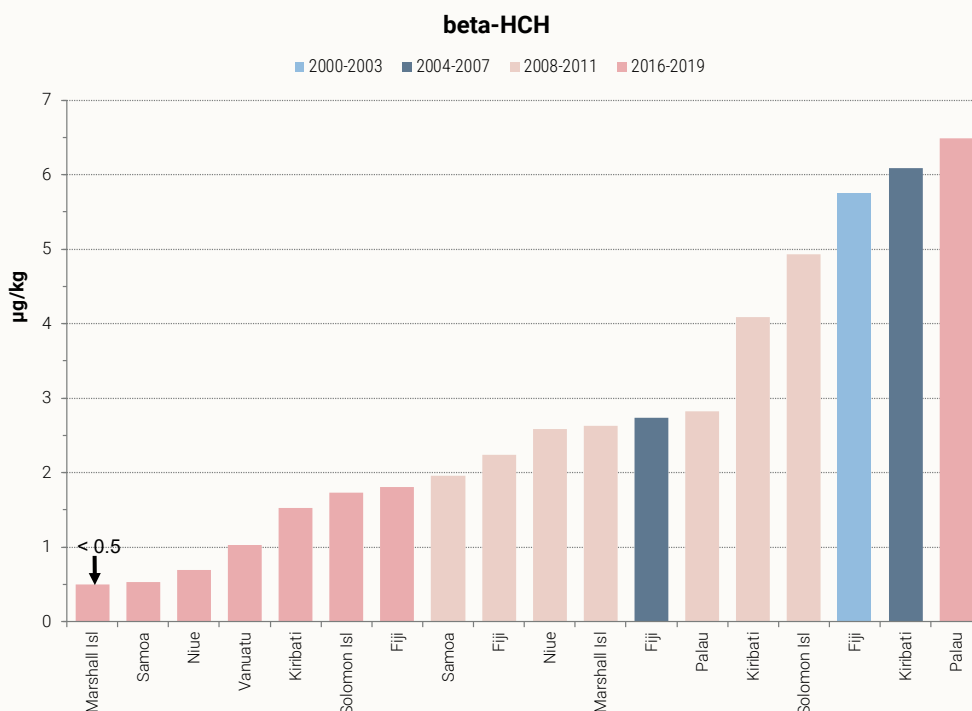
Figure 14 illustrates the results of beta-HCH for the four Asian countries participating in the 2016-2019 round (there were no data for the period between 2000 and 2015). A range of 0.6 to 42 µg beta-HCH /kg lipid was found.



**Figure 14:** Concentrations of beta-HCH in human milk (µg beta-HCH/kg lipid) from Asian countries in the period 2016-2019 (no data available for previous surveys between 2000 and 2015)

### 3.2.2.3. The Pacific Islands

Figure 15 illustrates the beta-HCH results for the Pacific Islands countries. All of these countries in all periods had beta-HCH concentrations in the range of low background contamination, between <0.5 and 6.5 µg/kg lipid. Overall, the countries in this region had the lowest beta-HCH concentrations in human milk.



**Figure 15:** Concentrations of beta-HCH in human milk (µg beta-HCH/kg lipid) from The Pacific Islands countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.2.2.4. The Group of Latin America and the Caribbean

Figure 16 illustrates the beta-HCH results for GRULAC countries that participated during the period 2016-2019 and levels found in previous surveys, if applicable. The

results ranged from 0.7 µg beta-HCH/kg lipid in Colombia (2019) to 29.7 µg beta-HCH/kg lipid in Uruguay (2009). In countries where beta-HCH concentrations above 5 µg/kg were found in samples submitted before 2011 (Uruguay in 2009, Barbados in 2010, Peru in 2011) a considerable downward trend was observed until 2019.

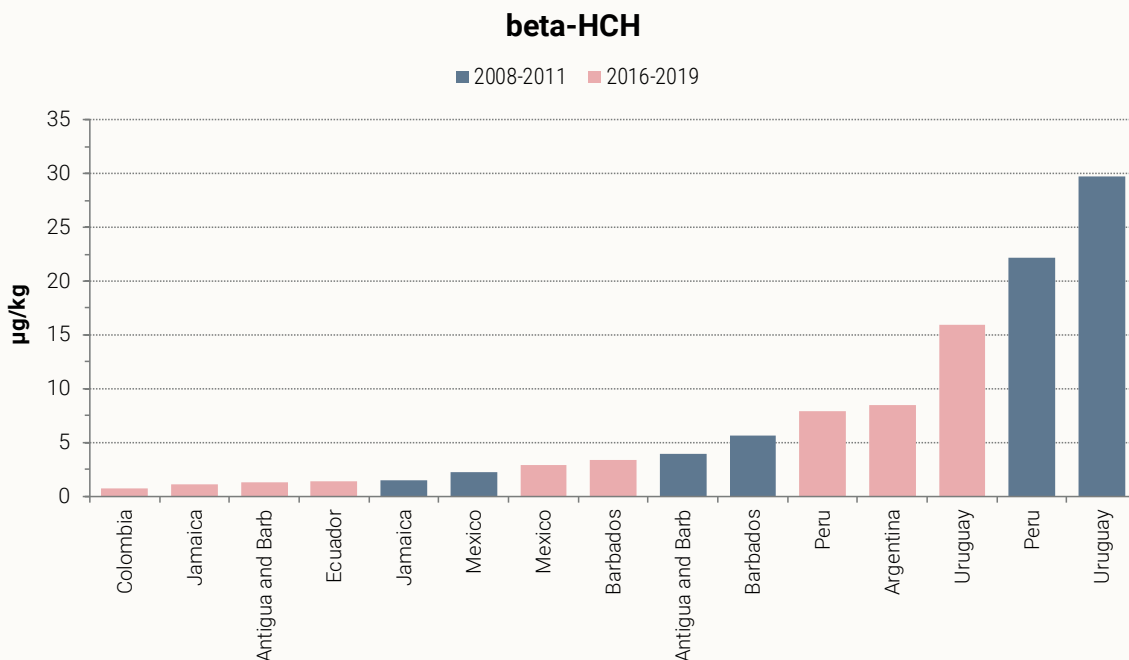


Figure 16: Concentrations of beta-HCH in human milk (µg beta-HCH/kg lipid) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period



Photo: ©Pexels / Livier Garcia

Coffee fruit harvesting

### 3.2.3. Hexachlorobenzene (HCB)

The maximum levels of HCB in the 82 countries participating between 2000 and 2019, and therefore the ranges, were much lower than those found for DDT and beta-HCH, with a minimum of about 1-2 µg/kg lipid in some countries and a maximum of 154 µg/kg lipid.

#### 3.2.3.1. Africa

HCB concentrations in all African countries during the whole period between 2000 and 2019 were in the range of low background concentrations, varying between 1 and 5 µg/kg lipid (Figure 17). A downward trend at the upper range of this background contamination can be observed when results from Nigeria and Mauritius (2008-2011) and Egypt (2000-2003) are compared with these countries' results in the period 2016-2019. In most other countries with repeated participation, concentrations were quite stable at low concentrations of approximately 2-3 µg/kg lipid.

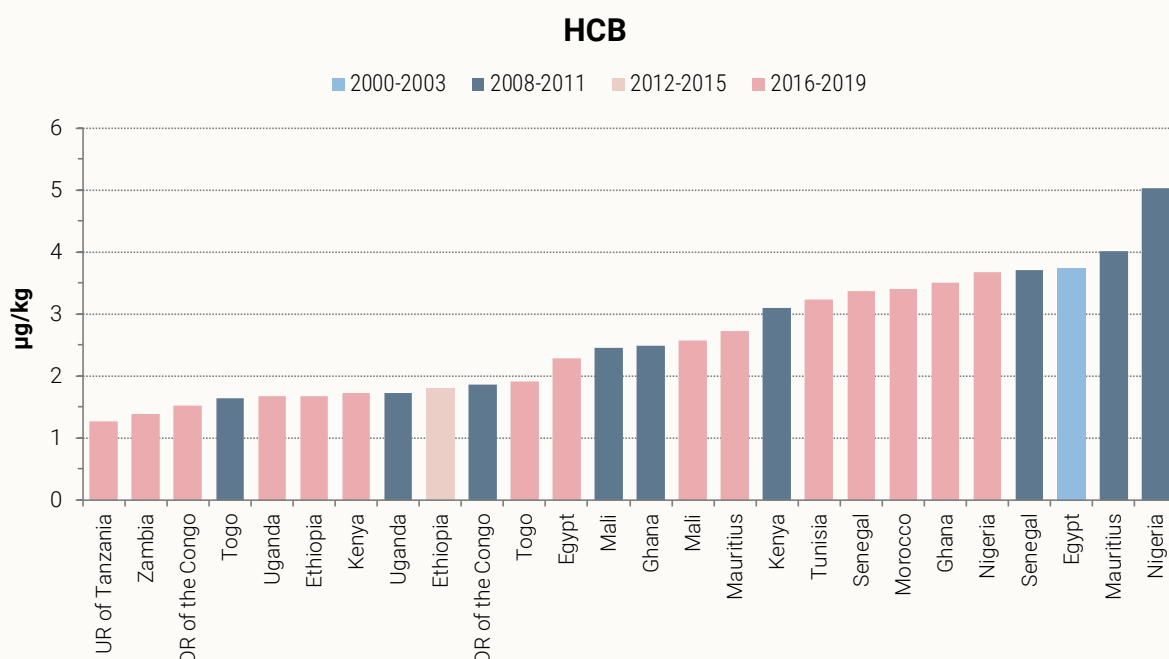


Figure 17: Concentrations of HCB in human milk (µg HCB/kg lipid) from African countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

#### 3.2.3.2. Asia

Figure 18 illustrates the HCB results for the four Asian countries participating in the 2016-2019 round (there are no data for the period between 2000 and 2015). A range of 2.5-34 µg HCB/kg lipid was found.

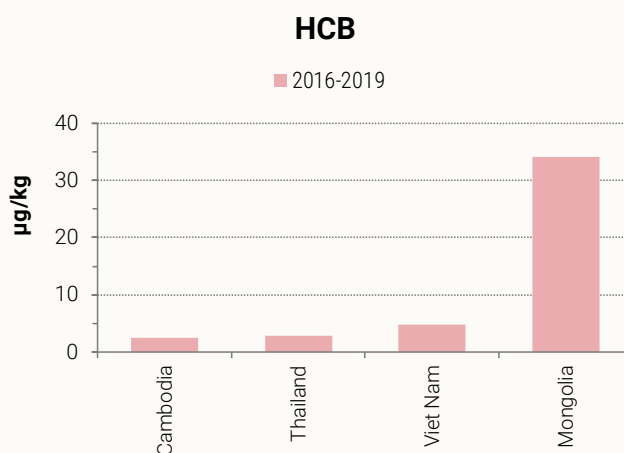


Figure 18: Concentrations of HCB in human milk (µg beta-HCB/kg lipid) from Asian countries in the period 2016-2019 (no data available for previous surveys between 2000 and 2015)

### 3.2.3.3. The Pacific Islands

Figure 19 illustrates the HCB results for the Pacific Islands countries. A downward trend can be observed between 2008-2011 and 2016-2019. Whereas average background

contamination in the period 2008-2011 was about 6 µg HCB/kg lipid, it dropped to about 3 µg/kg lipid in the period 2016-2019. Fiji participated four times between 2000 and 2019, with quite stable background concentrations of about 3-5 µg/kg lipid.

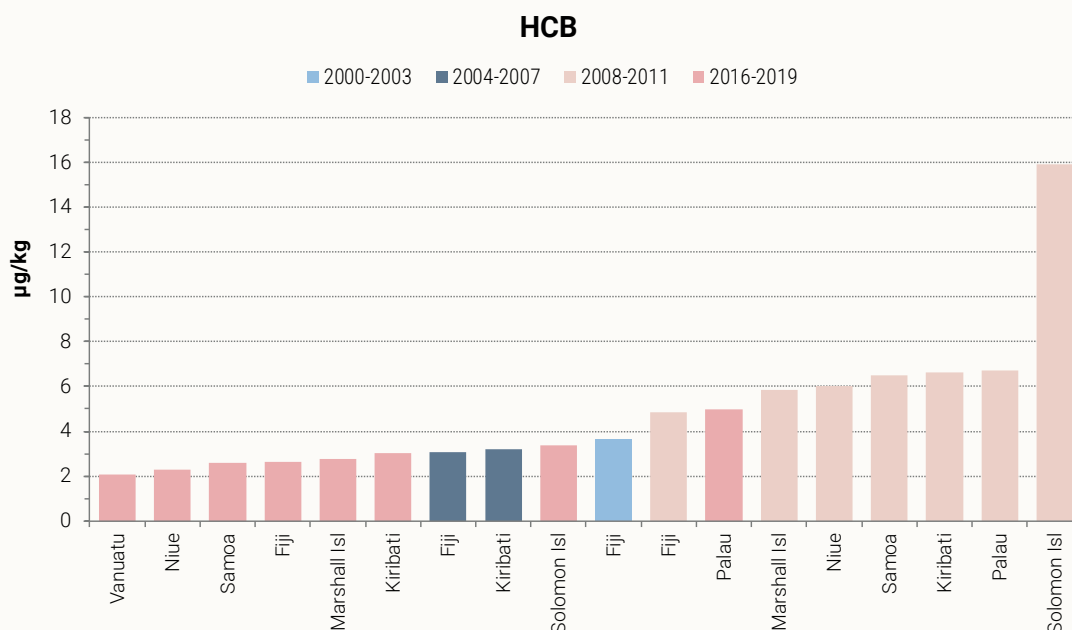


Figure 19: Concentrations of HCB in human milk (µg HCB/kg lipid) from the Pacific Islands countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.2.3.4. The Group of Latin America and the Caribbean

Figure 20 illustrates the HCB results for the GRULAC countries participating in 2016-2019 and levels found in previous surveys, if applicable. The results ranged from 3 µg/

kg lipid in Barbados (2018) to 14 µg/kg lipid in Uruguay (2009). Two countries at the upper end of this distribution participated in the period 2008-2011: downward trends were observed in Uruguay between 2009 and 2019 and in Mexico between 2011 and 2017.

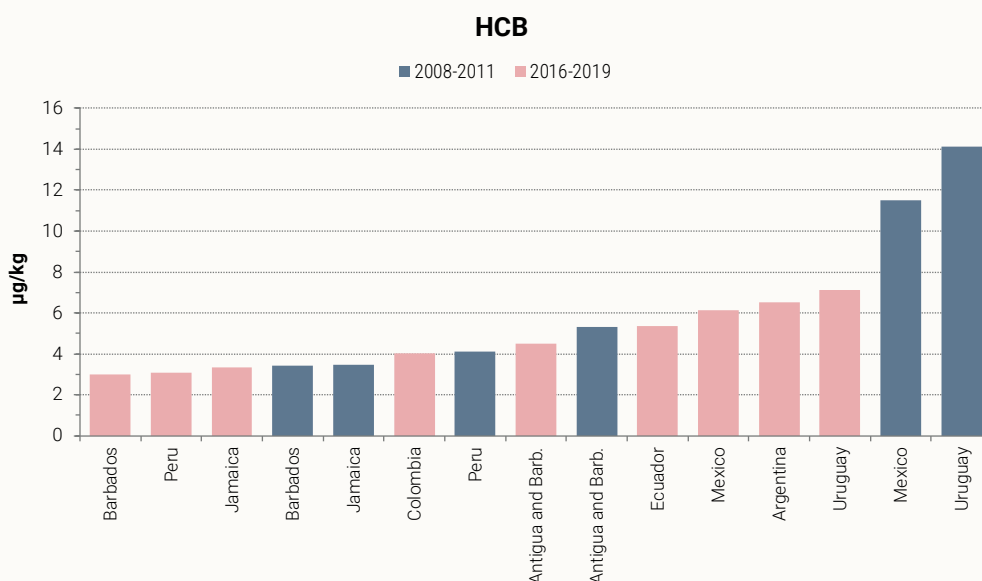


Figure 20: Concentrations of HCB in human milk (µg HCB/kg lipid) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

### 3.2.4. Aldrin, dieldrin

Aldrin, which is rapidly metabolized to dieldrin, is not found in humans and was not detected in any of the samples (<0.5 µg/kg lipid). Dieldrin levels were mostly in a low background range below 5 µg/kg lipid. As a maximum for the 36 countries, 11.2 µg/kg was found in Mali in 2009, decreasing to 1.7 µg/kg in 2019.

### 3.2.5. Chlordane

According to the definition of pesticide residue in food, the sum parameter “chlordane complex” comprises cis-chlordane (= “alpha chlordane”) and trans-chlordane (= “gamma-chlordane”) (both more relevant for food of plant origin) and the metabolite oxychlordane (relevant for food of animal origin). These were also the recommended analytes according to the *Guidance on the Global Monitoring Plan (GMP)* for POPs as of 2007. Later, cis- and trans-nonachlor, which are impurities in chlordane production, were added to the list of recommended analytes for chlordane in the GMP guidance. Thus the “chlordane group” comprises cis- and trans-chlordane, cis- and trans-nonachlor, and oxychlordane.

In the period 2016-2019 chlordane levels in the 36 countries were in a low background range below 5 µg/kg lipid. As a maximum for these countries, 11.7 µg/kg was found in Senegal in 2009, decreasing to 4.1 µg/kg in 2019.

### 3.2.6. Endrin

Endrin is rapidly metabolized to endrin ketone. The “endrin complex” is the sum of endrin and endrin ketone. Endrin and endrine ketone were not detected in any sample from the 36 countries in the period 2000-2019 (limit of quantification 0.5 µg/kg lipid).

### 3.2.7. Heptachlor

Heptachlor is rapidly metabolized to heptachlorepoxide. Cis-heptachlorepoxide can be found in humans. The “heptachlor complex” is the sum of heptachlor, cis-heptachlorepoxide and trans-heptachlorepoxide. In the few cases where there were quantifiable residues in samples from the 36 countries during the period 2000-2019, only cis-heptachlorepoxide was found, at concentrations below 5 µg/kg lipid.

### 3.2.8. Mirex

In most samples no mirex was detected (limit of quantification 0.5 µg/kg lipid). A few samples had concentrations

of up to 3 µg/kg lipid and one, from Uruguay, had a slightly higher level (9.8 µg/kg lipid in 2009). Uruguay subsequently showed a downward trend in 2019 to 2.9 µg/kg lipid.

### 3.2.9. Toxaphene

Toxaphene is a complex mixture of chlorinated bornanes and chlorinated camphens comprising about 16,000 congeners/isomers. Marker compounds to be monitored are the congeners Parlar 26 (P26), Parlar 50 (P50) and Parlar 62 (P62) as a basis for the sum parameter “toxaphene complex”. This was calculated through application of the “lower bound approach”, where only analytical results above the limit of quantification (0.5 µg/kg lipid) are used.

In most samples neither P26, P50 nor P62 were detected. In a few samples toxaphene was found in a range of between 0.5 and 4 µg toxaphene complex/kg lipid.

### 3.2.10. Chlordecone

Chlordecone was not detected in any of the samples submitted (limit of quantification 0.5 µg/kg lipid).

### 3.2.11. Endosulfan

The sum parameter “endosulfan complex” comprises the analytes alpha-endosulfan, beta-endosulfan and endosulfan sulfate.

Almost no samples had measurable residues above the limit of quantification (0.5 µg/kg lipid). The 2009 sample from Nigeria had 6.3 µg endosulfan complex/kg lipid, but residues in the 2019 sample were below the limit of quantification.

### 3.2.12. Pentachlorophenol (PCP), pentachloranisol (PCA)

Pentachlorophenol (PCP) was listed in Annex A of the Stockholm Convention in 2015 (Stockholm Convention 2019). PCP does not bioaccumulate, whereas its metabolite, pentachloroanisol (PCA) can be found after the use of PCP. PCA is therefore the recommended analyte for human milk.

As expected, no sample from the period 2016-2019 had PCP residues and most samples had no residues of PCA (limit of quantification 0.5 µg/kg lipid). One sample had a trace of PCA (Fiji, 1.1 µg/kg lipid in 2019), while another sample had a high level of 33.3 µg/kg lipid (Vanuatu in 2018).

### 3.2.13. Dicofol

Dicofol was listed in Annex A of the Stockholm Convention in 2019 (Stockholm Convention 2019). Therefore, it was not included in the list of 23 POPs that were requested to be analysed when the survey started in 2016. On a voluntary basis the analysis of dicofol was undertaken in order to obtain a complete picture of all the POPs covered by the Convention. In nearly all samples submitted between 2017 and 2019 dicofol was not detected (limit of quantification 0.5 µg/kg lipid). Only one sample, from Ethiopia in 2019, had a measurable level of dicofol at 3 µg/kg lipid.

### 3.2.14. Pentachlorobenzene (PeCB)

In most samples submitted between 2008 and 2019 no PeCB was found (limit of quantification 0.5 µg/kg lipid). In the remaining samples low levels of between 0.5 µg/kg and 1.2 µg/kg lipid were found.

### 3.2.15. Hexachlorobutadiene (HCBD)

The production and use of HCBD (e.g. formerly for various technical purposes) are prohibited through its listing in Annex A (elimination) of the Stockholm Convention in 2015; it can also be formed as an unintentional by-product and is therefore listed in 2017 in Annex C (unintentional

production) (Stockholm Convention 2019). In none of the samples submitted during the period 2016-2019 was HCBD detected above the limit of quantification (0.5 µg/kg lipid).

## 3.3. Polybrominated diphenyl ethers (Σ PBDE<sub>6</sub>)

In human milk samples from 79 of the 82 countries surveyed in the periods between 2000 and 2019, considerable variation in PBDE concentrations was observed. Of all samples, 80 per cent were in a range below 5 ng Σ PBDE<sub>6</sub>/g lipid. The highest concentration (223 ng Σ PBDE<sub>6</sub>/g lipid) was found in the period 2000-2003.

For the 36 countries participating in the period 2016-2019, as covered by this report, results for the six recommended analytes and the sum parameter Σ PBDE<sub>6</sub> are presented in Table S7.

### 3.3.1. Africa

Figure 21 illustrates the results for the African region between 2008 and 2019. In all countries, at all times, Σ PBDE<sub>6</sub> concentrations were approximately between 0.5 ng/g lipid and 3 ng/g lipid.

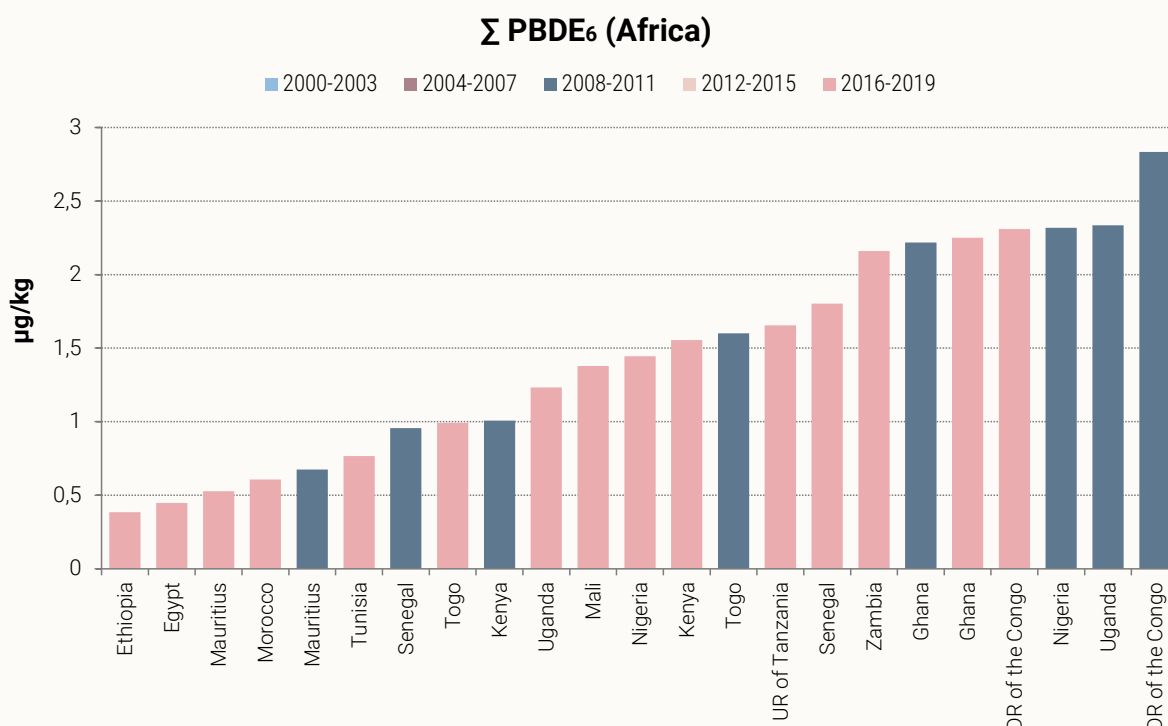
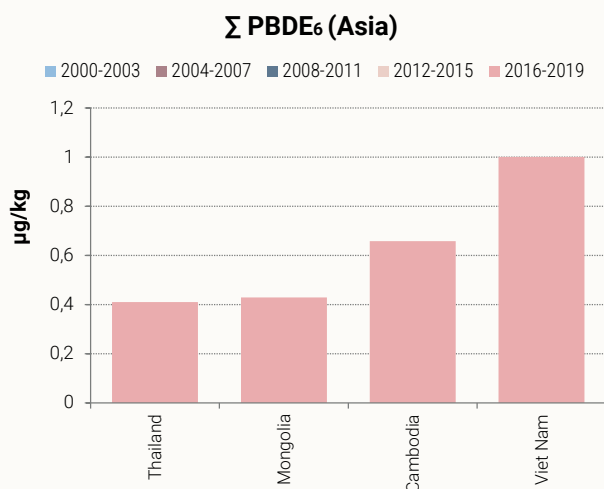


Figure 21: Concentrations of Σ PBDE<sub>6</sub> in human milk (µg Σ PBDE<sub>6</sub>/kg lipid) from African countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period



### 3.3.2. Asia

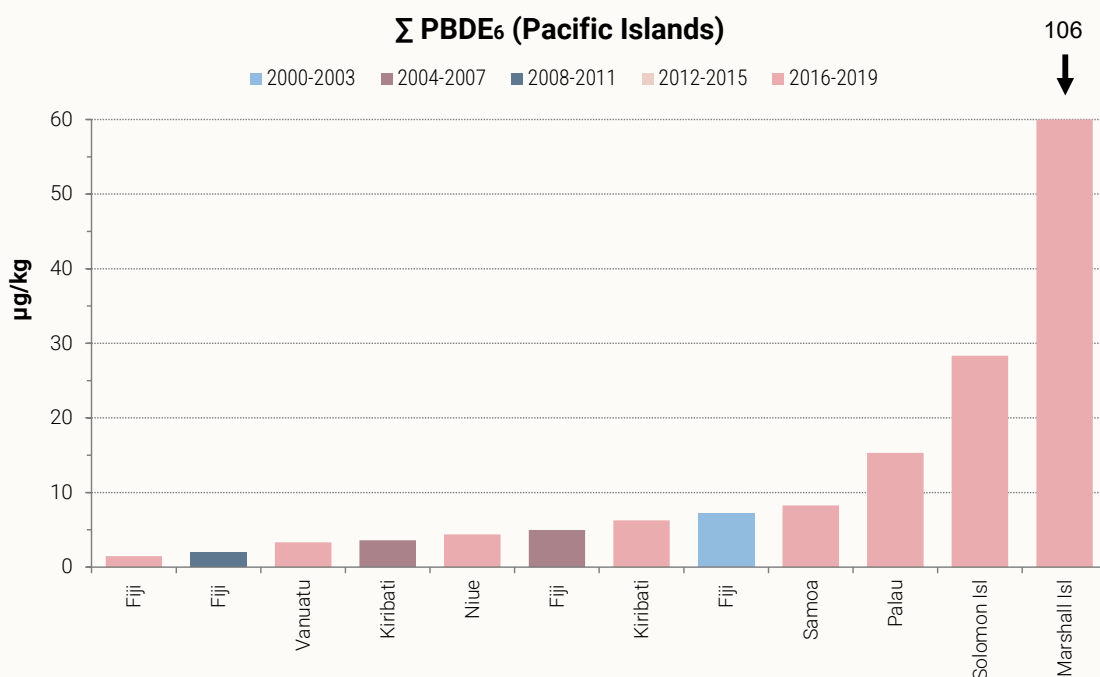
Figure 22 illustrates the results for the Asia region. Results are available for four countries between 2016 and 2019.  $\Sigma$  PBDE<sub>6</sub> concentrations were  $\leq 1$  ng/g lipid.



**Figure 22:** Concentrations of  $\Sigma$  PBDE<sub>6</sub> in human milk ( $\mu\text{g } \Sigma$  PBDE<sub>6</sub>/kg lipid) from Asian countries in the period 2016-2019 (no data available for previous surveys between 2000 and 2015)

### 3.3.3. The Pacific Islands

Most samples from the Pacific Islands countries had concentrations below 5 ng  $\Sigma$  PBDE<sub>6</sub>/g lipid (Figure 23). In five samples from the period 2016-2019, and one from an earlier period, there were higher levels:  $>5$  ng/g lipid, with a maximum of 107 ng  $\Sigma$  PBDE<sub>6</sub>/g in 2019 in the Marshall Islands.



**Figure 23:** Concentrations of  $\Sigma$  PBDE<sub>6</sub> in human milk ( $\mu\text{g } \Sigma$  PBDE<sub>6</sub>/kg lipid) from the Pacific Islands countries in the period 2016-2019 in comparison to three previous surveys (with an indication of their periods)

### 3.3.4. The Group of Latin America and the Caribbean (GRULAC)

Figure 24 illustrates the  $\Sigma$  PBDE<sub>6</sub> results for GRULAC countries participating during the period 2016-2019 and levels found in previous surveys, if applicable. The results ranged

from 0.4  $\mu\text{g}/\text{kg}$  lipid in Uruguay (2019) to 19  $\mu\text{g}/\text{kg}$  lipid in Antigua and Barbuda (2008). In the case of the two countries at the upper end of this distribution, downward trends were observed in Antigua and Barbuda between 2008 and 2018 and in Mexico between 2011 and 2017.

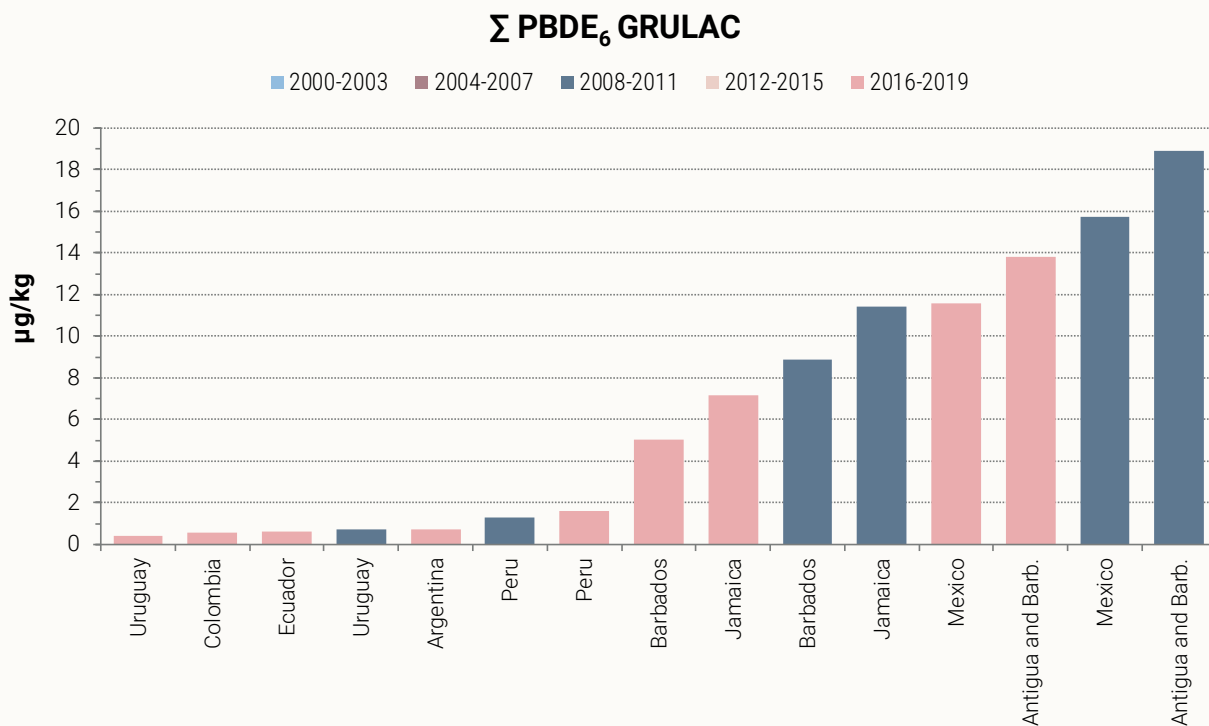


Figure 24: Concentrations of  $\Sigma$  PBDE<sub>6</sub> in human milk ( $\mu\text{g}$   $\Sigma$  PBDE<sub>6</sub>/kg lipid) from GRULAC countries in the period 2016-2019 and comparison to one previous survey conducted in the 2008-2011 period

### 3.4. Decabromodiphenyl ether (BDE-209)

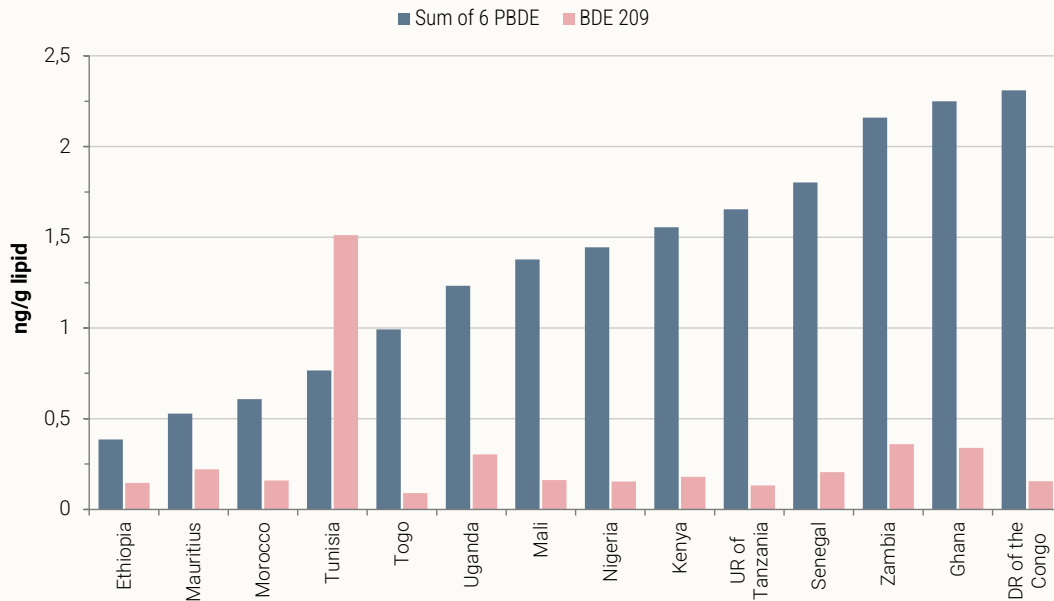
Decabromodiphenyl ether (BDE-209) was listed in Annex A of the Stockholm Convention in 2017 (Stockholm Convention 2019). It was included in the analysis of human milk samples for the period 2016-2019. In addition to the results of the six recommended analytes and the resulting sum parameter  $\Sigma$  PBDE<sub>6</sub> for the 36 countries covered by this report and the period 2016-2019, Table S7 presents also PBDE 209 and  $\Sigma$  PBDE<sub>7</sub>.

BDE-209 was quantified in 41 pooled human milk samples from 40 countries (including 6 self-funded countries - Germany submitted two pooled samples; and 33 project countries. In Egypt and Fiji, BDE-209 was not quantified and in Niue there were insufficient sample amounts for PBDE analyses). The concentration of BDE-209 during

this period were in the range between <0.06 ng/g and 5.92 ng/g, with a median of 0.21 ng/g and a 90th percentile of 1.53 ng/g. The median for the contribution of BDE-209 to the sum of 7 PBDEs ( $\Sigma$  PBDE<sub>7</sub> =  $\Sigma$  PBDE<sub>6</sub> + BDE-209) was 13 per cent, but ranged from 3 per cent to 66 per cent. This large difference in the contribution of BDE-209 to the sum of 7 PBDEs could possibly be explained by differences in local sources of emission and the use of different commercial PBDE mixtures as flame retardants.

In 13 countries out of 14 in Africa (excluding Egypt in which BDE-209 was not quantified) BDE-209 was not quantified, BDE-209 concentrations were below 0.4 ng/g lipid, with a contribution between 6 per cent and 66 per cent to the sum of 7 PBDEs. As a maximum, 1.51 ng BDE-209/g lipid was found in Tunisia, exceeding the  $\Sigma$  PBDE<sub>6</sub> concentration. In these 14 countries,  $\Sigma$  PBDE<sub>6</sub> levels were in the range of background contamination, at between 0.39 ng/g lipid and 2.31 ng/g lipid (Figure 25).

### Σ PBDE<sub>6</sub> and BDE 209 (African Group)

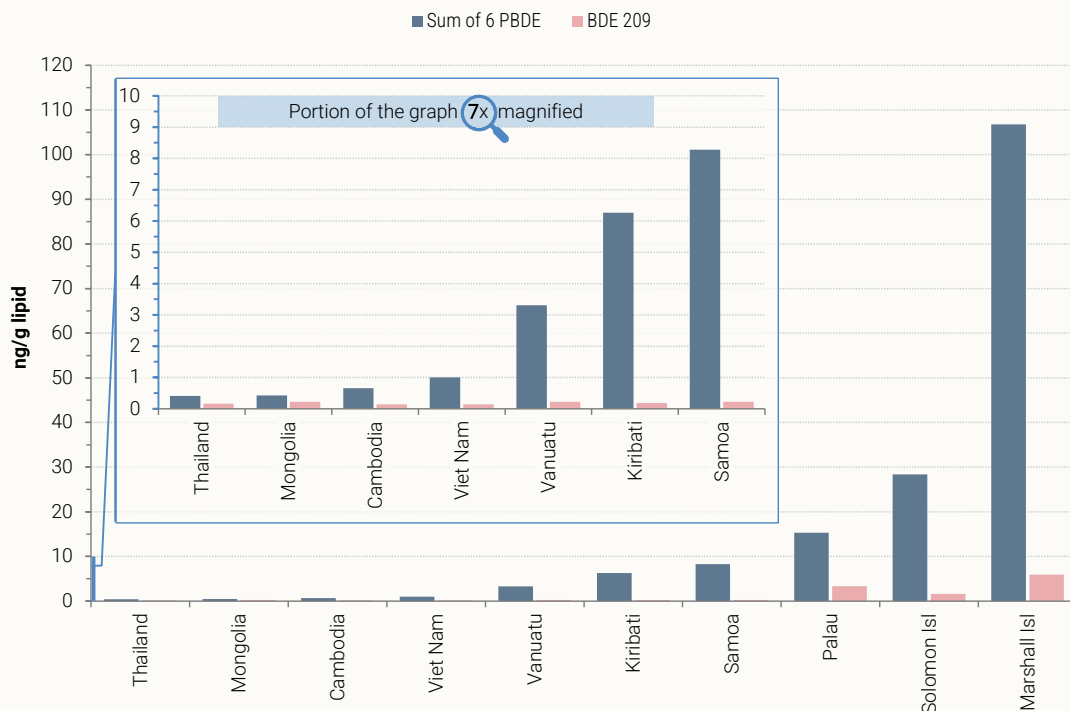


**Figure 25:** Concentration of BDE-209 and Σ PBDE<sub>6</sub> in human milk from countries in the African Group for the period 2016-2019 (expressed as ng Σ PBDE<sub>6</sub>/g lipid and ng BDE-209/g lipid, respectively)

In the four Asian countries the BDE-209 concentrations were below 0.3 ng/g lipid, with Σ PBDE<sub>6</sub> levels in the range of background contamination below 1 ng/g lipid. With increasing Σ PBDE<sub>6</sub> levels in the Pacific Islands countries higher BDE-209 concentrations were also found, with a maximum

of 5.92 ng BDE-209/g lipid in the Marshall Islands, contributing 5 per cent to Σ PBDE<sub>7</sub> (113 ng/g lipid). In Palau, BDE-209 results (3.31 ng/g lipid) contributed 18 per cent to Σ PBDE<sub>7</sub> (15.3 ng/g lipid). *Figure 26* illustrates the findings for these two regions, combined as “Asia-Pacific Group”.

### Σ PBDE<sub>6</sub> and BDE 209 (Asia-Pacific Group)

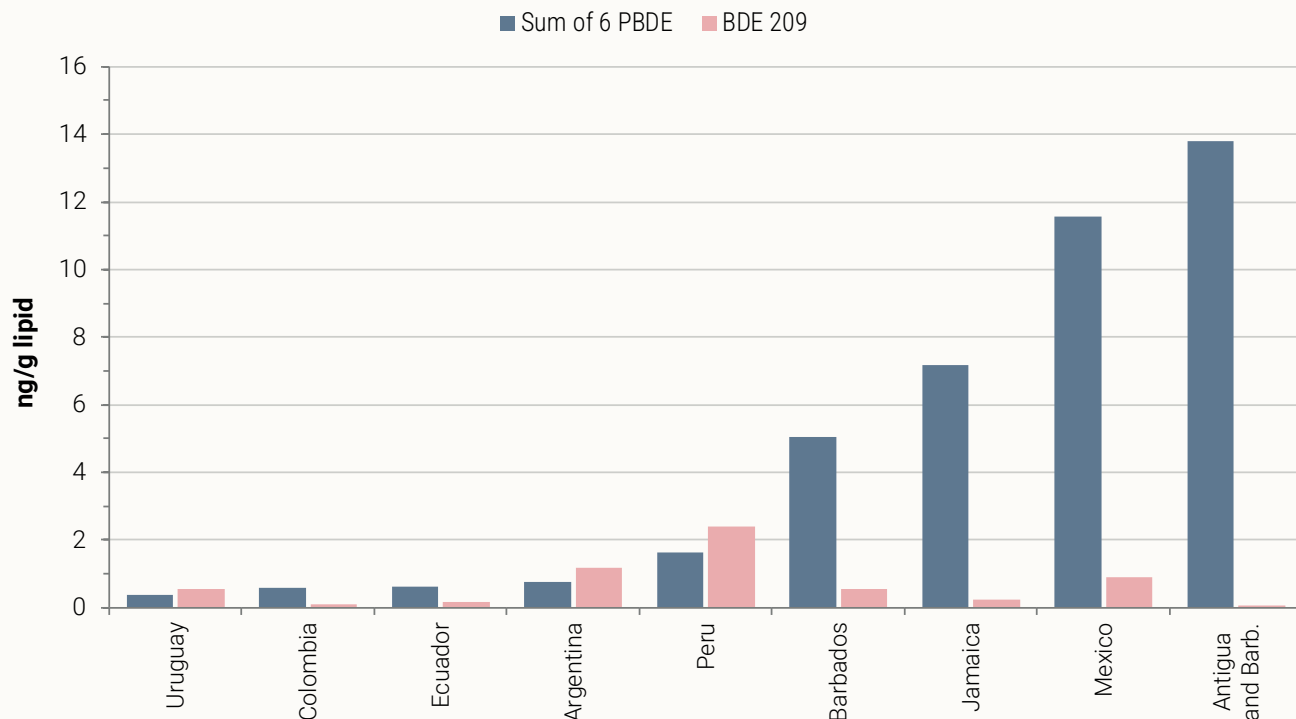


**Figure 26:** Concentration of BDE-209 and Σ PBDE<sub>6</sub> in human milk from countries in the Asia-Pacific Group for the period 2016-2019 (expressed as ng Σ PBDE<sub>6</sub>/g lipid and ng BDE-209/g lipid, respectively)

In most GRULAC countries BDE-209 concentrations were below 1 ng/g lipid, with a maximum of 2.4 ng BDE-209/g lipid in Peru. In many cases  $\Sigma$  PBDE<sub>7</sub> levels were in the range of a background contamination below 2 ng/g lipid in

five countries; the maximum  $\Sigma$  PBDE<sub>7</sub> concentration was found in Antigua and Barbuda (13.8 ng  $\Sigma$  PBDE<sub>6</sub>/g lipid), with BDE-209 levels below the limit of quantification (< 0.06 ng/g lipid) (Figure 27).

### $\Sigma$ PBDE<sub>6</sub> and BDE 209 GRULAC



**Figure 27:** Concentration of BDE-209 and  $\Sigma$  PBDE<sub>6</sub> in human milk from GRULAC countries for the period 2016-2019 (expressed as ng  $\Sigma$  PBDE<sub>6</sub>/g lipid and ng BDE-209/g lipid, respectively)

### 3.5. Hexabromocyclododecane (HBCD)

Technical products of hexabromocyclododecane (usual abbreviations HBCD or HBCD; here HBCD) predominantly contain the three stereoisomers  $\alpha$ -HBCD,  $\beta$ -HBCD and  $\gamma$ -HBCD. Whereas  $\gamma$ -HBCD is the main compound in technical HBCD,  $\alpha$ -HBCD is more persistent in the environment and biota, including humans.

The  $\alpha$ -HBCD levels of 102 pooled samples from 72 countries, collected between 2006 and 2019, ranged between < 0.1 ng/g lipid and 15 ng/g lipid (median: 0.5 ng/g lipid; 90 per cent of all results below 2 ng/g lipid).  $\beta$ -HBCD and  $\gamma$ -HBCD were in nearly all samples below the limit of

quantification (LOQ) for 90 per cent of the samples: < 0.1 ng/g lipid) or around the LOQ (max: 0.8 ng/g lipid). As a consequence,  $\alpha$ -HBCD is the predominant stereoisomer in human milk. Therefore, the sum parameter “sum of the three stereoisomers” is in close agreement with the  $\alpha$ -HBCD concentrations only.

For the 36 countries and their participation in the period 2016-2019 as covered by this report, the results of  $\alpha$ -HBCD,  $\beta$ -HBCD and  $\gamma$ -HBCD are presented in Table S8. In all surveys and in all countries of the African Group (Figure 28), the Asia-Pacific Group (Figure 29) and GRULAC Countries (Figure 30), the  $\alpha$ -HBCD levels were below 2 ng/g lipid.  $\beta$ -HBCD and  $\gamma$ -HBCD were in all samples below the limit of quantification (LOQ) for all these samples: < 0.1 ng/g lipid).

### alpha-HBCDD (African Group)

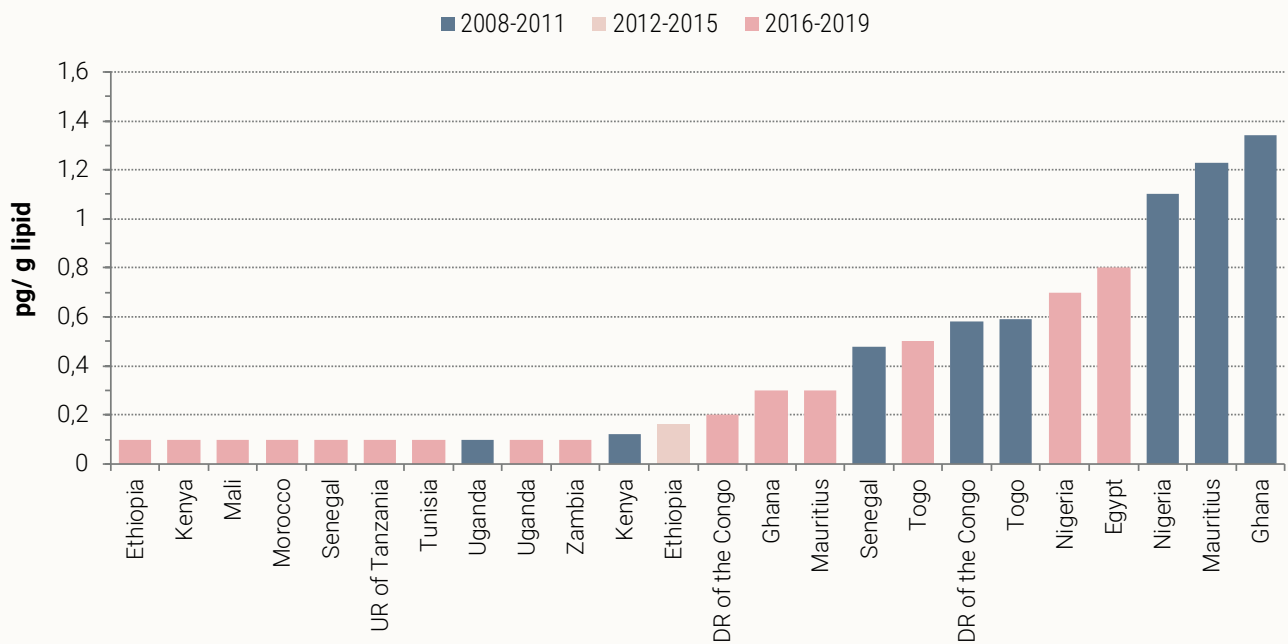


Figure 28: Concentrations of  $\alpha$ -HBCD in human milk ( $\mu\text{g } \alpha\text{-HBCD/kg lipid}$ ) from African countries in the period 2016-2019 in comparison to two previous surveys (with an indication of their periods)

### alpha-HBCDD (Asia-Pacific Group)

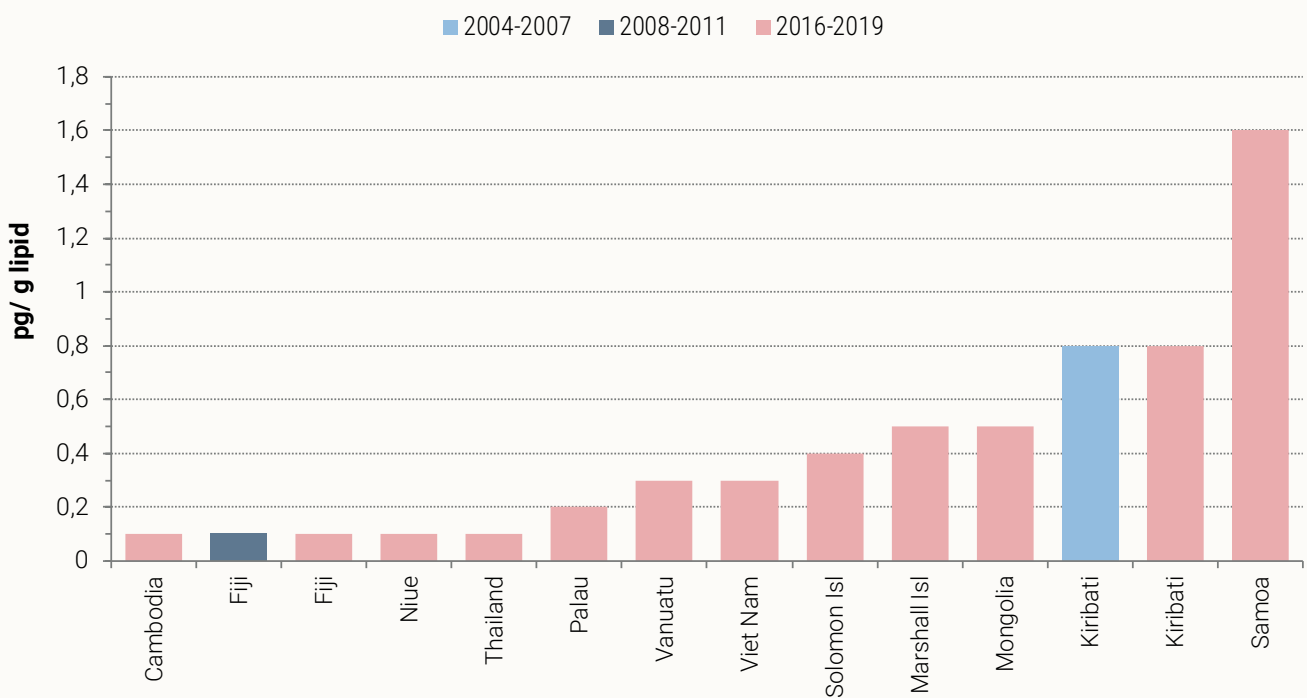
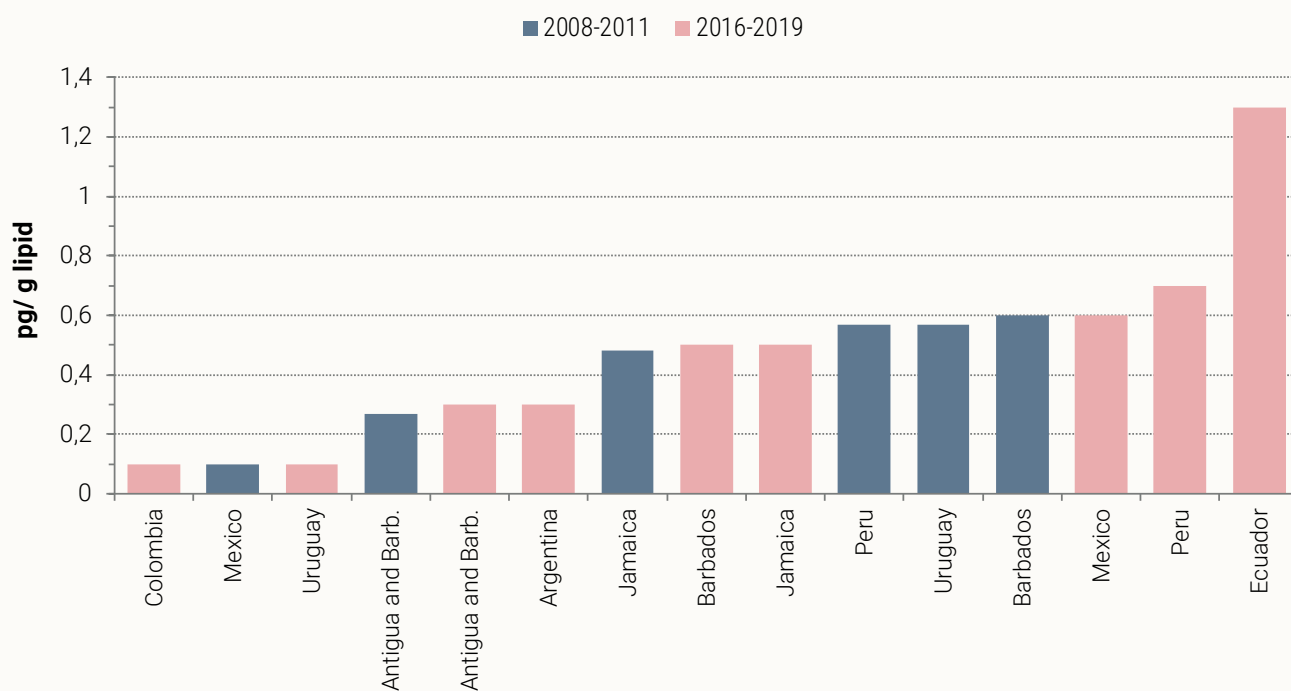


Figure 29: Concentration of  $\alpha$ -HBCD in human milk ( $\mu\text{g } \alpha\text{-HBCD/kg lipid}$ ) from countries in the Asia-Pacific Group for the period 2016-2019 in comparison to two previous surveys (with an indication of their periods)

## alpha-HBCDD (GRULAC)



**Figure 30:** Concentrations of  $\alpha$ -HBCD in human milk ( $\mu\text{g } \alpha\text{-HBCD/kg lipid}$ ) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

### 3.6. Hexabromobiphenyl (PBB 153)

Hexabromobiphenyl (PBB 153) was below the limit of quantification (0.5 ng/g lipid) in 106 of 110 pooled samples from 69 countries collected between 2009 and 2019. In four samples, low concentrations between 1.0 ng/g lipid and 1.7 ng/g lipid were found, among them those from the Democratic Republic of the Congo (2009) and Samoa (2011 and 2019).

### 3.7. Chlorinated paraffins (CPs)

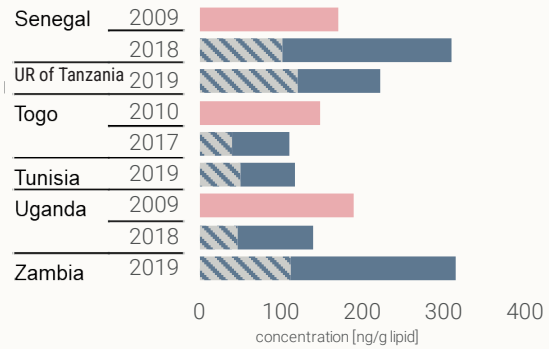
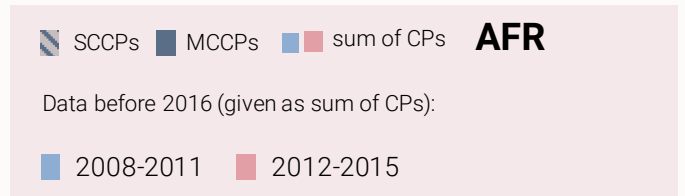
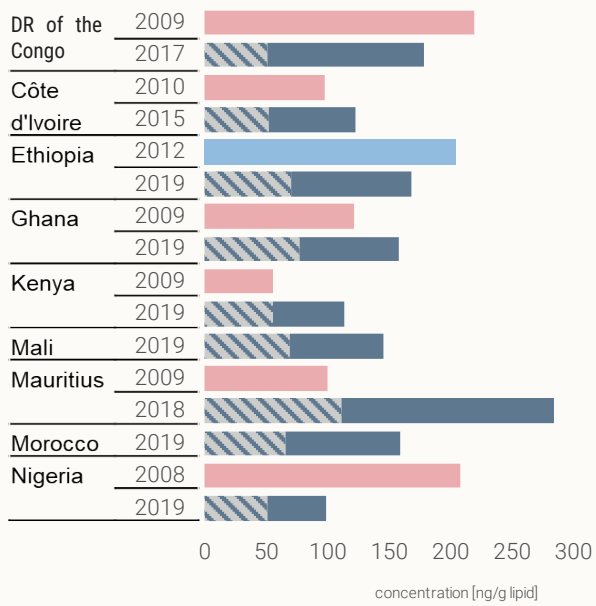
Chlorinated paraffins (CPs) are very complex mixtures of several million individual compounds. Contrary to medium-chain CPs (MCCPs,  $C_{14}\text{-}C_{17}$ ) and long-chain CPs (LCCPs,  $C_{18}\text{-}C_{30}$ ) investigated, the third subgroup, so far only short-chain chlorinated paraffins (SCCPs,  $C_{10}\text{-}C_{13}$ ) have been listed in 2017 in Annex A (elimination) (Stockholm Convention 2019). It has been proposed that MCCPs be listed and their inclusion is under review.

In 84 country-wide pooled human milk samples collected between 2009 and 2019 in 57 countries, the CP concentrations were determined. Until 2015 only total CP content

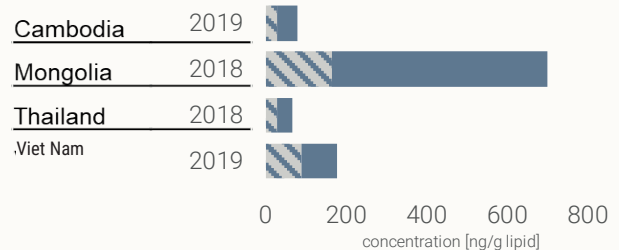
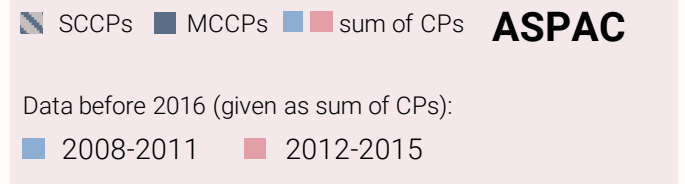
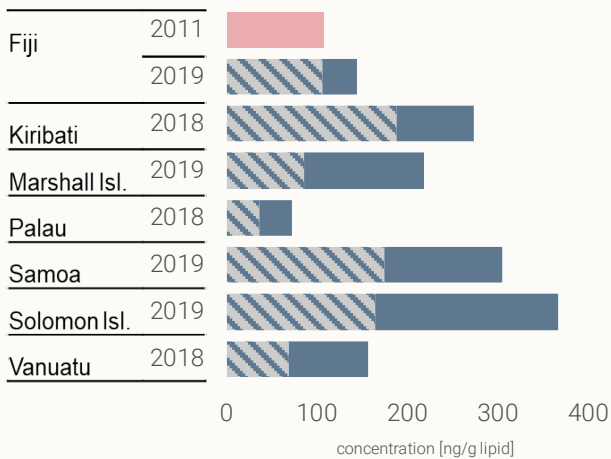
was determined. In light of ongoing efforts to also add other CP groups to the annexes of the Stockholm Convention and the glaring lack of data on the general background contamination world-wide, later on SCCPs and MCCPs were determined and the presence of LCCPs ( $C_{18}\text{-}C_{20}$  only) was investigated. CPs were present in all 84 samples, ranging 8.7-700 ng/g lipid (Krätschmer, Malisch and Vetter 2021).

For the 36 countries and their participation in the period 2016-2019 as covered by this report, the CP results are presented in *Table S9*. Within each geographical area, a wide range of CP levels was found (*Figures 31-33*). The samples from Mongolia (2018, 700 ng/g lipid CPs) marked the upper end of the concentration range.

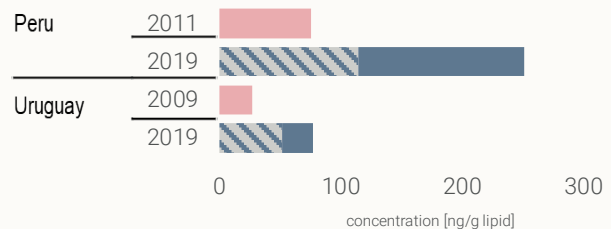
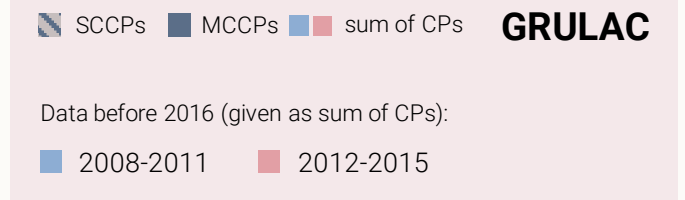
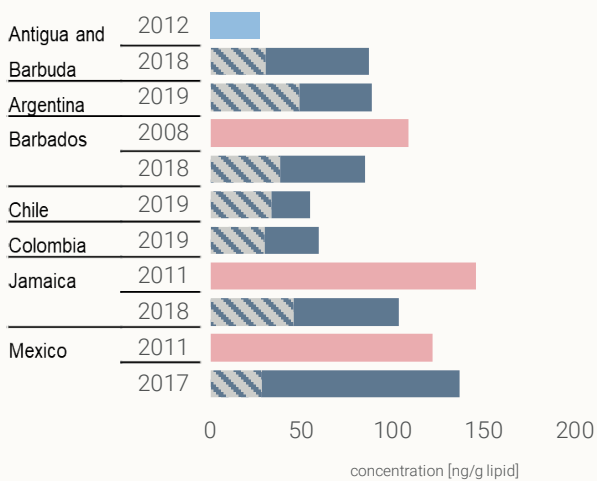
Regarding the relation between SCCPs and MCCPs, MCCP levels at least equalled SCCP levels in most samples, contributing 24-85 per cent to the total CP levels. In 36 of the 57 countries where distinct data was available, MCCPs even surpassed SCCPs. SCCPs and MCCPs dominated the share of POPs grouped as industrial chemicals and by-products in most areas, as shown in Chapter 4 (Summary and conclusions).



**Figure 31:** Concentrations of SCCPs, MCCPs and sum of CPs results for samples of human milk from countries of the African Group (AFR) in the period 2016-2019 and comparison to previous surveys, if applicable



**Figure 32:** Concentrations of SCCPs, MCCPs and sum of CPs results for samples of human milk from countries of the Asia-Pacific Group (ASPAC) in the period 2016-2019 and comparison to previous surveys, if applicable



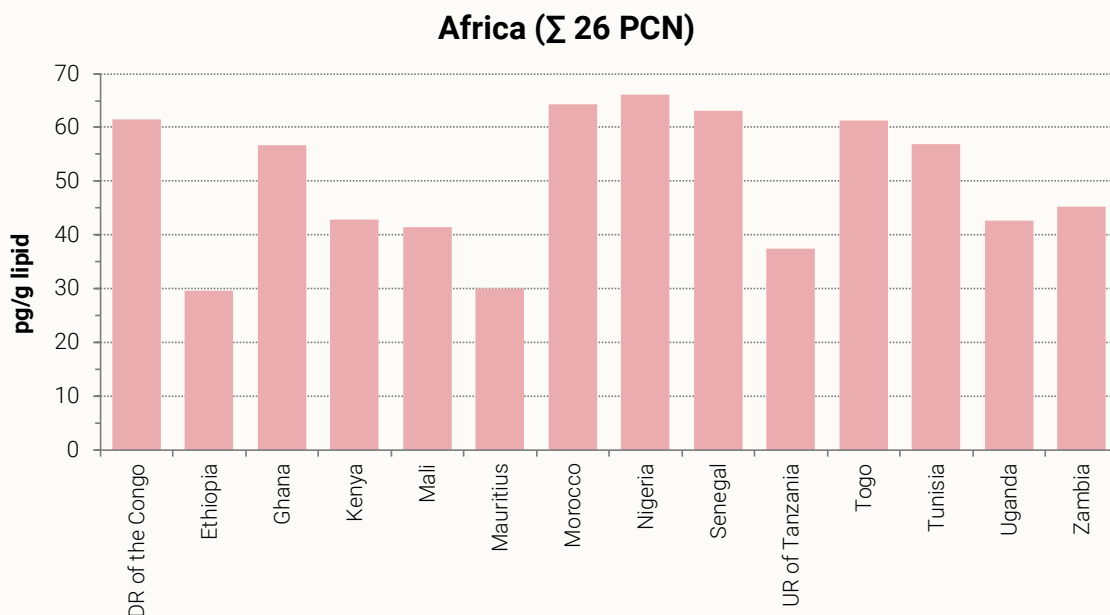
**Figure 33:** Concentrations of SCCPs, MCCPs and sum of CPs results for pooled country samples of human milk from GRULAC countries in the period 2016-2019 and comparison to previous surveys, if applicable

### 3.8. Polychlorinated naphthalenes (PCNs)

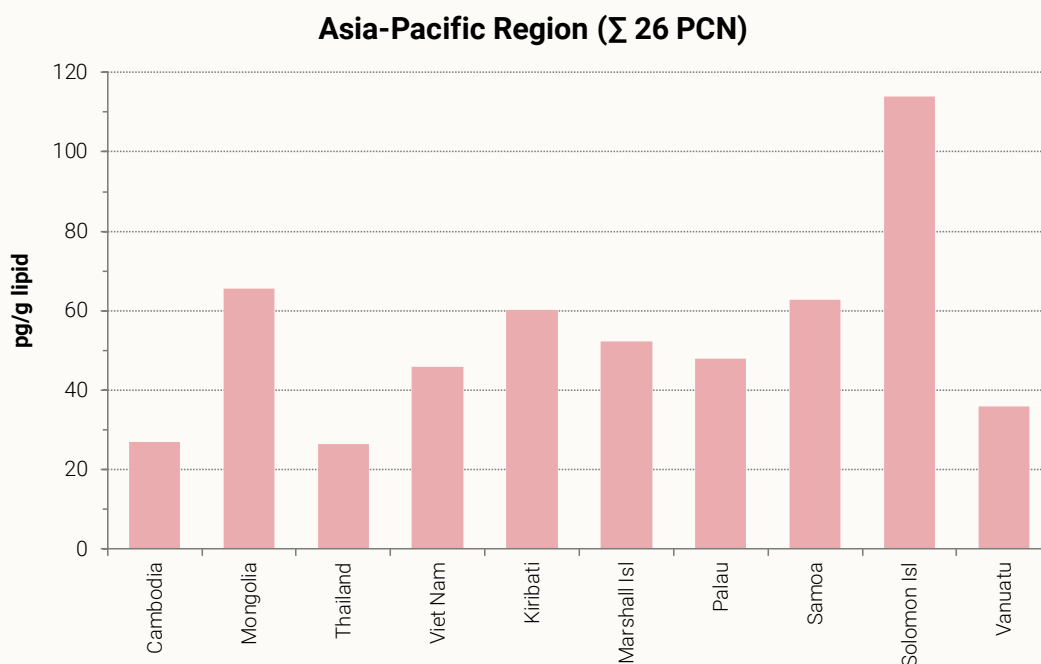
There are theoretically 75 PCN congeners (mono-chlorinated to octa-chlorinated) and, in practice, all occur at varying concentrations in technical products or are formed during thermal reactions. The GMP guidance (UNEP 2019) does not specify any specific congeners for analysis yet (“congeners to be decided”). For the WHO/UNEP-coordinated human milk survey in 2016-2019 a set of 26 PCN congeners were used. These

congeners were chosen based on toxicological characteristics, reported levels of occurrence, congener patterns, and the availability of analytical standards at times of method development and validation. Results are presented in *Tables S10-S12*.

The median concentration of  $\Sigma 26$  PCNs in pooled samples submitted by 39 countries in the period of 2016-2019 in the period 2016-2019 was 55 pg/g (picogram per gram) lipid (range 27-170 pg/g). *Figures 34 to 36* illustrate the findings in countries in the Africa, Asia-Pacific and GRU-LAC regions.

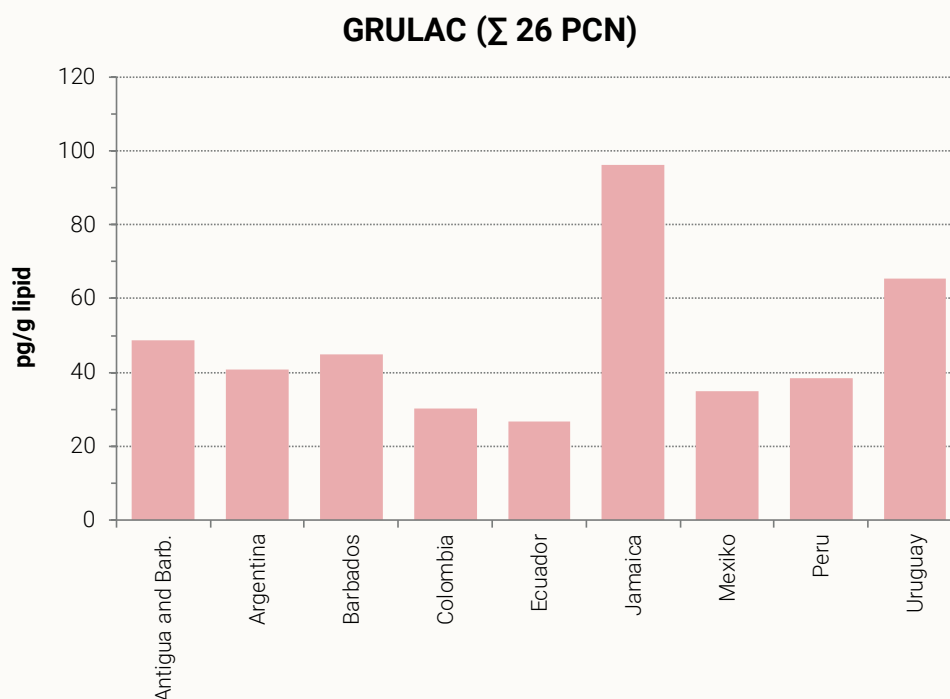


**Figure 34:** Results of the 2016-2019 survey for PCN concentrations (sum of 26 congeners) in human milk in countries from African countries (pg  $\Sigma$  26 PCN/g lipid)  
**Note:** Does not include Egypt due to lack of sufficient samples.



**Figure 35:** Results of the 2016-2019 survey for PCN concentrations (sum of 26 congeners) in human milk in countries from the Asia-Pacific region (pg  $\Sigma$  26 PCN/g lipid)





**Figure 36:** Results of the 2016-2019 survey for PCN concentrations (sum of 26 congeners) in human milk in GRULAC countries (pg  $\Sigma$  26 PCN/g lipid)

In June 2005, Geneva Switzerland, a World Health Organization (WHO)-International Programme on Chemical Safety expert meeting took place. Based on published data, there was agreement by the expert panel on the re-evaluation of toxic equivalency factors for dioxins and dioxin-like compounds assessment in 2005 that PCNs definitely should be considered for inclusion in the TEF concept, as among other adverse biological effects, PCNs also show dioxin-like toxicity (Van den Berg *et al.* 2006). This was estimated by calculating the toxic equivalents (TEQs) in these samples using two sets of relative effect potency (REP) values: i) a set used in earlier human exposure studies

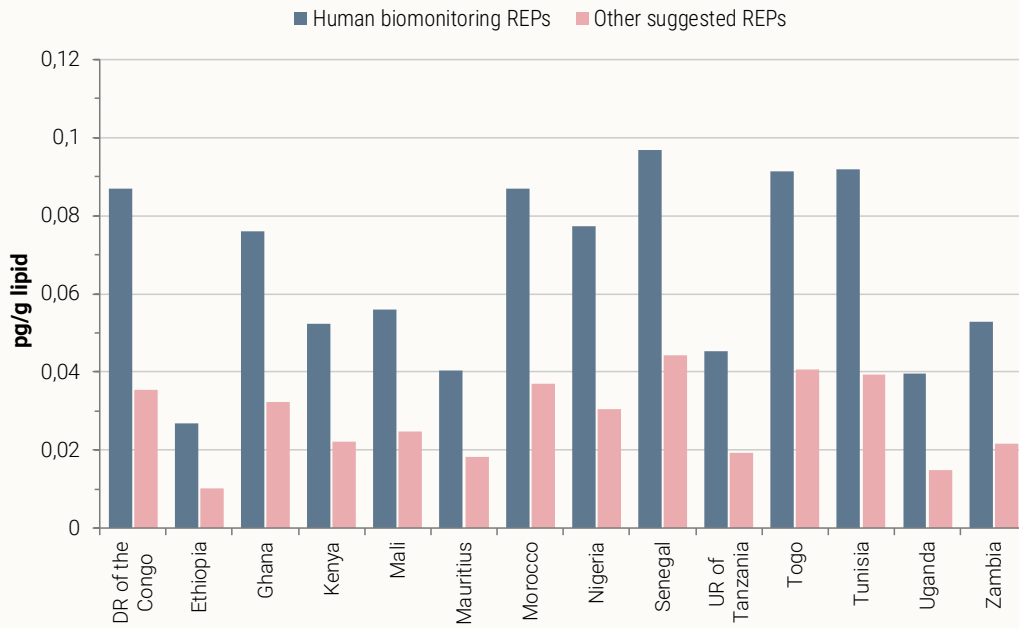
(e.g. Fernandes, Rose and Falandysz *et al.* 2017; Zacs *et al.* 2021); and ii) REPs suggested by Falandysz *et al.* (2014).

Figures 37 to 39 illustrate the results for countries in the Africa, Asia-Pacific and GRULAC regions. Finally, the contribution of PCNs to the overall sum of toxic equivalents is of interest. In comparison to PCDD/PCDF and DL-PCB (calculated as WHO<sub>2005</sub>-TEQ), the contribution of PCN-TEQ was on average between 1 and 2 per cent, with a wider range up to 5 per cent for the 39 countries participating in the 2016-2019 study.



PCNs can bioaccumulate in the tissues of aquatic organisms, including fish, when these are consumed by people the pollutants can enter the human food chain.

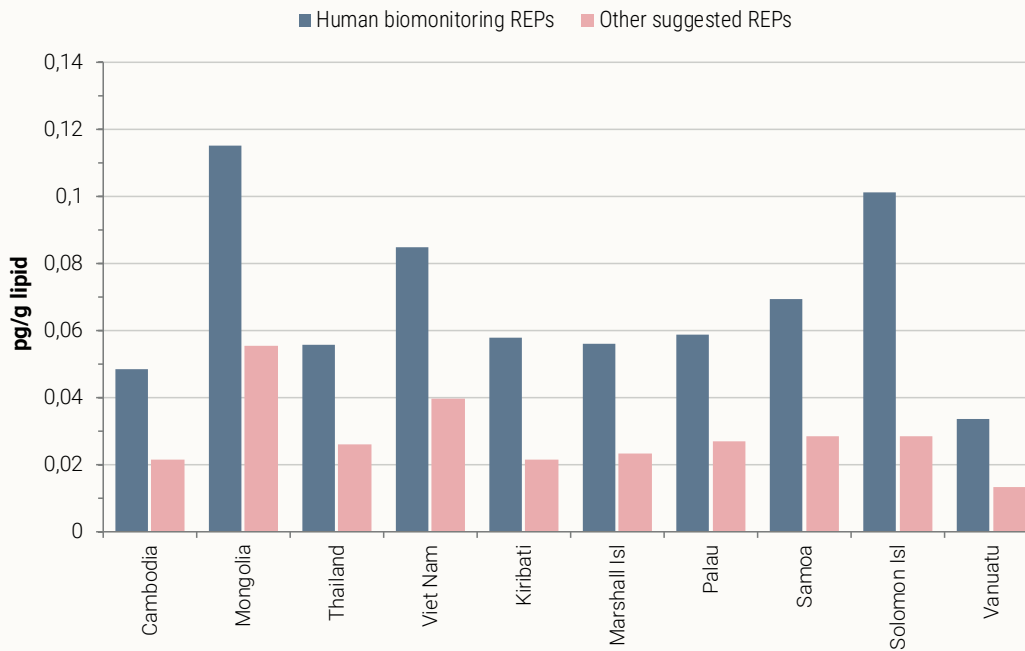
### Africa, PCN-TEQ



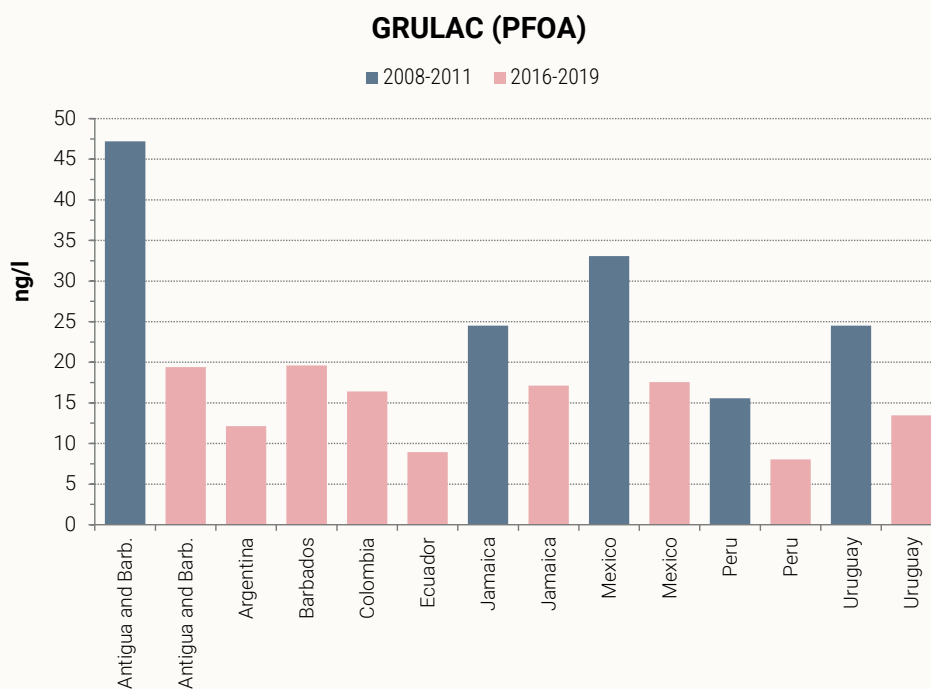
**Figure 37:** Results of the 2016-2019 survey for PCN-TEQ concentrations in human milk in African countries (pg PCN-TEQ/g lipid), if calculated with (i) REPs as used in human biomonitoring studies and (ii) other suggested REPs (Falandysz *et al.* 2014)

**Note:** Does not include Egypt due to lack of sufficient samples.

### Asia-Pacific, PCN-TEQ



**Figure 38:** Results of the 2016-2019 survey for PCN-TEQ concentrations in human milk in countries from the Asia-Pacific region (pg PCN-TEQ/g lipid), if calculated with (i) REPs as used in human biomonitoring studies and (ii) other suggested REPs (Falandysz *et al.* 2014)



**Figure 39:** Results of the 2016-2019 survey for PCN-TEQ concentrations in human milk in GRULAC countries (pg PCN-TEQ/g lipid), if calculated with (i) REPs as used in human biomonitoring studies and (ii) other suggested REPs (Falandysz *et al.* 2014)

### 3.9. Perfluorinated alkane substances (PFAS)

The targeted compounds were the two PFAS, perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), listed in Annexes B and A, respectively (Stockholm Convention 2019), and the one PFAS, perfluorohexane sulfonic acid (PFHxS), recommended to be listed.<sup>3</sup> In contrast to the lipophilic chlorinated and brominated POPs, which are reported on a lipid basis, the more polar PFAS data are reported on a product basis, e.g. as pg/g fresh weight (f.w.) or on volume basis, e.g. as nanograms per litre (ng/L).

The results of the UNEP/GEF GMP-2 project in the Africa, Asia, the Pacific Islands and GRULAC regions have been presented and discussed in two peer reviewed publications:

- Data on the regional occurrence of these substances in human milk for the Global Monitoring Plan under the Stockholm Convention in 2016-2019 comprise 44 human milk samples collected in 42 countries (Niue and Germany have two samples), including 35 countries of the UNEP/GEF GMP-2 project (does not include Egypt due to lack of sufficient sample material). PFOS was quantifiable in 35 out of the 43 pooled

samples across a wide range (total PFOS between <6.2 pg/g and 212 pg/g, calculated as sum of L-PFOS and br-PFOS); PFOA was quantified in all 44 samples in a quite narrow range (6.20 pg/g-37.4 pg/g); PFHxS was quantifiable in only four samples (max. 111 pg/g). Branched PFOS isomers had a share of 16 per cent of the total PFOS, on average, with a maximum of 33 per cent (Fiedler and Sadia 2021).

- Data on 101 samples, consisting of 86 national pools and 15 pools from States in Brazil obtained between 2008 and 2019, were used to estimate temporal trends as well. It was concluded that the goal to achieve a 50 per cent decrease in concentrations within 10 years had been met by Antigua and Barbuda, Kenya, and Nigeria for PFOS and by Antigua and Barbuda for PFOA. In a few cases increases were observed: in one country for PFOS and in four countries for PFOA (Fiedler *et al.* 2022).

With regard to the assessment of time trends, two aspects should be noted:

- There is no stipulation of a quantitative goal for the rate of reduction/decrease in POPs levels. The Convention's objectives are either to eliminate or to reduce production, use and releases, depending on the annex where a chemical is listed, but the rate of decline is nowhere specified or required. However, the 50 per

<sup>3</sup> In June 2022 the Conference of the Parties amended Annex A to the Convention as follows: "List perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds in Annex A without specific exemptions" (Decision SC-10/13) (Stockholm Convention 2022).

cent decrease rate in levels of POPs within a 10-year period was proposed as a quantitative objective for the ability of temporal studies to provide reliable monitoring information for the Parties to the Stockholm Convention.

- For the evaluation of time trends by Fiedler *et al.* (2022), samples were grouped into three five-year periods: 2005-2009, 2010-2014 and 2015-2019. However, samples were obtained between 2008 and 2019, with three samples from 2008 and 14 samples from 2009. Thus the first period (2005-2009) is rather a one- or two-year period comprising the years 2008 and, mainly, 2009. Three equal four-year periods (2008-2011, 2012-2015 and 2016-2019), reflecting more closely the rounds in the WHO/UNEP-coordinated exposure studies, seem more appropriate and were used in this report. These definitions of time periods result in significant differences. Comparison of average concentrations of PFOS, PFOA and PFHxS in the periods 2005-2009, 2010-2014 and 2015-2019 seems to indicate an increase from 2005-2009 to 2010-2014 and afterwards a decrease to 2015-2019 for these three analytes. However, grouping the countries relevant for this report into the three equal four-year periods which reflect quite closely the periods of the surveys shows decreasing trends for their participation in the 2008-2011 period to the period 2016-2019 (see the following).

For the findings in countries in the Africa, Asia-Pacific and GRULAC regions the data base of the GMP Data Warehouse was used, providing PFAS data on a volume basis (as ng/L). Results for the period 2016-2019 are presented in *Table S13*. Many African and GRULAC countries, and Fiji, participated

during the period 2008-2011. As an indication of temporal tendencies, these results were included in the illustration of the 2016-2019 findings for  $\Sigma$  PFOS (*Figures 40-42*) and for PFOA (*Figures 43-45*).

In the 14 African countries (excluding Egypt due to lack of sample material)  $\Sigma$  PFOS concentrations between 2008 and 2019 were in a range <6.4-31.5 ng/L, with downward tendencies in nine countries with availability of data for 2008-2011 as well. In these nine countries with repeated participation,  $\Sigma$  PFOS concentrations decreased by 45 per cent (as median) from 2008-2011 to 2016-2019. Overall, levels decreased from 23.7 ng/L as median (range 9.4-31.5 ng/L) in 2008-2011 to 10.6 ng/L as median (range <6.4-22.5 ng/L) in 2016-2019.

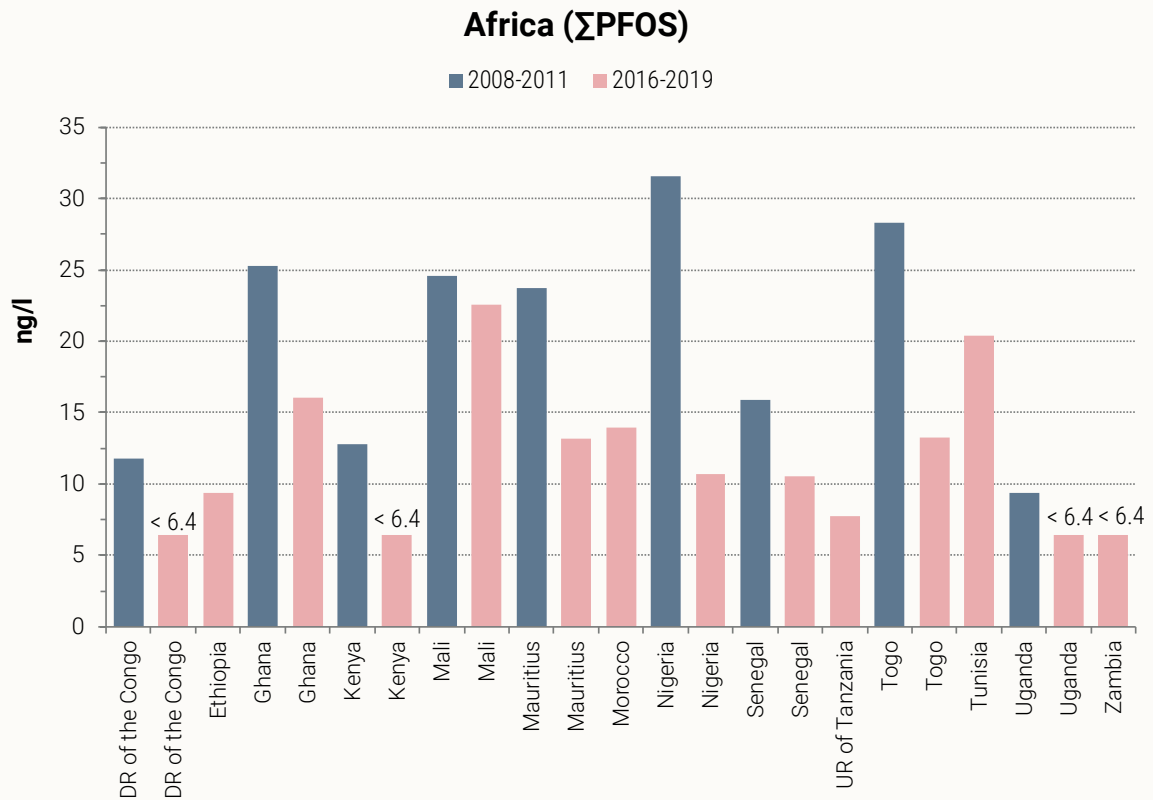
From the 12 countries in the Asia-Pacific region only data from Fiji were available for the period 2008-2011, showing a reduction of nearly 80 per cent from 27 ng/L to <6.4 ng/L. In the period 2016-2019 most countries had  $\Sigma$  PFOS concentrations in the range <6.4-30 ng/L (median 17.7 ng/L); however, the  $\Sigma$  PFOS concentrations in Kiribati exceeded this range by an order of magnitude (218 ng/L).

In the nine GRULAC countries  $\Sigma$  PFOS concentrations between 2008 and 2019 were in a range <6.4-58.6 ng/L, with downward tendencies in five countries that had data available for 2008-2011 as well. In these five countries with repeated participation,  $\Sigma$  PFOS concentrations decreased by 54 per cent (as median) between 2008-2011 and 2016-2019. Overall, levels decreased from 36.0 ng/L as median (range 9.6-58.6 ng/L) in 2008-2011 to 12.2 ng/L as median (range <6.4-41.7 ng/L) in 2016-2019.



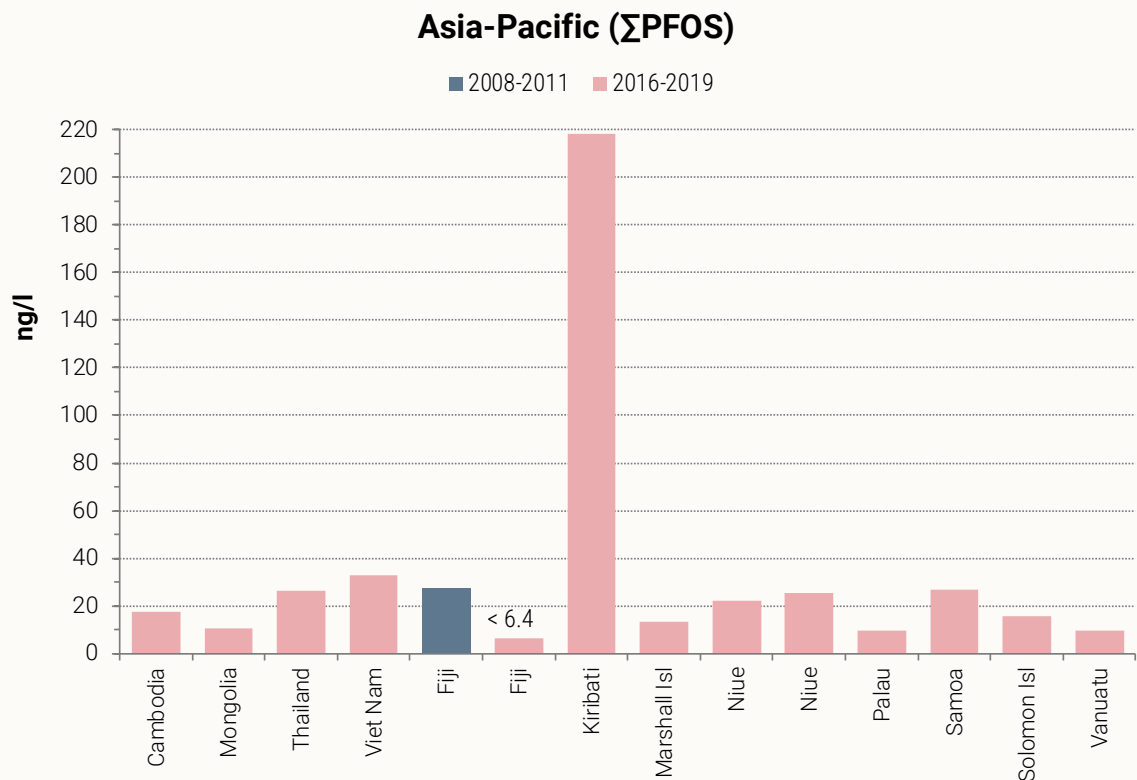
Photo: ©Pexels / Caio

PFASs are commonly used in the production of nonstick coatings for cookware, such as frying pans, griddles, and bakeware.

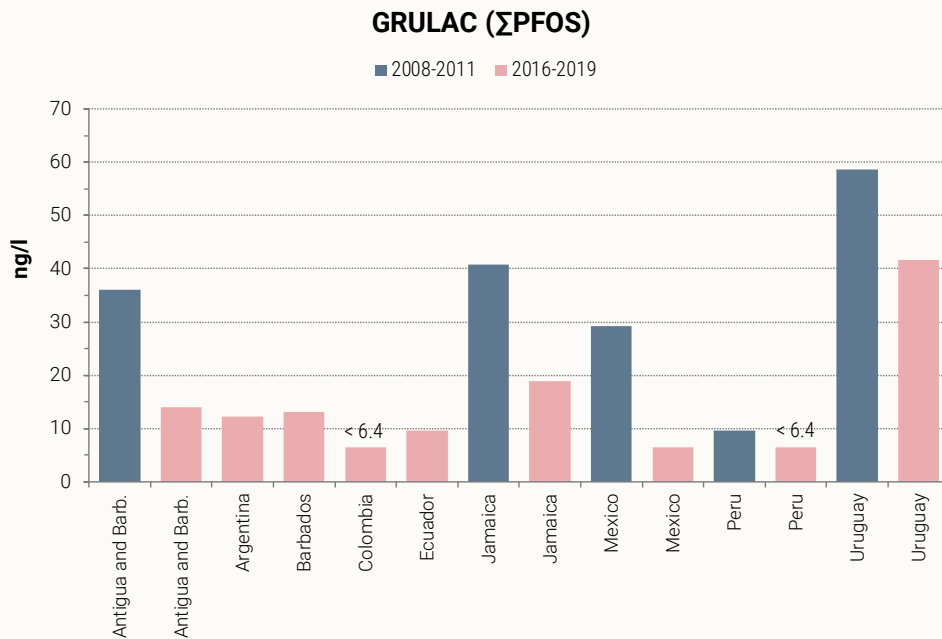


**Figure 40:** Concentrations of  $\Sigma$  PFOS in human milk (ng  $\Sigma$  PFOS/L) from African countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period)

**Note:** Does not include Egypt due to lack of sufficient samples.



**Figure 41:** Concentrations of  $\Sigma$  PFOS in human milk (ng  $\Sigma$  PFOS/L) from Asia-Pacific countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

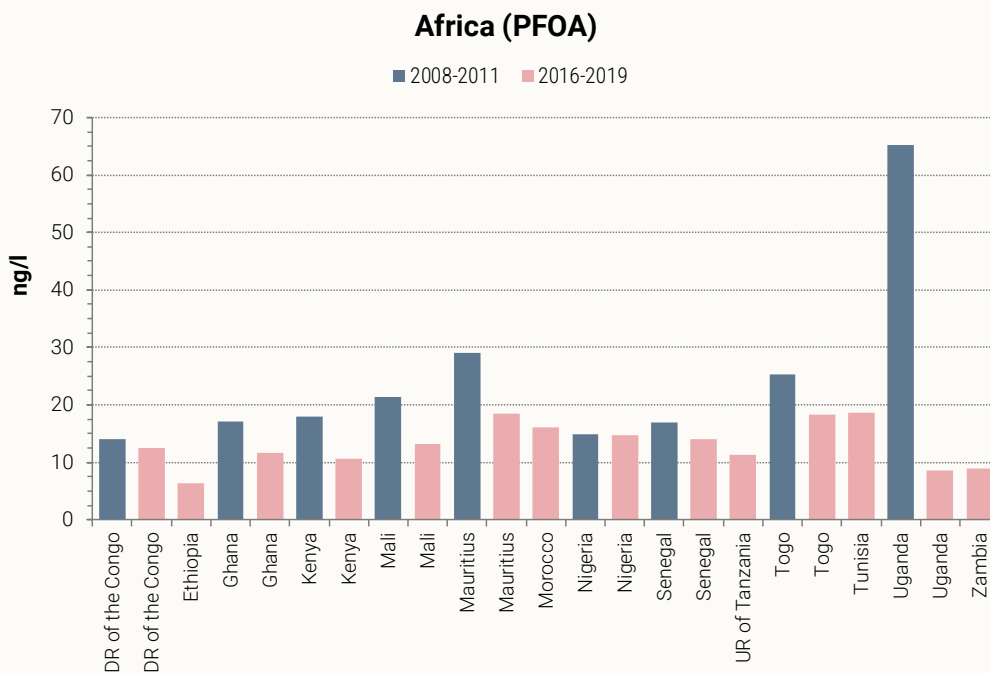


**Figure 42:** Concentrations of Σ PFOS in human milk (ng Σ PFOS/L) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period)

In the 14 African countries (Egypt not included due a lack of sufficient samples) PFOA concentrations between 2008 and 2019 were in a range 6.4-65.3 ng/L, with downward tendencies in nine countries with availability of data for 2008-2011. In these nine countries with repeated participation PFOA concentrations decreased by 32 per cent (as median) from 2008-2011 to 2016-2019. Overall, levels decreased from 18.0 ng/L as median (range 14.0-65.3

ng/L) in 2008-2011 to 12.8 ng/L as median (range 6.4-18.6 ng/L) in 2016-2019.

From the 12 countries in the Asia-Pacific region only data from Fiji were available for 2008-2011, showing a reduction of 70 per cent from 36 ng/L to 10.5 ng/L. In the period 2016-2019 all countries had PFOA concentrations in a range between 10 and 33 ng/L (median 15.1 ng/L).



**Figure 43:** Concentrations of PFOA in human milk (ng PFOA/L) from African countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period)

**Note:** Does not include Egypt due to lack of sufficient samples.

### Asia-Pacific (PFOA)

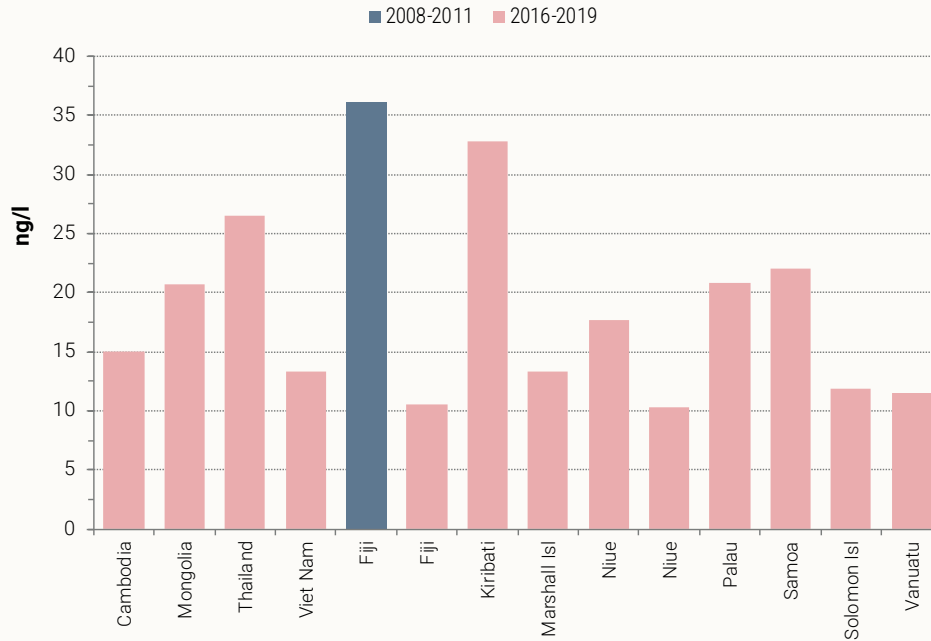


Figure 44: Concentrations of PFOA in human milk (ng PFOA/L) from Asia-Pacific countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

### GRULAC (PFOA)

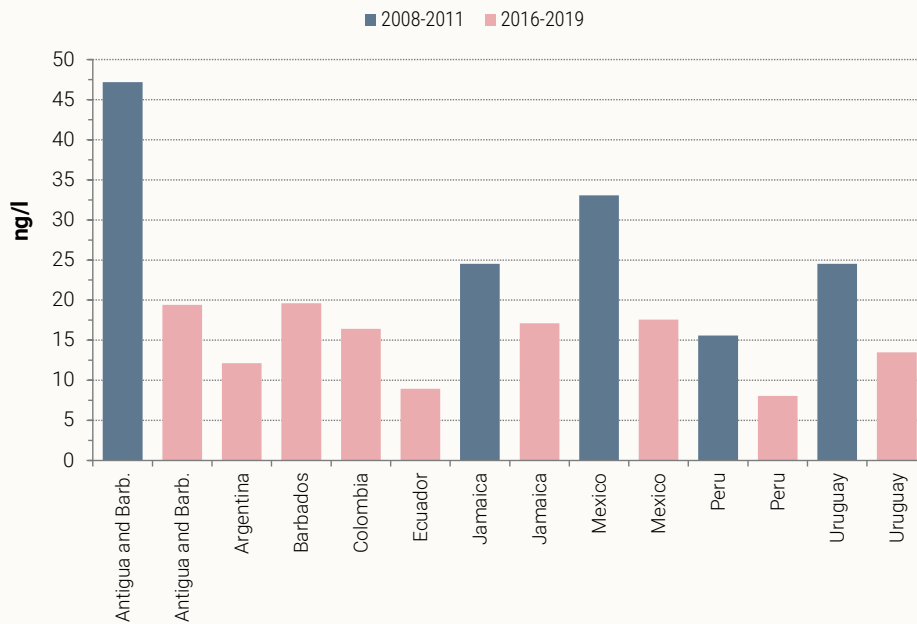


Figure 45: Concentrations of PFOA in human milk (ng PFOA/L) from GRULAC countries in the period 2016-2019 in comparison to one previous survey conducted in the 2008-2011 period

In the nine GRULAC countries, PFOA concentrations between 2008 and 2019 were in a range 8.0-47.2 ng/L, with downward tendencies in five countries with availability of data for 2008-2011. In these five countries with repeated participation PFOA concentrations decreased by 48 per cent (as median) between 2008-2011 and 2016-2019. Overall, levels decreased from 24.5 ng/L as median (range 15.6-47.2 ng/L) in 2008-2011 to 16.4 ng/L as

median (range 8.0-19.6 ng/L) in 2016-2019.

In the period 2016-2019, in 33 of 35 countries in the Africa, Asia-Pacific and GRULAC regions (excluding Egypt due to a lack of sufficient samples), PFHxS concentrations were below the limit of quantification (5.5 ng/L). One country had PFHxS concentrations slightly above the LOQ (Thailand, 7.5 ng/L), whereas another country had more than 10 times higher levels (Kiribati, 115 ng/L).

# SECTION 4

## Summary and conclusions





## 4. SUMMARY AND CONCLUSIONS

This report presents the findings of the WHO/UNEP-coordinated survey on persistent organic pollutants (POPs) in human milk as a core matrix under the GMP of the Stockholm Convention, performed in 2016-2019 in 36 countries in the Africa, Asia, the Pacific Islands and GRULAC regions. To ascertain comparability of results with previous WHO/UNEP-coordinated exposure studies performed between 2000 and 2019, human milk samples were collected following WHO- and UNEP-designed protocols under the supervision of a national coordinators in each country. A large number of individual samples was collected, and from equal amounts of the individual samples a pooled sample was prepared that was considered to be representative for the country. By performing the analysis at the Reference Laboratories (for chlorinated and brominated POPs in the period 2000-2019 at CVUA Freiburg, Germany, and for perfluoroalkane substances in the period 2009-2019 at Örebro University, Sweden), a high degree of reliability could be achieved.

By 2019 the POPs listed under the Stockholm Convention had increased to 30 chemicals or groups of chemicals (28 chlorinated or brominated, two perfluorinated). As no multi-method exists that would allow the determination of all POPs of interest by one method, various analytical methods with comprehensive quality control were applied. The collection of human milk, as a non-invasive sampling method, and the preparation of pooled samples that are considered representative of a country have a number of advantages. The most important are:

- This approach is very cost-effective;
- Due to the relatively high fat content of human milk and the large volume of the pooled (composite) sample, sufficient sample material was available to apply different methods for determination of all 30 presently listed POPs by 2019, as well as the two POPs proposed for listing: medium-chain chlorinated paraffins (MCCPs) and perfluorohexane sulfonic acid (PFHxS).<sup>4</sup> Thus results for this complete set of 30 POPs and two additional ones (chemicals of interest) proposed for listing under the Convention are available for this core matrix for the 2016-2019 period.

<sup>4</sup> In June 2022 the Conference of the Parties amended Annex A to the Convention as follows: "List perfluorohexane sulfonic acid (PFHxS), its salts and PFHxS-related compounds in Annex A without specific exemptions" (Decision SC-10/13) (Stockholm Convention 2022).

The project aimed at supporting the Convention's implementation by providing data to the effectiveness evaluation as required under Article 16. Temporal tendencies in POPs concentrations are indicated for 24 of the 36 countries with repeated participation in WHO/UNEP-coordinated exposure studies by comparing the 2016-2019 results with previous ones. Moreover, this survey contributes to the derivation of statistically significant time trends in the UN regional groups and globally on the basis of all 82 countries during the period 2000-2019. This places it amongst the largest the longest running global studies on human exposure to POPs.

Another important aspect is the share of the individual 30 POPs and two additional chemicals in total POPs concentrations. For discussion of this aspect, the following differentiation is necessary:

The lipophilic chlorinated and brominated POPs are reported on a lipid basis. Here, dioxin-like compounds (PCDDs, PCDFs, and dioxin-like PCBs contributing to toxic equivalents [TEQ], as well as polychlorinated naphthalenes [PCNs], which, according to peer reviewed publications, also have dioxin-like toxicity) have to be determined in the picogram per gram (pg/g) range, whereas the other chlorinated and brominated POPs are usually determined in the nanogram per gram (ng/g) (= microgram per kilogram [ $\mu\text{g}/\text{kg}$ ]) range.

The more polar perfluorinated alkane *substances* data are usually reported on a product basis (as pg/g fresh weight) or on a volume basis (ng/L).

During the whole period between 2000 and 2019 Africa had the widest variation in contamination of human milk with total TEQ (from contributions of PCDDs, PCDFs and DL-PCBs) observed in any group of countries (1.29-49 pg WHO<sub>2005</sub>-TEQ/g). Most of the African countries that participated in the 2016-2019 round had participated for the first time during the period 2008-2011; in these countries WHO<sub>2005</sub>-TEQ concentrations fell on average by about 22 per cent (range 14-40 per cent) until the period 2016-2019. The highest declining rate (about 60 per cent) was observed in Egypt between 2001-2002 and 2019.

None of the four Asian countries that participated in the 2016-2019 round had participated between 2000 and 2015. Thus temporal tendencies could not be derived for these countries.

All samples from the Pacific Islands countries submitted between 2000 and 2015 were in the range of approximately 3-6 pg WHO<sub>2005</sub>-TEQ/g lipid. In nearly all samples from

the period 2016-2019 concentrations were below 4 pg/g. Downward tendencies were observed in nearly all countries with repeated participation. Only one sample, from the Marshall Islands (2019), had a substantially higher concentration. The range of total TEQ concentrations in the 2016-2019 survey was 1.29-11.6 pg WHO<sub>2005</sub>-TEQ/g lipid (median 3.59 pg WHO<sub>2005</sub>-TEQ/g lipid).

The range for the other chlorinated and brominated POPs found in the 2016-2019 survey is shown in *Figures 46 and 47*. By far the highest concentrations were found for DDT and chlorinated paraffins (including both short-chain chlorinated paraffins [SCCPs] and MCCPs). The maximum concentration found for DDT (7100 ng/g lipid) was a factor of 10 higher than the maximum for the CPs (700 ng/g lipid for the sum of SCCP and MCCP). However, the median for CP concentrations (143 ng  $\Sigma$  SCCPs + MCCPs /g lipid [ $\Sigma$ SMCCPs /g lipid]) was higher than the median for DDT concentrations (128 ng/g lipid). The high CP concentrations were caused predominantly by MCCPs (median 83 ng/g lipid; maximum 536 ng/g lipid), with SCCP concentrations of 61 ng/g lipid as median and 188 ng/g lipid as maximum. PCBs follow next in the ranking of decreasing levels, with concentrations an order of magnitude lower on average than the CP concentrations (median 7.31 ng

NDL-PCB/g lipid [nanograms of non-dioxin-like PCB per gram of lipid] maximum 90 ng/g lipid).

In contrast to the decreasing tendencies for DDT and NDL-PCBs as “old legacy POPs” in most countries, concentrations of CPs as “emerging POPs” showed increasing tendencies in many countries.

Median concentrations of between 1 ng/g lipid and 3 ng/g lipid were found for  $\Sigma$  PBDE<sub>6</sub> (as well as  $\Sigma$  PBDE<sub>7</sub>, including BDE-209), beta-HCH and HCB; maximum concentrations between 10 ng/g lipid and 110 ng/g lipid were found for pentachloranisole, nonachlor, beta-HCH and  $\Sigma$  PBDE<sub>6</sub>. Concentrations of other chlorinated and brominated POPs were frequently below LOQ (0.5 ng/g lipid) or, if quantifiable, below 10 ng/g lipid. For polychlorinated naphthalenes (PCNs), which, according to peer reviewed publications, also have dioxin-like properties, the quite low concentrations for  $\Sigma$  PCN<sub>26</sub> could be assessed for a possible contribution as dioxin-like compounds to TEQs. Consequently, the contribution of PCNs to the overall sum of toxic equivalents is of interest.

Concentrations below 5 ng/g lipid can be seen as background concentrations. Background concentrations are defined as that portion of measured human milk levels

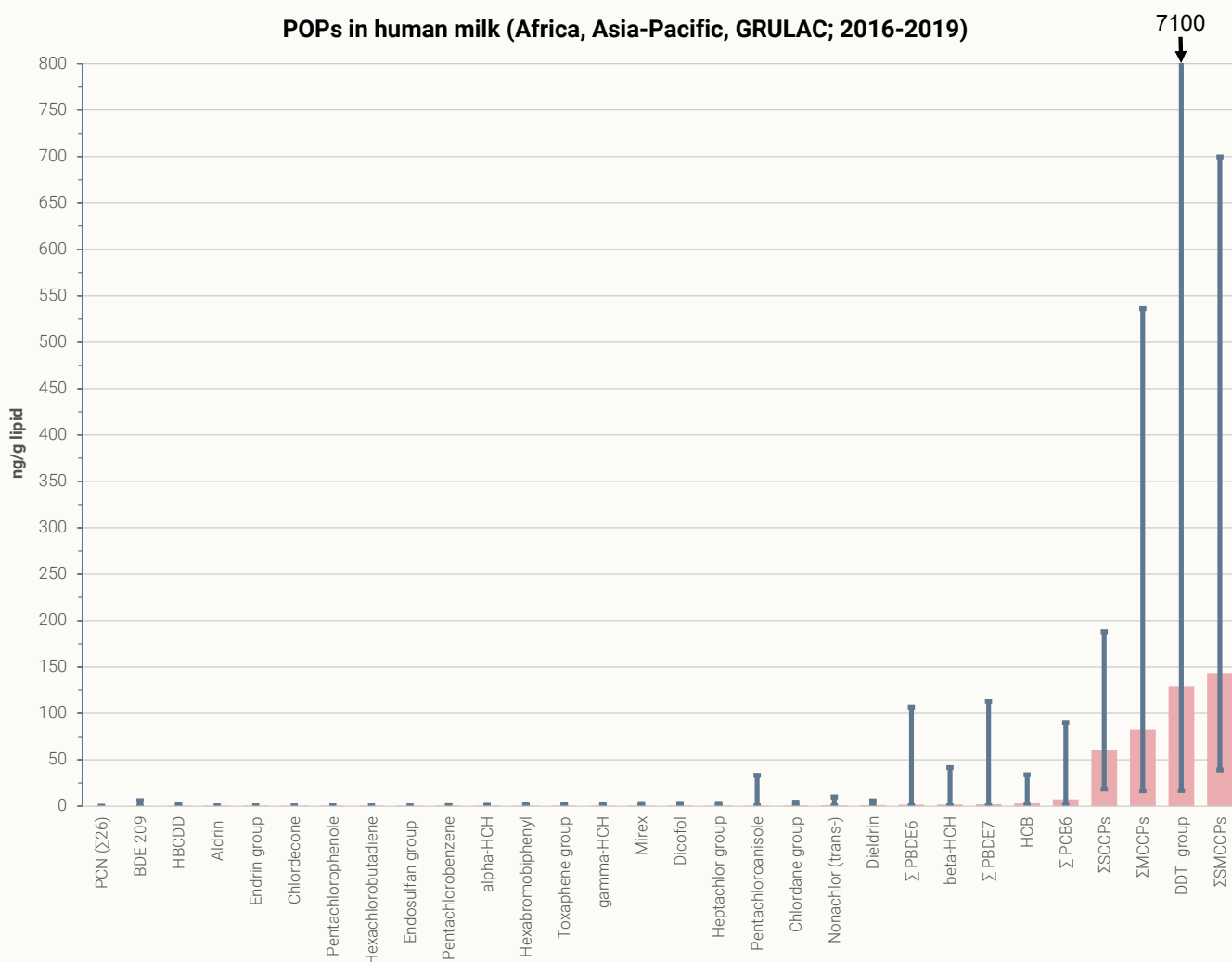


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found in the absence of specific sources and therefore not attributable to a known exposures, such as use of the chemical of interest or emissions within the study area.

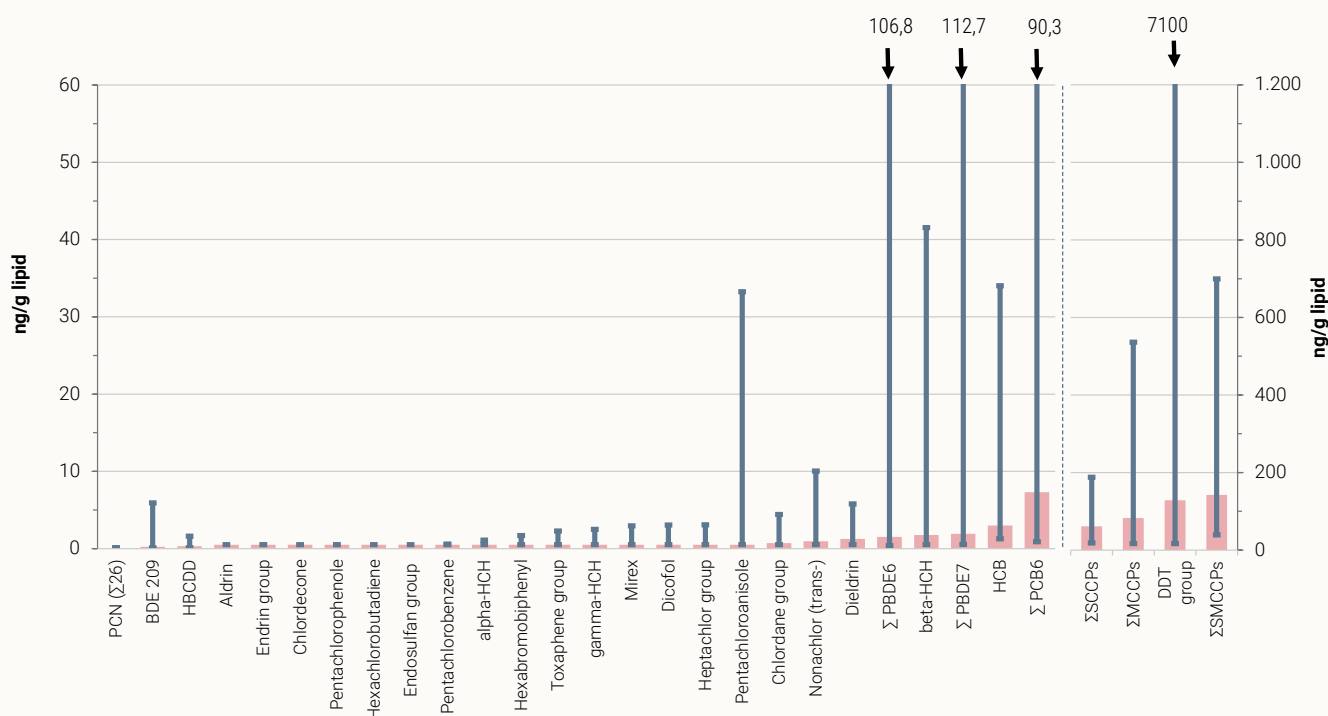
In contrast to findings of high concentrations (e.g. following the use of chemicals), after a sufficiently long withdrawal period for many POPs the levels are described as low background levels. However, the term “background level” does not in itself imply any level of safety. With

respect to potential adverse effects, risk assessments need to consider many factors, including the toxicity of the chemical of interest and the concentration range found. Performance of relevant research is encouraged, allowing WHO to provide guidance in regard to the discussion of a balance of potential adverse effects against positive health aspects for (breast fed) infants.



**Figure 46:** Range of concentrations of lipophilic chlorinated and brominated POPs in human milk (ng/g lipid) from 36 countries in the period 2016-2019 (median with error bars indicating the minimum and maximum)

## POPs in human milk (Africa, Asia-Pacific, GRULAC; 2016-2019)



**Figure 47:** Range of concentrations of lipophilic chlorinated and brominated POPs in human milk (ng/g lipid) from 36 countries in the period 2016-2019 (median with error bars indicating the minimum and maximum) (This figure is normalized to 100 ng/g lipid as maximum value, allowing a visual comparison also at lower concentration ranges)

The ranking of *SCCPs* and *MCCPs* among the broad spectrum of 28 recommended chlorinated and brominated analytes, as listed until 2019, is shown in *Figure 48*. The Convention POPs were sorted into two groups:

- Pesticides – aldrin, chlordane, chlordecone, DDT, dicofol; dieldrin, endosulfan, endrin, heptachlor,  $\alpha$ -HCH,  $\beta$ -HCH;  $\gamma$ -HCH; mirex, pentachlorobenzene, pentachlorophenol (including pentachloroanisole) and toxaphene;
- Others – Industrial chemicals and by-products – hexabromobiphenyl (HBB), HBCD, HCB, hexachlorobutadiene; PBDE; PCB, PCDD, PCDF and PCN.

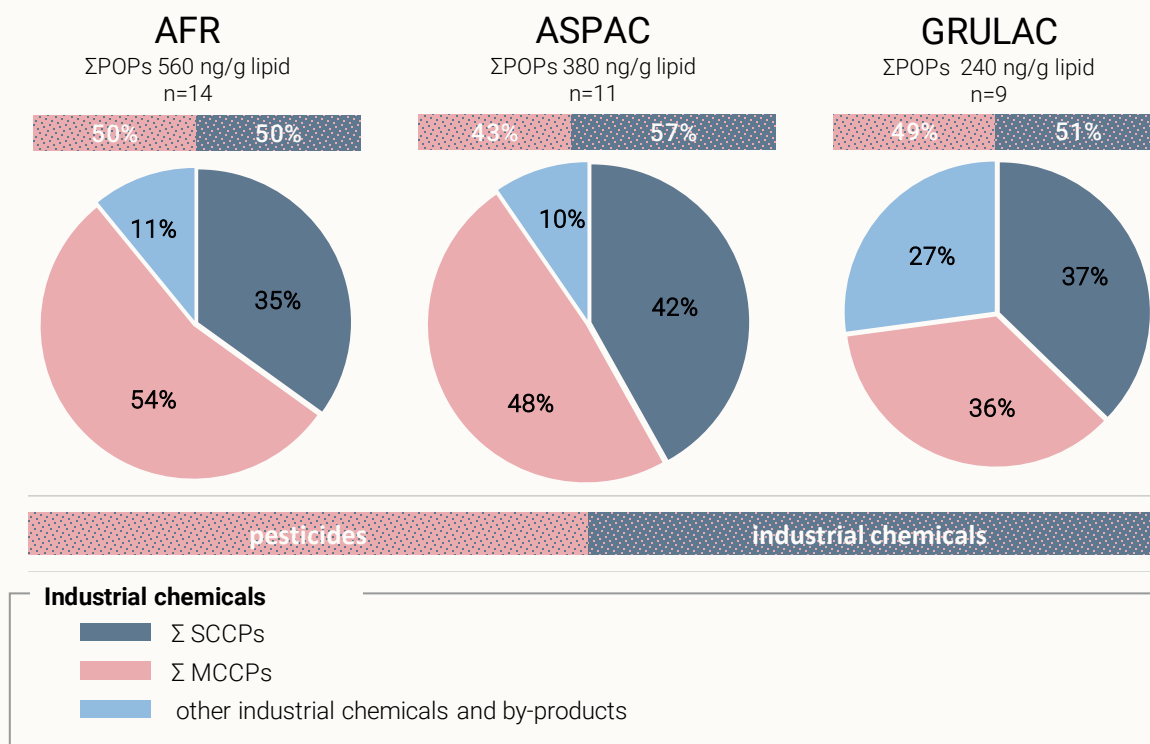
SCCPs and MCCPs (the latter proposed for listing under the Convention) dominated the share of POPs grouped as industrial chemicals and by-products in most areas.

Individual samples (from individual donors) can provide information on the distribution of exposures and on factors possibly contributing to exposure. Compared to pooled samples, they can span a broad range of concentrations.

If significantly elevated levels are found in pooled samples, a follow-up is usually recommended; if levels are quite low, no particular additional effort would seem to be necessary. This approach is much more efficient and cost-effective than analysing hundreds of individual samples to obtain an overview of POPs background levels in a certain country.

Given this design of the studies presented here, the dominance of SCCPs and MCCPs in comparison to concentrations of most other POPs in all UN regional groups are a cause for concern. If the sample pools with human milk from donors without any known major contamination sources nearby already show this consistently, with in some cases a high abundance of CPs, individual samples might be markedly higher (e.g. in the local population close to emission spots, or as result of exposure to consumer products or in the domestic environment).

The lactational intake of SCCPs and MCCPs by the breast-fed infant on the microgram scale, resulting from the mother's dietary and environmental exposure, should therefore call for follow-up studies and for further (or, in the case of MCCPs, any) regulatory efforts.



**Figure 48:** Median sum of all POPs analysed in pooled human milk samples from 2016-2019, sorted by UN regions and broken down into Stockholm Convention POPs groups (bar charts) and further into components of the "industrial chemicals" POPs group (pie charts). AFR: Africa, ASPAC: Asia-Pacific Group, GRULAC: Group of Latin America and the Caribbean.

In most countries during the period 2016-2019, concentrations of PFOS and PFOA (the two listed PFAS) were in a range between <6 and 30 ng/L. However, Σ PFOS concentrations in Kiribati exceeded this range by an order of magnitude (218 ng/L). In comparison to 2008-2011, decreasing tendencies were observed. Concentrations of PFHxS, a PFAS recommended for listing under the Convention, was below the limit of quantification (5.5 ng/L) in nearly all countries in 2016-2019. One country, Kiribati, had PFHxS concentrations more than 10 times higher (115 ng/L).

Taking into consideration the gender and age-differentiated windows of susceptibility and exposure to these harmful chemicals, it is recommended that future studies

should better understand the linkages between different social roles (including those related to gender, age and socioeconomic status) and POPs exposure, as well as ways to minimize exposure of vulnerable groups to POPs. For example, specific messaging or health campaigns targeting women of childbearing age on the importance of avoiding commonly known pollutants might help mitigate the issue to some extent.

In addition, strengthened collaboration between the environment and health sectors is imperative to facilitate comprehensive and efficient policy development. Through this, further downward trends in concentrations may be observed with time.

# SECTION 5

## Overall conclusions

The concept of WHO/UNEP-coordinated exposure studies with standardized protocols for preparation of pooled samples considered representative of a country or subgroup within a country, provides reliable data for human milk samples. The studies have taken place since 2000 and are both the largest and longest-running global study on human exposure to POPs.

This concept has allowed determination of all 30 listed POPs and two additional chemicals recommended for listing by 2019. The consideration of countries with repeated participation provides the best possible data base for the assessment of temporal trends. It is highly recommended to continue with this monitoring effort in order to secure enough data for a proper assessment of time trends and health effects in the future.

Furthermore, while monitoring is crucial for risk prevention, given the health impacts of many POPs, strengthening the collaboration between the environment and health sectors is also necessary to support cross-cutting and effective policy making.



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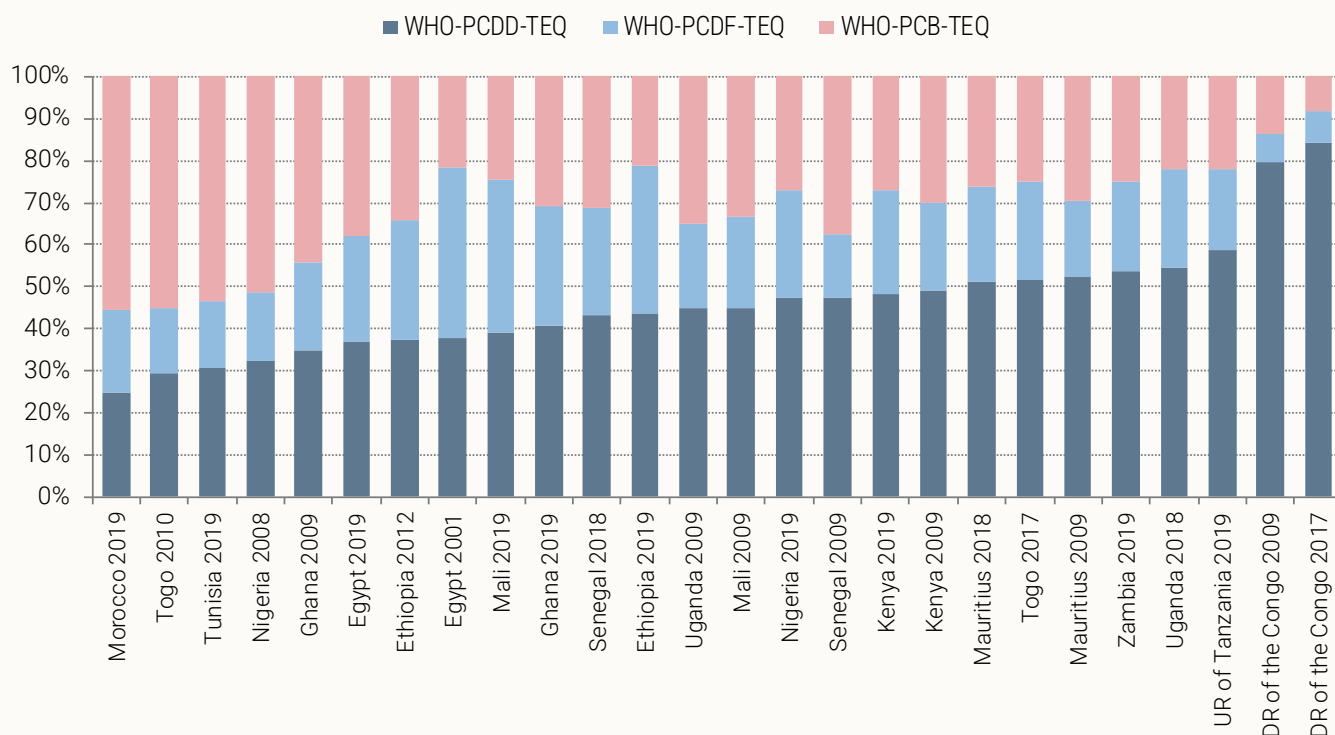
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# APPENDIX

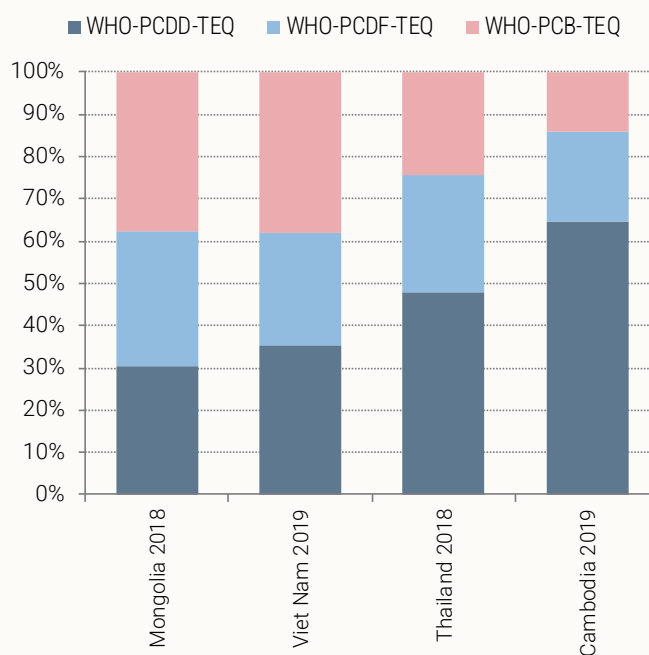
## Supplementary Figures

### WHO-TEQ (2005) Contribution - African group



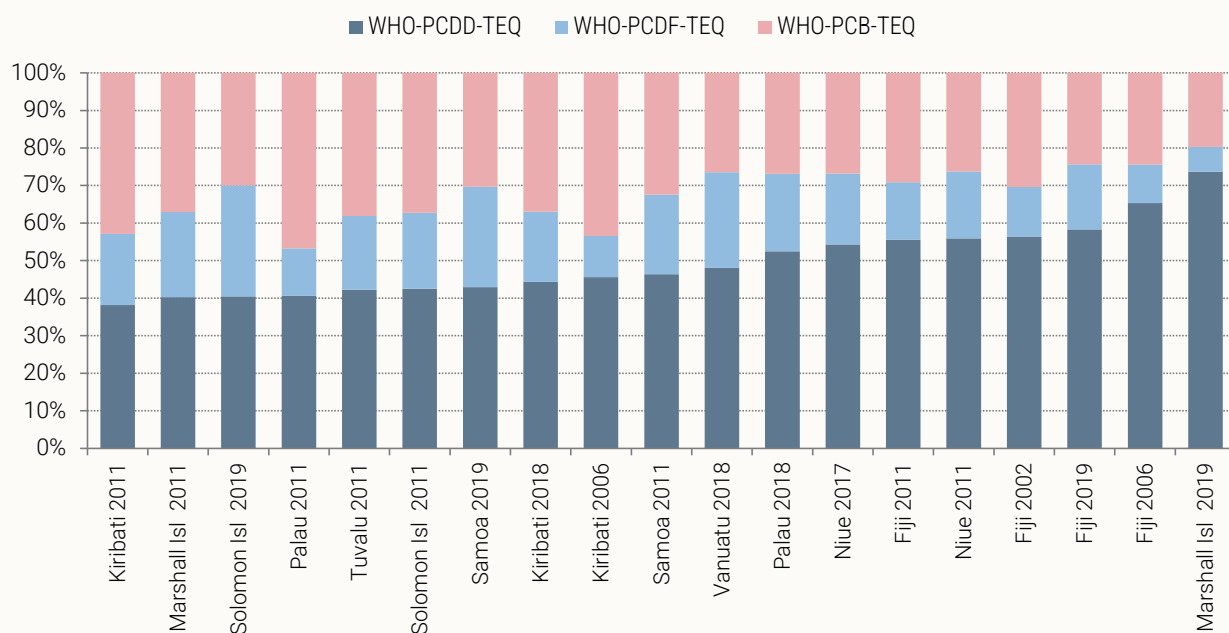
**Figure S1:** Relative contribution (%) of toxic equivalents of PCDD (WHO-PCDD-TEQ [2005]), PCDF (WHO-PCDF-TEQ [2005]) and dioxin-like PCB (WHO-PCB-TEQ [2005]) to total TEQ (WHO-PCDD/PCDF-PCB-TEQ [2005]) in human milk samples from countries of the African Group and year of submission

### WHO-TEQ (2005) Contribution - Asia



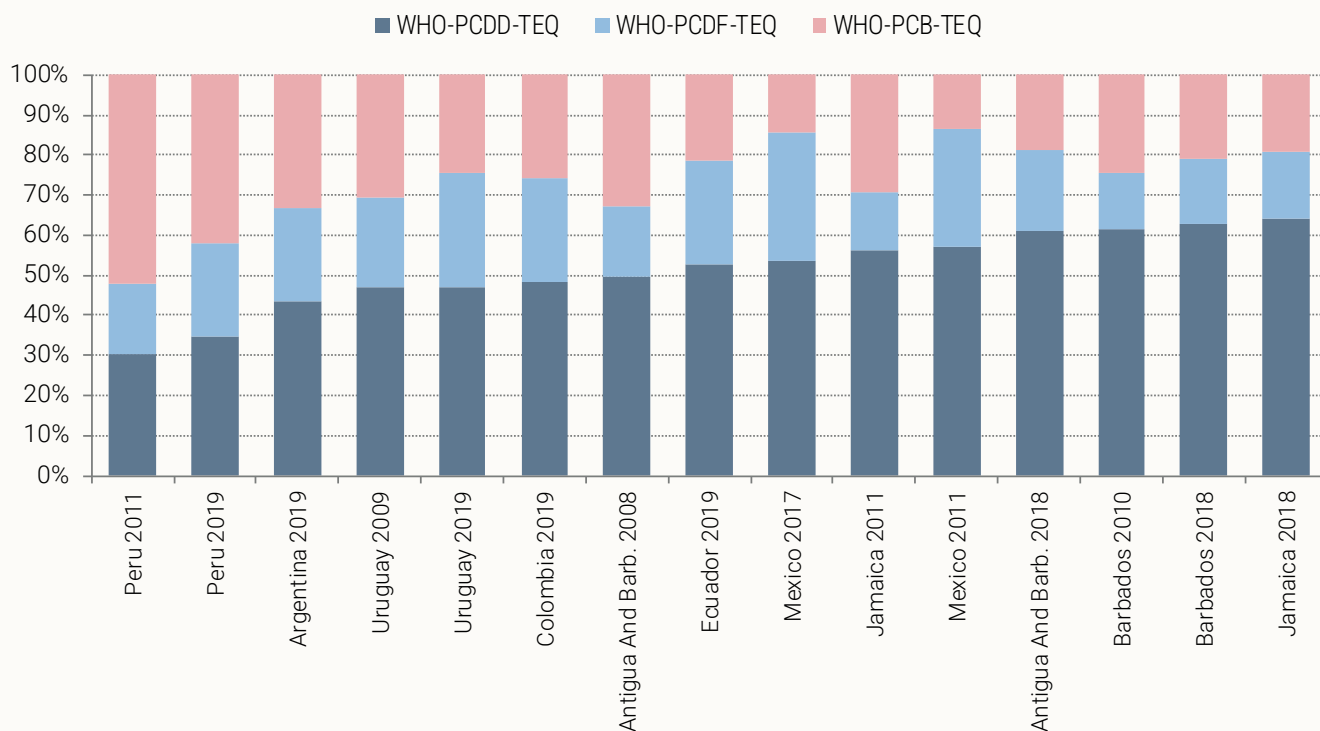
**Figure S2:** Relative contribution (%) of toxic equivalents of PCDD (WHO-PCDD-TEQ [2005]), PCDF (WHO-PCDF-TEQ [2005]) and dioxin-like PCB (WHO-PCB-TEQ [2005]) to total TEQ (WHO-PCDD/PCDF-PCB-TEQ [2005]) in human milk samples from Asian countries and year of submission

### WHO-TEQ (2005) Contribution - Pacific Islands



**Figure S3:** Relative contribution (%) of toxic equivalents of PCDD (WHO-PCDD-TEQ [2005]), PCDF (WHO-PCDF-TEQ [2005]) and dioxin-like PCB (WHO-PCB-TEQ [2005]) to total TEQ (WHO-PCDD/PCDF-PCB-TEQ [2005]) in human milk samples from Pacific Islands countries and year of submission

### WHO-TEQ (2005) Contribution - Latin America and Caribbean



**Figure S4:** Relative contribution (%) of toxic equivalents of PCDD (WHO-PCDD-TEQ [2005]), PCDF (WHO-PCDF-TEQ [2005]) and dioxin-like PCB (WHO-PCB-TEQ [2005]) to total TEQ (WHO-PCDD/PCDF-PCB-TEQ [2005]) in human milk samples from GRULAC countries and year of submission

## Supplementary Tables

**Table S1:** Implementation of the project “UNEP/GEF GMP2” – support of participating countries with glassware for collection of samples and receipt of the pooled samples at CVUA Freiburg

Total	Samples Received	No	Country	Shipment Of Glassware	Shipment Of Samples	Amount Of Milk Sample
<b>1. Africa</b>						
1	1	1	DR of the Congo	12/2016	11/2017	1150 ml
2	2	2	Egypt	12/2016	10/2019	250 ml
3	3	3	Ethiopia	12/2016	07/2019	1400 ml
4	4	4	Ghana	12/2016	08/2019	2000 ml
5	5	5	Kenya	12/2016	08/2019	1200 ml
6	6	6	Mali	12/2016	06/2019	1200 ml
7	7	7	Mauritius	12/2016	06/2018	1200 ml
8	8	8	Morocco	12/2016	06/2019	1250 ml
9	9	9	Nigeria	12/2016	03/2019	1500 ml
10	10	10	Senegal	12/2016	03/2018	1200 ml
11	11	11	UR of Tanzania	12/2016	07/2019	1350 ml
12	12	12	Togo	12/2016	11/2017	1250 ml
13	13	13	Tunisia	12/2016	07/2019	1300 ml
14	14	14	Uganda	01/2017	06/2018	1200 ml
15	15	15	Zambia	01/2017	07/2019	1200 ml
<b>2. Asia</b>						
16	16	1	Cambodia	01/2017	06/2019	1700 ml
17			Indonesia	02/2017	-	
18			Lao Pdr	01/2017	-	
19	17	2	Mongolia	01/2017	08/2018	1100 ml
20		5	Philippines	02/2017	-	
21	18	3	Thailand	12/2017	09/2018	1500 ml
22	19	4	Viet Nam	01/2017	03/2019	1500 ml
<b>3. the Pacific Islands</b>						
23	20	1	Fiji	11/2016	03/2019	160 ml
24	21	2	Kiribati	11/2016	06/2018	700 ml
25	22	3	Marshall Islands	11/2016	10/2019	700 ml
26	23	4	Niue	11/2016	12/2017	270 ml
27	24	5	Palau	11/2016	10/2018	1250 ml
28	25	6	Samoa	11/2016	09/2019	600 ml
29	26	7	Solomon Islands	11/2016	02/2019	1200 ml
30			Tuvalu	11/2016	-	
31	27	8	Vanuatu	02/2017	03/2018	1500 ml
<b>4. Grulac</b>						
32	28	1	Antigua and Barbuda	01/2017	04/2018	600 ml
33	29	2	Argentina	01/2017	01/2019	1300 ml
34	30	3	Barbados	01/2017	02/2018	1000 ml
35			Brazil	02/2017	-	
36			Chile	01/2017	-	
37	31	4	Colombia	02/2017	01/2019	1200 ml
38	32	5	Ecuador	01/2017	02/2019	1100 ml
39	33	6	Jamaica	01/2017	04/2018	400 ml
40	34	7	Mexico	01/2017	11/2017	1250 ml
41	35	8	Peru	01/2017	04/2019	1500 ml
42	36	9	Uruguay	01/2017	03/2019	1400 ml

**Table S2:** Complexes of DDT, chlordane, heptachlor, endrin and endosulfan and correction factors for molecular weight used for the calculation of the sum parameters

	Correction factors
<b>DDT</b>	
op <sup>1</sup> -DDT	1
pp <sup>1</sup> -DDT	1
op <sup>1</sup> -DDD	1.108
pp <sup>1</sup> -DDD	1.108
op <sup>1</sup> -DDE	1.115
pp <sup>1</sup> -DDE	1.115
<b>DDT complex *)</b>	
*) sum of all detected analytes, calculated as DDT	
<b>Chlordane</b>	
cis-chlordane (alpha-chlordane)	1
trans-chlordane (gamma-chlordane)	1
Oxychlordane	0.967
cis-nonachlor	0.923
trans-nonachlor	0.923
<b>Chlordane complex (cis+trans+oxy *):</b>	
*) residue definition according to GMP Guidance, 2007, and in food legislation: sum of cis- and trans-chlordane and oxychlordane, calculated as chlordane (without nonachlor)	
<b>Chlordane group (all 5 analytes **):</b>	
**) according GMP Guidance, 2019: sum of all 5 recommended analytes (including nonachlor)	
<b>Heptachlor</b>	
Heptachlor	1
cis-heptachlor epoxide	0.959
trans-heptachlor epoxide	0.959
<b>Heptachlor complex *)</b>	
*) sum of all detected analytes, calculated as heptachlor	
<b>Endrin</b>	
Endrin	1
Endrin ketone	1
<b>Endrin complex *)</b>	
*) sum of all detected analytes, calculated as endrin	
<b>Endosulfan</b>	
alpha-endosulfan	1
beta-endosulfan	1
endosulfan sulfate	0.962
<b>Endosulfan complex *)</b>	
*) sum of all detected analytes, calculated as endosulfan	

**Table S3:** Concentrations of PCDD, PCDF and PCB in human milk samples of the 2016-2019 period from Africa

Country →	Units ↓	DR of the Congo	Egypt	Ethiopia	Ghana	Kenya	Mali	Mauritius	Morocco	Nigeria	Senegal	UR of Tanzania	Togo	Tunisia	Uganda	Zambia
		2017	2019	2019	2019	2019	2019	2018	2019	2019	2018	2019	2017	2019	2018	2019
WHO-PCDD/F-TEQ (2005 / LB)	pg/ g lipid	9,97	5,59	1,02	2,68	1,52	2,90	1,54	2,95	2,51	5,74	1,68	3,31	2,24	1,24	1,37
WHO-PCDD/F-TEQ (2005 / UB)	pg/ g lipid	9,97	5,59	1,02	2,68	1,52	2,90	1,54	2,96	2,52	5,74	1,68	3,32	2,24	1,24	1,37
WHO-PCB-TEQ (2005 / LB)	pg/ g lipid	0,89	3,45	0,27	1,20	0,57	0,95	0,55	3,70	0,93	2,62	0,47	1,10	2,57	0,35	0,46
WHO-PCB-TEQ (2005 / UB)	pg/ g lipid	0,89	3,45	0,27	1,20	0,57	0,95	0,55	3,70	0,93	2,62	0,47	1,10	2,57	0,35	0,46
WHO-PCDD/F-PCB-TEQ (2005 / LB)	pg/ g lipid	10,86	9,03	1,29	3,88	2,09	3,85	2,10	6,65	3,44	8,36	2,15	4,41	4,82	1,59	1,83
WHO-PCDD/F-PCB-TEQ (2005 / UB)	pg/ g lipid	10,86	9,04	1,29	3,88	2,09	3,85	2,10	6,66	3,45	8,36	2,15	4,42	4,82	1,59	1,83
<b>PCDD/PCDF</b>																
2,3,7,8-TCDD	pg/ g lipid	0,71	1,04	0,18	0,33	0,21	0,29	0,17	0,37	0,22	0,53	0,14	0,25	0,28	0,16	0,10
1,2,3,7,8-PeCDD	pg/ g lipid	1,44	2,05	0,30	0,69	0,55	0,63	0,51	0,99	0,82	1,48	0,58	0,87	0,85	0,48	0,35
1,2,3,4,7,8-HxCDD	pg/ g lipid	0,95	0,55	0,12	0,24	0,27	0,28	0,28	0,36	1,14	0,86	0,33	0,50	0,46	0,20	0,26
1,2,3,6,7,8-HxCDD	pg/ g lipid	5,96	1,37	0,39	1,91	1,14	2,43	1,91	1,54	2,15	7,29	2,07	3,76	2,10	1,13	1,55
1,2,3,7,8,9-HxCDD	pg/ g lipid	56,01	0,28	0,29	1,33	0,62	1,21	0,76	0,46	1,63	3,14	1,37	2,91	0,53	0,55	1,07
1,2,3,4,6,7,8-HpCDD	pg/ g lipid	61,97	1,23	1,28	18,37	4,33	17,63	8,75	3,62	8,39	44,07	14,60	41,10	3,56	3,63	20,77
OCDD	pg/ g lipid	323,36	2,91	9,20	73,37	30,96	65,96	42,73	28,38	50,60	153,22	59,41	115,63	18,82	21,81	128,35
2,3,7,8-TCDF	pg/ g lipid	0,72	1,32	0,32	1,50	0,39	2,61	0,22	0,42	0,43	3,78	0,28	0,94	0,27	0,26	0,19
1,2,3,7,8-PeCDF	pg/ g lipid	0,39	0,96	0,19	0,46	0,16	0,54	0,15	0,32	0,26	0,76	0,16	0,45	0,17	0,19	0,15
2,3,4,7,8-PeCDF	pg/ g lipid	1,69	5,34	1,00	2,47	1,11	2,91	1,07	3,23	2,08	4,21	0,87	2,31	1,81	0,80	0,80
1,2,3,4,7,8-HxCDF	pg/ g lipid	0,84	2,56	0,43	0,74	0,51	0,99	0,45	1,08	0,83	1,74	0,40	0,93	0,73	0,36	0,44
1,2,3,6,7,8-HxCDF	pg/ g lipid	0,77	1,52	0,40	0,70	0,49	0,89	0,40	1,13	0,76	1,72	0,50	0,89	0,65	0,32	0,45
1,2,3,7,8,9-HxCDF	pg/ g lipid	0,08	<0,047	0,10	0,04	0,06	0,06	0,05	<0,027	<0,021	0,06	0,08	<0,049	0,02	0,06	0,03
2,3,4,6,7,8-HxCDF	pg/ g lipid	0,37	0,84	0,21	0,35	0,24	0,33	0,21	0,58	0,43	0,76	0,21	0,34	0,36	0,17	0,20
1,2,3,4,6,7,8-HpCDF	pg/ g lipid	1,07	0,39	0,31	1,14	0,56	0,91	0,59	0,84	0,67	2,39	0,81	1,39	0,52	0,29	0,83
1,2,3,4,7,8,9-HpCDF	pg/ g lipid	0,10	0,06	0,03	0,14	0,07	0,08	0,04	<0,010	0,04	0,18	<0,009	0,09	0,03	0,04	0,12
OCDF	pg/ g lipid	0,16	<0,00803	0,04	0,79	0,14	0,15	0,22	0,10	0,05	0,32	0,20	0,46	0,10	0,09	0,49
<b>Dioxin-like PCB</b>																
<b>Non-ortho PCB</b>																
PCB 77	pg/g lipid	3,70	6,50	1,81	2,48	2,47	2,46	5,29	5,05	3,00	3,91	2,07	3,38	1,95	3,90	2,26
PCB 81	pg/g lipid	1,11	4,25	0,65	1,24	1,00	0,86	< 0,49	1,36	1,17	< 0,62	0,81	< 0,52	0,89	0,78	0,94
PCB 126	pg/g lipid	7,15	30,36	2,22	9,75	4,84	7,80	3,86	31,26	7,16	19,66	3,76	8,67	21,39	2,85	3,75
PCB 169	pg/g lipid	3,13	11,96	1,38	4,89	1,98	3,72	4,12	11,87	4,43	12,91	2,30	4,82	9,06	1,44	1,31
<b>Mono-ortho PCB</b>																
PCB 105	pg/g lipid	382,7	337,6	43,5	364,2	152,3	256,3	205,8	793,6	420,2	686,5	146,7	328,1	465,6	130,4	330,1
PCB 114	pg/g lipid	67,7	36,7	< 1,21	57,9	12,7	29,8	30,8	159,6	45,8	104,3	< 2,37	51,4	64,7	20,2	43,5
PCB 118	pg/g lipid	1230,7	761,7	157,0	1330,9	371,1	957,4	732,0	3480,8	1385,7	3308,0	494,9	1300,2	2309,2	401,6	819,8
PCB 123	pg/g lipid	<11,73	9,5	< 1,22	< 5,28	3,7	8,5	< 4,74	25,0	11,1	19,4	< 2,11	< 10,22	< 8,83	< 1,65	11,1
PCB 156	pg/g lipid	578,8	294,5	19,3	552,5	113,9	485,5	314,0	1714,9	514,0	2976,2	128,5	775,9	1575,0	114,2	215,3
PCB 157	pg/g lipid	82,2	70,6	< 2,34	88,0	28,7	66,8	55,2	174,7	111,9	370,3	26,2	133,1	212,8	26,7	46,2
PCB 167	pg/g lipid	149,7	95,7	8,4	181,1	36,2	152,5	86,2	628,9	159,2	940,0	< 4,50	256,8	586,4	30,3	39,8
PCB 189	pg/g lipid	69,3	53,6	5,4	55,5	7,7	64,1	30,3	259,9	58,1	394,4	< 2,29	108,9	192,2	8,3	18,4
<b>Indicator-PCB</b>																
PCB 28	ng/g lipid	0,3816	0,1833	0,2225	0,2711	0,5807	0,2510	0,3615	0,3942	0,2918	0,4160	0,3319	0,2600	0,3895	0,2576	1,5473
PCB 52	ng/g lipid	0,1357	0,1267	0,0642	0,0413	0,0655	0,1120	0,1060	0,1608	0,1236	0,1414	0,0527	0,1188	0,1236	0,0946	0,0534
PCB 101	ng/g lipid	0,2441	0,0857	0,0363	0,1545	0,0558	0,1761	0,1271	0,3993	0,2346	0,2982	0,0546	0,2207	0,1803	0,1052	0,0814
PCB 138	ng/g lipid	3,2757	0,7394	0,1902	3,6249	0,6146	3,3709	1,4392	13,2337	3,5343	22,0858	0,9531	5,4024	12,2814	0,6295	1,0601
PCB 153	ng/g lipid	5,7074	1,3995	0,2521	6,3213	0,7243	5,9305	2,3988	26,7014	6,8011	40,4226	1,3006	9,5680	23,2685	0,8455	1,5311
PCB 180	ng/g lipid	4,2230	1,1760	0,1313	3,3313	0,4336	4,3050	1,4367	19,8030	3,5222	26,9201	0,7004	6,7353	16,7316	0,5306	0,8042
<b>Sum 6 Indicator PCB</b>	ng/g lipid	13,9674	3,7106	0,8966	13,7444	2,4745	14,1455	5,8692	60,6924	14,5076	90,2840	3,3933	22,3052	52,9748	2,4628	5,0775

**Table S4:** Concentrations of PCDD, PCDF and PCB in human milk samples of the 2016-2019 period from Asia and the the Pacific Islands

Country →		Cambodia	Mongolia	Thailand	Viet Nam	Fiji	Kiribati	Marshall Islands	Niue	Palau	Samoa	Solomon Islands	Vanuatu
Year ↓	Units ↓	2019	2018	2018	2019	2019	2018	2019	2017	2018	2019	2019	2018
WHO-PCDD/F-TEQ (2005 / LB)	pg/ g lipid	3,92	2,24	1,80	2,62	2,19	1,92	9,32	1,29	2,63	2,30	1,73	1,43
WHO-PCDD/F-TEQ (2005 / UB)	pg/ g lipid	3,92	2,24	1,80	2,62	2,20	1,92	9,32	1,29	2,63	2,30	1,73	1,43
WHO-PCB-TEQ (2005 / LB)	pg/ g lipid	0,66	1,35	0,58	1,62	0,71	1,13	2,29	0,47	0,97	1,00	0,74	0,52
WHO-PCB-TEQ (2005 / UB)	pg/ g lipid	0,66	1,35	0,58	1,62	0,71	1,13	2,29	0,47	0,97	1,00	0,74	0,52
WHO-PCDD/F-PCB-TEQ (2005 / LB)	pg/ g lipid	4,58	3,58	2,38	4,24	2,90	3,05	11,61	1,76	3,59	3,30	2,47	1,95
WHO-PCDD/F-PCB-TEQ (2005 / UB)	pg/ g lipid	4,58	3,58	2,38	4,24	2,90	3,05	11,61	1,76	3,60	3,30	2,48	1,95
<b>PCDD/PCDF</b>													
2,3,7,8-TCDD	pg/ g lipid	0,42	0,36	0,22	0,39	0,31	0,31	1,18	0,19	0,25	0,40	0,36	0,29
1,2,3,7,8-PeCDD	pg/ g lipid	1,21	0,55	0,71	0,80	0,89	0,69	4,98	0,43	1,03	0,73	0,47	0,47
1,2,3,4,7,8-HxCDD	pg/ g lipid	0,79	0,36	0,30	0,67	0,44	0,41	2,81	0,23	0,65	0,36	0,19	0,20
1,2,3,6,7,8-HxCDD	pg/ g lipid	5,72	0,89	1,00	1,43	1,92	1,57	12,53	1,49	3,45	1,32	0,80	0,85
1,2,3,7,8,9-HxCDD	pg/ g lipid	1,50	0,32	0,38	0,61	1,68	0,85	5,24	1,01	1,17	0,58	0,39	0,39
1,2,3,4,6,7,8-HpCDD	pg/ g lipid	43,98	1,29	3,00	2,75	8,30	6,15	28,38	5,39	6,37	3,92	2,52	2,67
OCDD	pg/ g lipid	250,25	12,29	22,94	28,46	36,60	39,35	174,02	26,97	45,18	45,44	47,21	23,64
2,3,7,8-TCDF	pg/ g lipid	0,32	0,26	0,31	0,54	0,40	0,58	0,27	0,13	0,20	0,65	0,59	0,55
1,2,3,7,8-PeCDF	pg/ g lipid	0,32	0,16	0,25	0,47	0,20	0,24	0,17	0,11	0,18	0,32	0,30	0,23
2,3,4,7,8-PeCDF	pg/ g lipid	1,89	2,66	1,55	2,54	1,11	1,29	1,51	0,73	1,64	2,05	1,71	1,13
1,2,3,4,7,8-HxCDF	pg/ g lipid	1,39	1,45	0,55	1,21	0,37	0,43	1,04	0,37	0,89	0,74	0,61	0,35
1,2,3,6,7,8-HxCDF	pg/ g lipid	1,09	1,18	0,59	1,07	0,44	0,45	1,10	0,34	0,79	0,79	0,55	0,37
1,2,3,7,8,9-HxCDF	pg/ g lipid	0,14	0,07	0,12	0,07	<0,018	<0,02	0,04	0,02	<0,035	0,03	<0,019	<0,014
2,3,4,6,7,8-HpCDF	pg/ g lipid	0,44	0,49	0,26	0,50	0,35	0,21	0,35	0,18	0,40	0,33	0,26	0,19
1,2,3,4,6,7,8-HpCDF	pg/ g lipid	5,19	0,62	0,65	0,78	0,70	0,54	1,93	0,64	1,36	0,61	0,69	0,44
1,2,3,4,7,8,9-HpCDF	pg/ g lipid	0,30	0,06	0,05	0,07	0,04	0,04	0,13	0,05	0,05	0,03	0,05	0,04
OCDF	pg/ g lipid	0,43	0,10	0,08	<0,005	0,12	0,12	0,20	0,08	0,21	0,05	0,09	0,08
<b>Dioxin-like PCB</b>													
<b>Non-ortho PCB</b>													
PCB 77	pg/g lipid	3,28	5,06	< 0,16	3,24	9,99	7,75	4,70	3,99	3,54	3,98	5,51	3,79
PCB 81	pg/g lipid	0,75	1,17	< 0,16	1,66	< 0,37	1,42	1,25	< 0,12	< 0,69	1,10	1,36	< 0,35
PCB 126	pg/g lipid	4,88	9,71	3,96	11,08	5,81	8,77	18,00	3,47	7,26	7,88	4,61	3,79
PCB 169	pg/g lipid	4,92	6,64	5,28	6,96	2,60	6,42	8,36	3,00	3,37	5,50	8,52	3,46
<b>Mono-ortho PCB</b>													
PCB 105	pg/g lipid	109,0	709,6	115,3	2085,5	252,2	329,4	1329,1	133,0	935,2	248,5	122,9	179,6
PCB 114	pg/g lipid	29,6	297,9	< 2,17	248,5	33,6	< 19,34	198,0	42,5	103,6	32,8	9,9	25,4
PCB 118	pg/g lipid	340,9	2919,8	420,1	5990,8	891,3	1080,3	4600,4	514,4	2739,4	757,8	427,9	594,4
PCB 123	pg/g lipid	< 1,77	21,4	< 1,65	75,2	12,0	20,8	39,7	13,0	< 21,40	< 2,42	< 1,69	< 2,56
PCB 156	pg/g lipid	145,2	1312,5	138,3	1052,8	205,6	271,1	1149,8	201,8	531,5	309,9	130,9	182,0
PCB 157	pg/g lipid	31,1	320,1	31,2	236,6	47,7	67,0	242,5	46,3	129,7	64,9	16,0	31,9
PCB 167	pg/g lipid	36,1	214,7	28,2	263,3	72,6	115,0	370,1	81,1	160,3	84,9	50,6	52,8
PCB 189	pg/g lipid	18,7	45,4	12,5	45,5	23,6	22,2	95,7	31,0	< 10,56	23,1	18,0	< 8,00
<b>Indicator-PCB</b>													
PCB 28	ng/g lipid	0,1430	0,5524	0,2768	0,5045	0,2458	0,5516	0,30	0,27	0,33	0,31	0,21	0,23
PCB 52	ng/g lipid	0,2506	0,1925	0,0616	0,0993	0,1413	0,1659	0,21	0,16	0,23	0,19	0,06	0,12
PCB 101	ng/g lipid	0,1190	0,1800	0,0637	0,1411	0,1863	0,2116	0,70	0,13	0,36	0,14	0,07	0,17
PCB 138	ng/g lipid	0,8558	4,6464	0,8088	5,9306	1,1524	1,5912	5,54	1,20	2,52	1,37	0,51	0,78
PCB 153	ng/g lipid	1,4120	7,5519	1,4190	6,2354	2,0049	2,4130	9,71	1,79	3,53	2,26	1,05	1,43
PCB 180	ng/g lipid	0,8993	2,4812	0,6246	1,8508	0,7187	1,0710	6,91	0,98	1,64	1,42	0,64	0,89
<b>Sum 6 Indicator PCB</b>	ng/g lipid	3,6797	15,6042	3,2544	14,7617	4,4494	6,0044	23,37	4,52	8,61	5,68	2,55	3,61

**Table S5:** Concentrations of PCDD, PCDF and PCB in human milk samples of the 2016-2019 period from the Group of Latin American and Caribbean Countries

Country →		Antigua and Barbuda	Argentina	Barbados	Colombia	Ecuador	Jamaica	Mexico	Peru	Uruguay
Year ↓	Units ↓	2018	2019	2018	2019	2019	2018	2017	2019	2019
WHO-PCDD/F-TEQ (2005 / LB)	pg/ g lipid	2,37	3,13	3,08	1,89	1,99	4,18	3,48	2,17	4,29
WHO-PCDD/F-TEQ (2005 / UB)	pg/ g lipid	2,37	3,13	3,09	1,89	1,99	4,18	3,48	2,17	4,29
WHO-PCB-TEQ (2005 / LB)	pg/ g lipid	0,55	1,56	0,83	0,66	0,55	1,00	0,58	1,58	1,40
WHO-PCB-TEQ (2005 / UB)	pg/ g lipid	0,55	1,56	0,83	0,66	0,55	1,00	0,58	1,58	1,40
WHO-PCDD/F-PCB-TEQ (2005 / LB)	pg/ g lipid	2,92	4,68	3,91	2,55	2,54	5,18	4,06	3,74	5,69
WHO-PCDD/F-PCB-TEQ (2005 / UB)	pg/ g lipid	2,92	4,68	3,91	2,55	2,54	5,18	4,06	3,74	5,69
<b>PCDD/PCDF</b>										
2,3,7,8-TCDD	pg/ g lipid	0,29	0,29	0,36	0,25	0,23	0,43	0,40	0,28	0,61
1,2,3,7,8-PeCDD	pg/ g lipid	1,11	1,10	1,42	0,77	0,82	1,75	1,19	0,77	1,54
1,2,3,4,7,8-HxCDD	pg/ g lipid	0,43	0,36	0,84	0,36	0,52	1,22	0,71	0,30	0,53
1,2,3,6,7,8-HxCDD	pg/ g lipid	2,25	5,07	3,24	1,17	1,37	6,04	3,32	1,46	3,45
1,2,3,7,8,9-HxCDD	pg/ g lipid	0,68	0,66	1,62	0,46	0,51	2,15	1,02	0,41	0,81
1,2,3,4,6,7,8-HpCDD	pg/ g lipid	4,04	2,79	9,15	2,19	3,99	17,46	6,30	3,34	3,08
OCDD	pg/ g lipid	24,45	6,32	39,79	11,46	25,01	102,83	23,71	15,91	10,74
2,3,7,8-TCDF	pg/ g lipid	0,34	0,36	0,18	0,29	0,25	0,42	0,58	0,54	0,71
1,2,3,7,8-PeCDF	pg/ g lipid	0,18	0,29	0,15	0,14	0,18	0,23	0,50	0,28	0,42
2,3,4,7,8-PeCDF	pg/ g lipid	1,32	2,45	1,29	1,42	1,46	1,67	2,67	1,98	3,82
1,2,3,4,7,8-HxCDF	pg/ g lipid	0,58	1,18	0,75	0,71	0,69	1,21	1,80	0,92	1,40
1,2,3,6,7,8-HxCDF	pg/ g lipid	0,54	1,13	0,88	0,70	0,71	1,04	1,60	0,79	1,40
1,2,3,7,8,9-HxCDF	pg/ g lipid	<0,016	0,06	<0,02	0,06	<0,017	<0,03	0,08	0,04	0,13
2,3,4,6,7,8-HxCDF	pg/ g lipid	0,26	0,63	0,36	0,37	0,32	0,56	0,75	0,34	0,85
1,2,3,4,6,7,8-HpCDF	pg/ g lipid	0,53	1,52	1,82	0,77	1,10	2,61	1,44	0,49	0,96
1,2,3,4,7,8,9-HpCDF	pg/ g lipid	<0,01	0,09	<0,04	0,03	0,06	0,11	0,12	0,03	0,10
OCDF	pg/ g lipid	0,10	0,08	<0,04	0,08	0,96	0,19	0,12	0,12	0,08
<b>Dioxin-like PCB</b>										
<b>Non-ortho PCB</b>										
PCB 77	pg/g lipid	3,97	2,88	4,75	2,60	2,57	7,43	3,47	3,84	2,76
PCB 81	pg/g lipid	0,64	1,42	< 1.25	0,88	1,24	< 0.75	< 0.23	0,95	1,24
PCB 126	pg/g lipid	4,09	12,43	6,12	5,35	4,12	7,09	4,67	13,03	10,92
PCB 169	pg/g lipid	2,45	4,22	4,07	2,99	3,64	3,59	2,43	4,33	7,09
<b>Mono-ortho PCB</b>										
PCB 105	pg/g lipid	331,5	1060,5	409,4	175,4	139,2	928,1	180,5	942,2	407,1
PCB 114	pg/g lipid	< 20.74	151,9	< 53.36	32,4	26,0	157,8	53,7	147,1	101,4
PCB 118	pg/g lipid	1225,0	3515,4	1616,3	585,0	461,7	3295,9	661,4	2525,6	1569,5
PCB 123	pg/g lipid	< 4.33	48,9	< 13.37	7,4	8,4	31,4	< 7.57	40,5	18,2
PCB 156	pg/g lipid	446,5	917,9	612,8	232,1	128,7	989,9	238,1	665,6	660,5
PCB 157	pg/g lipid	100,2	185,4	127,7	42,6	37,2	240,1	47,6	129,1	157,3
PCB 167	pg/g lipid	152,7	300,4	228,8	72,9	50,7	301,4	61,2	217,1	179,3
PCB 189	pg/g lipid	39,0	50,9	77,0	26,9	11,6	58,2	< 15.23	54,8	44,0
<b>Indicator-PCB</b>										
PCB 28	ng/g lipid	0,28	1,20	0,49	0,42	0,18	0,60	0,62	0,30	0,49
PCB 52	ng/g lipid	0,12	0,16	0,19	0,07	0,04	0,24	0,11	0,08	0,07
PCB 101	ng/g lipid	0,13	0,29	0,18	0,08	<0,01	0,40	0,08	0,12	0,10
PCB 138	ng/g lipid	2,98	5,01	5,09	1,35	0,75	5,28	1,07	3,55	3,05
PCB 153	ng/g lipid	5,06	6,77	8,75	2,31	1,26	7,11	1,58	5,34	4,64
PCB 180	ng/g lipid	2,50	3,25	4,56	1,69	0,77	2,77	1,00	3,29	2,52
<b>Sum 6 Indicator PCB</b>	ng/g lipid	11,08	16,68	19,26	5,91	3,01	16,40	4,46	12,68	10,87

**Table S6:** Concentrations of organochlorine pesticides and industrial contaminants in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, the Pacific Islands and GRULAC countries

Region	Country	o,p'-DDD	p,p'-DDD	o,p'-DDE	p,p'-DDE	o,p'-DDT	p,p'-DDT	DDT group	alpha HCH	beta HCH	gamma HCH	HCB
Africa	DR of the Congo	nd	nd	nd	160	2,2	14,8	195	0,5	0,9	nd	1,5
Africa	Egypt	nd	1	nd	12	nd	1,9	17	0,6	19,7	0,7	2,3
Africa	Ethiopia	1,2	7	2,4	5000	150,0	1400,0	7100	nd	nd	nd	1,7
Africa	Ghana	nd	nd	nd	66	nd	2,9	77	nd	1,5	nd	3,5
Africa	Kenya	nd	nd	nd	70	0,7	5,0	84	nd	1,6	nd	1,7
Africa	Mali	nd	1	2,9	458	7,0	24,3	543	nd	4,3	nd	2,6
Africa	Mauritius	nd	1	0,6	467	4,9	32,8	559	nd	5,3	nd	2,7
Africa	Morocco	nd	1	nd	197	2,6	13,6	238	nd	3	nd	3,4
Africa	Nigeria	nd	nd	nd	86	nd	3,0	99	nd	2,2	nd	3,7
Africa	Senegal	nd	1	nd	156	1,6	14,5	191	nd	11,7	1,8	3,4
Africa	UR of Tanzania	nd	nd	nd	331	0,6	9,3	379	nd	2,0	nd	1,3
Africa	Togo	nd	1	0,6	543	3,3	20,1	630	1,1	0,9	nd	1,9
Africa	Tunisia	nd	nd	nd	84,4	nd	2,2	96	nd	8,1	nd	3,2
Africa	Uganda	nd	nd	nd	92	nd	3,9	107	nd	nd	nd	1,7
Africa	Zambia	nd	nd	1,6	413	8,6	79,6	549	nd	2,7	nd	1,4
Asia	Cambodia	nd	nd	nd	80,4	nd	3,1	93	nd	0,6	nd	2,5
Asia	Mongolia	nd	nd	nd	38	nd	3,4	45	nd	41,6	nd	34,0
Asia	Thailand	nd	nd	nd	416	nd	8,6	473	nd	0,8	nd	2,8
Asia	Viet Nam	nd	1	nd	135	nd	14,4	166	nd	3,3	nd	4,8
The Pacific Islands	Fiji	nd	nd	nd	90,7	0,6	3,4	105,0	nd	1,8	2,5	2,7
The Pacific Islands	Kiribati	nd	nd	nd	71,2	nd	5,2	84,6	nd	1,5	nd	3,0
The Pacific Islands	Marshall Islands	nd	nd	nd	26,5	nd	1,3	30,8	nd	nd	nd	2,8
The Pacific Islands	Niue	nd	nd	nd	161,0	1,3	5,0	185,8	nd	0,7	nd	2,3
The Pacific Islands	Palau	nd	nd	nd	55,4	nd	6,5	68,2	nd	6,5	0,6	5,0
The Pacific Islands	Samoa	nd	nd	nd	109,5	nd	2,9	124,9	nd	0,5	nd	2,6
The Pacific Islands	Solomon Islands	nd	3,7	nd	1185,3	2,3	58,1	1386,1	nd	1,7	nd	3,5
The Pacific Islands	Vanatu	nd	nd	nd	114,7	nd	4,1	132,0	nd	1,0	nd	2,1
GRULAC	Antigua and Barb.	nd	nd	nd	54,5	nd	2,3	63,1	nd	1,3	0,8	4,5
GRULAC	Argentina	nd	nd	nd	134,8	nd	4,5	154,8	nd	8,5	nd	6,5
GRULAC	Barbados	nd	nd	nd	89,7	nd	2,8	102,9	0,8	3,4	nd	3,0
GRULAC	Colombia	nd	nd	nd	69,1	nd	2,5	79,6	nd	0,7	nd	4,0
GRULAC	Ecuador	nd	1,5	nd	322,9	nd	14,4	376,2	nd	1,4	nd	5,4
GRULAC	Jamaica	nd	nd	nd	80,8	0,9	8,0	98,9	nd	1,2	nd	3,3
GRULAC	Mexico	nd	nd	nd	538,0	0,9	21,5	622,2	nd	2,9	nd	6,1
GRULAC	Peru	nd	nd	nd	184,0	nd	3,5	208,6	nd	8,0	nd	3,1
GRULAC	Uruguay	nd	nd	nd	38,6	nd	3,1	46,1	nd	15,9	nd	7,1



**Table S6: (continued)** Concentrations of organochlorine pesticides and industrial contaminants in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, the Pacific Islands and GRULAC countries

Region	Country	Aldrin	Dieldrin	Heptachlor	Heptachlor-epoxide cis	Heptachlor-epoxide trans	Heptachlor group	alpha-endosulfan	beta-endosulfan	Endosulfan sulfat	Endosulfan group
Africa	DR of the Congo	nd	0,8	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Egypt	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Ethiopia	nd	nd	nd	nd	nd	nd	nd	nd	0,54	0,5
Africa	Ghana	nd	1,4	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Kenya	nd	1,3	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Mali	nd	1,7	nd	0,6	nd	0,6	nd	nd	nd	nd
Africa	Mauritius	nd	0,5	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Morocco	nd	1,8	nd	3,2	nd	3,1	nd	nd	nd	nd
Africa	Nigeria	nd	1,2	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Senegal	nd	1,9	nd	0,6	nd	0,5	nd	nd	nd	nd
Africa	UR of Tanzania	nd	2,1	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Togo	nd	0,9	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Tunisia	nd	1,0	nd	0,5	nd	nd	nd	nd	nd	nd
Africa	Uganda	nd	0,6	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Zambia	nd	2,0	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Cambodia	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Mongolia	nd	0,5	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Thailand	nd	0,6	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Viet Nam	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Fiji	nd	1,8	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Kiribati	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Marshall Islands	nd	0,8	nd	0,6	nd	0,6	nd	nd	nd	nd
The Pacific Islands	Niue	nd	3,2	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Palau	nd	1,4	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Samoa	nd	0,9	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Solomon Islands	nd	0,9	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Vanatu	nd	1,8	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Antigua and Barb.	nd	1,9	nd	1,1	nd	1,0	nd	nd	nd	nd
GRULAC	Argentina	nd	0,7	nd	1,0	nd	0,9	nd	nd	nd	nd
GRULAC	Barbados	nd	5,8	nd	1,2	nd	1,2	nd	nd	nd	nd
GRULAC	Colombia	nd	2,4	nd	0,7	nd	0,6	nd	nd	nd	nd
GRULAC	Ecuador	nd	0,7	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Jamaica	nd	2,1	nd	0,8	nd	0,8	nd	nd	nd	nd
GRULAC	Mexico	nd	1,5	nd	1,0	nd	1,0	nd	nd	nd	nd
GRULAC	Peru	nd	2,0	nd	0,7	nd	0,7	nd	nd	nd	nd
GRULAC	Uruguay	nd	2,4	nd	1,9	nd	1,8	nd	nd	nd	nd

**Table S6: (continued)** Concentrations of organochlorine pesticides and industrial contaminants in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, Pacific Islands and GRULAC countries

Region	Country	alpha-chlordane	gamma-chlordane	oxy-chlordane	Chlordane group	trans-Nonachlor	cis-Nonachlor	Endrin	Endrin ketone	Endrin group	Parlar 26	Parlar 50	Parlar 62	Parlar (toxaphene) group	Mirex
Africa	DR of the Congo	nd	nd	0,5	0,5	0,7	nd	nd	nd	nd	nd	0,6	nd	0,6	nd
Africa	Egypt	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Ethiopia	nd	nd	0,6	0,6	0,6	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Ghana	nd	nd	nd	nd	1,0	nd	nd	nd	nd	nd	0,6	nd	0,6	nd
Africa	Kenya	nd	nd	nd	nd	0,6	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Mali	nd	nd	1,3	1,2	1,1	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Mauritius	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Morocco	nd	nd	4,4	4,2	3,2	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Nigeria	nd	nd	0,9	0,9	1,9	0,6	nd	nd	nd	0,8	1,5	nd	2,3	nd
Africa	Senegal	nd	nd	4,2	4,1	1,8	nd	nd	nd	nd	nd	0,6	nd	0,6	nd
Africa	UR of Tanzania	nd	nd	nd	nd	0,5	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Togo	nd	nd	2,5	2,5	1,7	nd	nd	nd	nd	nd	0,7	nd	0,7	nd
Africa	Tunisia	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Uganda	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Africa	Zambia	nd	nd	1,3	1,2	2,0	nd	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Cambodia	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Mongolia	nd	nd	2,0	1,9	0,50	nd	nd	nd	nd	nd	nd	nd	nd	nd
Asia	Thailand	nd	nd	1,8	1,8	2,6	nd	nd	nd	nd	nd	nd	nd	nd	0,9
Asia	Viet Nam	nd	nd	nd	nd	0,6	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Fiji	nd	nd	nd	nd	0,6	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Kiribati	nd	nd	nd	nd	1,0	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Marshall Islands	nd	nd	0,9	0,9	1,6	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Niue	nd	nd	nd	nd	0,9	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Palau	nd	nd	1,2	1,2	1,8	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Samoa	nd	nd	nd	nd	1,0	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Solomon Islands	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
The Pacific Islands	Vanatu	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Antigua and Barb.	nd	nd	1,4	1,4	1,9	nd	nd	nd	nd	nd	0,7	nd	0,7	nd
GRULAC	Argentina	nd	nd	1,9	1,8	1,1	nd	nd	nd	nd	nd	nd	nd	nd	2,3
GRULAC	Barbados	nd	nd	4,6	4,4	10,0	1,3	nd	nd	nd	nd	0,6	nd	0,6	0,7
GRULAC	Colombia	nd	nd	1,4	1,4	2,4	nd	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Ecuador	nd	nd	1,0	0,9	0,8	nd	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Jamaica	nd	nd	1,9	1,8	3,1	0,6	nd	nd	nd	nd	0,6	nd	0,6	nd
GRULAC	Mexico	nd	nd	1,9	1,8	1,7	nd	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Peru	nd	nd	0,6	0,6	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
GRULAC	Uruguay	nd	nd	2,1	2,0	1,3	nd	nd	nd	nd	nd	nd	nd	nd	2,9

**Table S6: (continued)** Concentrations of organochlorine pesticides and industrial contaminants in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, Pacific Islands and GRULAC countries

Region	Country	Chlordecone	Pentachloro-phenole	Pentachloro-anisole	Dicofol; p,p-	Pentachloro-benzene	Hexachloro-1,3-Butadiene
Africa	DR of the Congo	nd	nd	nd	nd	nd	nd
Africa	Egypt	na	na	nd	nd	nd	nd
Africa	Ethiopia	nd	nd	nd	3,0	nd	nd
Africa	Ghana	nd	nd	nd	nd	nd	nd
Africa	Kenya	nd	nd	nd	nd	nd	nd
Africa	Mali	nd	nd	nd	nd	nd	nd
Africa	Mauritius	nd	nd	nd	nd	nd	nd
Africa	Morocco	nd	nd	nd	nd	nd	nd
Africa	Nigeria	nd	nd	nd	nd	nd	nd
Africa	Senegal	nd	nd	nd	nd	nd	nd
Africa	UR of Tanzania	nd	nd	nd	nd	nd	nd
Africa	Togo	nd	nd	nd	nd	nd	nd
Africa	Tunisia	nd	nd	nd	nd	nd	nd
Africa	Uganda	nd	nd	nd	nd	nd	nd
Africa	Zambia	nd	nd	nd	nd	nd	nd
Asia	Cambodia	nd	nd	nd	nd	nd	nd
Asia	Mongolia	nd	nd	nd	nd	nd	nd
Asia	Thailand	nd	nd	nd	nd	nd	nd
Asia	Viet Nam	nd	nd	nd	nd	nd	nd
The Pacific Islands	Fiji	nd	nd	1,1	nd	nd	nd
The Pacific Islands	Kiribati	nd	nd	nd	nd	nd	nd
The Pacific Islands	Marshall Islands	nd	nd	nd	nd	nd	nd
The Pacific Islands	Niue	na	na	nd	nd	nd	nd
The Pacific Islands	Palau	nd	nd	nd	nd	nd	nd
The Pacific Islands	Samoa	nd	nd	nd	nd	nd	nd
The Pacific Islands	Solomon Islands	nd	nd	nd	nd	nd	nd
The Pacific Islands	Vanatu	nd	nd	33,3	nd	nd	nd
GRULAC	Antigua and Barb.	nd	nd	nd	nd	nd	nd
GRULAC	Argentina	nd	nd	nd	nd	nd	nd
GRULAC	Barbados	nd	nd	nd	nd	nd	nd
GRULAC	Colombia	nd	nd	nd	nd	nd	nd
GRULAC	Ecuador	nd	nd	nd	nd	nd	nd
GRULAC	Jamaica	nd	nd	nd	nd	nd	nd
GRULAC	Mexico	nd	nd	nd	nd	0,6	nd
GRULAC	Peru	nd	nd	nd	nd	nd	nd
GRULAC	Uruguay	nd	nd	nd	nd	nd	nd

**Table S7:** Concentrations of PBDE in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, the Pacific Islands and GRULAC countries

Region	Country	BDE 47	BDE 99	BDE 100	BDE 153	BDE 154	BDE 183	BDE 209	Σ PBDE6	Σ PBDE7
Africa	DR of the Congo	1,36	0,33	0,21	0,32	0,03	0,07	0,16	2,31	2,47
Africa	Egypt	0,13	0,13	0,05	0,08	0,02	0,03	n.a.	0,45	n.a.
Africa	Ethiopia	0,09	0,04	0,02	0,12	0,01	0,10	0,15	0,39	0,53
Africa	Ghana	1,18	0,33	0,24	0,36	0,04	0,10	0,34	2,25	2,59
Africa	Kenya	0,77	0,19	0,17	0,32	0,02	0,08	0,18	1,56	1,74
Africa	Mali	0,61	0,17	0,12	0,34	0,03	0,11	0,16	1,38	1,54
Africa	Mauritius	0,12	0,05	0,05	0,26	0,01	0,04	0,22	0,53	0,75
Africa	Morocco	0,22	0,08	0,06	0,18	0,01	0,05	0,16	0,61	0,77
Africa	Nigeria	0,63	0,19	0,19	0,34	0,03	0,08	0,15	1,45	1,60
Africa	Senegal	0,71	0,42	0,20	0,35	0,05	0,07	0,21	1,80	2,01
Africa	UR of Tanzania	0,82	0,28	0,15	0,25	0,03	0,13	0,13	1,66	1,79
Africa	Togo	0,41	0,15	0,11	0,23	0,03	0,07	0,09	0,99	1,08
Africa	Tunisia	0,26	0,10	0,08	0,21	0,01	0,10	1,51	0,77	2,28
Africa	Uganda	0,67	0,20	0,10	0,18	0,02	0,05	0,30	1,23	1,54
Africa	Zambia	1,06	0,33	0,18	0,39	0,03	0,18	0,36	2,16	2,52
Asia	Cambodia	0,24	0,09	0,06	0,22	0,02	0,03	0,14	0,66	0,80
Asia	Mongolia	0,15	0,05	0,03	0,15	0,01	0,03	0,22	0,43	0,65
Asia	Thailand	0,15	0,07	0,04	0,12	0,01	0,02	0,16	0,41	0,57
Asia	Viet Nam	0,16	0,10	0,08	0,55	0,03	0,08	0,14	1,00	1,14
Asia	Fiji	0,68	0,17	0,12	0,36	0,02	0,12	n.a.	1,47	n.a.
Asia	Kiribati	3,66	0,98	0,79	0,66	0,10	0,08	0,18	6,26	6,45
Asia	Marshall Islands	63,12	19,53	11,02	11,43	1,56	0,14	5,92	106,79	112,70
Asia	Niue	2,56	0,69	0,51	0,53	0,06	0,03	n.a.	4,39	n.a.
Asia	Palau	9,66	1,65	1,99	1,86	0,11	0,05	3,31	15,33	18,64
Asia	Samoa	3,56	0,67	0,73	2,50	0,11	0,71	0,23	8,28	8,50
Asia	Solomon Islands	14,32	9,84	2,45	1,05	0,62	0,07	1,62	28,35	29,97
Asia	Vanuatu	2,11	0,47	0,33	0,32	0,04	0,04	0,22	3,31	3,53
GRULAC	Antigua and Barb.	7,24	1,67	1,61	3,12	0,12	0,04	n.d. < 0.06	13,79	13,79
GRULAC	Argentina	0,32	0,10	0,10	0,17	0,02	0,03	1,16	0,74	1,91
GRULAC	Barbados	2,72	0,85	0,54	0,74	0,06	0,13	0,54	5,04	5,58
GRULAC	Colombia	0,20	0,10	0,05	0,19	0,02	0,04	0,10	0,59	0,69
GRULAC	Ecuador	0,15	0,04	0,05	0,27	0,01	0,08	0,16	0,60	0,77
GRULAC	Jamaica	4,17	1,06	0,92	0,93	0,07	0,04	0,25	7,18	7,43
GRULAC	Mexico	7,67	1,62	1,20	0,97	0,08	0,03	0,91	11,58	12,49
GRULAC	Peru	1,03	0,19	0,16	0,19	0,02	0,04	2,40	1,62	4,03
GRULAC	Uruguay	0,15	0,08	0,03	0,11	0,01	0,02	0,57	0,39	0,96

**Table S8:** Concentrations of HBCDD in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, the Pacific Islands and GRULAC countries

Region	Country	alpha-HBCDD	beta-HBCDD	gamma-HBCDD
Africa	DR of the Congo	0,20	< 0.1	< 0.1
Africa	Egypt	0,80	< 0.1	< 0.1
Africa	Ethiopia	< 0.1	< 0.1	< 0.1
Africa	Ghana	0,30	< 0.1	< 0.1
Africa	Kenya	< 0.1	< 0.1	< 0.1
Africa	Mali	< 0.1	< 0.1	< 0.1
Africa	Mauritius	0,30	< 0.1	< 0.1
Africa	Morocco	< 0.1	< 0.1	< 0.1
Africa	Nigeria	0,70	< 0.1	< 0.1
Africa	Senegal	0,10	< 0.1	< 0.1
Africa	UR of Tanzania	< 0.1	< 0.1	< 0.1
Africa	Togo	0,50	< 0.1	< 0.1
Africa	Tunisia	< 0.1	< 0.1	< 0.1
Africa	Uganda	< 0.1	< 0.1	< 0.1
Africa	Zambia	< 0.1	< 0.1	< 0.1
Asia	Cambodia	< 0.1	< 0.1	< 0.1
Asia	Mongolia	0,50	< 0.1	< 0.1
Asia	Thailand	< 0.1	< 0.1	< 0.1
Asia	Viet Nam	0,30	< 0.1	< 0.1
The Pacific Islands	Fiji	< 0.1	< 0.1	< 0.1
The Pacific Islands	Kiribati	0,80	< 0.1	< 0.1
The Pacific Islands	Marshall Islands	0,50	< 0.1	< 0.1
The Pacific Islands	Niue	< 0.1	< 0.1	< 0.1
The Pacific Islands	Palau	0,20	< 0.1	< 0.1
The Pacific Islands	Samoa	1,60	< 0.1	< 0.1
The Pacific Islands	Solomon Islands	0,40	< 0.1	< 0.1
The Pacific Islands	Vanuatu	0,30	< 0.1	< 0.1
GRULAC	Antigua and Barbuda	0,30	< 0.1	< 0.1
GRULAC	Argentina	0,30	< 0.1	< 0.1
GRULAC	Barbados	0,50	< 0.1	< 0.1
GRULAC	Colombia	< 0.1	< 0.1	< 0.1
GRULAC	Ecuador	1,30	< 0.1	< 0.1
GRULAC	Jamaica	0,50	< 0.1	< 0.1
GRULAC	Mexico	0,60	< 0.1	< 0.1
GRULAC	Peru	0,70	< 0.1	< 0.1
GRULAC	Uruguay	< 0.1	< 0.1	< 0.1

**Table S9:** Concentrations of CPs in human milk samples (ng/g lipid) of the 2016-2019 period from African, Asian, the Pacific Islands and GRULAC countries

Region	Country	ΣSCCPs	ΣMCCPs	ΣSMCCPs
Africa	DR of the Congo	50	128	178
Africa	Egypt	n.a.	n.a.	n.a.
Africa	Ethiopia	70	98	168
Africa	Ghana	77	80	158
Africa	Kenya	56	57	113
Africa	Mali	69	76	145
Africa	Mauritius	111	172	284
Africa	Morocco	66	93	159
Africa	Nigeria	51	47	98
Africa	Senegal	102	207	309
Africa	UR of Tanzania	121	101	222
Africa	Togo	40	71	111
Africa	Tunisia	51	67	118
Africa	Uganda	46	94	140
Africa	Zambia	112	203	315
Asia	Cambodia	29	50	79
Asia	Mongolia	164	536	700
Asia	Thailand	18	24	43
Asia	Viet Nam	89	88	177
The Pacific Islands	Fiji	70	25	95
The Pacific Islands	Kiribati	188	85	273
The Pacific Islands	Marshall Islands	86	132	218
The Pacific Islands	Niue	n.a.	n.a.	n.a.
The Pacific Islands	Palau	24	23	48
The Pacific Islands	Samoa	175	130	305
The Pacific Islands	Solomon Islands	107	129	236
The Pacific Islands	Vanuatu	69	87	156
GRULAC	Antigua and Barb.	31	56	87
GRULAC	Argentina	32	25	58
GRULAC	Barbados	38	46	85
GRULAC	Colombia	33	20	53
GRULAC	Ecuador	20	19	39
GRULAC	Jamaica	46	58	103
GRULAC	Mexico	28	109	137
GRULAC	Peru	114	137	251
GRULAC	Uruguay	34	17	50

n.a. = not analyzed (not sufficient sample material)

**Table S10:** Concentrations of PCNs in human milk samples (pg/g lipid) of the 2016-2019 period from African countries

UN Region →	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa	Africa
Country →	DR of the Congo	Ethiopia	Ghana	Kenya	Mali	Mauritius	Morocco	Nigeria	Senegal	UR of Tanzania	Togo	Tunisia	Uganda	Zambia
PCN 27	n.d. < 0.062	n.d. < 0.092	n.d. < 0.057	n.d. < 0.070	0,24	n.d. < 0.064	0,20	n.d. < 0.041	0,10	n.d. < 0.069	0,17	0,09	n.d. < 0.054	0,11
PCN 28/36	3,30	2,03	3,86	2,17	3,07	2,59	3,07	3,67	6,27	2,70	3,45	1,51	8,33	2,78
PCN 31	n.d. < 0.087	n.d. < 0.129	n.d. < 0.079	n.d. < 0.098	n.d. < 0.125	n.d. < 0.090	n.d. < 0.095	n.d. < 0.058	n.d. < 0.077	n.d. < 0.096	n.d. < 0.135	n.d. < 0.085	n.d. < 0.076	n.d. < 0.094
PCN 42	2,72	2,02	2,82	2,72	2,44	1,91	5,37	2,78	3,62	2,59	2,81	1,84	1,68	3,03
PCN 46	1,72	1,40	1,11	1,89	2,21	1,16	3,97	1,30	1,50	0,96	1,45	0,84	0,90	1,65
PCN 48	1,25	0,90	2,17	1,32	1,46	0,56	2,17	1,68	1,84	0,83	1,44	1,04	0,93	1,01
PCN 49	n.d. < 0.081	n.d. < 0.127	n.d. < 0.066	n.d. < 0.091	n.d. < 0.103	n.d. < 0.070	n.d. < 0.065	n.d. < 0.050	n.d. < 0.065	n.d. < 0.077	n.d. < 0.130	n.d. < 0.057	n.d. < 0.078	n.d. < 0.101
PCN 50	0,29	0,21	0,23	0,30	0,22	0,11	0,17	0,19	0,14	0,17	0,13	0,10	0,18	0,29
PCN 52/60	22,92	10,44	22,44	14,38	14,48	10,89	23,30	29,52	21,76	14,76	24,79	25,59	13,55	16,11
PCN 53	0,87	1,01	0,62	1,10	0,42	0,45	0,39	0,73	0,46	0,63	0,68	0,19	0,94	1,05
PCN 59	2,45	2,31	1,53	3,16	1,52	1,20	1,32	2,16	1,43	1,52	1,57	0,48	2,25	2,63
PCN 63	0,67	0,48	0,71	0,59	0,31	0,19	0,23	0,84	0,38	0,45	0,40	0,12	1,09	0,69
PCN 64/68	0,37	0,22	0,36	0,43	0,25	0,07	0,13	0,61	0,25	0,19	0,23	0,05	0,31	0,44
PCN 65	0,28	0,40	0,18	0,37	n.d. < 0.102	0,10	n.d. < 0.070	0,37	0,15	0,21	n.d. < 0.096	n.d. < 0.050	0,42	0,44
PCN 66/67	16,80	4,66	15,49	10,63	11,87	8,83	17,78	14,40	21,54	9,28	19,71	19,18	6,83	10,35
PCN 69	6,09	2,29	4,04	2,55	2,01	1,22	5,45	6,05	2,52	2,44	3,35	5,50	3,31	3,38
PCN 70	0,15	n.d. < 0.100	0,14	n.d. < 0.064	n.d. < 0.104	0,07	0,15	0,13	0,16	n.d. < 0.081	0,15	n.d. < 0.051	n.d. < 0.064	n.d. < 0.065
PCN 71/72	0,70	0,75	0,50	0,79	0,50	0,32	0,16	1,16	0,45	0,39	0,35	n.d. < 0.048	0,96	0,78
PCN 73	1,01	0,40	0,53	0,45	0,50	0,27	0,45	0,59	0,55	0,24	0,53	0,33	0,93	0,42
PCN 74	n.d. < 0.041	n.d. < 0.054	n.d. < 0.042	0,05	n.d. < 0.056	n.d. < 0.040	n.d. < 0.041	n.d. < 0.031	n.d. < 0.041	n.d. < 0.046	n.d. < 0.051	n.d. < 0.031	0,06	0,03
PCN 75	n.d. < 0.054	n.d. < 0.108	n.d. < 0.066	n.d. < 0.052	n.d. < 0.103	n.d. < 0.082	n.d. < 0.068	n.d. < 0.044	n.d. < 0.054	n.d. < 0.078	n.d. < 0.095	n.d. < 0.061	n.d. < 0.053	n.d. < 0.058
Sum PCN - 26 congeners (LB)	61,6	29,6	56,8	42,9	41,5	29,9	64,3	66,2	63,1	37,4	61,2	56,9	42,7	45,2
Sum PCN- 13 Congeners *)	48,8	19,8	44,0	30,4	30,0	22,0	47,7	53,1	47,5	27,9	49,6	50,8	26,9	32,6

1) Egypt: not analyzed (not sufficient sample material)

\*) PCNs 52/60, 53, 66/67, 64/68, 69, 71/72, 73, 74, 75 (LB)

**Table S11:** Concentrations of PCNs in human milk samples (pg/g lipid) of the 2016-2019 period from Asian and the Pacific Islands countries

UN Region →	Asia	Asia	Asia	Asia	The Pacific Islands	The Pacific Islands	The Pacific Islands	The Pacific Islands	The Pacific Islands	The Pacific Islands
Country →	Cambodia	Mongolia	Thailand	Viet Nam	Kiribati	Marshall Isl.	Palau	Samoa	Solomon Isl	Vanuatu
PCN 27	n.d. < 0.042	0,19	n.d. < 0.109	n.d. < 0.048	n.d. < 0.054	n.d. < 0.073	0,13	n.d. < 0.081	0,21	n.d. < 0.070
PCN 28/36	3,76	6,39	1,18	4,66	6,98	11,45	12,87	7,06	2,90	5,87
PCN 31	n.d. < 0.059	n.d. < 0.130	n.d. < 0.153	n.d. < 0.067	n.d. < 0.076	n.d. < 0.102	n.d. < 0.067	n.d. < 0.113	n.d. < 0.111	n.d. < 0.097
PCN 42	0,73	8,26	0,84	3,57	2,66	2,17	2,13	2,44	2,92	1,85
PCN 46	0,37	4,83	n.d. < 0.184	0,58	0,92	0,79	1,07	1,24	1,89	1,14
PCN 48	0,97	0,77	0,57	1,28	1,22	0,74	0,78	1,54	1,36	1,18
PCN 49	n.d. < 0.045	n.d. < 0.088	n.d. < 0.085	n.d. < 0.052	n.d. < 0.077	n.d. < 0.093	n.d. < 0.047	n.d. < 0.118	n.d. < 0.132	n.d. < 0.084
PCN 50	0,08	0,21	0,13	0,21	0,29	0,13	0,16	0,41	1,12	0,17
PCN 52/60	7,37	13,67	9,24	13,05	26,17	18,15	13,93	22,96	38,80	13,59
PCN 53	0,06	0,69	n.d. < 0.089	0,11	0,78	0,61	0,35	0,98	5,25	0,61
PCN 59	0,33	1,45	0,25	0,44	2,34	1,53	0,91	3,97	9,95	1,53
PCN 63	0,25	0,12	0,16	0,24	0,81	0,43	0,23	1,11	5,74	0,38
PCN 64/68	0,16	n.d. < 0.066	n.d. < 0.119	n.d. < 0.053	0,37	0,26	0,15	0,42	2,37	0,18
PCN 65	n.d. < 0.062	n.d. < 0.072	n.d. < 0.130	n.d. < 0.058	0,42	0,23	0,08	0,63	3,88	0,18
PCN 66/67	10,31	27,41	12,61	19,16	10,09	11,18	13,06	13,31	12,04	6,32
PCN 69	1,84	0,99	1,03	1,62	5,77	3,53	1,47	4,60	18,56	2,48
PCN 70	0,12	n.d. < 0.074	n.d. < 0.132	0,25	n.d. < 0.063	n.d. < 0.072	0,07	0,18	n.d. < 0.138	n.d. < 0.091
PCN 71/72	n.d. < 0.059	0,15	n.d. < 0.124	0,19	0,77	0,57	0,20	1,16	5,87	0,32
PCN 73	0,53	0,51	0,37	0,64	0,55	0,38	0,40	0,70	0,93	0,26
PCN 74	0,06	0,04	n.d. < 0.073	n.d. < 0.039	n.d. < 0.037	n.d. < 0.049	n.d. < 0.037	0,07	0,15	n.d. < 0.053
PCN 75	n.d. < 0.075	n.d. < 0.079	n.d. < 0.169	n.d. < 0.054	n.d. < 0.063	n.d. < 0.077	n.d. < 0.054	n.d. < 0.087	n.d. < 0.083	n.d. < 0.094
Sum PCN (LB)	26,9	65,7	26,4	46,0	60,1	52,2	48,0	62,8	113,9	36,0
Sum PCN- 13 Congeners *)	20,3	43,5	23,2	34,8	44,5	34,7	29,6	44,2	84,0	23,8

1) Fiji, Niue: not analyzed (not sufficient sample material)

\*) PCNs 52/60, 53, 66/67, 64/68, 69, 71/72, 73, 74, 75 (LB)



**Table S12:** Concentrations of PCNs in human milk samples (pg/g lipid) of the 2016-2019 period from GRULAC countries

UN Region →	GRULAC	GRULAC	GRULAC	GRULAC	GRULAC	GRULAC	GRULAC	GRULAC	GRULAC
Country →	Antigua and Barb.	Argentina	Barbados	Colombia	Ecuador	Jamaica	Mexiko	Peru	Uruguay
PCN 27	n.d. < 0.069	0,08	n.d. < 0.069	0,19	n.d. < 0.065	n.d. < 0.263	0,20	n.d. < 0.081	0,19
PCN 28/36	13,90	2,41	20,48	3,59	1,28	22,51	6,62	1,41	4,51
PCN 31	n.d. < 0.096	n.d. < 0.070	n.d. < 0.096	n.d. < 0.077	n.d. < 0.091	n.d. < 0.367	0,16	n.d. < 0.113	n.d. < 0.058
PCN 42	2,61	1,18	1,68	1,00	0,94	4,97	1,09	1,05	2,25
PCN 46	1,02	0,65	0,82	0,82	0,57	2,14	0,56	0,66	0,97
PCN 48	0,64	0,98	0,66	0,84	0,61	1,26	1,49	1,35	1,63
PCN 49	n.d. < 0.087	n.d. < 0.060	n.d. < 0.063	n.d. < 0.046	n.d. < 0.061	n.d. < 0.307	n.d. < 0.054	n.d. < 0.082	n.d. < 0.053
PCN 50	0,08	0,18	0,09	0,11	0,10	0,31	0,19	0,19	0,20
PCN 52/60	15,25	12,94	9,69	9,05	9,60	31,14	9,03	13,93	25,08
PCN 53	0,33	0,35	0,25	0,21	0,20	1,68	0,27	0,31	0,56
PCN 59	0,90	0,57	0,51	0,51	0,64	3,90	0,56	0,81	1,33
PCN 63	0,24	0,33	0,16	0,18	0,20	1,30	0,22	0,23	0,35
PCN 64/68	0,09	0,18	n.d. < 0.070	n.d. < 0.056	0,10	0,40	0,25	n.d. < 0.069	0,12
PCN 65	0,12	n.d. < 0.058	n.d. < 0.076	n.d. < 0.061	n.d. < 0.062	n.d. < 0.323	0,09	n.d. < 0.076	0,13
PCN 66/67	11,42	18,24	8,91	12,11	10,21	17,61	12,12	16,50	24,76
PCN 69	1,68	1,88	1,19	1,27	1,93	6,55	1,39	1,75	2,52
PCN 70	n.d. < 0.064	0,07	0,08	0,07	n.d. < 0.063	n.d. < 0.329	0,15	n.d. < 0.077	0,12
PCN 71/72	0,18	0,18	0,10	n.d. < 0.058	0,18	1,82	0,26	0,10	0,31
PCN 73	0,33	0,47	0,21	0,40	0,25	0,52	0,33	0,29	0,40
PCN 74	n.d. < 0.035	n.d. < 0.037	n.d. < 0.042	n.d. < 0.048	n.d. < 0.036	n.d. < 0.177	n.d. < 0.029	n.d. < 0.042	n.d. < 0.027
PCN 75	n.d. < 0.061	n.d. < 0.067	n.d. < 0.078	n.d. < 0.075	n.d. < 0.076	n.d. < 0.351	n.d. < 0.057	n.d. < 0.090	n.d. < 0.049
Sum PCN (LB)	48,80	40,69	44,84	30,36	26,81	96,11	34,99	38,59	65,42
Sum PCN- 13 Congeners *)	29,3	34,2	20,4	23,0	22,5	59,7	23,7	32,9	53,7

\*) PCNs 52/60, 53, 66/67, 64/68, 69, 71/72, 73, 74, 75 (LB)

**Table S13:** Concentrations of PFAS in human milk samples (ng/l) of the 2016-2019 period from GRULAC countries (data from GMP Data Warehouse; note that data were published as pg/g f.w., including results for L-PFOS and br-PFOS, see Fiedler and Sadia, 2021)

Region	Country	Year	PFOS *) (ng/l)	PFOA (ng/l)	PFHxS (ng/l)
Africa	DR of Congo	2017	< 6,4	12,5	< 5,8
Africa	Ethiopia	2019	9,4	6,4	< 5,8
Africa	Ghana	2019	16,0	11,7	< 5,8
Africa	Kenya	2019	< 6,4	10,7	< 5,8
Africa	Mali	2019	22,5	13,2	< 5,8
Africa	Mauritius	2018	13,2	18,5	< 5,8
Africa	Morocco	2019	14,0	16,2	< 5,8
Africa	Nigeria	2019	10,7	14,8	< 5,8
Africa	Senegal	2018	10,5	14,0	< 5,8
Africa	UR of Tanzania	2019	7,7	11,3	< 5,8
Africa	Togo	2017	13,2	18,3	< 5,8
Africa	Tunisia	2019	20,4	18,6	< 5,8
Africa	Uganda	2018	< 6,4	8,6	< 5,8
Africa	Zambia	2019	< 6,4	8,9	< 5,8
Asia and Pacific	Cambodia	2019	17,7	15,1	< 5,8
Asia and Pacific	Fiji	2019	< 6,4	10,5	< 5,8
Asia and Pacific	Kiribati	2018	218,2	32,8	115
Asia and Pacific	Marshall Isl	2019	13,3	13,3	< 5,8
Asia and Pacific	Mongolia	2018	10,5	20,7	< 5,8
Asia and Pacific	Niue (2017)	2017	22,3	17,6	< 5,8
Asia and Pacific	Niue (2019)	2019	25,6	10,3	< 5,8
Asia and Pacific	Palau	2018	9,8	20,8	< 5,8
Asia and Pacific	Samoa	2019	26,8	22,1	< 5,8
Asia and Pacific	Solomon Isl	2019	15,8	11,9	< 5,8
Asia and Pacific	Thailand	2018	26,3	26,6	7,5
Asia and Pacific	Vanuatu	2018	9,5	11,5	< 5,8
Asia and Pacific	Viet Nam	2019	33,0	13,3	< 5,8
GRULAC	Antigua and Barbuda	2018	13,9	19,4	< 5,8
GRULAC	Argentina	2019	12,2	12,1	< 5,8
GRULAC	Barbados	2018	13,1	19,6	< 5,8
GRULAC	Colombia	2019	< 6,4	16,4	< 5,8
GRULAC	Ecuador	2019	9,6	8,9	< 5,8
GRULAC	Jamaica	2018	18,9	17,1	< 5,8
GRULAC	Mexico	2017	6,5	17,6	< 5,8
GRULAC	Peru	2019	< 6,4	8,0	< 5,8
GRULAC	Uruguay	2019	41,7	13,5	< 5,8

\* sum of L-PFOS and br-PFOS



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