Time Trends in Human Milk Derived from WHO- and UNEP-Coordinated Exposure Studies, Chapter 3: Perfluoroalkyl Substances (PFAS)

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Abstract

Temporal trends of perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), and perfluorohexane sulfonic acid (PFHxS) were assessed using 86 pooled human milk samples from 59 countries from all United Nations Regional Groups collected between 2008 and 2019 as part of the WHO/UNEP-coordinated exposure studies. The primary objective of these temporal studies is to provide monitoring data for the effectiveness evaluation of the Stockholm Convention on Persistent Organic Pollutants (POPs). General temporal trends were estimated using data from all participating countries by grouping into three equal four-year periods (2008–2011, 2012–2015, and 2016–2019) reflecting the performance of three rounds of the studies. A more precise approach is the use of data from 24 countries with repeated participation in the WHO/UNEP-coordinated exposure studies, 22 of them in different periods and two in the same period. However, there were no Western European countries with multiple participation, and only two countries from the Asia-Pacific Region with one country submitting two samples in the same period.

The country-specific PFOS data showed decreasing tendencies in 19 of the 22 countries with participation in different periods, quite constant levels in two countries and increasing tendencies in one country (from its first participation in 2010 to the second participation in 2015). For the two countries with repeated

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participation in the same period, it does not seem appropriate to derive country-specific temporal tendencies. An overall decrease of PFOS concentrations over 10 years of 48% and 52%, respectively, was calculated by the Theil–Sen method (1) using all samples and (2) using samples from countries with repeated participation. PFOA concentrations showed decreasing tendencies in 17 countries of the 22 countries with participation in different periods, quite constant levels in two countries and increasing tendencies in three countries (from their first participation between 2009 and 2011 to the second participation in 2015). An overall decrease over 10 years for PFOA concentrations of 42% and 47%, respectively, was calculated by the Theil–Sen method (1) using all samples and (2) using samples from countries with repeated participation. The estimation of general temporal trends for PFHxS was not possible, as in 84% of the samples PFHxS concentrations were below the limit of quantification (5.7 ng/L).

Keywords

Time trends · Human milk biomonitoring · Stockholm Convention on Persistent Organic Pollutants · Perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), Perfluorohexane sulfonic acid (PFHxS) · Global WHO/UNEP studies · UN Regional Groups

1 Introduction

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 (Malisch et al. 2023a). The number of analytes in these studies gradually expanded as new POPs were listed in the annexes of the Stockholm Convention, including perfluorooctane sulfonic acid (PFOS) (UNEP 2009; UNEP 2019a), perfluorooctanoic acid (PFOA) (UNEP 2019b), and perfluorohexane sulfonic acid (PFHxS) (UNEP 2019c; UNEP 2022). PFOS, PFOA, and PFHxS were included as targeted compound in samples of the studies collected between 2008 and 2019 and analysed at the Örebro University, Örebro, Sweden.

Data on the regional occurrence of these substances during 2016–2019 comprise 44 human milk samples collected in 42 countries. PFOS was quantifiable in 36 samples across a wide range (total PFOS between <6.2 pg/g and 212 pg/g, calculated as sum of linear PFOS [L-PFOS] and branched isomers [br-PFOS]); PFOA was quantified in all 44 samples in a narrower range (6.20 pg/g–37.4 pg/g); PFHxS was quantifiable in only four samples (max. 111 pg/g). Branched PFOS isomers on average had a share of 16% of the total PFOS with a maximum of 33% (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. It was explained that the GMP guidance (UNEP 2019d) stipulates the goal to achieve 50% reduction of POPs concentrations in the core matrices over a 10-year period. It was concluded that the Stockholm Convention goal of 50% reduction in 10 years was achieved for PFOS by Antigua and Barbuda, Kenya, and Nigeria and for PFOA by Antigua and Barbuda, only. In a few cases, increases were observed; in one country for PFOS, in four countries for PFOA (Fiedler et al. 2022).

In Part IV of this compendium on WHO/UNEP-coordinated exposure studies on human milk, three specific articles assess time trends. Two approaches can be used: (1) temporal trends derived from all participating countries; (2) temporal trends derived only from countries with multiple participation in WHO/UNEP-coordinated exposure studies. The first article "Time Trends in Human Milk Derived from WHO- and UNEP-Coordinated Exposure Studies, Chapter 1" introduced various aspects and presented results for polychlorinated biphenyls (PCB), polychlorinated dibenzo-p-dioxins (PCDD), and polychlorinated dibenzofurans (PCDF) (Malisch et al. 2023b), the second article (Chapter 2) on DDT [dichlorodiphenyltrichloroethane], beta-hexachlorocyclohexane (beta-HCH), and hexachlorobenzene (HCB) (Malisch et al. 2023c). In addition, temporal trends for polybrominated diphenyl ethers (PBDE) were assessed as part of the presentation of findings of polybrominated substances (Schächtele et al. 2023). This article (as Chapter 3 of the three specific articles on time trends) presents the assessment of temporal trends of perfluoroalkyl substances (PFAS).

2 Material and Methods

2.1 Source of Data

Per- and polyfluoroalkyl substances (PFAS) were analysed at the Örebro University, Örebro, Sweden and the analytical methods and results published (Sadia et al. 2020; Fiedler and Sadia 2021; Fiedler et al. 2022). The results were presented on product basis (as pg/g fresh weight). *(PFAS have different physical and chemical properties than the chlorinated and brominated POPs listed by the Convention, which are reported on lipid basis.)*

The data belong to UNEP and the sample-submitting countries and are publicly available in the Data Warehouse of the Stockholm Convention Global Monitoring Plan (GMP DWH) (GMP DWH 2020). For this article, the data base of the GMP DWH was used providing PFAS data on volume basis (as ng/L). Data of 86 pooled samples obtained from 59 countries between 2008 and 2019 were used.

2.2 Methods of Statistical Data Treatment: Trends vs. Tendencies

For methods of statistical data treatment, see subsection 2.4 in the preceding article "Time Trends in Human Milk Derived from WHO- and UNEP-Coordinated Exposure Studies, Chapter 1" on time trends for PCB, PCDD, and PCDF (Malisch et al. 2023b). This included the non-parametric linear Theil–Sen trend estimator and the median method to derive decreases (*decrease rate constants*) that are expected to show exponential trends (as commonly observed in cases after stop of production and application of a chemical rather than unrealistic linear trends). Non-detects were substituted by 0.707 multiple of the detection limit.

We differentiate between *trends* as statistically significant decrease (requiring *p*-values <0.05) and changes of concentrations indicating *tendencies* as statistically not significant decrease. Simulations show that Theil–Sen p is never below 0.05 for fewer than 5 data points. As for all countries with repeated participation only two data points were available, the observed changes of PFAS concentrations in these countries are statistically not significant and therefore only indicate tendencies. However, on regional and global basis, data from countries with repeated participation can be combined to provide more than 5 data points allowing to derive statistically significant trends.

The median method is based on tendencies of individual countries. Thus, the inclusion of countries in the median method is not possible, if a country did not participate at least twice. Therefore, the results of the median method applied for the UN regional groups comprising all countries are the same as for the UN regional groups comprising only countries with repeated participation. However, the results of the worldwide median method are calculated as median trend of the five regional trends and not based on individual countries trends. Therefore, the results of the worldwide median method computed for PFAS concentrations using all countries are not the same as using only countries with repeated participation.

As PFOS was listed under the Stockholm Convention already in 2009, PFOA only in 2019, and PFHxS was through the assessment procedure but not yet through the process of adoption, the Hites method of break point search was used to check whether a break point between possibly increasing and decreasing trends in the samples collected between 2009 and 2019 could be found (Hites 2019).

3 Results and Discussion

The assessment of temporal trends is a key element for the effectiveness evaluation of the Stockholm Convention on Persistent Organic Pollutants (POPs) and human milk is a core matrix for this purpose. As the first step, time trends can be derived based on data from all participating countries. However, levels among countries are often highly variable. Therefore, a more precise approach is the assessment of temporal trends by considering only countries with repeated participation in the WHO/UNEP-coordinated exposure studies. Because levels correlate within countries more closely than among countries, this allows more certainty in drawing conclusions on time trends which are not potentially influenced by individual results from countries submitted just for a single round and seems optimal for the evaluation of the effectiveness for the purpose of Article 16 of the Convention. However, it should be noted that typically only very few time points from most individual countries are available, which prevents from deriving statistically significant temporal trends in these cases. Yet, the existing data can still indicate decreasing or increasing tendencies in POP concentrations. Nevertheless, pooling data in regions allows to derive statistically significant time trends in the UN regional groups and globally.

To provide reliable monitoring information for the Parties to the Stockholm Convention, the GMP guidance document proposed a quantitative objective for temporal studies: These studies should be able to detect a 50% decrease in the levels of POPs within a 10-year period (UNEP 2015; UNEP 2019d). However, in distinction from this goal for abilities of temporal studies to detect changes over time, there is no stipulation of a quantitative goal for the rate of reduction/decrease in POPs levels, e.g. in the core matrices by the GMP guidance document or by the Convention. 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.

For the evaluation of time trends, the 86 pooled samples obtained from 59 countries between 2008 and 2019 can be grouped, e.g. into three 5-year periods: 2005–2009, 2010–2014, 2015–2019 (Fiedler et al. 2022). However, the samples were obtained between 2008 and 2019, with 3 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, e.g. 2008–2011, 2012–2015, and 2016–2019 reflect more closely the performance of rounds in the WHO/UNEP-coordinated exposure studies (fifth, sixth, and seventh survey) (Malisch et al. 2023a), seem to be more appropriate and were used in this article.

The regional distribution of samples and the regional distribution within each of the three periods varied considerably, with most samples originating from the African region. There was no sample from Western European and Others Group (WEOG) in the first period (2008–2011). The second period (2012–2015) has the lowest number of samples in all UN Regions except the Eastern European Group (EEG) (Table 1). Note that two countries submitted two samples in different years of the same period: Chile (2008, 2011) and Niue (2017, 2019; the sample of 2017 was sent for analysis of all POPs and the sample of 2019 only for determination of PFAS). Furthermore, Germany submitted two sub-pools in 2019.

Table 1 Regional distribution of national pools and their distribution within each of three periods

	Countries	Number of national samples			
	(all)	2008–2011	2012–2015	2016–2019	Total
Africa	16	11	2	14	27
Asia-Pacific[a]	16	5	0	13	18
GRULAC[b]	12	8	4	9	21
EEG	8	3	7	2	12
WEOG[c]	7	0	2	6	8
Sum	59	27	15	44	86

[a]1 Country with two samples in the same period (Niue, 2017 and 2019)
[b]1 Country with two samples in the same period (Chile, 2008 and 2011)
[c]1 Country with two sub-pools (Germany, 2019)

Table 2 Regional distribution of national pools obtained from countries with repeated participation and their distribution within each of three periods

	Countries	Number of national samples (repeated participation)			
	(repeated participation)	2008–2011	2012–2015	2016–2019	Total
Africa	11	11	2	9	22
Asia-Pacific[a]	2	1	0	3	4
GRULAC[b]	7	8	1	5	14
EEG	4	3	4	1	8
WEOG	0	0	0	0	0
Sum	24	23	7	18	48

[a]1 country with two samples in the same period (Niue, 2017 and 2019)
[b]1 country with two samples in the same period (Chile, 2008 and 2011)

Table 3 Regional distribution of years between samplings

	Countries with repeated participation	Countries with data for 2008–2011 and 2016–2019	Years between samplings	Countries with data for two subsequent periods	Years between samplings	Countries with data for the same period	Years between samplings
Africa	11	9	7–11	2	4–5	0	–
Asia-Pacific	2	1	8	0	–	1	2
GRULAC	7	5	6–10	1	4	1	3
EEG	4	0	–	4	5–6	0	–
WEOG	0	0	–	0	–	0	–
Sum	24	15		7		2	

24 countries participated repeatedly between 2008 and 2019 submitting in total 48 samples (Table 2). 22 of these countries submitted samples in different periods, two countries in the same period (Chile, 2008 and 2011, and Niue, 2017 and 2019). Most samples came from African countries ($N = 22$), followed by the Group of Latin America and the Caribbean Countries (GRULAC) ($N = 14$). There were no WEOG countries with repeated participation in these three periods.

With regard to the length of the time-series, the GMP guidance document considers it naïve to expect monitoring of POPs to reveal temporal trends with any confidence within a sampling period of five years unless the changes are very large. More suitable would be a period of at least 10–15 years to detect significant changes of moderate size (5% /year) (UNEP 2019d). Table 3 gives an overview of the number of years between the samplings in the UN regions. For 15 countries, PFAS results are available for the first (2008–2011) and third (2016–2019) period with 6–11 years between samplings. In 7 countries, data are available for two subsequent periods (either [i] 2008–2011 and 2012–2015 or [ii] 2012-–2015 and 2016–2019) with 4–6 years between samplings. In two countries, samples were collected in the same period, with 2–3 years between samplings.

3.1 PFOS

PFOS was listed in Annex B of the Convention in 2009 (UNEP 2009).

3.1.1 Estimation of Temporal Trends by Differentiation into Three Periods between 2008 and 2019

As PFAS analysis was initiated with samples submitted in 2008, time trends for PFOS can be derived beginning with the 2008–2011 period. As a starting point, descriptive parameters summarizing the results of 86 national pools from 59 countries were used as basis for the first *general* estimation of temporal trends with differentiation into three equal four-year periods between 2008 and 2019. The median of the PFOS concentrations increased from 24.6 ng/L in the 2008–2011 period ($N = 27$) to 36.0 ng/L in the 2012–2015 period ($N = 15$) and then decreased to 13.6 ng/L in the 2016–2019 period ($N = 44$). The highest maximum level (218 ng/L) was found in the 2016–2019 period (Fig. 1).

Thus, the summarizing descriptive parameters seem to indicate an increase of the PFOS concentrations after listing in 2009 until the 2012–2015 period. However, it has to be checked whether these fluctuations of the median are likely due to the result of participation of different countries in different rounds of the WHO/UNEP-coordinated exposure studies. Therefore, as described for PCDD/PCDF, PCB, DDT, beta-HCH, and HCB in the first two chapters of the three specific articles on assessment of time trends, also for PFOS a more precise evaluation based on country-specific results is necessary.

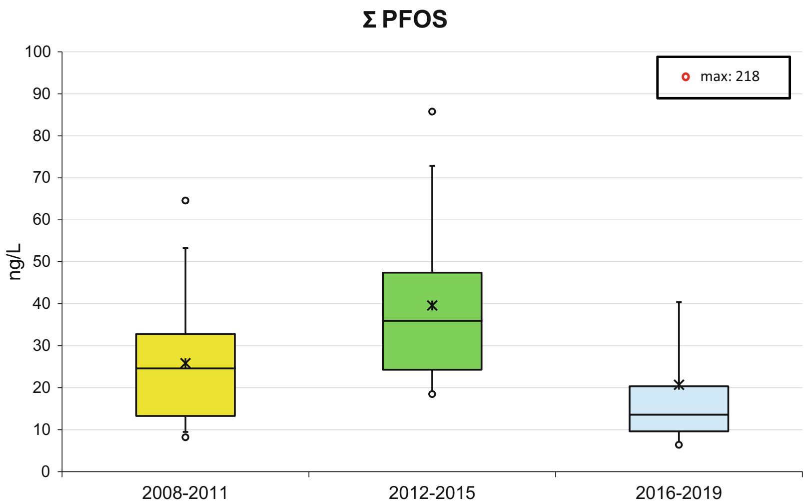

Fig. 1 Range of ∑PFOS concentrations (ng/L) in all samples over three periods ($N = 27$ in 2008–2011; $N = 15$ in 2012–2015; $N = 44$ in 2016–2019) [box plot; minimum and maximum: as circles; fifth and 95th percentile: as whiskers; lower (25–50%) and upper (50–75%) quartiles, separated by the line for the median: as box; mean: as asterisk]

3.1.2 African Group

In the 27 national pools from 16 African countries, ∑PFOS concentrations between 2008 and 2019 ranged <6.4–45.9 ng/L, with downward tendencies from the 2008–2011 period to 2016–2019 in 9 countries with repeated participation (decrease by 44% as median of the observed decrease between the first and second submission [without normalization, e.g. to a 10-year period]), with quite constant concentrations between 2011 and 2015 in Niger and increasing concentrations from 2010 (33.3 ng/L) to 2015 (45.9 ng/L) in Côte d'Ivoire (Fig. 2).

Figure 3 illustrates the temporal tendencies in countries with multiple participation using the Theil–Sen method (comprising all individual pooled samples of countries with repeated participation and assuming exponential trends, see subsection 2.2). The overall decrease per 1 year and 10 years is given in Table 4. The limited number of samples did not allow to determine a statistically significant decrease for countries ($p \sim 1.000$) (for statistical significance of *trends* requiring p-values <0.05 and changes of concentrations indicating *tendencies*, see subsection 2.2). As median of 11 countries, the levels of ∑PFOS in all African countries decreased within a 10-year period by 48%. This is in line with the statistically significant ($p < 0.001$) decrease over 10 years of 50.3% for this UN region derived by countries with multiple participation and calculated by the Theil–Sen method (see Table 5 in the following).

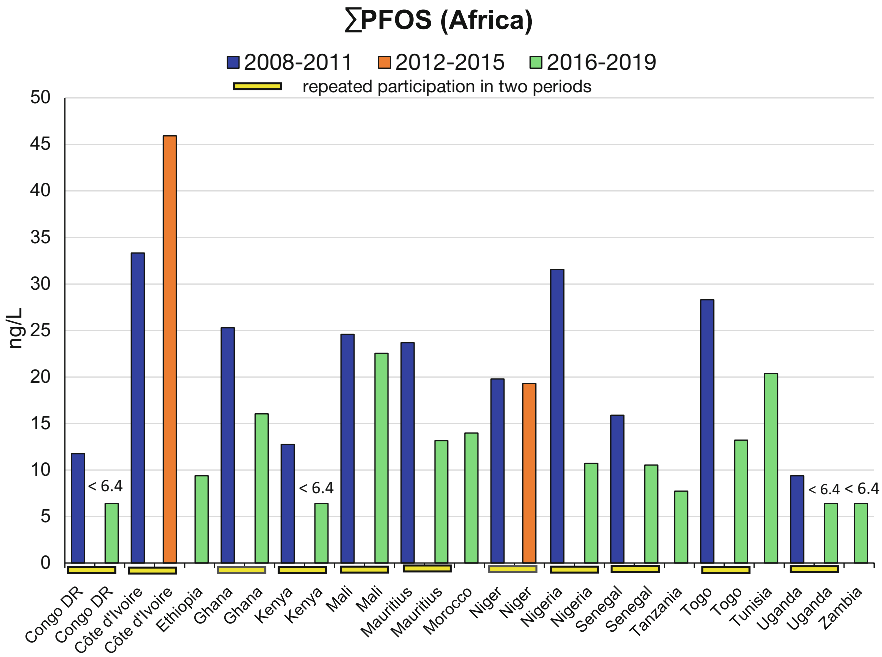

Fig. 2 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from African countries over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

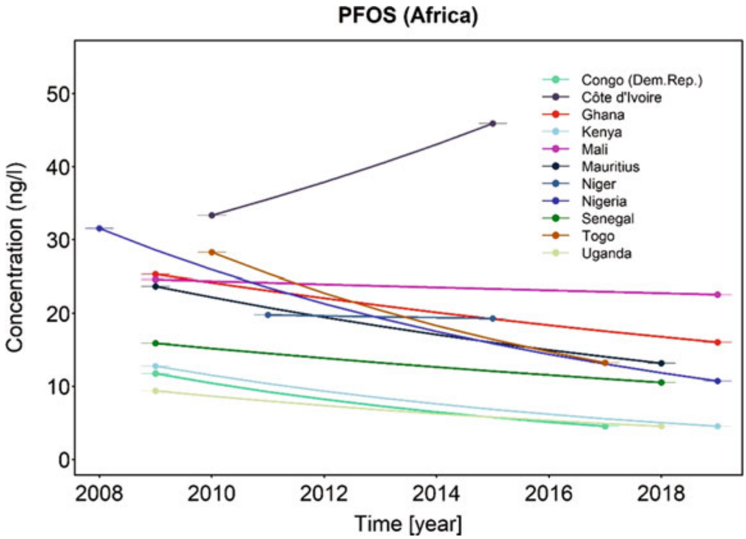

Fig. 3 Temporal tendencies of ∑PFOS concentrations (ng ∑PFOS/L) for African countries with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 4 Overall decrease (%) of ∑PFOS concentrations per 1 year and 10 years in African countries (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Congo (DR)	11.2	69.7	1.000
Côte d'Ivoire	−6.6	−89.7	1.000
Ghana	4.5	36.6	1.000
Kenya	9.9	64.6	1.000
Mali	0.9	8.3	1.000
Mauritius	6.3	48.0	1.000
Niger	0.7	6.3	1.000
Nigeria	9.3	62.5	1.000
Senegal	4.5	36.7	1.000
Togo	10.3	66.3	1.000
Uganda	7.8	55.5	1.000
Median	6.3	48.0	

Statistically significant time trends can be derived in the UN regional groups by pooling of data from countries. The time trends of ∑PFOS concentrations in the African region derived by the Theil–Sen method are illustrated by Fig. 4 for all 27 national pools from 16 countries and by Fig. 5 for 22 national pools from 11 countries with multiple participation. An overall decrease over 10 years for

Table 5 Overall decrease (%) of PFOS concentrations in the African Group computed (1) using all samples and (2) using samples from countries with repeated participation

African Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	16	6.8	6.3	50.3	48.0	<0.001
Repeatedly	11	6.4	6.3	48.5	48.0	≤0.001

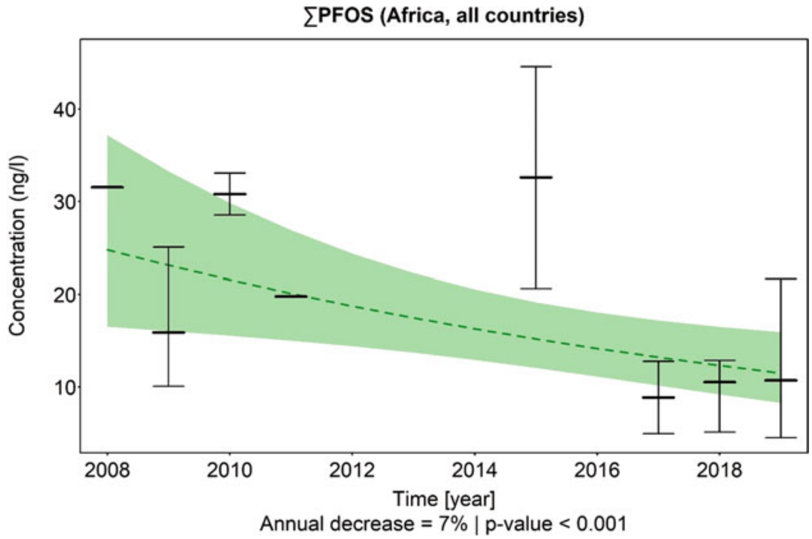

Fig. 4 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in all African countries (27 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

PFOS concentrations in the African Group of 50.3% and 48.5%, respectively, was determined by using (1) all samples and (2) samples from countries with repeated participation (Table 5).

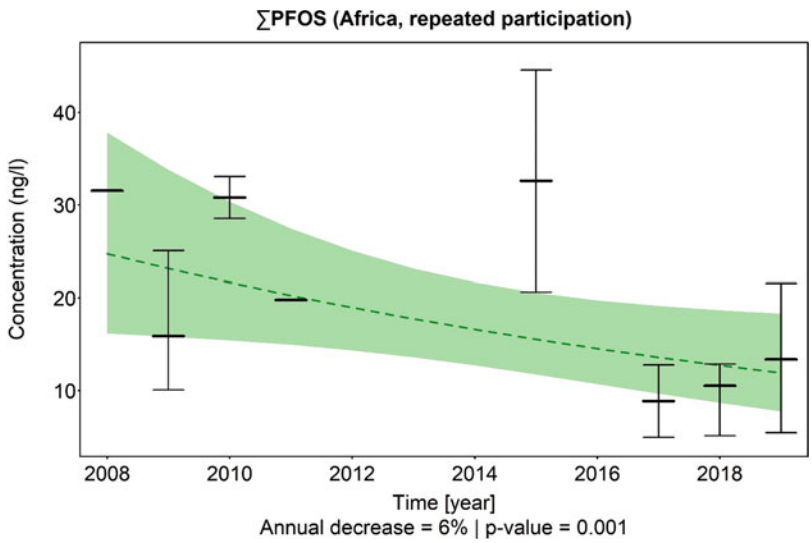

Fig. 5 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in African countries with repeated participation (22 national pools, 11 countries)

3.1.3 Asia-Pacific Group

In the most recent period (2016–2019), the pooled samples from 12 countries had ∑PFOS concentrations in the range < 6.4–30 ng/L (median 17.7 ng/L); however, the ∑PFOS concentrations in the sample from Kiribati exceeded this range by an order of magnitude (218 ng/L). For comparison, in the period 2008–2011, ∑PFOS concentrations were in the range 8.2–27 ng/L (median 13.8 ng/L; $N = 5$); however, different countries participated in these two periods. From the 16 countries of the Asia-Pacific region, data covering two periods were available only for Fiji showing a reduction of 76% from 2011 (27 ng/L) to 2019 (<6.4 ng/L) (Fig. 6).

Figure 7 illustrates the temporal tendencies observed in the two countries with multiple participation using the Theil–Sen method (comprising the individual pooled samples and assuming exponential trends, see subsection 2.2). The limited number of samples did not allow to determine a statistically significant decrease for Fiji ($p \sim 1.000$) (Table 6). The short period between the two samplings in Niue (2017 and 2019) should not be used for calculation of temporal trends over a 10 year-period.

Temporal trends of ∑PFOS concentrations in the Asia-Pacific region were assessed by the Theil–Sen method combining all 18 national pools from 16 countries (Fig. 8). As explained above, regional trends cannot be derived based on countries with multiple participation due to lack of a sufficient number of countries. Overall rates for changes over time were computed by the Theil–Sen method using all samples and showed a statistically not significant increase of 25% over 10 years

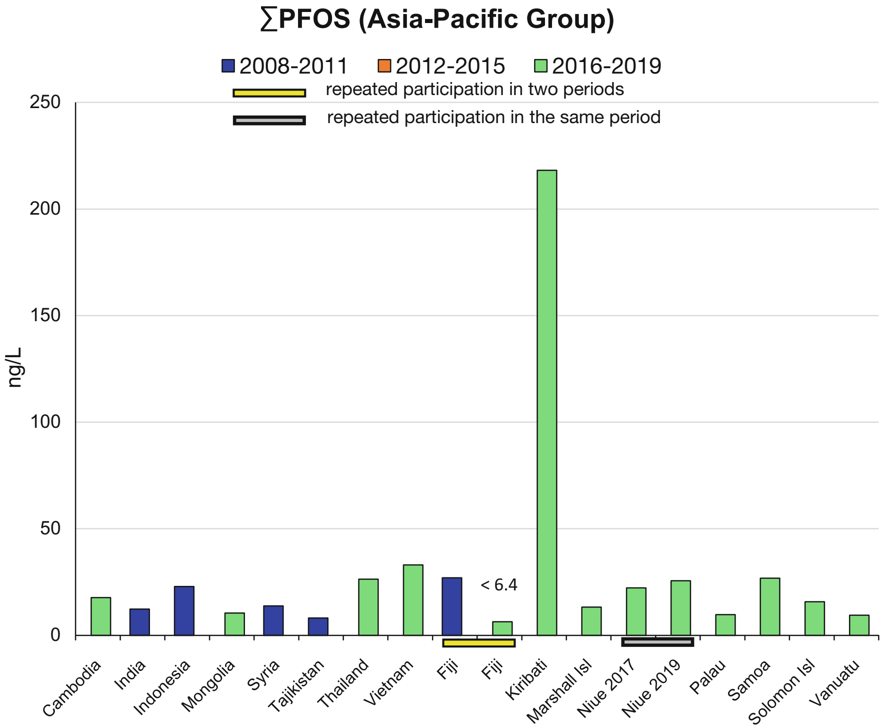

Fig. 6 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from Asia-Pacific countries over time (period 2008–2011 in blue, period 2012–2015 in orange [however, no samples in this period], and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

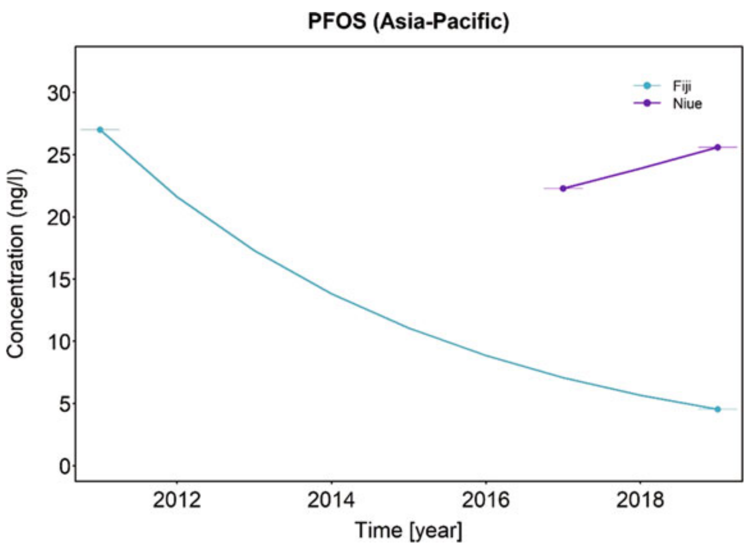

Fig. 7 Temporal tendencies of ∑PFOS concentrations (ng ∑PFOS/L) for countries of the Asia-Pacific Group with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 6 Overall decrease (%) of ∑PFOS concentrations per 1 year and 10 years in human milk from Fiji (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Fiji	20.0	89.3	1.000

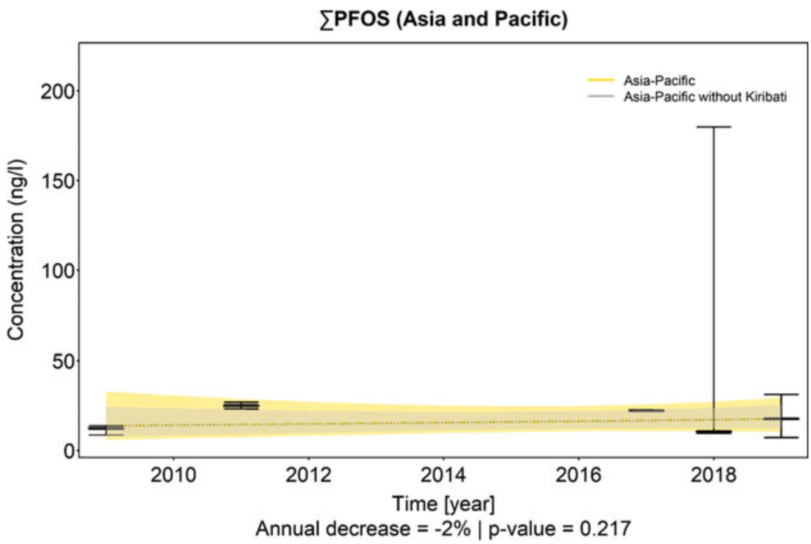

Fig. 8 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in countries of the Asia-Pacific group (18 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

Table 7 Overall decrease (%) of PFOS concentrations in the Asia-Pacific Group computed using all samples (n.a. = not applicable). Negative decreases are to be read as increase

Asia-Pacific Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All samples	16	−2.3	n.a.	−24.9	n.a.	0.217

(Table 7). Only two countries (Fiji and Niue) provided more than one value, however, the short period between the two samplings in Niue (2017 and 2019) should not be used for calculation of temporal trends over a 10 year-period. Therefore, in the Asia-Pacific Group the median method cannot be applied.

3.1.4 Group of Latin American and Caribbean Countries (GRULAC)

In 21 national pools from 12 GRULAC countries, ∑PFOS concentrations between 2008 and 2019 were in a range < 6.4–67.3 ng/L (maximum in one of three samples from Brazil, 2012), with downward tendencies in all 6 countries with repeated participation in different periods. In the five countries with participation in the 2008–2011 and 2016–2019 periods (Antigua-Barbuda, Jamaica, Mexico, Peru, and Uruguay), ∑PFOS concentrations decreased by 54% as median of the observed decreases between the first and second submission (without normalization, e.g. to a 10-year period). In Haiti as country with participation in two subsequent periods (2008–2011 and 2012–2015), ∑PFOS concentrations decreased by 45% from 2011 to 2015 (Fig. 9). The two samples from Chile were submitted in the same period. With regard to the short period between the collection of samples (2008 and 2011) and to observations discussed above (related to Table 1), it does not seem to be appropriate to derive temporal tendencies from these two samples.

Overall, levels decreased from 32.7 ng/L as median (range 9.6–58.6 ng/L; $N = 8$) in 2008–2011 to 12.2 ng/L as median (range < 6.4–41.7 ng/L; $N = 9$) in 2016–2019, with a higher median of 37.9 ng/L in the 2012–2015 period (range 21.7 to 67.3 ng/L;

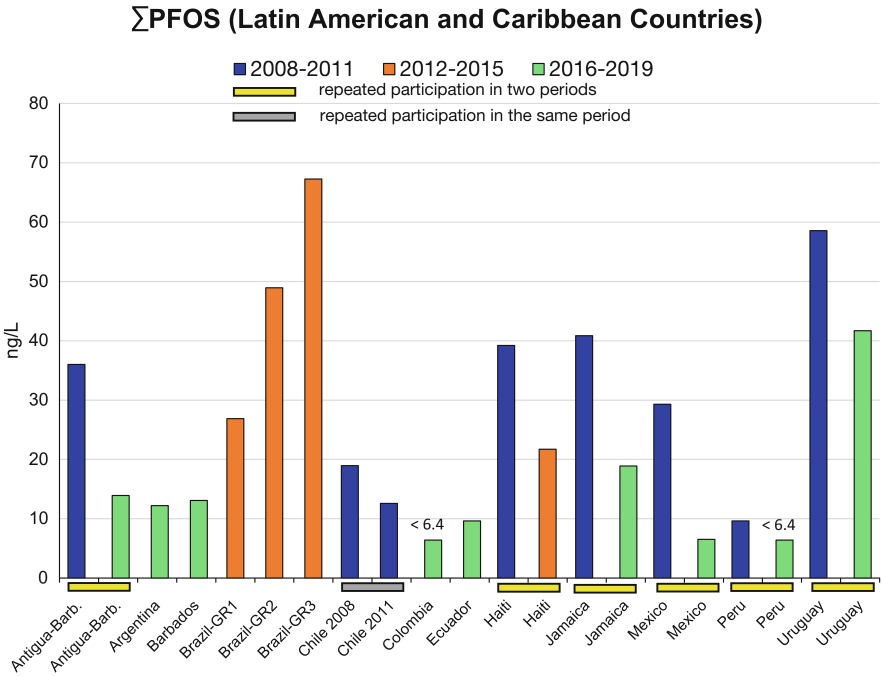

Fig. 9 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from countries of the Group of Latin America and the Caribbean over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods are marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

$N = 4$). The observed increase from 2008–2011 to 2012–2015 demonstrates the limitations of this first *general* estimation of temporal trends, if different countries are included in different periods: The four data for concentrations in 2012–2015 are dominated by three results from one country (Brazil). Whereas in Haiti a downward tendency was observed from 2011 to 2015, temporal tendencies cannot be derived for Brazil, as only data for three samples of 2012 are available. Thus, this UN region is an example that in contrast to a general estimation of time trends from all participating countries for a region, the assessment of temporal trends from countries with repeated participation is a more precise approach because levels among countries are often highly variable and single sample contributions have a significant effect on the regional results in a certain period.

Figure 10 illustrates the temporal tendencies in countries with multiple participation using the Theil–Sen method (comprising all individual pooled samples and assuming exponential trends, see subsection 2.2). The overall decreases per 1 year and 10 years are given in Table 8. The limited number of samples did not allow to determine statistically a significant decrease for countries ($p \sim 1.000$). As median of the six countries with repeated participation in different periods, the levels of ∑PFOS in all Latin American and Caribbean countries decreased within a 10-year period by 64%. This is in line with the statistically significant ($p < 0.001$) decrease over 10 years of 66% for this UN region derived by the Theil–Sen method (see Table 9 in the following).

Time trends of ∑PFOS concentrations in this region derived by the Theil–Sen method are illustrated by Fig. 11 for all 21 national pools from 12 countries and by Fig. 12 for 12 national pools from 6 countries with multiple participation. Statistically significant decreases over 10 years of 70% and 66%, respectively, were

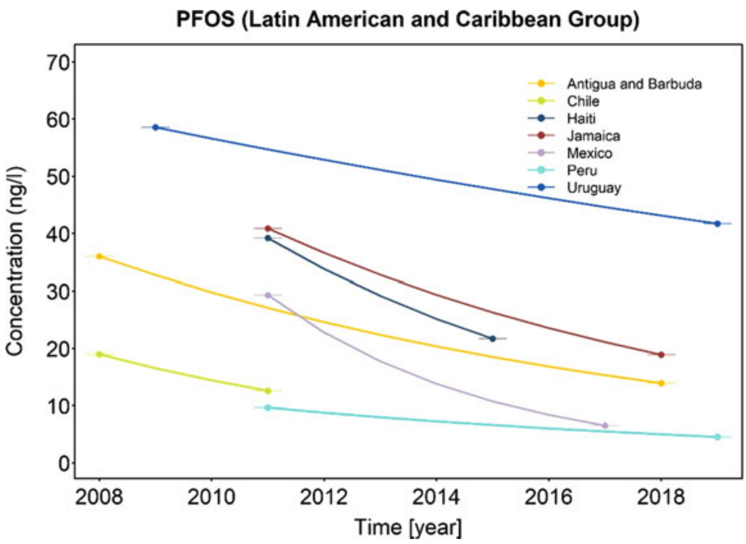

Fig. 10 Temporal tendencies of ∑PFOS concentrations (ng ∑PFOS/L) for countries of the Group of Latin America and the Caribbean with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 8 Overall decrease (%) of ∑PFOS concentrations per 1 year and 10 years in countries of the Group of Latin America and the Caribbean (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Antigua-Barb.	9.1	61.3	1.000
Haiti	13.7	77.2	1.000
Jamaica	10.4	66.8	1.000
Mexico	22.2	91.8	1.000
Peru	9.0	61.2	1.000
Uruguay	3.3	28.8	1.000
Median	9.7	64.2	

Table 9 Overall decrease (%) of PFOS concentrations in countries of the Group of Latin America and the Caribbean computed (i) using all samples and (ii) using samples from countries with repeated participation in different periods

Latin America and Caribbean	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	12	11.3	10.4	69.8	64.2	<0.001
Repeatedly	6	10.0	9.8	66.0	64.2	0.003

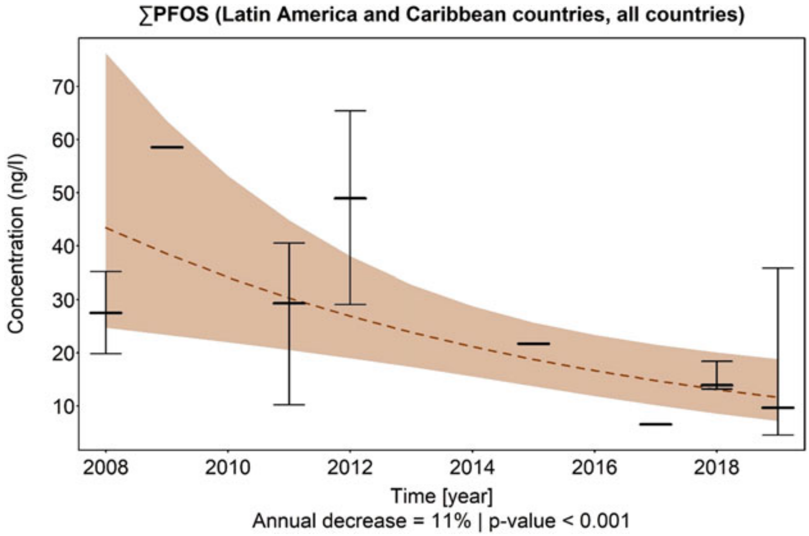

Fig. 11 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in countries of the Group of Latin America and the Caribbean (21 national pools, 12 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

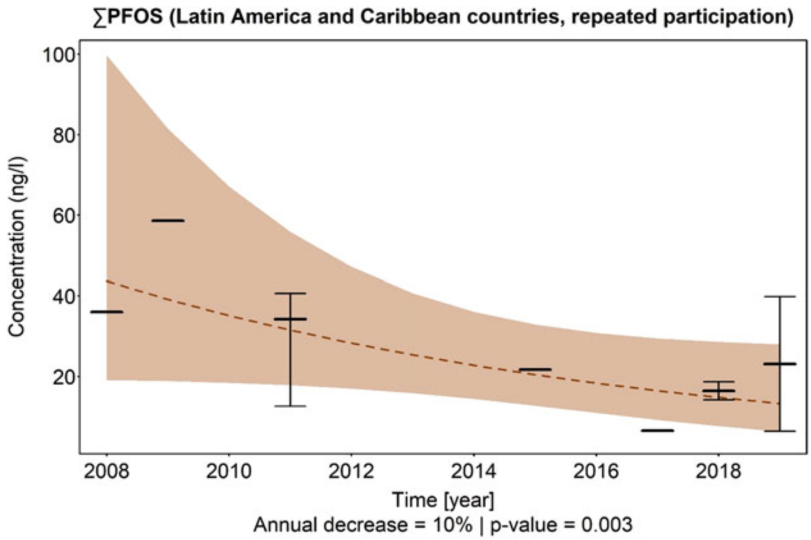

Fig. 12 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in countries of the Group of Latin America and the Caribbean with repeated participation in different periods (12 national pools, 6 countries)

achieved computed (1) using all samples and (2) using samples from countries with repeated participation (Table 9).

3.1.5 Eastern European Group

In 12 national pools from 8 Eastern European countries, ∑PFOS concentrations between 2008 and 2019 were in the range 12.6–85.5 ng/L, with constant concentrations in one country with multiple participation (Moldova, 2009 and 2015) and downward tendencies in 3 countries. In the Czech Republic, ∑PFOS concentrations decreased by 58% from 2014 to 2019, in Georgia by 39% from 2009 to 2014, and in Lithuania by 47% from 2009 to 2015 (calculated as observed decrease rates between the first and second submission [without normalization, e.g. to a 10-year period]) (Fig. 13).

Based on a quite low number of countries, overall levels decreased from 35.0 ng/L as median ($N = 3$; range 32.2–64.6 ng/L) in 2008–2011 to 13.9 ng/L as median ($N = 2$; range 12.6–15.3 ng/L) in 2016–2019, with a slightly higher median of 36.0 ng/L and a higher maximum concentration ($N = 7$; range 18.5 to 85.8 ng/L) in the 2012–2015 period. As discussed above for the Latin American and Caribbean countries, again these obviously (slightly) increasing tendencies from 2008–2011 to 2012–2015 demonstrate the limitations of this first *general* estimation of temporal trends, if different countries are included in different periods: Three countries (Bulgaria, Croatia, and Romania) are included with single participation only in the 2012–2015 period and ∑PFOS concentrations between 29.4 and 85.8 ng/L (mean:

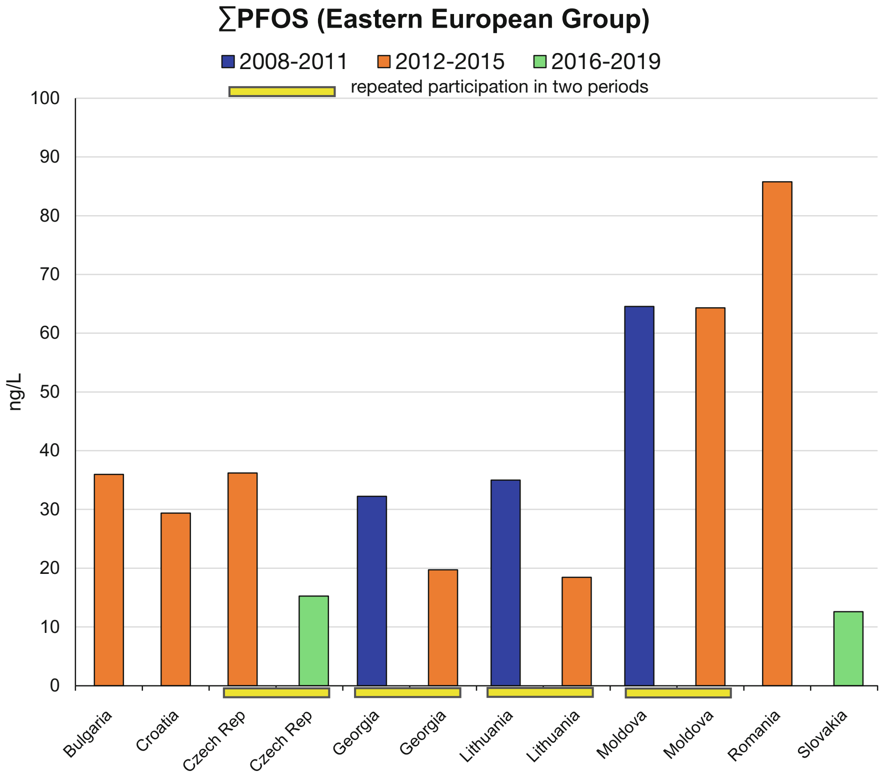

Fig. 13 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from countries of the Eastern European Group over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

50.4 ng/L). This range is higher than for the four countries with multiple participation in the 2012–2015 period (range 18.5–64.3 ng/L, mean: 34.7 ng/L).

Thus, also this UN region is an example that levels among countries are often highly variable and single contributions might have a significant effect on the regional results in a certain period, whereas temporal trends can be derived more precisely on countries with multiple participation.

Figure 14 illustrates the temporal tendencies in the four countries with multiple participation using the Theil–Sen method (comprising all individual pooled samples and assuming exponential trends, see subsection 2.2). Overall decreases per 1 year and 10 years are given in Table 10. The limited number of samples did not allow to determine statistically significant decreases for countries ($p \sim 1.000$). As median of the four countries, the levels of ∑PFOS in these Eastern European countries decreased within a 10-year period by 64%. This is in line with the statistically significant ($p < 0.001$) decrease over 10 years of 56% for this UN region derived from countries with multiple participation and calculated by the Theil–Sen method (see Table 11 in the following).

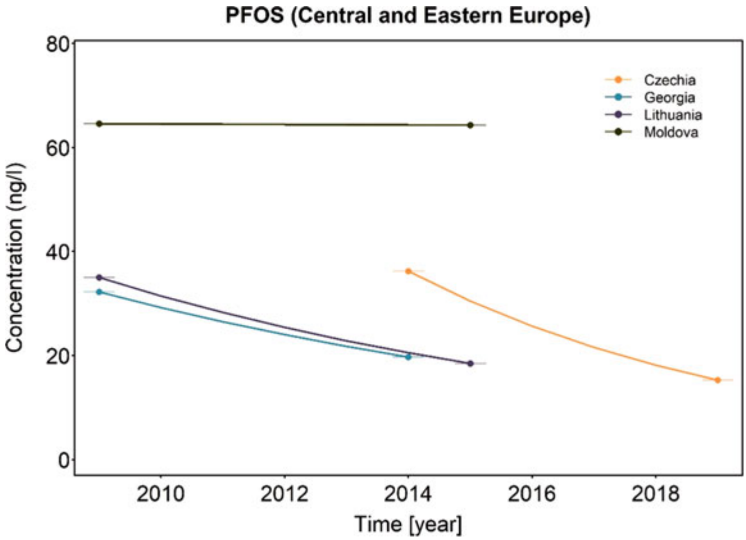

Fig. 14 Temporal tendencies of ∑PFOS concentrations (ng ∑PFOS/L) for countries of the Eastern European Group with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 10 Overall decrease (%) of ∑PFOS concentrations per 1 year and 10 years in countries of the Eastern European Group (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Czechia	15.9	82.2	1.000
Georgia	9.4	62.6	1.000
Lithuania	10.1	65.6	1.000
Moldova	0.1	0.7	1.000
Median	9.7	64.1	

Table 11 Overall decrease (%) of PFOS concentrations in the Eastern European Group (EEG) computed (1) using all samples and (2) using samples from countries with repeated participation

Eastern European Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	8	9.4	9.7	62.6	64.1	0.002
Repeatedly	4	8	9.7	56.4	64.1	0.052

Statistically significant time trends can be derived in this UN regional group by pooling of data from countries. The time trends of ∑PFOS concentrations derived by the Theil–Sen method are illustrated by Fig. 15 for all 12 national pools from 8 countries and by Fig. 16 for the 8 national pools from 4 countries with multiple

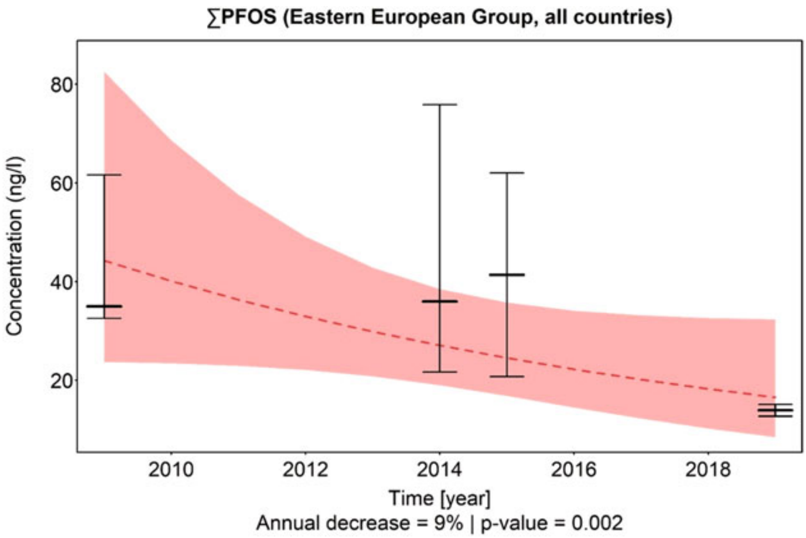

Fig. 15 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in countries of the Eastern European Group (12 national pools, 8 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

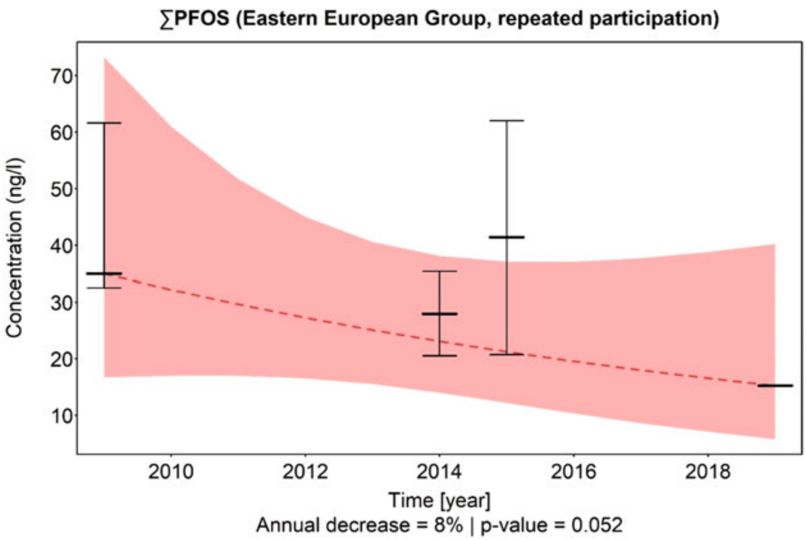

Fig. 16 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in countries of the Eastern European Group with repeated participation (8 national pools, 4 countries)

participation. Overall decreases over 10 years for PFOS concentrations of 62.6% and 56.4%, respectively, were calculated (1) using all samples and (2) using samples from countries with repeated participation (Table 11).

Although the significance of both trend estimates is relatively high (p-value 0.002 and 0.052, respectively), changes in trend slopes may be expected in the period 2011–2015 after listing of PFOS in 2009 in 2009 in Annex B of the Convention. Using the Hites method of break point search (Hites 2019) on all countries in the Eastern European Group, a break point was found for PFOS, although, the result was non-significant.

3.1.6 Western European and Others Group (WEOG)

In 8 national pools from 7 Western European countries, ∑PFOS concentrations between 2014 and 2019 were in the range 13.9–52.9 ng/L. There were no countries with multiple participation (Fig. 17). Statistically significant time trends can be derived in this UN regional group by pooling of all data from countries. The time trends derived by the Theil–Sen method are illustrated by Fig. 18. An overall decrease over 10 years of 79.1% was computed using all samples (Table 12).

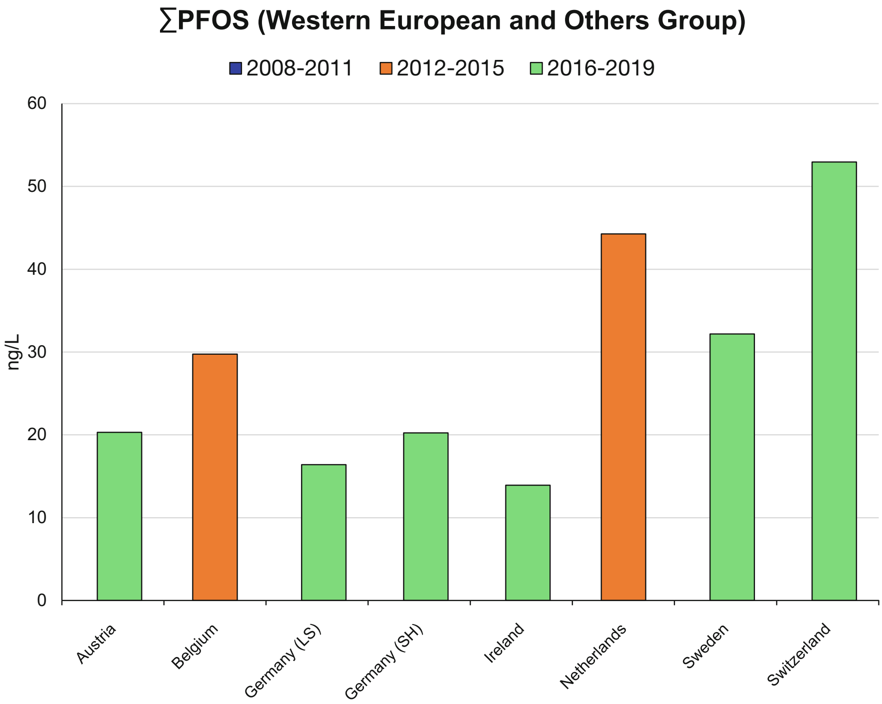

Fig. 17 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from Western European countries over time (period 2008–2011 in blue [however, no samples in this period], period 2012–2015 in orange, and period 2016–2019 in green)

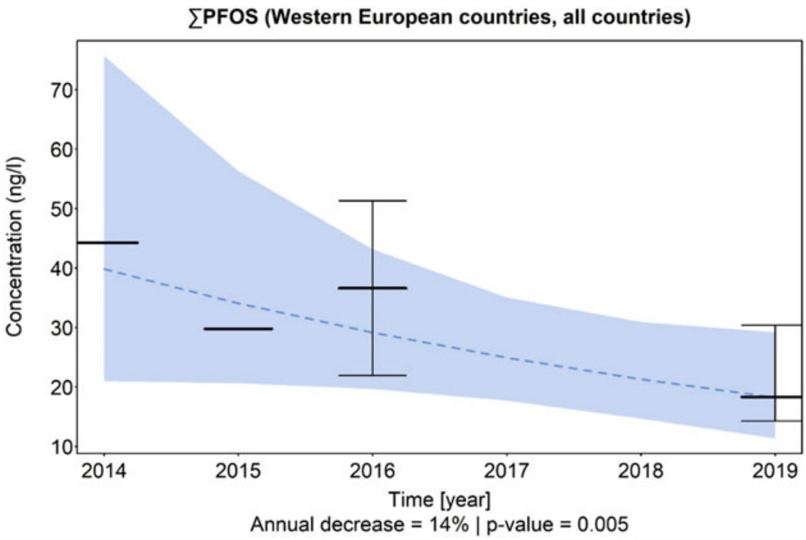

Fig. 18 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) in Western European countries (8 national pools, 7 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

Table 12 Overall decrease (%) of PFOS concentrations in Western European countries (computed using all samples)

Western Europe and Others Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All samples	7	14.5	–	79.1	–	0.005

3.1.7 Worldwide

The time trends of ∑PFOS concentrations derived by the Theil–Sen method from all 86 national pools from 59 countries are illustrated by Fig. 19 and for the 48 national pools from 24 countries with multiple participation by Fig. 20. The results from both approaches were comparable: Statistically significant decreases over 10 years for PFOS concentrations of 48.3% and 52.2%, respectively, were computed (1) using all samples and (2) using samples from countries with repeated participation (Table 13).

As described in subsection 3.1.1 above, the summarizing descriptive parameters seem to indicate an increase of the PFOS concentrations from 2008–2011 to 2012–2015 with a subsequent decrease to 2016–2019. The impact of countries

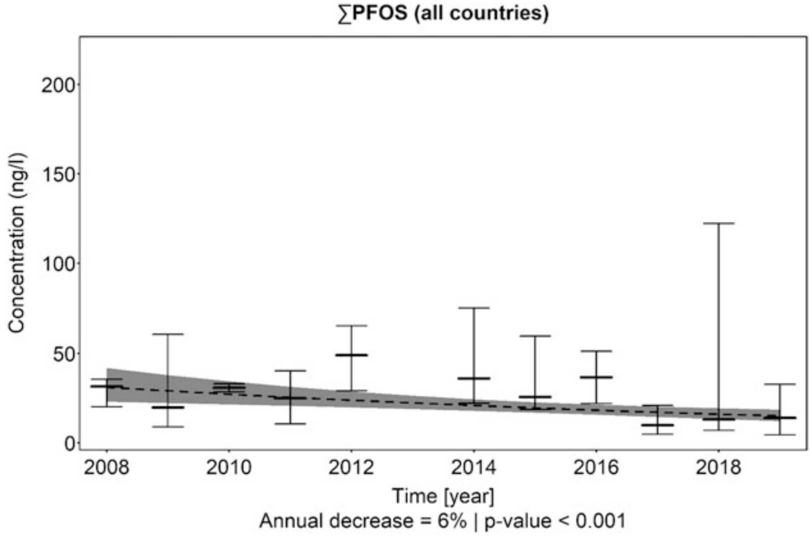

Fig. 19 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) using data from all countries (86 national pools, 59 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

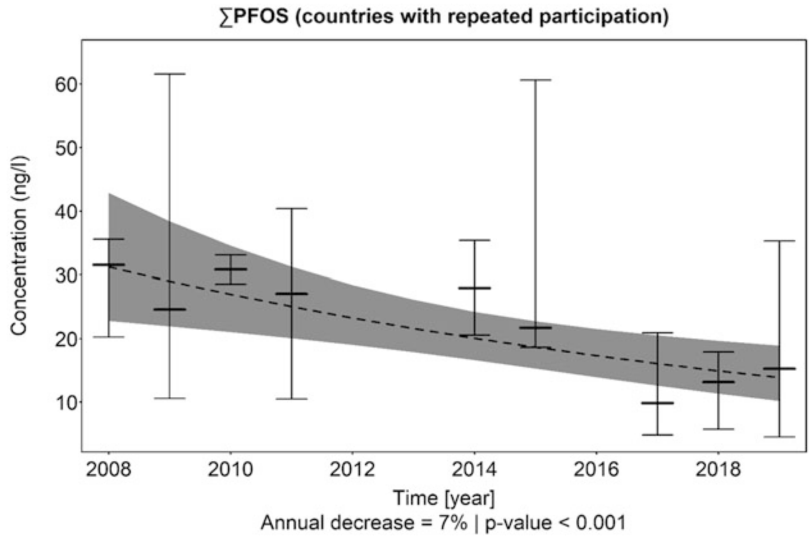

Fig. 20 Theil–Sen exponential trends of ∑PFOS concentrations in human milk (ng ∑PFOS/L) using data from countries with repeated participation (48 national pools, 24 countries)

Table 13 Overall decrease (%) of ∑PFOS concentrations worldwide computed (1) using all samples and (2) using samples from countries with repeated participation

Worldwide	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	59	6.4	9.4	48.3	62.6	<0.001
Repeatedly	24	7.1	7.2	52.2	52.6	<0.001

Table 14 Comparison of median, minimum, and maximum ∑PFOS concentrations (ng/L) in samples from countries participating in the 2012–2015 period: (1) countries with participation also in the 2008–2011 or 2016–2019 period; (2) countries participating only in the 2012–2015 period

	Countries with multiple participation	Countries with single participation only in 2012–2015 period
N	7	8
Median	21.7	40.1
Min	18.5	26.9
Max	64.3	85.8

participating in the 2012–2015 period on the obvious increase after listing in 2009 is assessed in the following in more details.

The lowest number of samples was available for this period (in total: 15 national pools). Seven samples came from countries with repeated participation (Côte d'Ivoire, Czech Republic, Georgia, Haiti, Lithuania, Moldova, Niger) and eight samples from countries with single participation (three samples from Brazil; Belgium, Bulgaria, Croatia, Netherlands, Romania). The range found in this period in all 15 samples (median 36.0 ng/L; range 18.5–85.8 ng/L) was clearly influenced by countries with single participation only in the 2012–2015 period: Minimum, median, and maximum concentrations were higher than in countries with multiple participation (Table 14).

The country-specific temporal trends for the seven countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019 were discussed above in the corresponding UN Regional Groups. Five of these countries had decreasing tendencies, one increasing and one quite constant concentration (Figs. 21 and 22). A decrease of 51% over 10 years was calculated for these seven countries, with a range between 6% and 82% for decrease rates over 10 years in Czechia, Georgia, Haiti, Lithuania, and Niger, quite constant concentrations in Moldova and an increase of 90% over 10 years in Côte d'Ivoire. As a result, the participation of different countries in the three rounds and the elevated contribution of samples from countries participating only in the 2012–2015 round explains the

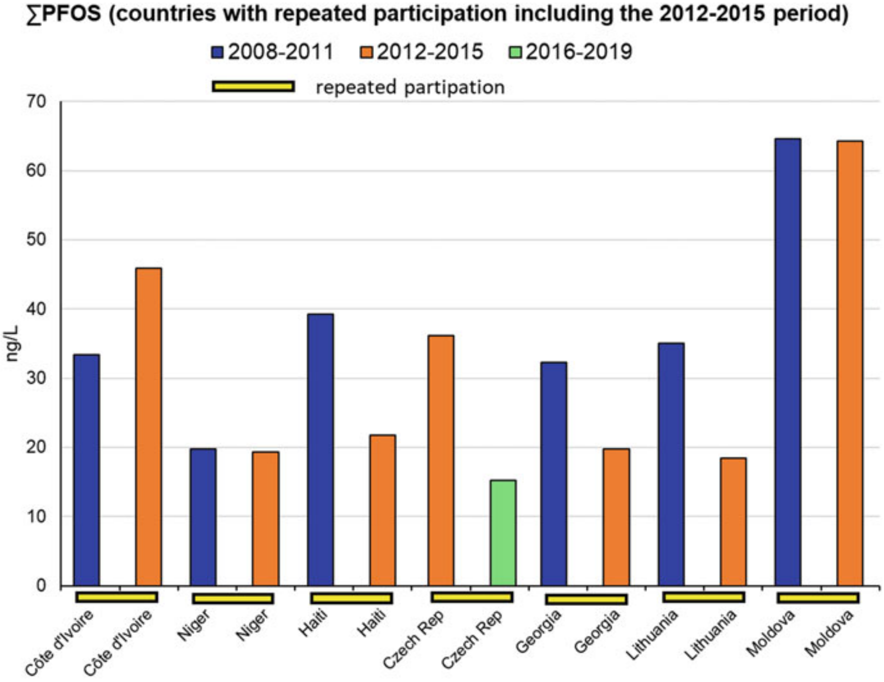

Fig. 21 Overview of the development of ∑PFOS concentrations in human milk (ng ∑PFOS/L) from seven countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019 (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods are marked by yellow rectangles)

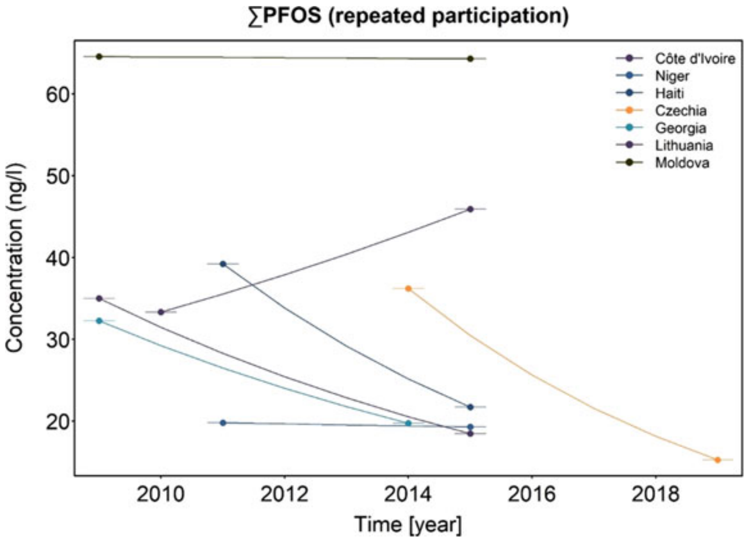

Fig. 22 Temporal tendencies of ∑PFOS concentrations (ng ∑PFOS/L) for seven countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019 using the Theil–Sen method

fluctuation of the descriptive parameters (e.g. median) between the three periods with an obvious maximum in the 2012–2015 period.

3.2 PFOA

PFOA was listed in Annex A of the Convention in 2019 (UNEP 2019b).

3.2.1 Estimation of Temporal Trends by Differentiation into Three Periods between 2008 and 2019

The median of the PFOA concentrations of 86 national pools from 59 countries increased from 21.5 ng/L in the 2008–2011 period ($N = 27$) to 32.1 ng/L in the 2012–2015 period ($N = 15$) and then decreased to 16.3 ng/L in the 2016–2019 period ($N = 44$). The highest maximum level (65.3 ng/L) was found in the 2008–2011 period (Fig. 23). However, as described above for PFOS, it has to be checked also for PFOA whether these fluctuations of the median are likely due to the result of participation of different countries in different rounds of the WHO/UNEP-coordinated exposure studies.

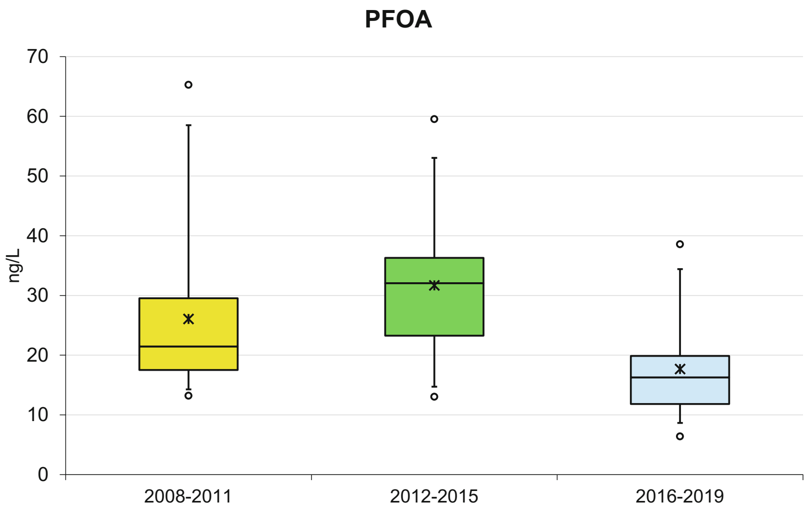

Fig. 23 Range of PFOA concentrations (ng/L) in three periods ($N = 27$ in 2008–2011; $N = 15$ in 2012–2015; $N = 44$ in 2016–2019) [box plot; minimum and maximum: as circles; fifth and 95th percentile: as whiskers; lower (25–50%) and upper (50–75%) quartiles, separated by the line for the median: as box; mean: as asterisk]

3.2.2 African Group

In the 16 African countries, PFOA concentrations between 2008 and 2019 were in a range 6.4–65.3 ng/L. In two of the 11 countries with repeated participation, PFOA concentrations remained quite constant (Côte d'Ivoire; Nigeria). In 9 countries, downward tendencies were observed, with a decrease by 31% as median of the observed decreases between the first and second submission (without normalization, e.g. to a 10-year period) and the highest decrease by 87% (Uganda, from 2008 to 2018) (Fig. 24).

Overall, levels decreased from 18.0 ng/L as median (range 14.0–65.3 ng/L; $N = 11$) in 2008–2011 to 12.8 ng/L as median (range 6.4–18.6 ng/L; $N = 14$) in 2016–2019.

Figure 25 illustrates the temporal tendencies in 11 countries with multiple participation using the Theil–Sen method (comprising all individual pooled samples and assuming exponential trends, see subsection 2.2). Overall decreases per 1 year and 10 years are given in Table 15. The limited number of samples did not allow to determine statistically significant decreases for countries ($p \sim 1.000$). As median of these countries, the levels of PFOA in African countries decreased within a 10-year period by 37%. This is in line with the statistically significant ($p < 0.001$) decrease over 10 years of 32.5% for this UN region derived by countries with multiple

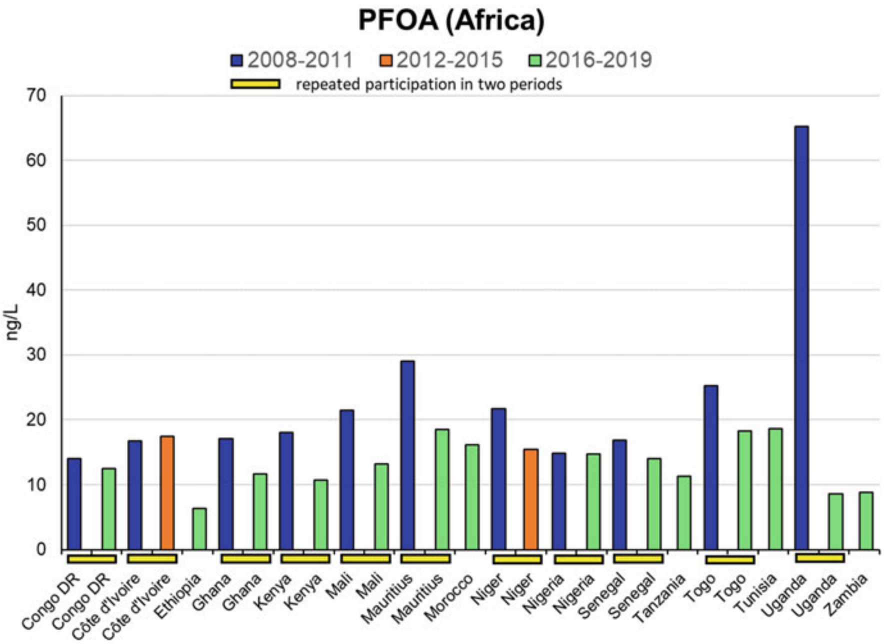

Fig. 24 Overview of the development of PFOA concentrations in human milk (ng/L) from African countries over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

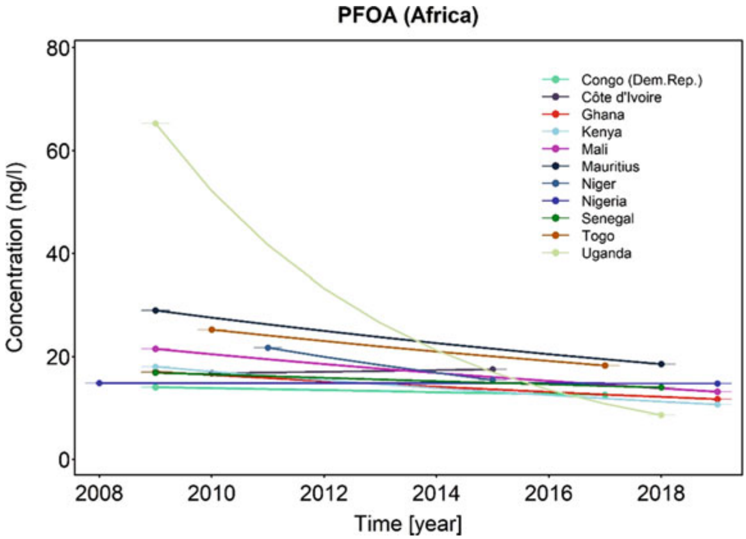

Fig. 25 Temporal tendencies of PFOA concentrations (ng/L) for African countries with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 15 Overall decrease (%) of PFOA concentrations per 1 year and 10 years in African countries (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Congo (DR)	1.4	13.4	1.000
Côte d'Ivoire	−0.9	−9.4	1.000
Ghana	3.7	31.5	1.000
Kenya	5.1	40.8	1.000
Mali	4.8	38.7	1.000
Mauritius	4.9	39.2	1.000
Niger	8.1	57.3	1.000
Nigeria	0.0	0.4	1.000
Senegal	2.1	18.8	1.000
Togo	4.5	37.0	1.000
Uganda	20.1	89.5	1.000
Median	4.5	37.0	

participation and calculated by the Theil–Sen method (see Table 16 in the following).

Statistically significant time trends can be derived in the African Regional Group by pooling of data from countries. The time trends of PFOA concentrations derived by the Theil–Sen method are illustrated by Fig. 26 for all 27 national pools from

Table 16 Overall decrease (%) of PFOA concentrations in the African Group computed (1) using all samples and (2) using samples from countries with repeated participation

African Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	16	4.5	4.5	36.9	37.0	<0.001
Repeatedly	11	3.9	4.5	32.5	37.0	<0.001

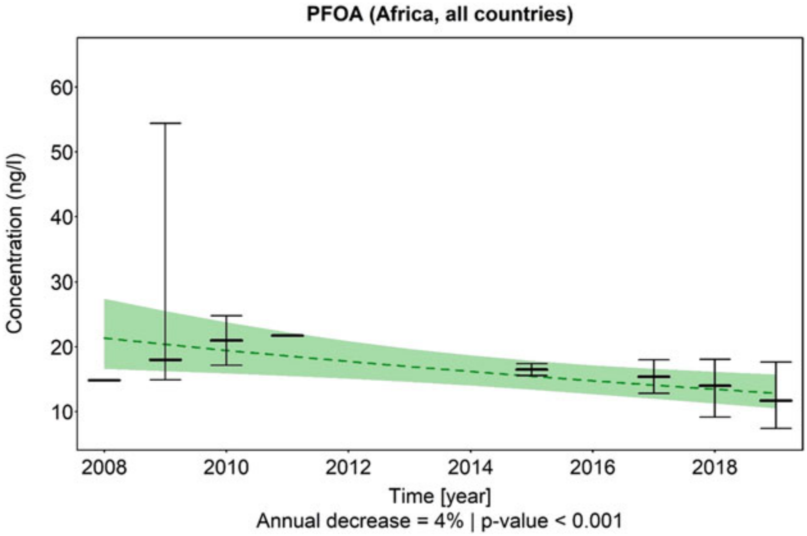

Fig. 26 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) in all African countries (27 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

16 countries and by Fig. 27 for 22 national pools from 11 countries with multiple participation. Overall decreases over 10 years for PFOA concentrations in the African Group of 37% and 33%, respectively, were achieved computed (i) using all samples and (ii) using samples from countries with repeated participation (Table 16).

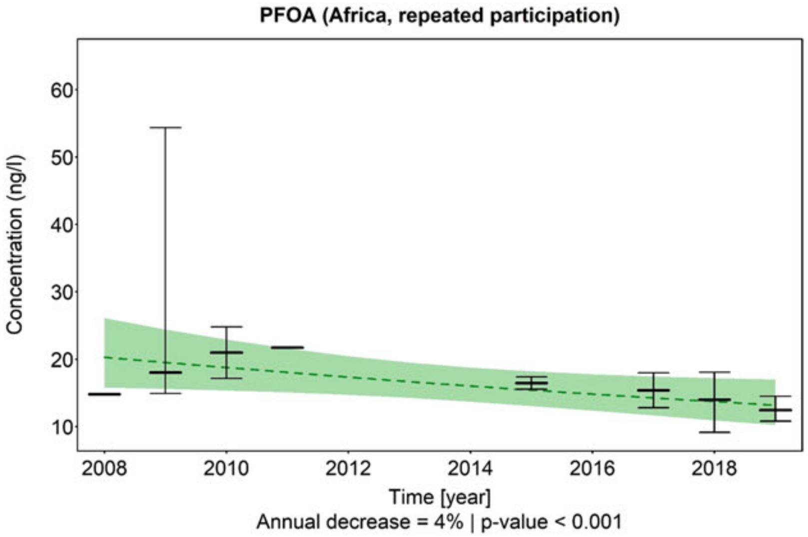

Fig. 27 Theil–Sen exponential trends of PFOA in human milk (ng/L) in African countries with repeated participation (22 national pools, 11 countries)

3.2.3 Asia-Pacific Group

From the 16 countries of the Asia-Pacific region, data covering two periods were available only for Fiji showing a reduction of 70% from 2011 (36 ng/L) to 2019 (10.5 ng/L). In the most recent period (2016–2019), all countries had PFOA concentrations in a range between 10 and 33 ng/L (median 15.1 ng/L; $N = 13$), in comparison to a range between 13 and 36 ng/L (median 20.6 ng/L; $N = 5$) in the 2008–2011 period (Fig. 28).

Figure 29 illustrates the temporal tendencies in the two countries with multiple participation using the Theil–Sen method. The limited number of samples did not allow to determine a statistically significant decrease for Fiji ($p \sim 1.000$) (Table 17). The short period between the two samplings in Niue (2017 and 2019) should not be used for calculation of temporal trends over a 10 year-period.

Statistically significant time trends of PFOA concentrations in the Asia-Pacific region were derived by the Theil–Sen method combining all 18 national pools from 16 countries (Fig. 30). As explained above, regional trends cannot be derived based on countries with multiple participation due to lack of a sufficient number of countries. An overall decrease over 10 years of 38.6% was computed by the Theil–Sen method using all samples (Table 18).

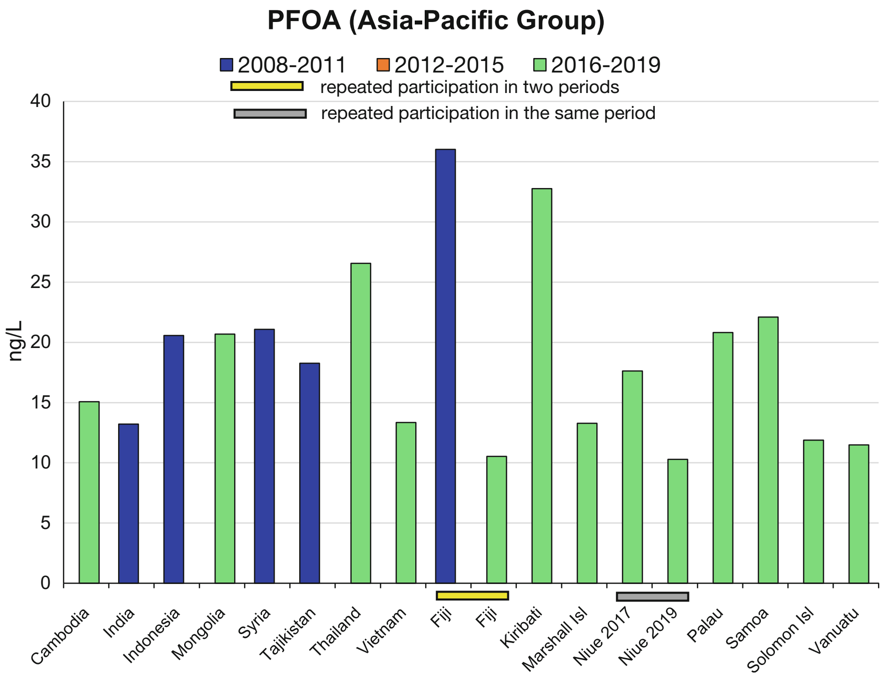

Fig. 28 Overview of the development of PFOA concentrations in human milk (ng PFOA/L) from countries of the Asia-Pacific Group over time (period 2008–2011 in blue, period 2012–2015 in orange [however, no samples in this period], and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

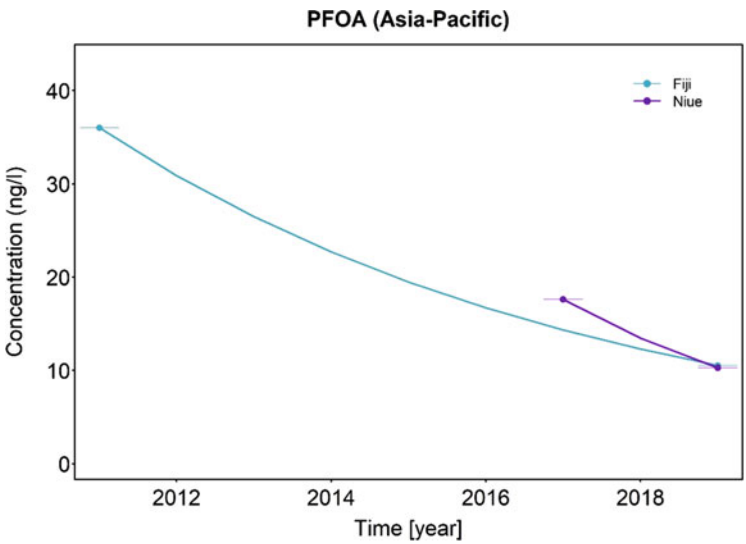

Fig. 29 Temporal tendencies of PFOA concentrations (ng/L) for countries of the Asia-Pacific Group with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 17 Overall decrease (%) of PFOA concentrations per 1 year and 10 years in Fiji (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Fiji	14.2	78.5	1.000

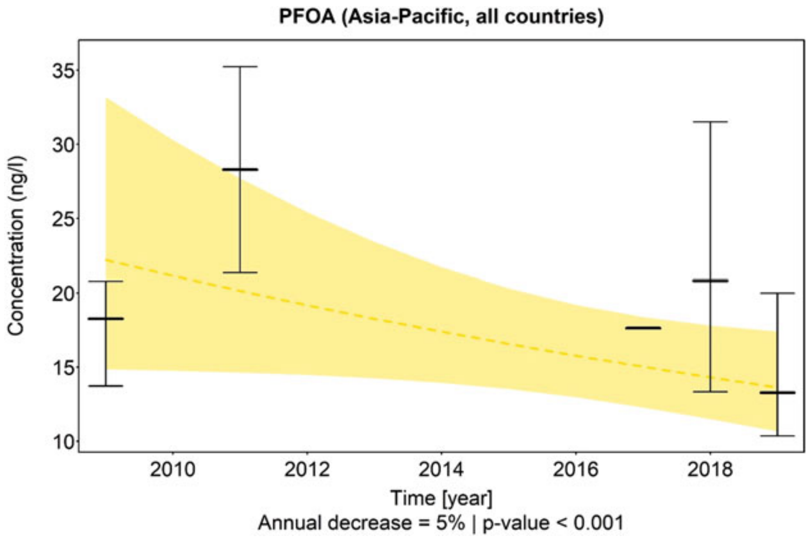

Fig. 30 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) in countries of the Asia-Pacific group (18 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

Table 18 Overall decrease (%) of PFOA concentrations in the Asia-Pacific Group computed using all samples (n.a. = not applicable)

Asia-Pacific Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All samples	16	4.8	n.a.	38.6	n.a.	<0.001

3.2.4 Group of Latin American and Caribbean Countries (GRULAC)

In the 12 GRULAC countries, PFOA concentrations between 2008 and 2019 were in the range 8.0–63.3 ng/L, with downward tendencies in 5 countries from the 2008–2011 period to the 2016–2019 period. In these 5 countries (Antigua-Barbuda, Jamaica, Mexico, Peru, and Uruguay), PFOA concentrations decreased by 47% as median of the observed decrease rates between the first and second submission (without normalization, e.g. to a 10-year period) from 2008–2011 to 2016–2019. The considerable differences in PFOA concentrations in the two samples from Chile (2008; 2011) collected in the same period are presumably caused by differences in the regional origin of these two samples, as concluded from discussion of WHO-TEQ and PCDD/PCDF patterns samples (Malisch et al. 2023d) and not the result of decreasing trends over a period of three years. In Haiti, an upward tendency was observed from 2011 to 2015 (Fig. 31).

Overall, levels decreased from 24.5 ng/L as median (range 15.6–63.3 ng/L; $N = 8$) in 2008–2011 to 16.4 ng/L as median (range 8.0–19.6 ng/L; $N = 9$) in 2016–2019, with a higher median of 29.9 ng/L in the 2012–2015 period (range 26.1 to 35.2 ng/L; $N = 4$). As explained above already for PFOS in this UN region, the inclusion of different countries in different periods limits the comparability of

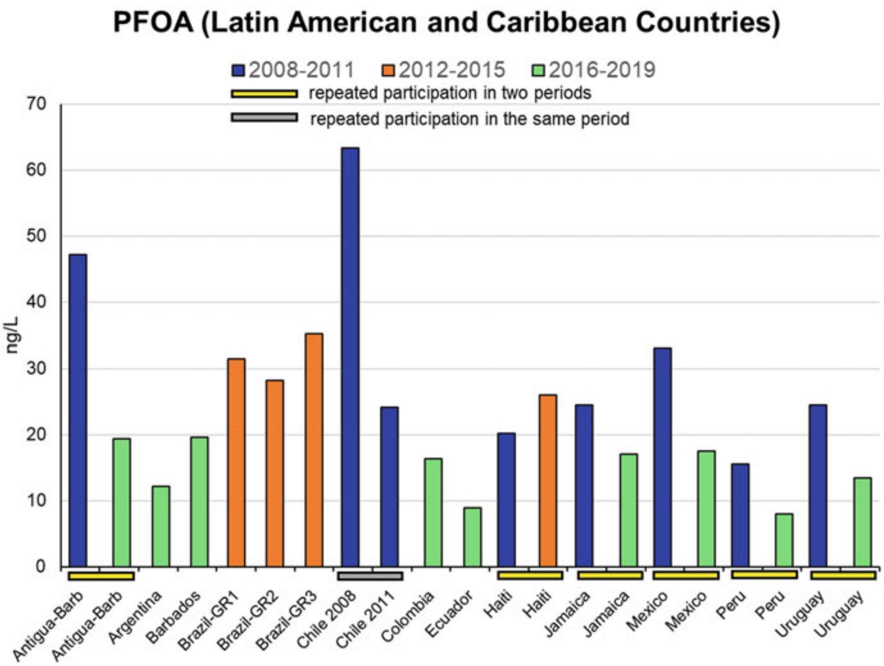

Fig. 31 Overview of the development of PFOA concentrations in human milk (ng/L) from Latin American and Caribbean countries over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

results. The four data for the 2012–2015 period were essentially influenced by the three results from one country (Brazil). However, temporal trends cannot be derived for Brazil, as only results for three samples of 2012 are available.

Figure 32 illustrates the temporal tendencies of PFOA in human milk from the seven countries with multiple participation using the Theil–Sen method. Overall decrease rates per 1 year and 10 years are given in Table 19. The limited number of samples did not allow to determine statistically significant decreases for these Latin American and Caribbean countries ($p \sim 1.000$). As median, the levels of PFOA in these countries decreased within a 10-year period by 51%. This is in line with the statistically significant ($p < 0.001$) decrease over 10 years of 65% for this UN region

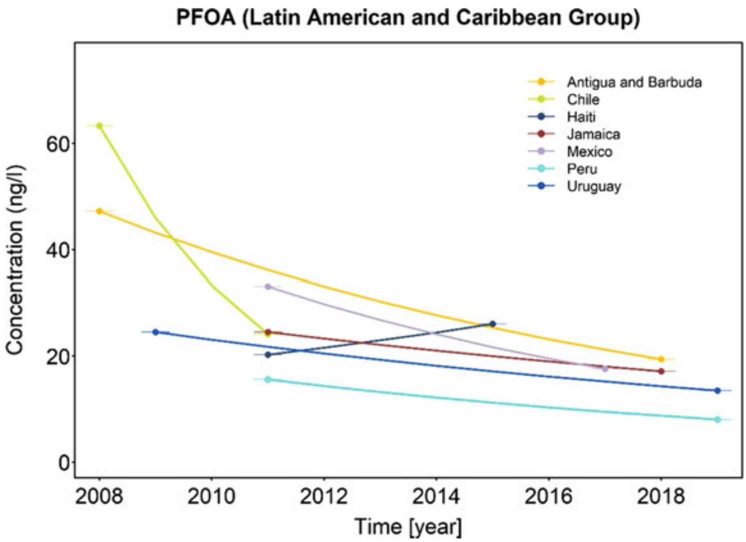

Fig. 32 Temporal tendencies of PFOA concentrations (ng/L) for Latin American and Caribbean countries with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 19 Overall decrease (%) of PFOA concentrations per 1 year and 10 years in Latin American and Caribbean countries (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Antigua-Barb.	8.5	58.9	1.000
Haiti	−6.5	−87.9	1.000
Jamaica	5.0	40.2	1.000
Mexico	10.0	65.2	1.000
Peru	7.9	56.2	1.000
Uruguay	5.8	45.1	1.000
Median	7.9	50.6	

derived by countries with multiple participation and calculated by the Theil–Sen method (see Table 20 in the following).

Time trends of PFOA concentrations in this region derived by the Theil–Sen method are illustrated by Fig. 33 for all 21 national pools from 12 countries and by Fig. 34 for 14 national pools from 7 countries with multiple participation. Statistically significant decreases over 10 years of 63% and 65%, respectively, were

Table 20 Overall decrease (%) of PFOA concentrations in Latin American and Caribbean countries computed (1) using all samples and (2) using samples from countries with repeated participation

Latin America and Caribbean	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	12	9.5	7.9	63.0	50.6	<0.001
Repeatedly	7	9.8	7.9	64.5	50.6	<0.001

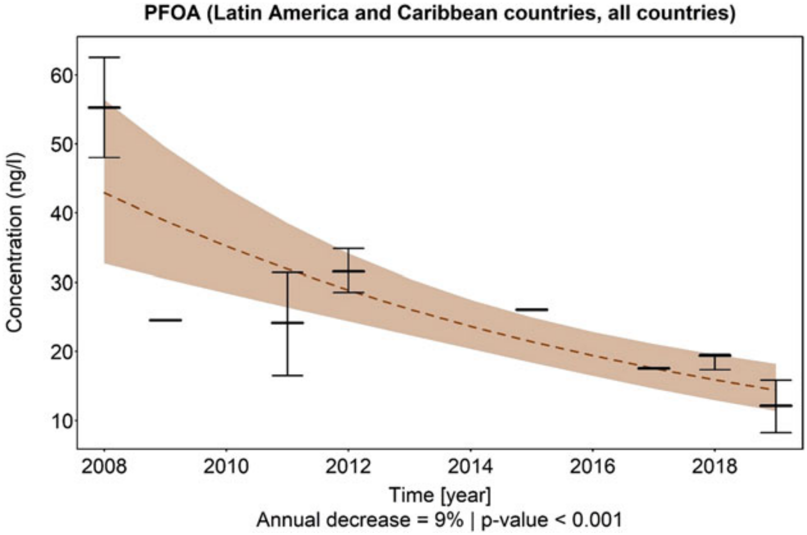

Fig. 33 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) in Latin American and Caribbean countries (21 national pools, 12 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

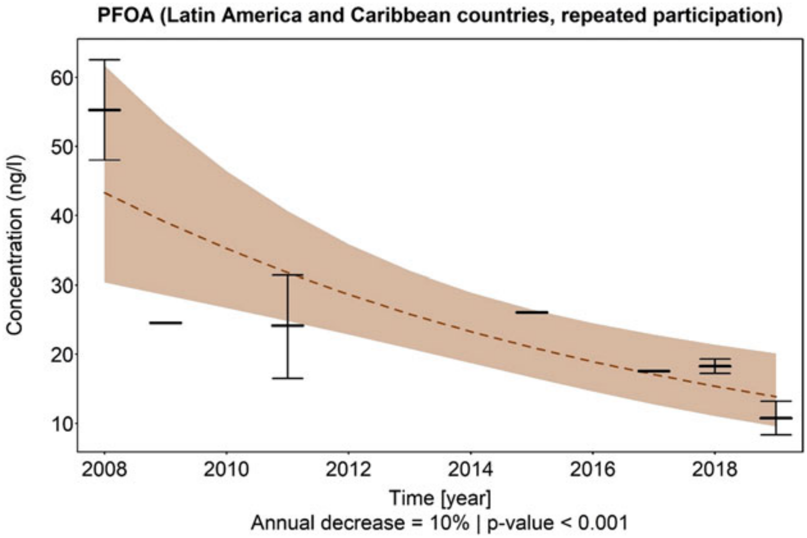

Fig. 34 Theil–Sen exponential trends of PFOA in human milk (ng/L) in Latin American and Caribbean countries with repeated participation (14 national pools, 7 countries)

achieved computed (1) using all samples and (2) using samples from countries with repeated participation (Table 20).

3.2.5 Eastern European Group

In 12 national pools from 8 Eastern European countries, PFOA concentrations between 2008 and 2019 were in the range 13.0–50.3 ng/L, with increasing concentrations from 2009 until 2015 by 12% in Lithuania, and by 67% in Moldova, with downward tendencies from 2009 until 2014 by 37% in Georgia, and with downward tendencies from 2014 to 2019 by 37% in the Czech Republic (Fig. 35). Overall, the summarizing parameters seem to indicate quite constant PFOA levels with 30.1 ng/L as median ($N = 3$; range 20.6–31.2 ng/L) in 2008–2011, 34.9 ng/L as median ($N = 7$; range 13.0–50.3 ng/L) in 2012–2015, and 27.5 ng/L as median ($N = 2$; range 25.4–29.5 ng/L) in 2016–2019.

Figure 36 illustrates the temporal tendencies in the four countries with multiple participation using the Theil–Sen method. Overall decreases per 1 year and 10 years are given in Table 21. The limited number of samples did not allow to determine statistically significant decreases for countries ($p \sim 1.000$). Whereas in the Czech Republic and Georgia an overall decrease over 10 years of 60% was found, a slight increase was observed in Lithuania and a clear increase in Moldova.

The inconsistent temporal tendencies of these four countries with multiple participation are reflected by the lack of statistical significance after pooling of data from

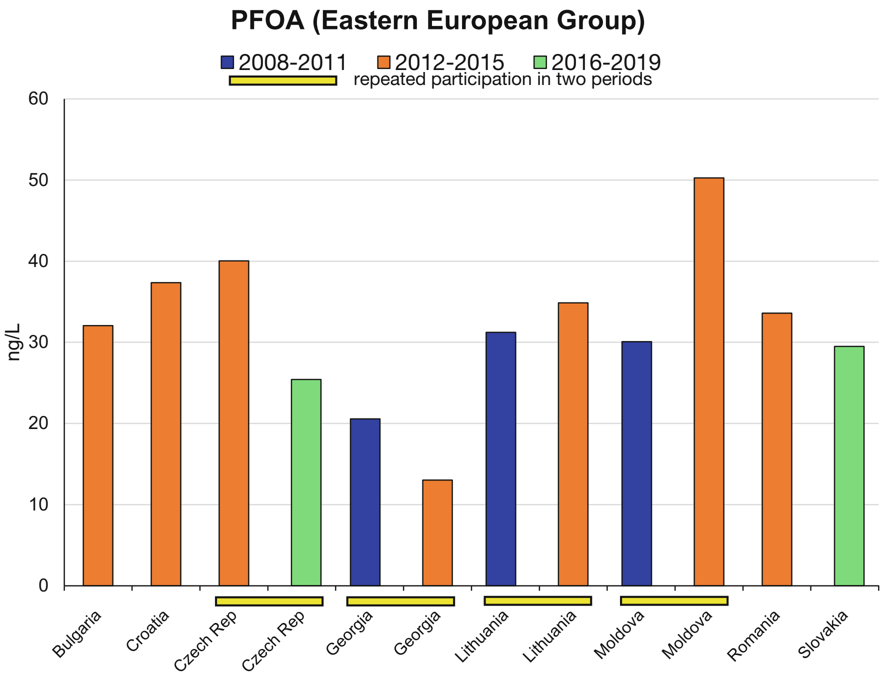

Fig. 35 Overview of the development of PFOA concentrations in human milk (ng/L) from countries of the Eastern European Group over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

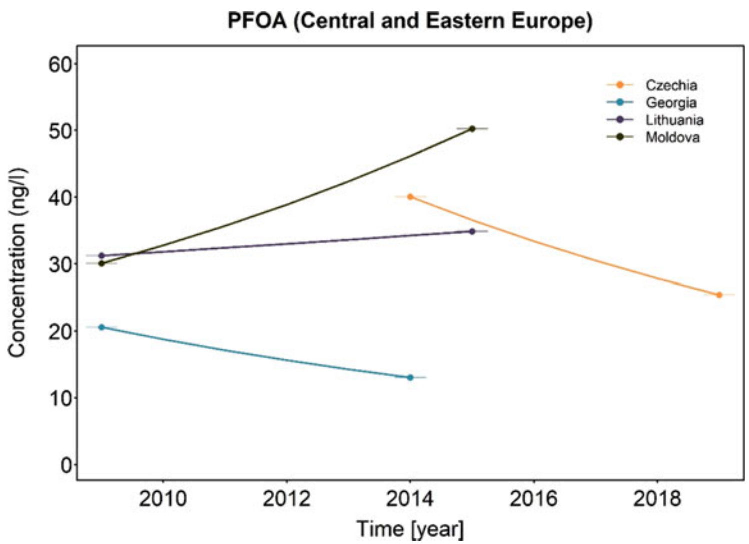

Fig. 36 Temporal tendencies of PFOA concentrations (ng/L) for countries of the Eastern European Group with repeated participation between 2008 and 2019 using the Theil–Sen method

Table 21 Overall decrease (%) of PFOA concentrations per 1 year and 10 years in human milk from countries of the Eastern European Group (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 10 years	Trend *p*-value overall
Czechia	8.7	59.7	1.000
Georgia	8.7	59.9	1.000
Lithuania	−1.9	−20.2	1.000
Moldova	−8.9	−135.3	1.000
Median	3.4	19.8	

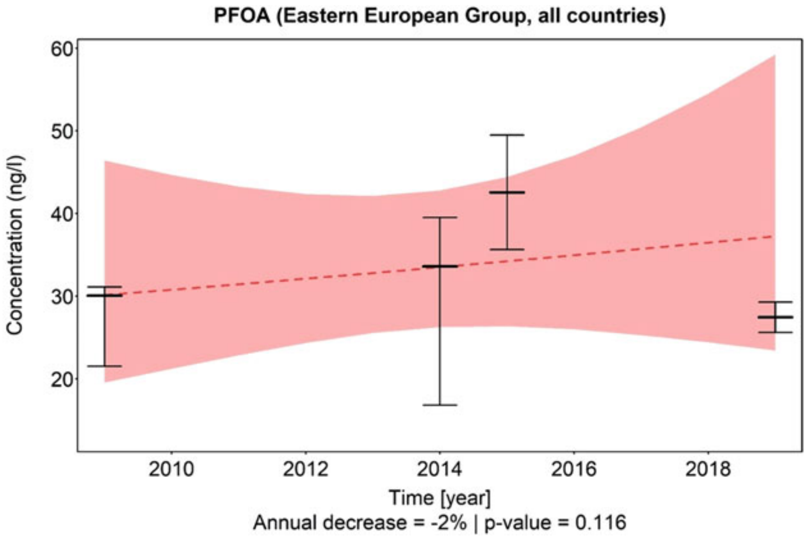

Fig. 37 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) from countries of the Eastern European Group (12 national pools, 8 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

these countries and use of the Theil–Sen method (Fig. 37 for all 12 national pools from 8 countries; Fig. 38 for the 8 national pools from 4 countries with multiple participation). Furthermore, discrepancies between the Theil–Sen method and the median method indicate difficulties in assessing statistically significant temporal trends for overall temporal tendencies over 10 years for PFOA (Table 22).

Using the Hites method of break point search (Hites 2019) on all countries in the Eastern European Group, a break point was found for PFOA, although, the result was non-significant.

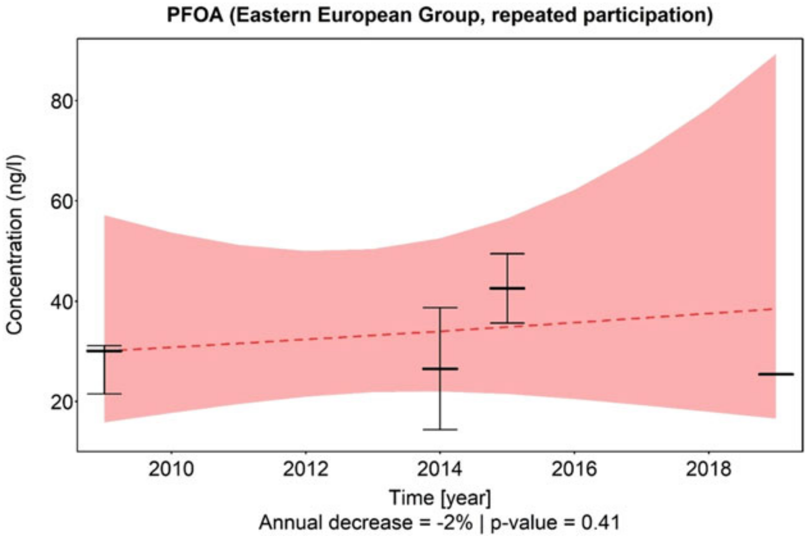

Fig. 38 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) in countries of the Eastern European Group with repeated participation (8 national pools, 4 countries)

Table 22 Overall decrease (%) of PFOA concentrations in the Eastern European Group (EEG) computed (1) using all samples and (2) using samples from countries with repeated participation. Negative decreases are to be read as increase

Eastern European Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	8	−2.1	3.6	−23.6	19.8	0.116
Repeatedly	4	−2.5	3.6	−27.9	19.8	0.41

3.2.6 Western European and Others Group (WEOG)

In eight national pools from seven Western European countries, PFOA concentrations between 2014 and 2019 were in a range 18.2–59.5 ng/L. There were no countries with multiple participation (Fig. 39). Time trends derived by the Theil–Sen method are illustrated by Fig. 40. No statistically significant time trend can be derived in this UN regional group by pooling of all data from countries. An overall decrease over 10 years of 58% was computed using all samples (Table 23).

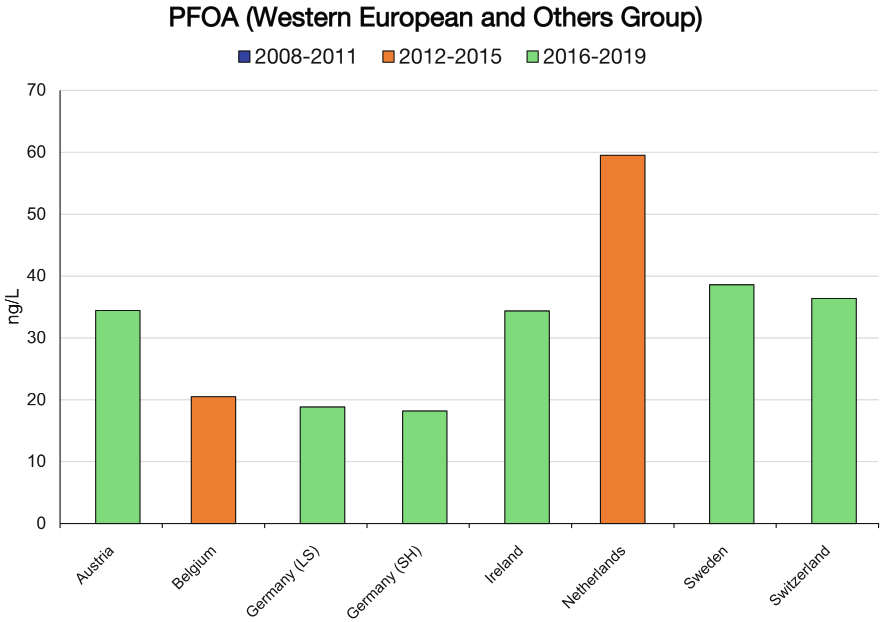

Fig. 39 Overview of the development of PFOA concentrations in human milk (ng/L) from countries of the Western European and Others Group over time (period 2008–2011 in blue [without samples in this period], period 2012–2015 in orange, and period 2016–2019 in green)

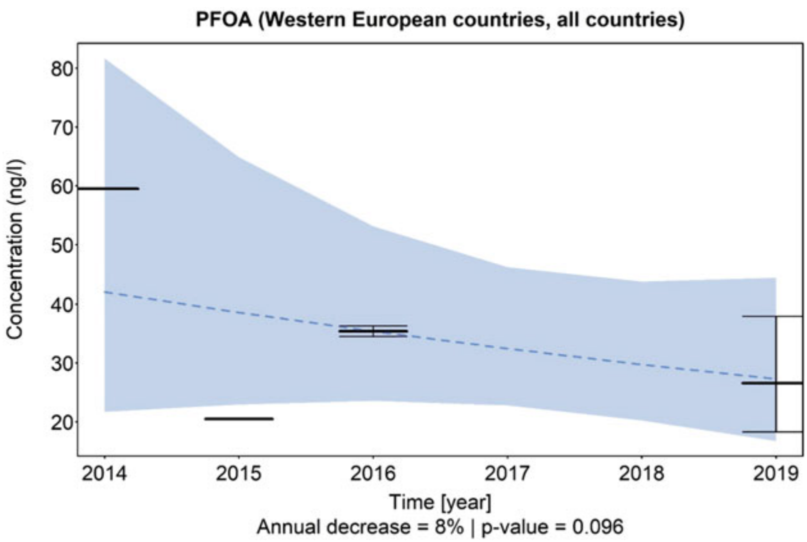

Fig. 40 Theil–Sen exponential trends of PFOA in human milk (ng/L) in countries of the Western European Group and Others Group (8 national pools, 7 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

Table 23 Overall decrease (%) of PFOA concentrations in Western European countries (computed using all samples)

Western Europe and Others Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All samples	7	8.3	–	58	–	0.096

3.2.7 Worldwide

The time trends of PFOA concentrations derived by the Theil–Sen method from all 86 national pools from 59 countries are illustrated by Fig. 41 and for the 48 national pools from 24 countries with multiple participation by Fig. 42. The results from both approaches were comparable: Statistically significant decreases over 10 years for PFOA concentrations of 41.7% and 46.6%, respectively, were calculated by the Theil–Sen method (1) using all samples and (2) using samples from countries with repeated participation (Table 24).

The summarizing descriptive parameters of Fig. 23 in subsection 3.2.1 seem to indicate an increase of PFOA concentrations from 2008–2011 to 2012–2015 with a subsequent decrease to 2016–2019. The lowest number of samples was available for

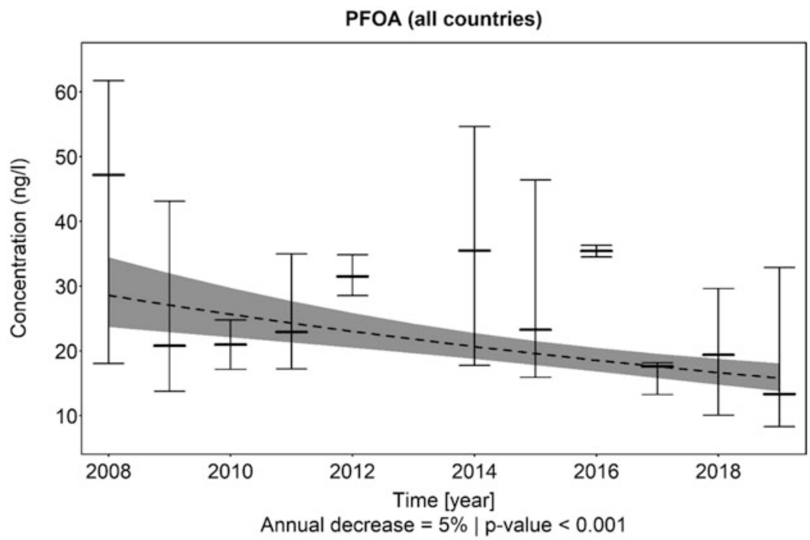

Fig. 41 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) worldwide using data from all countries (86 national pools, 59 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles.)

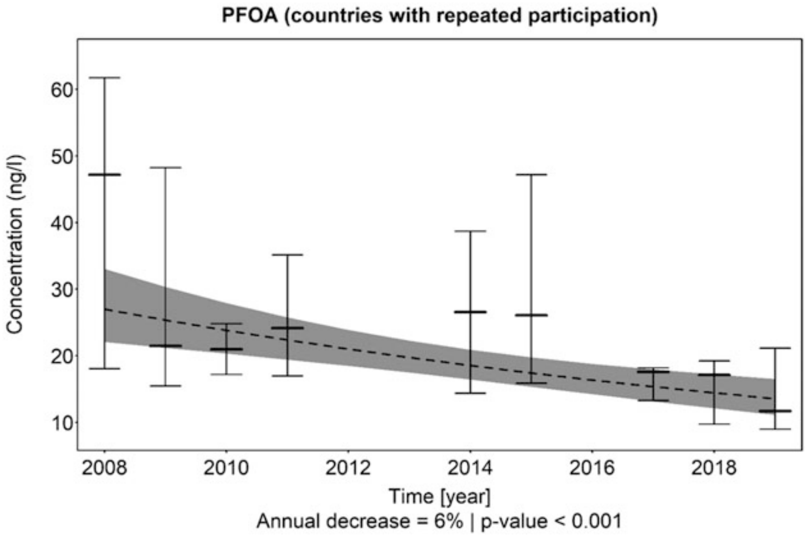

Fig. 42 Theil–Sen exponential trends of PFOA concentrations in human milk (ng/L) worldwide using data from countries with repeated participation (48 national pools, 24 countries)

Table 24 Overall decrease (%) of PFOA concentrations worldwide computed (1) using all samples and (2) using samples from countries with repeated participation

Worldwide	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall
		Theil–Sen method	Median method	Theil–Sen method	Median method	
All countries	59	5.3	4.8	41.7	38.6	<0.001
Repeatedly	24	6.1	6.9	46.6	51.0	<0.001

the 2012–2015 period (in total: 15 national pools). 7 samples came from countries with repeated participation (Côte d'Ivoire, Czech Republic, Georgia, Haiti, Lithuania, Moldova, Niger) and 8 samples from countries with single participation (three samples from Brazil; Belgium, Bulgaria, Croatia, Netherlands, Romania). The range found in this period in all 15 samples (median 36.0 ng/L; range 18.5–85.8 ng/L) was clearly influenced by countries with single participation only in the 2012–2015 period: Minimum, median, and maximum concentrations in the 2012–2015 period were higher in countries participating only in this period than in countries with multiple participation (Table 25). Therefore, the participation of different countries in the three rounds and the elevated contribution of samples

Table 25 Comparison of median, minimum, and maximum PFOA concentrations (ng/L) in the 2012–2015 period of (1) countries with participation also in the 2008–2011 or 2016–2019 period; (2) countries participating only in the 2012–2015 period

	Countries with multiple participation	Countries with single participation only in 2012–2015 period
N	7	8
Median	26.1	32.8
Min	13.0	20.5
Max	50.3	59.5

from countries participating only in the 2012–2015 round contributes considerably to the fluctuation of the descriptive parameters (e.g. median) between the three periods with an obvious maximum in the 2012–2015 period.

The country-specific temporal trends for the seven countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019 were discussed above in the corresponding UN Regional Groups. Inconsistent temporal tendencies of these seven countries were found: Three of these countries had clearly decreasing tendencies (decrease rates over 10 years between 57% and 60% in Niger, Czechia, and Georgia), two a slightly increasing tendency (Côte d'Ivoire, −9.4%; Lithuania, −20.2%), and two clearly increasing tendencies (Haiti, −87%; Moldova, −135%) (for illustration, see Figs. 43 and 44).

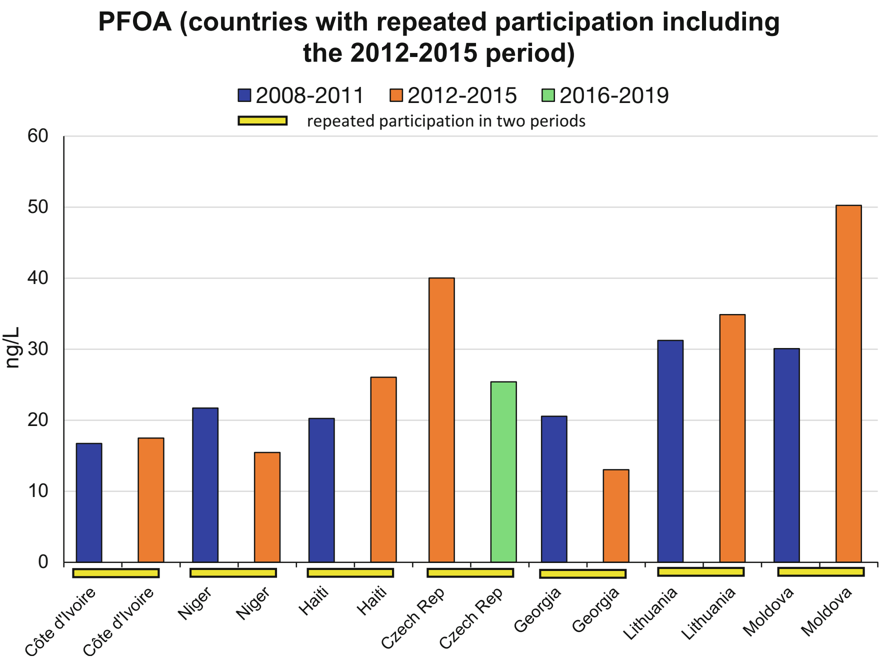

Fig. 43 Overview of the development of PFOA concentrations in human milk (ng/L) for countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019

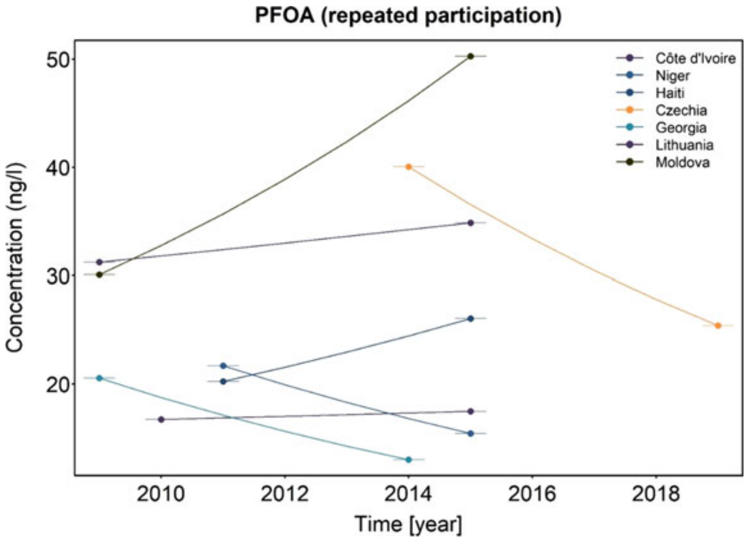

Fig. 44 Temporal tendencies of PFOA concentrations (ng/L) for countries with repeated participation including the 2012–2015 period as one of two periods between 2008 and 2019 using the Theil–Sen method

3.3 PFHxS

An initial indicative list of PFHxS, its salts and PFHxS-related compounds was presented to the Convention of Parties in 2019 (UNEP 2019c). PFHxS was then listed in Annex A of the Convention in 2022 (UNEP 2022).

In 84% of the 86 samples from 59 countries, PFHxS concentrations were below the limit of quantification (LOQ; 5.7 ng/L). 11 countries had PFHxS concentrations (slightly) above the LOQ (range 5.7–10 ng/l). Four samples had concentrations above 10 ng/L with one country more than 10-times higher levels (13.3 ng/L [Romania, 2014]; 17.9 ng/L [Sweden, 2019]; 35.8 ng/L [Haiti, 2011]; 115 ng/L [Kiribati, 2018]). The high rate of samples with concentrations below LOQ impedes the assessment of time trends.

3.3.1 African Group

From 27 samples of 16 countries, 25 samples were below LOQ (< 5.7 ng/L); one country had a concentration in the range of LOQ (Togo, 2010: 5.8 ng/L), one country above LOQ (Mali 2009: 7.8 ng/L) (Fig. 52 in the appendix). In nearly all countries with repeated participations, the PFHxS concentrations were below LOQ for both participations. Therefore, no temporal tendencies can be derived, neither in general between periods based on results of all countries, nor by combining data from all countries and use of the Theil–Sen method (Fig. 45) and country-specific for countries with repeated participation except for Mali (decrease from 2009 [7.8 ng/L]

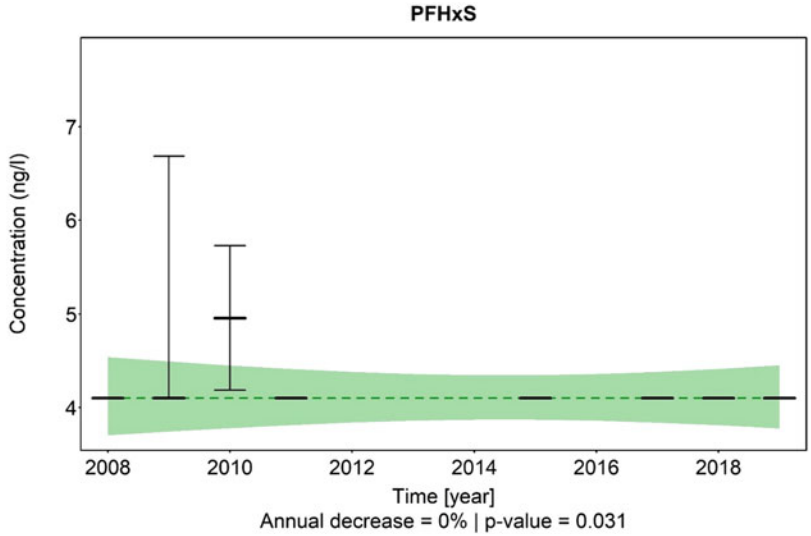

Fig. 45 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) from all African countries (27 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

to 2019 [<LOQ]). The changes in Togo from 5.8 ng/L in 2010 to <5.7 in 2017 are in the range of the LOQ and cannot be used to estimate temporal trends quantitatively.

3.3.2 Asia-Pacific Group

From 18 samples of 16 countries, 15 samples were below LOQ (< 5.7 ng/L); one country had a concentration in the range of the LOQ (Fiji, 2011: 5.9 ng/L), one country of 7.5 ng/L (Thailand, 2018), and one country 115 ng/L (Kiribati, 2018) (Fig. 53 in the appendix). The changes in Fiji from 5.9 ng/L in 2011 to <5.7 in 2019 are in the range of the LOQ and cannot be used to estimate any temporal tendencies. Both samples from Niue (collected in 2017 and 2019) were below LOQ. Therefore, no temporal tendencies can be derived, neither in general between periods based on results of all countries, nor by combining data from all countries and use of the Theil–Sen method (Fig. 46) nor country-specific for countries with repeated participation.

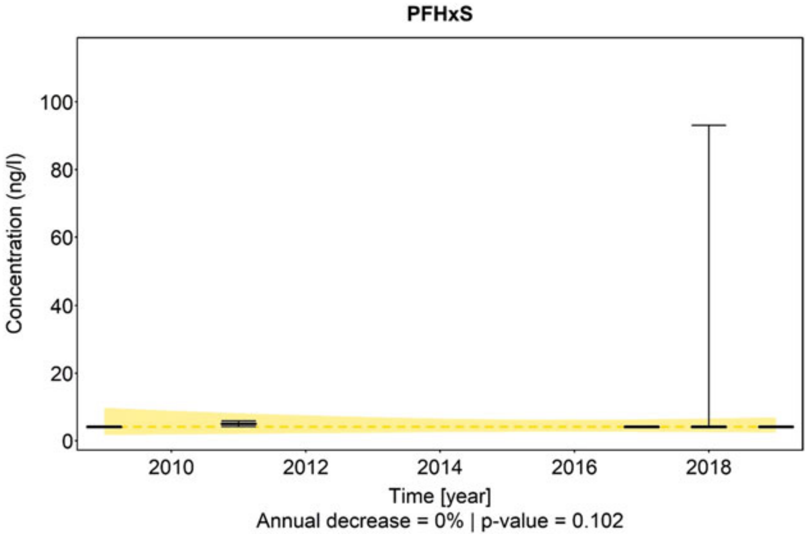

Fig. 46 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) in countries of the Asia-Pacific group (18 national pools, 16 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

3.3.3 Group of Latin American and Caribbean Countries (GRULAC)

From 21 samples of 12 countries, 17 samples were below LOQ (<5.7 ng/L). From six countries with repeated participation in two periods, in three countries the PFHxS concentrations were below LOQ in both periods. In two countries, PFHxS concentrations had decreasing tendencies from a concentration quite close to the LOQ to below LOQ (Antigua-Barbuda, 2008: 6.6 ng/L, 2018: <5.7 ng/L; Jamaica, 2011: 6.0 ng/L, 2018: <5.7 ng/L). In one country, a considerable decrease was found (Haiti, 2011: 35.8 ng/L, 2015: 8 ng/L) (Fig. 54 in the appendix). Therefore, temporal tendencies cannot be derived in general between periods based on results of all countries, nor by combining data from all countries and use of the Theil–Sen method (Fig. 47). On a country-specific basis, a considerable decrease was found in Haiti.

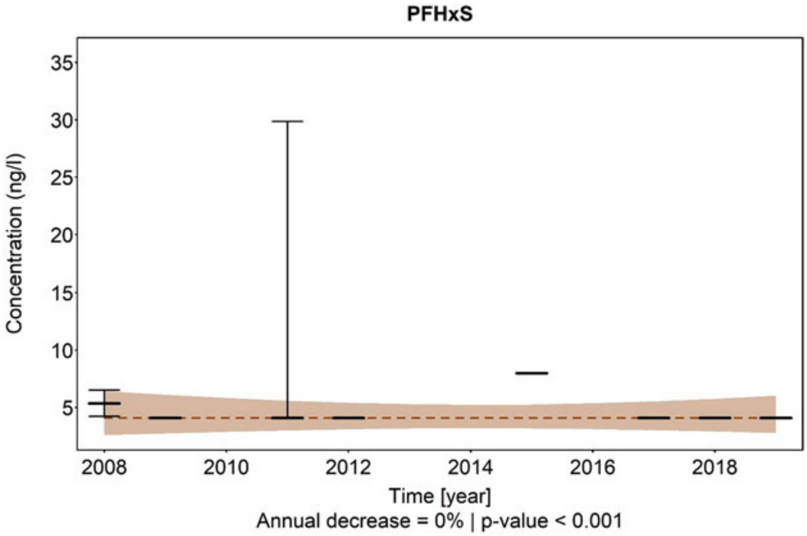

Fig. 47 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) in Latin American and Caribbean countries (21 national pools, 12 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

3.3.4 Eastern European Group

From 12 samples of 8 countries, 9 samples were below LOQ (<5.7 ng/L). One country had a PFHxS concentration in the range of the LOQ (Croatia, 2014: 6.1 ng/L) and two countries above (Moldova, 2015: 9.9 ng/L; Romania, 2014: 13.3 ng/L) (Fig. 55 in the appendix). Therefore, no general temporal tendencies can be derived by comparing periods, nor by combining data from all countries and use of the Theil–Sen method (Fig. 48). From four countries with repeated participation, an increasing tendency was observed in Moldova from 2009 to 2015 (from <LOQ to 9.9 ng/L), whereas in the other three countries (Czech Republic, Georgia, Lithuania) PFHxS concentrations below LOQ were found in both periods.

Using the Hites method of break point search (Hites 2019) on all countries in the Eastern European Group, a break point was found for PFHxS, although, the result was non-significant.

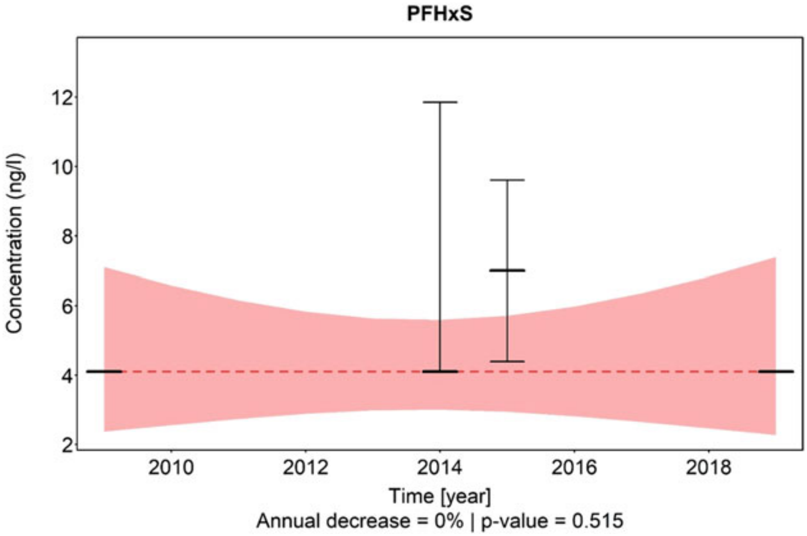

Fig. 48 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) in countries of the Eastern European Group (12 national pools, 8 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

3.3.5 Western European and Others Group (WEOG)

From 8 samples of 7 countries, 5 samples were below LOQ (<5.7 ng/L), three samples above (Netherlands, 2014: 8.6 ng/L; Sweden, 2019: 17.9 ng/L; Switzerland, 2016: 8.7 ng/L) (Fig. 56 in the appendix). No country participated repeatedly. Therefore, no temporal tendencies can be derived, neither in general between periods based on results of all countries, nor by combining data from all countries and use of the Theil–Sen method (Fig. 49) nor country-specific for countries with repeated participation.

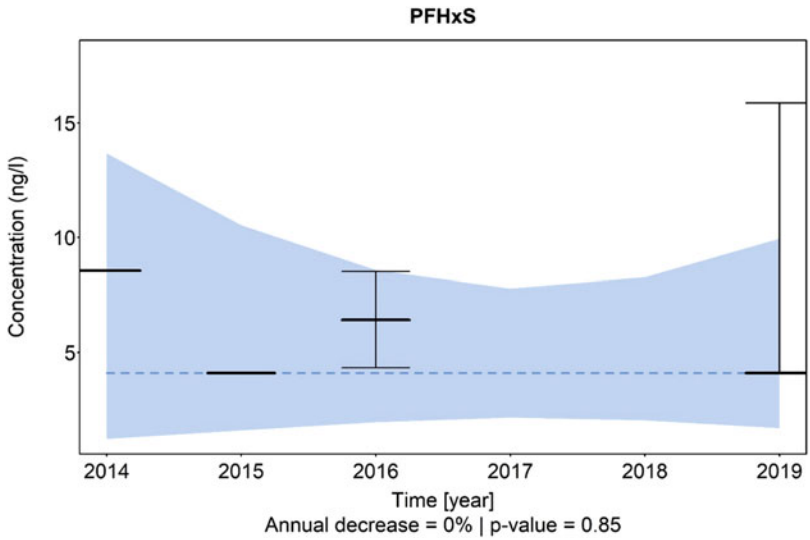

Fig. 49 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) in countries of the Western European Group and Others Group (8 national pools, 7 countries). (The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

3.3.6 Worldwide

Due to the very high percentage of samples below LOQ, no temporal trends could be derived, neither by combining all samples and use of Theil–Sen method (Fig. 50) nor by combining all samples from countries with repeated participation and use of Theil–Sen method (Fig. 51).

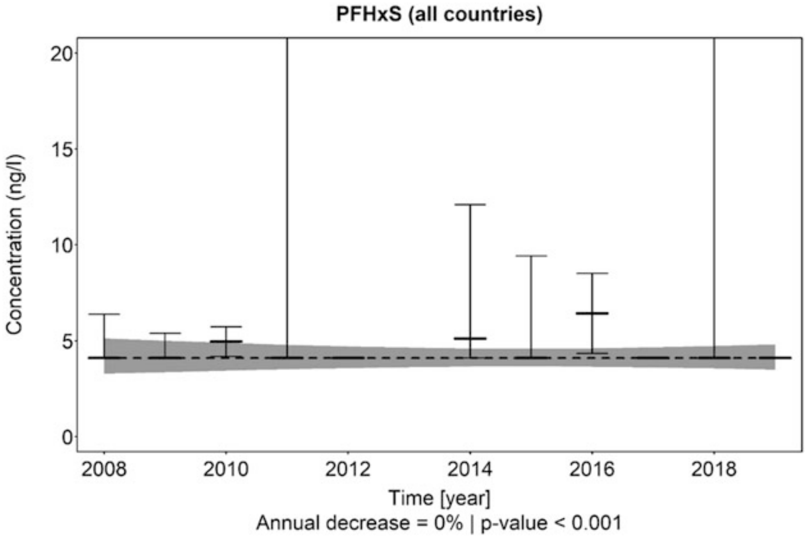

Fig. 50 Theil–Sen exponential trends of PFHxS concentrations in human milk (ng/L) worldwide using data from all countries (86 national pools, 59 countries). (The shaded are shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, hinges and whiskers show ranges between fifth and 95th percentiles. To replace left-censored values, values under LOQ were substituted by 1/sqrt(2) of the LOQ.)

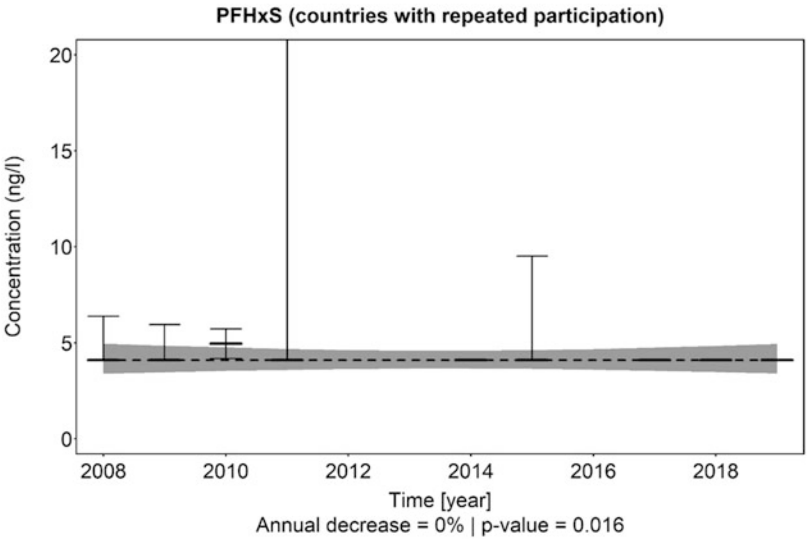

Fig. 51 Theil–Sen exponential trends of PFOA in human milk (ng/L) worldwide using data from countries with repeated participation (48 national pools, 24 countries)

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Disclaimer The authors alone are responsible for the views expressed in this publication, which do not necessarily represent the decisions, policy, or views of the World Health Organization and the United Nations Environment Programme.

Appendix

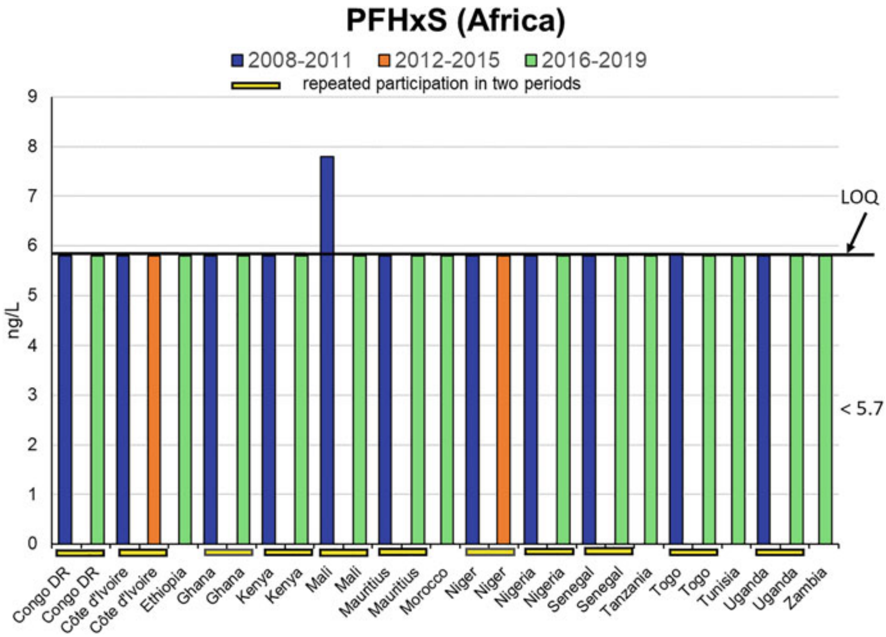

Fig. 52 Illustration of findings of PFHxS concentrations around the limit of quantification (LOQ) in human milk (ng/L) from African countries over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

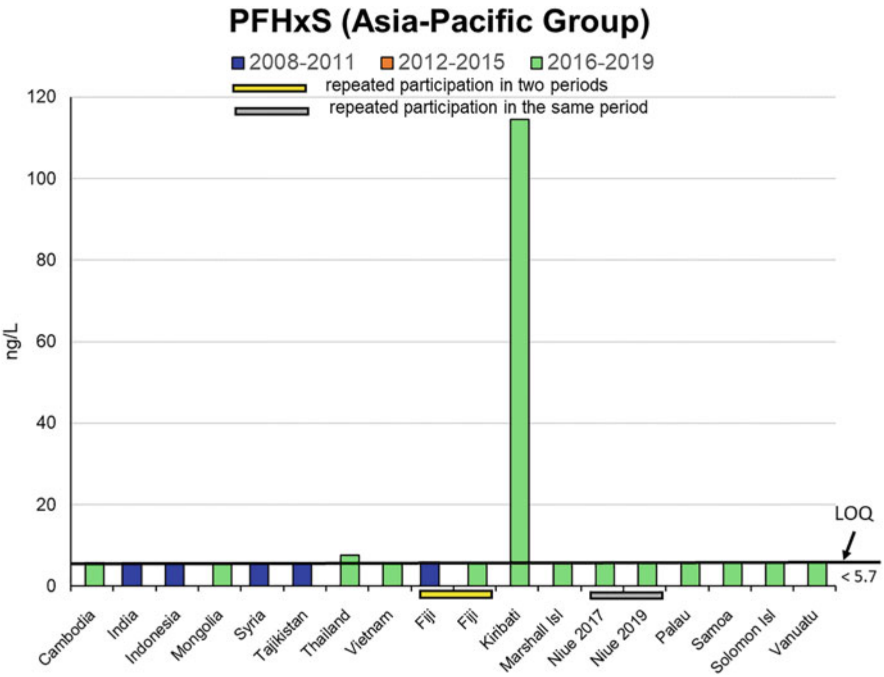

Fig. 53 Illustration of findings of PFHxS concentrations around the limit of quantification (LOQ) in human milk (ng/L) from countries of the Asia-Pacific Group over time (period 2008–2011 in blue, period 2012–2015 in orange [however, without samples in this period], and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

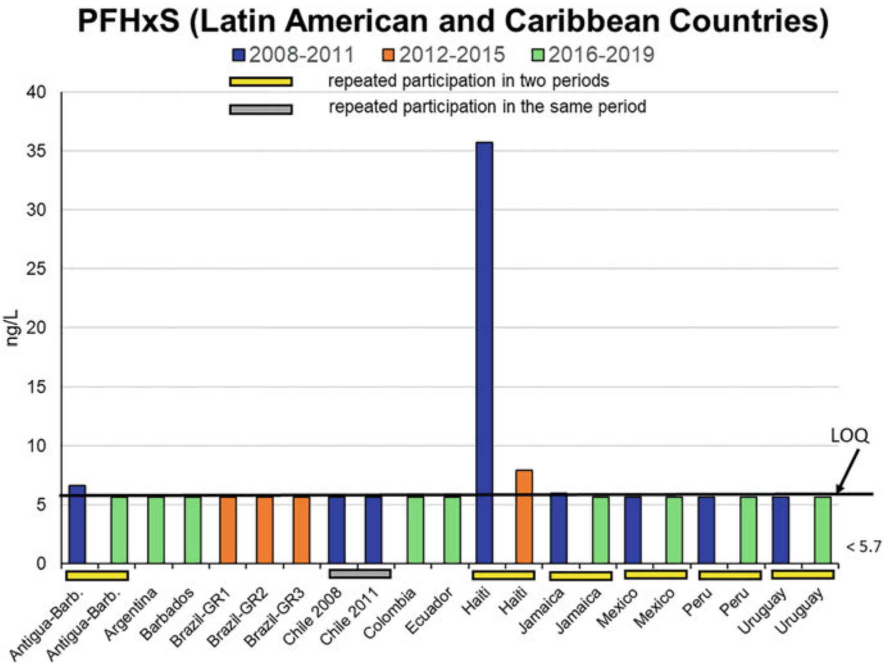

Fig. 54 Illustration of findings of PFHxS concentrations around the limit of quantification (LOQ) in human milk (ng/L) from Latin American and Caribbean countries over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles, with repeated participation in the same period by grey rectangles)

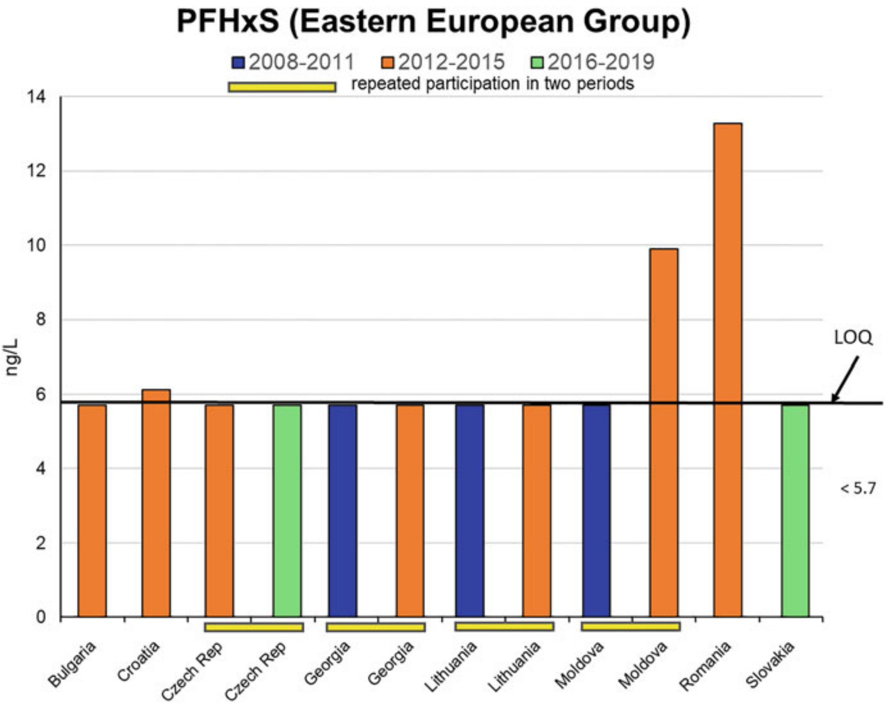

Fig. 55 Illustration of findings of PFHxS concentrations around the limit of quantification (LOQ) in human milk (ng/L) from countries of the Eastern European Group over time (period 2008–2011 in blue, period 2012–2015 in orange, and period 2016–2019 in green; countries with repeated participation in two of these periods marked by yellow rectangles)

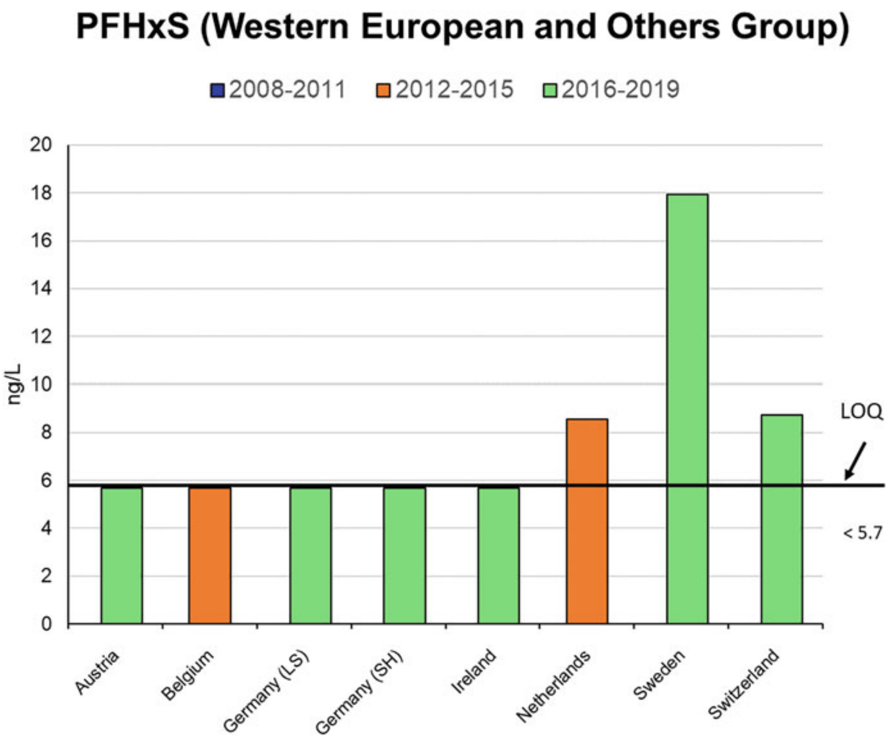

Fig. 56 Illustration of findings of PFHxS concentrations around the limit of quantification (LOQ) in human milk (ng/L) from countries of the Western European and Others Group over time (period 2008–2011 in blue [however, without samples in this period], period 2012–2015 in orange, and period 2016–2019 in green)

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