Time Trends in Human Milk Derived from WHO- and UNEP-Coordinated Exposure Studies, Chapter 2: DDT, Beta-HCH and HCB

Rainer Malisch, Björn Hardebusch, Ralf Lippold,
F. X. Rolaf van Leeuwen, Gerald Moy, Angelika Tritscher,
Kateřina Šebková, Jana Klánová, and Jiří Kalina

Abstract

Temporal trends of DDT ("DDT complex" as sum of p,p′-DDT, o,p′-DDT, p,p′-DDD and p,p′-DDE), beta-hexachlorocyclohexane (beta-HCH) and hexachlorobenzene (HCB) were assessed using pooled human milk samples from 44 countries from all United Nations Regional Groups based on their repeated participation in WHO/UNEP-coordinated exposure studies performed between 2000 and 2019. In contrast to a general estimation of time trends based on results from all countries, this is a more precise approach, because levels among countries are often highly variable. 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).

R. Malisch (✉) · B. Hardebusch · R. Lippold
State Institute for Chemical and Veterinary Analysis of Food (Chemisches und Veterinäruntersuchungsamt, CVUA), Freiburg, Germany
e-mail: pops@cvuafr.bwl.de

F. X. R. van Leeuwen
World Health Organization (WHO) European Centre for the Environment and Health at the National Institute of Public Health and the Environment (coordinating the human milk study from 2000 to 2003), Bilthoven, The Netherlands

G. Moy
World Health Organization, Global Environment Monitoring System/Food Contamination Monitoring and Assessment Programme (GEMS/Food) (coordinating the human milk study from 2004 to 2008), Geneva, Switzerland

A. Tritscher
World Health Organization, Department of Food Safety and Zoonoses (coordinating the human milk study from 2009 to 2011), Geneva, Switzerland

K. Šebková · J. Klánová · J. Kalina
RECETOX, Faculty of Science, Masaryk University, Brno, Czech Republic

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For DDT, an overall decrease over 10 years between 50% and 80% was achieved in Africa, the Asia-Pacific region and in Latin America and the Caribbean region, and at a global level. Slightly lower decreases were observed in European countries because DDT was banned much earlier in these countries and only residual levels were depleting. Western European countries had the lowest median and the lowest maximum DDT concentrations. This is an indication that the decrease might be faster in regions with higher concentrations, compared to a slower decrease in less contaminated regions. The frequency distribution of the country-specific decrease (*decrease rate constants*) confirms these findings.

For beta-HCH, an overall decrease over 10 years between 50% and 98% was achieved in all UN regions and at a global level. Country-specific decreases vary in the low background range (below 5 μg beta-HCH/kg lipid). Regarding HCB, all countries from Africa and many countries from the Pacific Islands and Latin America and the Caribbean were in the range of a low background contamination below 5 μg/kg lipid resulting in a wide range of reduction rates. In contrast, in countries with HCB concentrations above 30 μg/kg lipid in previous rounds, overall decreases over 10 years between 50% and 85% were observed.

Therefore, the reduction rates should be seen also in context with the concentration range: A differentiation of levels above or in the range of the background contamination seems to be advised. If high levels are found, sources might be detected which could be eliminated. However, at low background levels, other factors, e.g. contamination of feed and food by air via long-range transport and subsequent bioaccumulation, cannot be influenced locally.

Keywords

Time trends · Human milk biomonitoring · Stockholm Convention on Persistent Organic Pollutants · DDT · Beta-hexachlorocyclohexane (beta-HCH) · Hexachlorobenzene (HCB) · Global WHO/UNEP studies · UN Regional Groups

1 Introduction

The World Health Organization (WHO) and the United Nations Environment Programme (UNEP) performed between 1987 and 2019 seven exposure studies on human milk. 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. In this compendium, specific papers address various aspects of these human milk surveys, including in Part I a review of human milk surveys on POPs from a historical perspective (Fürst 2023), the overview of the WHO/UNEP-coordinated exposure studies (Malisch et al. 2023a) and a review of the Stockholm Convention, the Global Monitoring Plan (GMP) and its implementation in regional and global monitoring reports (Šebková 2023).

Worldwide trends in DDT concentrations in human breast milk were assessed compiling data since 1951 until the end of the 1990s (Smith 1999). A global overview on the spatial and temporal trends of Stockholm Convention POPs in breast milk reviews scientific publications between 1995 and 2011 (Fång et al. 2015). The regional and global monitoring reports for the GMP assess datasets in the core media—ambient air, human tissues (human breast milk or blood) and water for hydrophilic POPs, but also other media such as soil, biota, plants are used to support interpretation of observed levels and their trends. These reports are available at the homepage of the Stockholm Convention (>Implementation>Global Monitoring Plan>Monitoring Reports).

In three articles of Part IV of this compendium, time trends derived from the WHO/UNEP-coordinated exposure studies are evaluated, in the first article ("Time Trends in Human Milk Derived from WHO- and UNEP-coordinated Exposure Studies, Chapter 1") for polychlorinated biphenyls (PCB), polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) (Malisch et al. 2023b). This includes a clarification of quantitative goals of temporal studies and Convention's objectives: To provide reliable monitoring information for the Parties to the Stockholm Convention, as a quantitative objective for temporal studies the GMP guidance document proposed the ability to detect a 50% decrease in the levels of POPs within a 10-year period. However, 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 (UNEP 2015, 2019).

For chlorinated pesticides and industrial chemicals, the presentation and discussion of the 2000–2019 results in Part III includes a first *general* estimation of time trends in the five rounds for all countries participating over these 20 years (Malisch et al. 2023c). With regard to the concentrations found, most relevant were **DDT** (expressed as "DDT complex" and comprised of p,p′- and o,p′-isomers of DDT [dichlorodiphenyltrichloroethane] and the transformation products p,p′-DDE [dichlorodiphenyldichloroethylene] and p,p′-DDD [dichlorodiphenyl dichloroethane]), beta-hexachlorocyclohexane (**beta-HCH**) and hexachlorobenzene (**HCB**). As chapter 2 of the three articles on time trends, this paper presents the results of a more precise approach for the assessment of these three POPs by consideration of *results only from countries with repeated participation* in the studies.

2 General Aspects

2.1 Minimization of Sources of Variation; Samples and UN Regional Groups; Long-Term Quality Control

For minimization of possible sources of variation from the sampling design, in all rounds a number of individual samples were collected and pooled (mixed) samples prepared following a standardized protocol that was supervised by national coordinators. Equal aliquots of individual samples were combined to give composite samples, which are considered representative of the average levels of the analytes of interest in human milk for a certain country or subpopulation of a country at the time of sampling. For an overview of the scope, protocols for collection of samples, factors minimizing sources of variation for assessments of time trends, participation of countries and the five UN Regional Groups (African Group; Asia-Pacific Group; Group of Latin American and Caribbean Countries [GRULAC]; Eastern European Group; Western European and Others Group [WEOG]), see (Malisch et al. 2023a), for results of organochlorine pesticides determined in 163 pooled human milk samples from 82 countries, see (Malisch et al. 2023c).

For minimization of the variation from chemical analysis at the 2000–2019 WHO/UNEP-coordinated studies, the determination of organochlorine pesticides and industrial contaminants in the pooled samples was performed by the Reference Laboratory applying long-term analytical quality control (Hardebusch et al. 2023).

Due to the particular scope at the beginning of a study with regard to the expansion of analytes of interest over time, results from 119 pooled samples from 44 countries with repeated participation between 2000 and 2019 are available for various organochlorine pesticides and industrial chemicals, including DDT complex, beta-HCH and HCB as covered in this article. The detailed data for all samples is contained at the POPs Global Monitoring Plan Data Warehouse (GMP DWH) and can be publicly retrieved (GMP DWH 2020).

2.2 "DDT Complex" as Sum Parameter for DDT

The sum parameter "DDT complex" is calculated as sum of o,p′-DDT, p,p′-DDT, p,p′-DDE and p,p′-DDD using correction factors for molecular weight for the metabolites p,p′-DDE and p,p′-DDD. This term is used in combination with the unit to express concentrations; alternatively, the common short form "DDT" is used. The extent of the contribution of p,p′-DDT and p,p′-DDE to DDT complex is indicative of an older or more recent use of DDT (Malisch et al. 2023c).

2.3 Methods of Statistical Data Treatment: Trends vs. Tendencies

If a country had sent two or more pooled samples in a certain round, the median of these samples in this period has been used for the country in some summarizing figures and tables. These are identified as "country results" or "aggregated data". However, for the time trend analysis, data were not aggregated, but values of all individual pooled samples were used.

For methods of statistical data treatment (use of the non-parametric linear Theil–Sen trend estimator and of the median method to derive decrease rates expecting exponential trends [as commonly observed in cases after stop of production and application of a chemical rather than unrealistic linear trends] and for prevention of Simpson's paradox), see subsection 2.4 in the preceding chapter "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 includes the differentiation between *trends* as statistically significant decreases (*decrease rate constants)* requiring *p*-values <0.05 and changes of concentrations indicating *tendencies* as statistically not significant decreases. As Theil–Sen p is never below 0.05 for less than 5 data points and for most countries only less than 5 data points were available, statistically significant trends could be derived only for regions (combining data from countries) and few countries, showing on 95% confidence level whether the trend is not caused by random variance in the data. In addition, for some countries, based on statistically significant reduction rates and participation also in the decade after 2010, a prognosis of the estimated concentrations in 2025 was derived. However, for most countries, only two or three data points are available. In these cases, the observed changes of the concentrations do not allow to draw statistically significant conclusions on trends and therefore indicate tendencies.

2.4 Background Concentration

As explained in the chapter on findings of organochlorine pesticides and industrial chemicals (Malisch et al. 2023c), a background concentration is defined as that portion of the measured human milk levels that is found in the absence of specific sources, e.g., the chemical of interest was not used or emitted within the study area, or after a sufficiently long phase-out period (depending on the half-life). In contrast to findings of high concentrations, e.g. after use of chemicals, the low levels described as "background levels" are not attributable to a known emission source.

However, the term "background level" does not imply per se 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 measured concentration range. For human milk, potential adverse effects have to be balanced against its many known positive health aspects for breastfed infants.

3 DDT

3.1 Global level and comparison between UN Regional Groups

For DDT concentrations in human milk, large differences were found among the 119 pooled samples from 44 countries with repeated participation between 2000 and 2019: The range between a minimum of 17 μg DDT complex/kg lipid found in 2019 and a maximum of 23,500 μg DDT complex/kg lipid found in 2012 covered three orders of magnitude (median: 283 μg DDT complex/kg lipid). This is the same range as found in 134 country results (based on aggregated data) of 82 countries (as total number including one-time participants between 2000 and 2019), with a median of 255 μg DDT complex/kg lipid (Malisch et al. 2023c).

As recommended by the GMP guidance document (UNEP 2019), the Theil–Sen method was applied for power analysis of statistical trends. Statistically significant trends were derived for the UN Regional Groups by combination of data from countries. Basic results of the exponential trends calculated by this method comprise the overall decreases per 1 year and 10 years. Statistical differences between the Theil–Sen method and the additionally applied median method to derive time trends in different UNEP Regional Groups were insignificant on 95% confidence level, which shows that the Simpson's paradox caused by different sampling periods is weak in these cases (Table 1). For the country-specific results in the following subsections 3.2–3.6, also decrease rates per 5 years are shown, which are about 20% higher than the decrease rates per 4 years representing the average lengths of WHO/UNEP-coordinated exposure studies.

An overall decrease within a 10-year period between 50% and 80% was achieved for DDT levels in Africa, the Asia-Pacific Group and the Group of Latin American and Caribbean Countries, and at a global level. Lower reduction rates were observed in the Eastern European Group and the Western European and Others Group, which had banned DDT much earlier. Generally, the highest DDT concentrations in the five periods between 2000 and 2019 were found either in Africa or in the Asia-Pacific Group or in the Group Latin America and Caribbean Countries, whereas Western European and Others Group countries had the lowest median and lowest maximum of DDT concentrations. This is an indication that the decrease might be faster in regions with higher initial concentrations, compared to a slower decrease in less contaminated regions. All trends were statistically significant (p-value <0.001 and 0.017, respectively).

On a global level, the decrease over 10 years was nearly 60% calculated by the Theil–Sen method with use of all individual samples. The median method

Table 1 Overall decrease (%) of DDT concentrations (calculated as DDT complex) in human milk in the five UN Regional Groups and worldwide (computed using all samples from countries with repeated participation; for UN Regional Groups, in particular for "Others" in the "Western European and Others" UN Regional Group, see Malisch et al., 2023a and 2023c and subsection 3.6)

UN Regional Groups	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend p-value overall[a]
		Theil–Sen method	Median method	Theil–Sen method	Median method	
Africa	13	12.7	14.5	74.3	79.1	<0.001
Asia-Pacific	8	15.0	10.3	80.2	66.5	<0.001
Latin America and Caribbean	9	6.6	7.7	49.5	55.0	0.017
Eastern Europe	6	4.7	5.9	38.5	45.5	0.017
Western Europe and Others	8	4.8	6.4	39.1	48.6	<0.001
Global	44	8.4	6.6	58.3	49.5	<0.001

[a]For Theil–Sen method

(as median of the 5 UN Regional Groups and of the 44 countries) gave comparable results.

The exponential trends of DDT concentrations in human milk derived by the Theil–Sen method in the five UN regions and worldwide by combination of data from countries are illustrated in Fig. 1a–f. These figures are normalized according to the maximum concentration found in the respective UN Regional Groups. Thus, the different scales illustrate the different ranges between and within the UN Regional Groups. For a detailed discussion of the regional data, see the following subsections 3.2–3.6.

3.2 African Group

Figure 2 (for aggregated data, see subsection 2.3) and Fig. 3 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate temporal changes of DDT concentrations in 13 countries from Africa with repeated participation between 2000 and 2019. In Egypt, a reduction of 96% was observed from nearly 400 µg DDT complex/kg lipid in the 2000–2003 period to the 2016–2019 period, when with 17 µg DDT complex/kg lipid the lowest DDT concentration of all countries on a global level during the whole period 2000–2019 was found. Most countries participated for the first time in the 2008–2011 period; in these countries, DDT concentrations fell on average by about 70% until the period 2016–2019 (range between 59% and 93% decrease). The highest concentration of 23,500 µg DDT complex/kg lipid (or 23.5 mg/kg) found in Ethiopia in 2012 decreased by 70% until 2019. For discussion of use of DDT to combat mosquitos for malaria control and measures taken by Ethiopia to successfully reduce DDT levels, see (Gebremichael et al. 2013; Malisch et al. 2023c).

The overall decreases per 1 year, 5 years and 10 years are given in Table 2. The limited number of samples did not allow to determine statistically significant decrease rates (p ~ 1.000) (for statistical significance of *trends* requiring p-values <0.05 and changes of concentrations indicating *tendencies*, see subsection 2.3). On average, the levels of DDT in all African countries decreased within a 10-year period by nearly 80% (median 79%; range 63–95%). This is in line with the statistically significant ($p < 0.001$) decrease over 10 years for all African countries of 74% calculated by the Theil–Sen method (see Table 1 in Sect. 3.1).

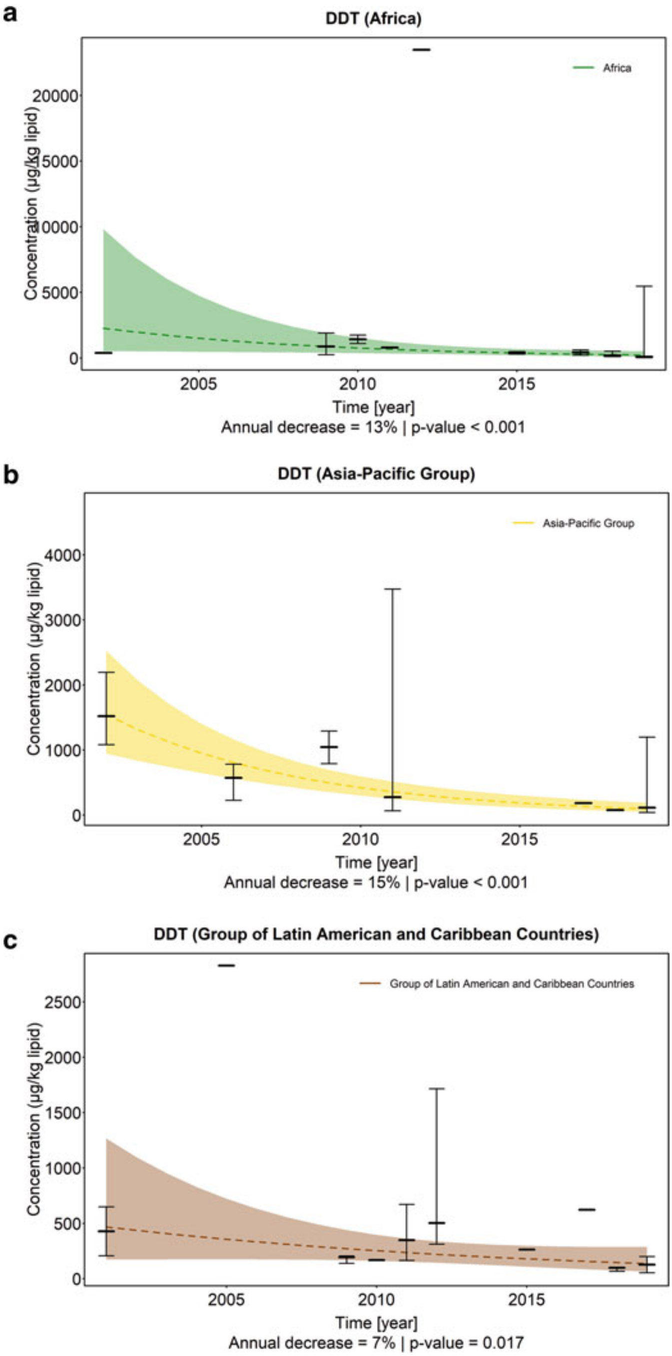

Fig. 1 (**a–f**) Theil–Sen exponential trends of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) worldwide and in the five UN regions. The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, whiskers show ranges between fifth and 95th percentiles

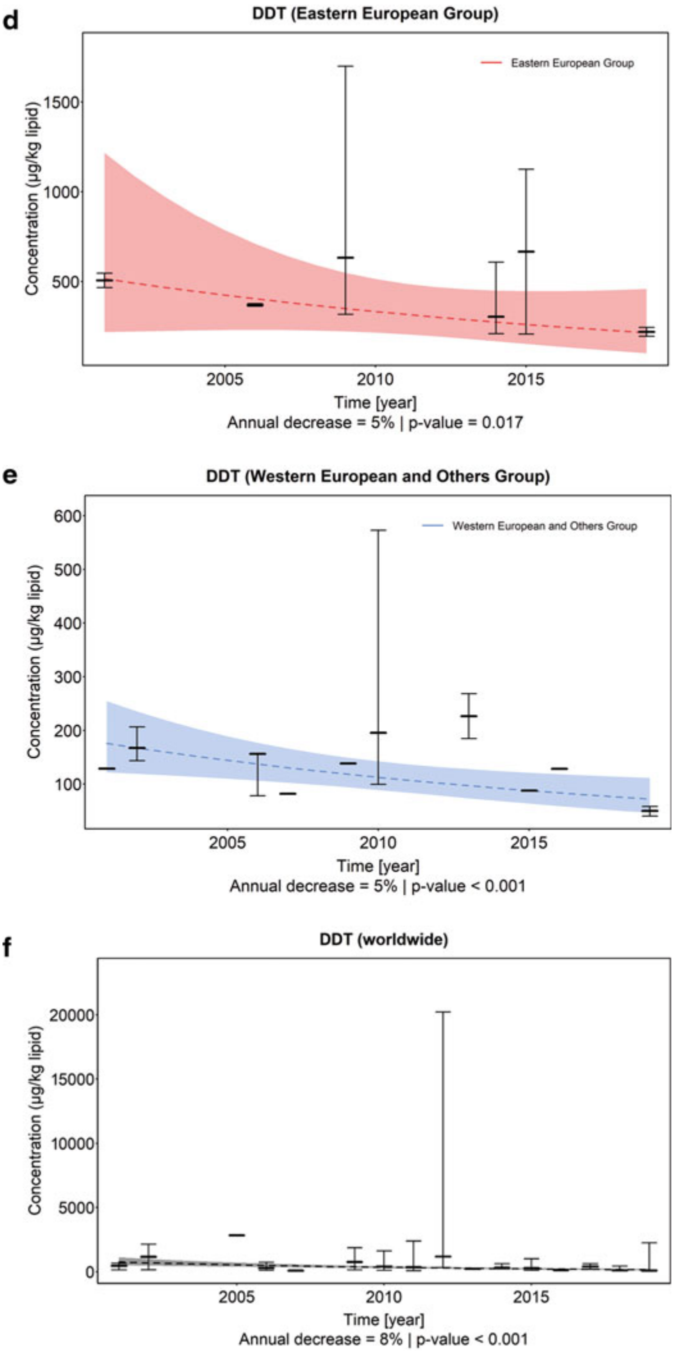

Fig. 1 (continued)

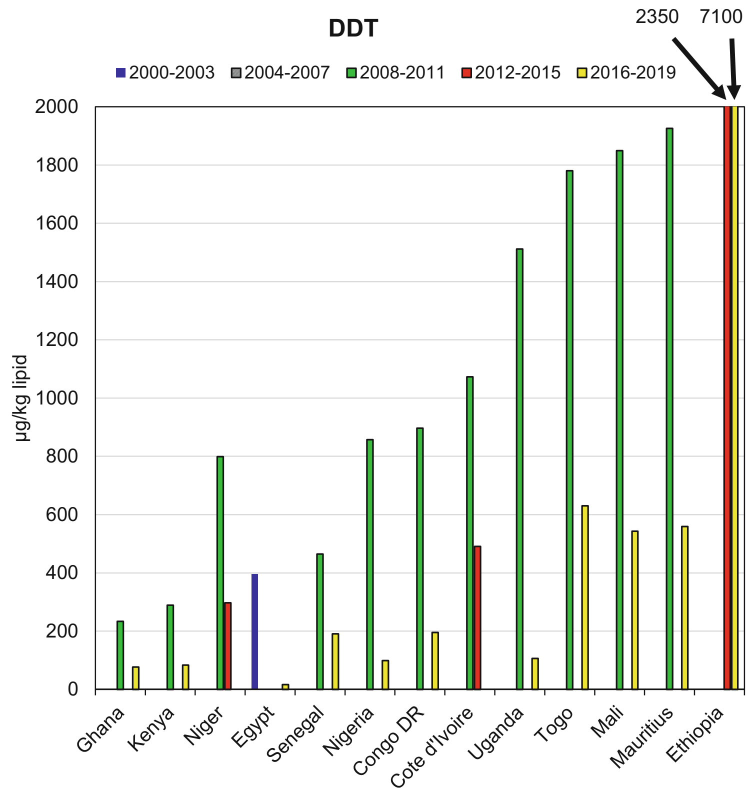

Fig. 2 Overview of the development of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid; aggregated data) over time in African countries with repeated participation between 2000 and 2019

3.3 Asia-Pacific Group

Figures 4 (for aggregated data) and 5 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of DDT concentrations in 8 countries from the Asia-Pacific Group with repeated participation between 2000 and 2019.

In Hong Kong SAR of China, DDT concentrations fell approximately by 35% from the 2002 level of 1580 µg DDT complex/kg lipid (median of 10 samples from different population subgroups, see Hui et al. 2008) to the 2009 level of 1040 µg DDT complex/kg lipid (median of 4 samples from different subgroups). In Fiji, a reduction of nearly 80% was observed from 2000–2003 to 2008–2011, which then was further reduced by about 60% until 2016–2019 to 8% of the initially found concentration (1340 µg DDT complex/kg lipid in 2001). Most other countries

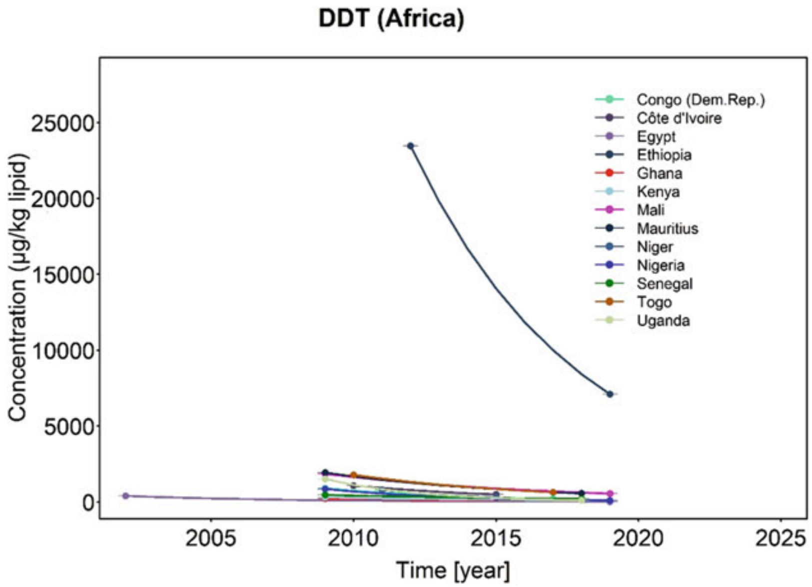

Fig. 3 Temporal tendencies of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) in African countries with repeated participation between 2000 and 2019 using the Theil–Sen method

Table 2 Overall decrease (%) of DDT concentrations (expressed as µg DDT complex/kg lipid) in human milk in African countries per 1 year, 5 years and 10 years (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Trend *p*-value overall
Congo (DR)	17.3	61.4	85.1	1.000
Côte d'Ivoire	14.5	54.3	79.1	1.000
Egypt	17.0	60.6	84.5	1.000
Ethiopia	15.7	57.4	81.9	1.000
Ghana	10.5	42.7	67.1	1.000
Kenya	11.7	46.2	71.0	1.000
Mali	11.5	45.8	70.6	1.000
Mauritius	12.8	49.7	74.7	1.000
Niger	21.9	71.0	91.6	1.000
Nigeria	19.4	66.0	88.5	1.000
Senegal	9.4	39.0	62.8	1.000
Togo	13.8	52.4	77.3	1.000
Uganda	25.5	77.1	94.7	1.000
Median	14.5	54.3	79.1	

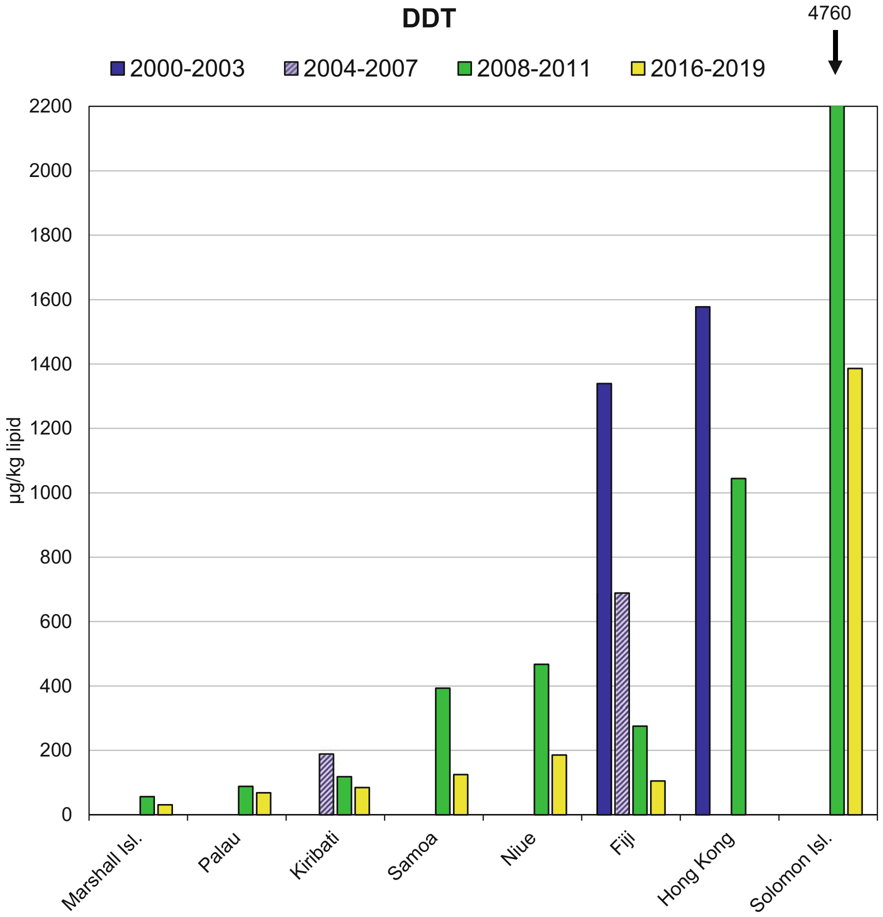

Fig. 4 Overview of the development of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid; aggregated data) over time in countries of the Asia-Pacific Group with repeated participation between 2000 and 2019

participated the first time in the period 2008–2011; Kiribati in 2004–2007. In all these countries, a decrease was observed in the following years (median of decrease: 60%, range 22–71%), including Solomon Islands with a decrease of the high DDT level in 2011 (4760 µg DDT complex/kg lipid) by 71% until 2019.

The overall decreases per 1 year, 5 years and 10 years are given in Table 3. The median of reduction rates in the DDT levels within a 10-year period in 8 countries was 65% (range 31–79%). Statistically significant was the decrease of 47% over 10 years in Hong Kong SAR of China and of 79% in Fiji ($p < 0.001$). The limited

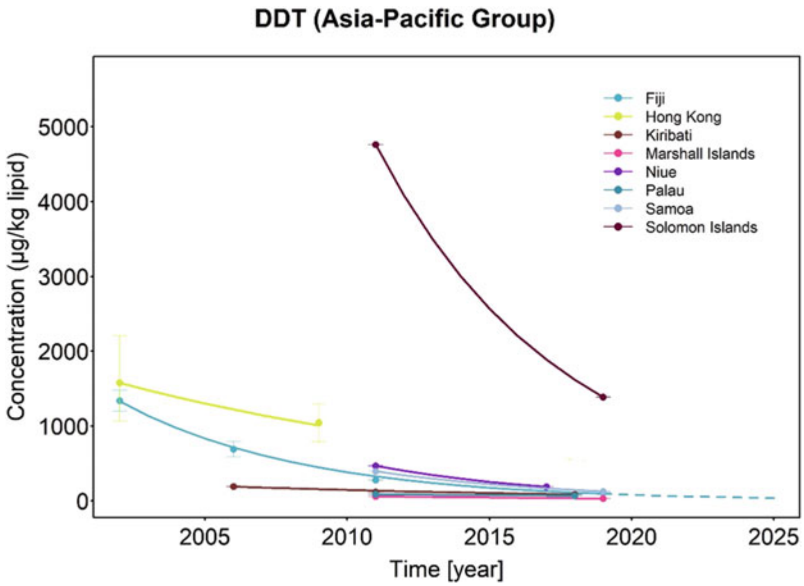

Fig. 5 Temporal tendencies of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) in countries of the Asia-Pacific Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with statistically significant time trends in Hong Kong SAR of China and Fiji)

Table 3 Overall decrease (%) of DDT concentrations in human milk in countries of the Asia-Pacific Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [µg/kg lipid]	Trend *p*-value overall
Fiji	14.5	54.2	79.0	36.6	<0.001
Hong Kong	6.2	27.4	47.3		<0.001
Kiribati	6.5	28.5	48.8		0.250
Marshall Isl.	7.2	31.3	52.8		1.000
Niue	14.2	53.6	78.5		1.000
Palau	3.6	16.6	30.5		1.000
Samoa	13.4	51.2	76.2		1.000
Solomon Isl.	14.3	53.8	78.6		1.000
Median	10.3	42.1	66.5		

number of samples did not allow to determine statistically significant decreases in the other countries ($p = 0.250$ and 1.000, respectively). For Fiji with a statistically significant decrease and participation also in the 2016–2019 round, in addition a prognosis about the probable concentrations in 2025 was made.

3.4 Group of Latin American and Caribbean Countries (GRULAC)

Figures 6 (for aggregated data) and 7 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of DDT concentrations in 9 Latin American and Caribbean countries with repeated participation between 2000 and 2019.

Based on comparison of median concentrations, in Brazil an increase by 17% was found from 2001 to 2012. In Haiti, DDT concentrations in human milk fell by 91% from 2004 (with 2830 μg DDT complex/kg lipid the highest concentration found in this UN Regional Group) to 2015. Most other countries participated for the first time in the period 2008–2011. In all these countries, a decrease was observed in the following years (median: 41%; range 11–67%).

The overall decreases per 1 year, 5 years and 10 years are given in Table 4. By most countries, a (statistically not significant) decrease in the DDT levels within a 10-year period was achieved (median for 8 countries with reduction rates per 10 years: 60%; range 17–91%). The (statistically not significant) increase calculated

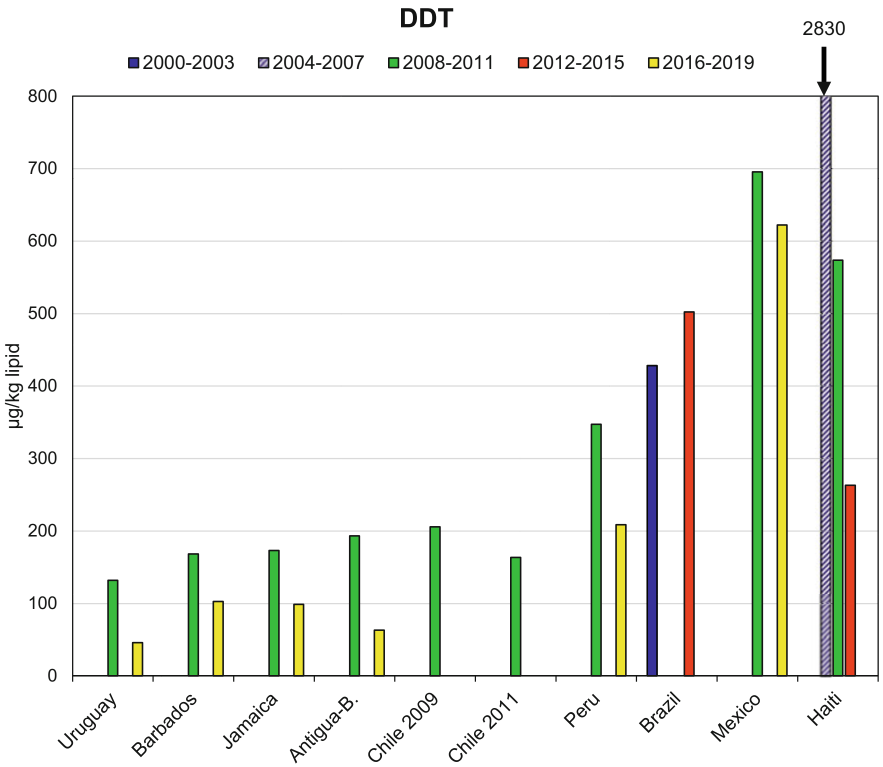

Fig. 6 Overview of the development of DDT concentrations in human milk (expressed as μg DDT complex/kg lipid; aggregated data) over time in Latin American and Caribbean Countries with repeated participation between 2000 and 2019

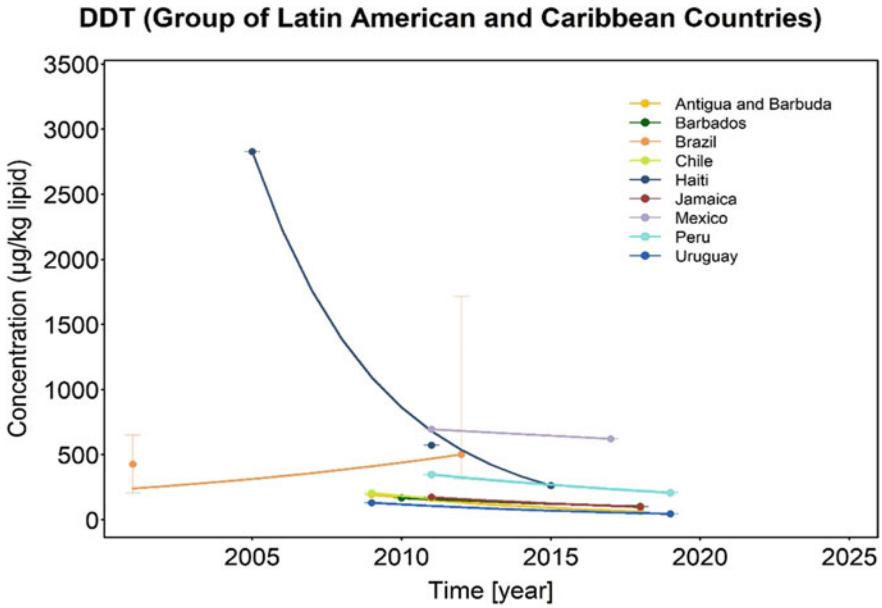

Fig. 7 Temporal tendencies of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) in Latin American and Caribbean Countries with repeated participation between 2000 and 2019 using the Theil–Sen method

Table 4 Overall decrease (%) of DDT concentrations in human milk in Latin American and Caribbean Countries per 1 year, 5 years and 10 years (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Trend *p*-value overall
Antigua-Barb	11.7	46.3	71.2	1.000
Barbados	6.0	26.5	46.0	1.000
Brazil	−6.9	−39.9	−95.8	0.220
Chile	10.8	43.6	68.2	1.000
Haiti	21.1	69.5	90.7	0.250
Jamaica	7.7	32.9	55.0	1.000
Mexico	1.8	8.8	16.9	1.000
Peru	6.2	27.3	47.1	1.000
Uruguay	10.0	40.9	65.1	1.000
Median	7.7	32.9	55.0	

for Brazil by the Theil–Sen method reflects the high variation within the two sampling periods (182 µg DDT complex/kg lipid and 675 µg DDT complex/kg lipid for the two analysed pooled samples from 2001; 291 µg DDT complex/kg lipid, 502 µg DDT complex/kg lipid and 1850 µg DDT complex/kg lipid for the three national pools from 2012). The median of the decreases (*decrease rate constants*) per 10 years of 55% for all countries is in line with the statistically significant decrease over 10 years for all GRULAC countries of 50% calculated by the Theil–Sen method (see Sect. 3.1).

3.5 Eastern European Group

Figures 8 (for aggregated data, see subsection 2.3) and 9 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of DDT concentrations in 6 countries of the Eastern European Group with repeated participation between 2000 and 2019. Overall decrease rates per 1 year, 5 years and 10 years are given in Table 5.

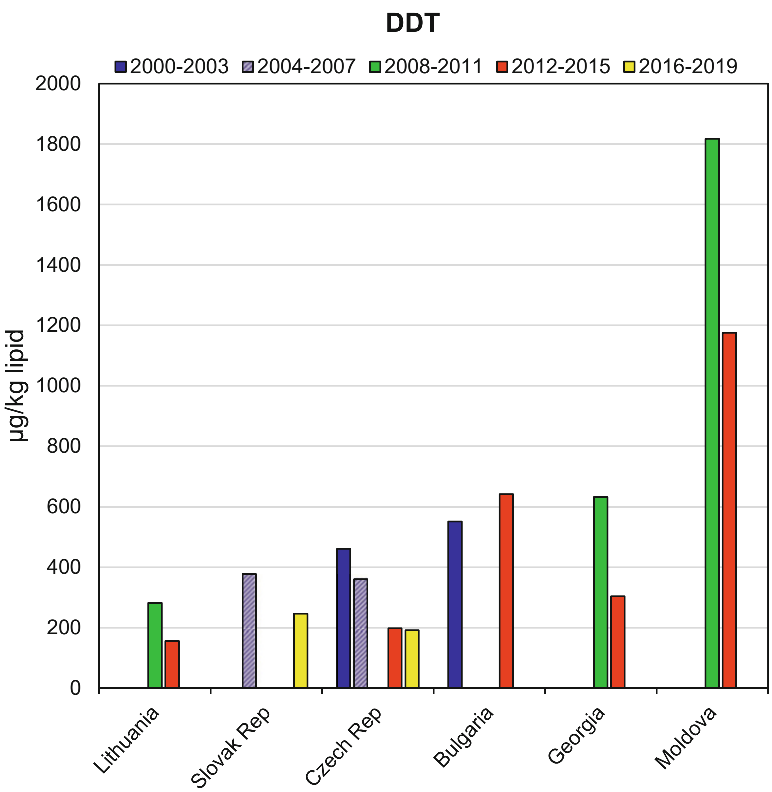

Fig. 8 Overview of the development of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid; aggregated data) over time in countries of the Eastern European Group with repeated participation between 2000 and 2019

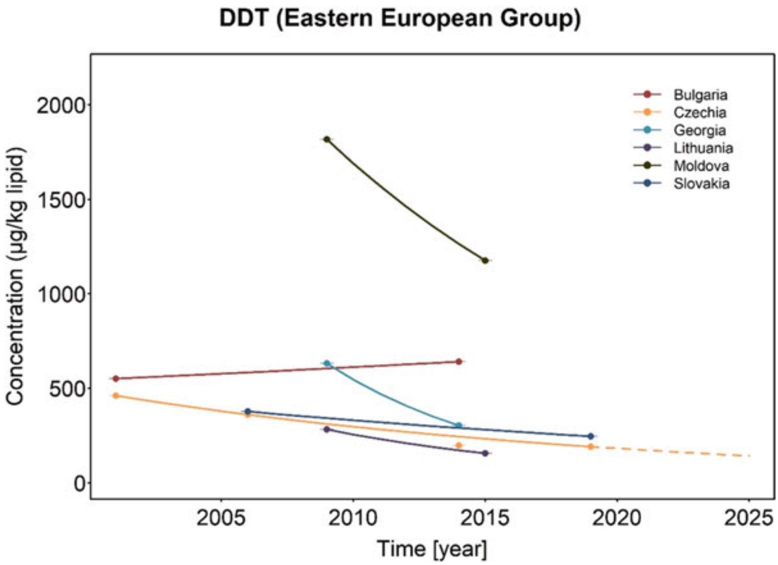

Fig. 9 Temporal tendencies of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) in countries of the Eastern European Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Czechia)

Table 5 Overall decrease (%) of DDT concentrations in human milk in countries of the Eastern European Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [µg/kg lipid]	Trend *p*-value overall
Bulgaria	−1.2	−6.0	−12.5		1.000
Czechia	4.8	21.7	38.6	143	0.031
Georgia	13.6	51.9	76.9		1.000
Lithuania	9.4	38.9	62.7		1.000
Moldova	7.0	30.4	51.6		1.000
Slovakia	3.2	15.2	28.0		1.000
Median	5.9	26.2	45.5		

A continuous decrease is observed in nearly all countries over all periods. Most data points are available for Czechia (Czech Republic) with four participations between the 2001 and 2019. Here, DDT concentrations in human milk fell from initially 461 µg DDT complex/kg lipid in 2001 by 57% until 2012 and then levelled out remaining quite constant until 2019. The highest concentration found in Moldova in 2009 (1820 µg DDT complex/kg lipid) decreased by 35% until 2015. In Bulgaria, DDT levels increased by 16% from 2001 to 2014.

The median of the reduction rates in the DDT levels within a 10-year period was 45%, with a wide range between countries. In Bulgaria, a slight increase was found, which was statistically not significant. Statistically significant was the decrease over 10 years of 39% in Czechia; for this country, also a prognosis of the estimated DDT concentration in 2025 was calculated. For the other Eastern European countries, the (statistically not significant) decrease over 10 years was between 28% and 77%.

3.6 Western European and Others Group (WEOG)

Figures 10 (for aggregated data) and 11 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of DDT concentrations in 8 countries of the Western European and Others Group with repeated participation between 2000 and 2019. Western European countries had in comparison to other UN regions quite low DDT concentrations, probably due to early bans on the use of DDT in agriculture implemented in most of these countries. A continuous decrease is observed in the WEOG region nearly over all periods. In Germany, which covers the whole period between 2000 and 2019, a

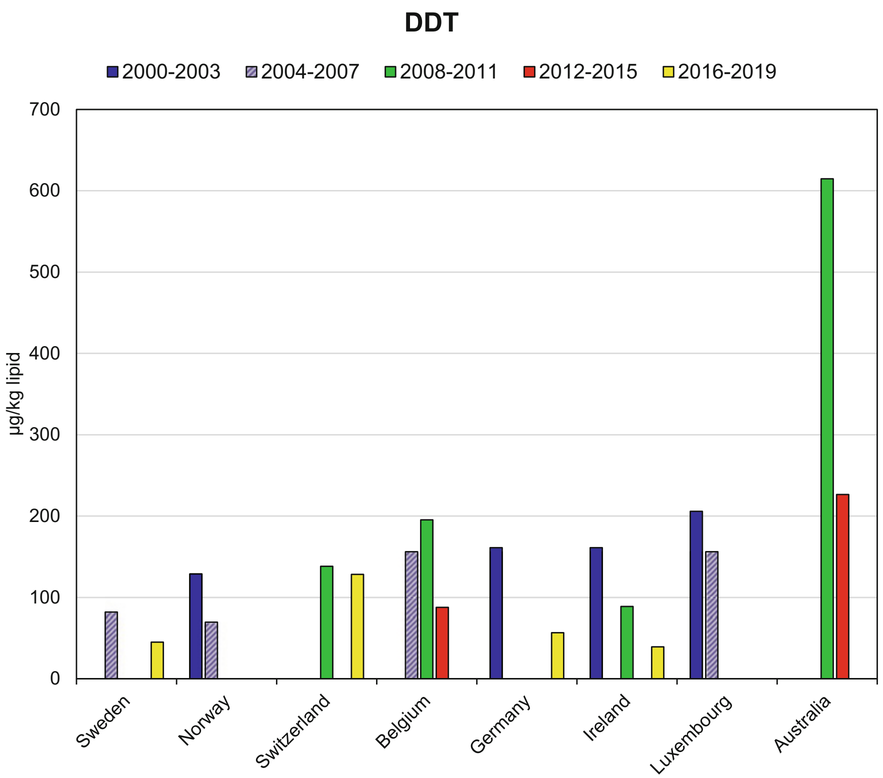

Fig. 10 Overview of the development of DDT concentrations in human milk (expressed as μg DDT complex/kg lipid; aggregated data) over time in countries of the Western European and Others Group with repeated participation between 2000 and 2019

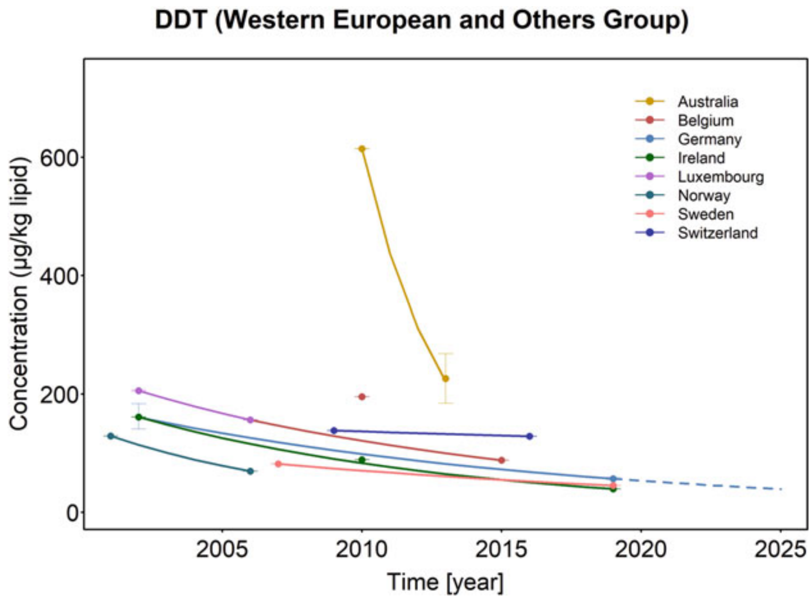

Fig. 11 Temporal tendencies of DDT concentrations in human milk (expressed as µg DDT complex/kg lipid) in countries of the Western European and Others Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Germany)

decrease of DDT concentrations in human milk from 161 µg DDT complex/kg in 2002 (as median of 4 samples) by 65% to 2019 was achieved, with a statistically significant decrease rate over 10 years of 46%. Based on this, also a prognosis of the estimated concentrations in 2025 was derived for Germany (Table 6).

Table 6 Overall decrease (%) of DDT concentrations in human milk in countries of the Western European and Others Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [µg/kg lipid]	Trend *p*-value overall
Australia	28.8	81.7	96.7		0.5
Belgium	6.2	27.4	47.3		0.5
Germany	5.9	26.4	45.8	39.3	0.008
Ireland	7.9	33.9	56.3		0.25
Luxembourg	6.7	29.2	49.9		0.5
Norway	11.6	46.0	70.8		1.000
Sweden	4.9	22.0	39.2		1.000
Switzerland	1.1	5.2	10.1		1.000
Median	6.4	28.3	48.6		

The median of the (statistically not significant) reduction rates for the other 6 Western European countries per 10 years was 49% (range 10–71%) (Table 6). With 97%, the highest decrease over 10 years was found in Australia, which had the highest DDT concentrations in human milk in the WEOG countries (615 μg DDT complex/kg; 2010). For comparison, p,p′-DDE concentrations of 311 ± 174 ng/g lipid were found in a comprehensive study in Australia in 2002–2003 (Mueller et al. 2008). These findings for temporal tendencies are again an indication that the decrease might be higher in areas with initially elevated concentrations and might get lower and finally level out over time when measures were taken to eliminate sources.

3.7 Dependence of Decrease (Decrease Rate Constants) on Concentration

The decrease (*decrease rate constants*) within a 10-year period have to be seen also in context with the concentration range: A differentiation of levels above or in the range of background contamination seems to be advised. If high levels are found, sources might be detected which could be eliminated. For source control, an understanding of country- and chemical-specific use is necessary. Directly after a ban or other measures taken to reduce discharges, concentrations of pesticides can be expected to decrease relatively fast in environmental samples (UNEP 2007, 2015, 2019). However, at background levels, other factors, e.g. contamination of feed and food by air via long-range transport and subsequent bioaccumulation, cannot be influenced locally, and some fluctuation over time might be expected when concentrations tend to level out.

DDT concentrations in human milk in the 44 countries with repeated participation between 2000 and 2019 comprise a wide range between 17 μg DDT complex/kg lipid and 23,500 μg DDT complex/kg lipid. This was grouped into four ranges (<200; 200–500; 500–2000; >2000) to check the dependence of the decrease rates on the "initial" (the first measured) concentration in a country. The lower end of the frequency distribution was derived from Western European countries, which had in comparison to other UN regions quite low DDT concentrations (many samples <200 μg DDT complex/kg lipid), probably due to early bans on the use of DDT in agriculture implemented in most of these countries. The upper end is related to a more recent use.

Table 7 shows, that the reduction rates over 10 year in the upper part for all three samples with concentrations above 2000 μg DDT complex/kg lipid (decrease in the range 79–91%) are considerably higher than in the lower part (for 13 samples; range 10–71% for decrease of samples <200 μg DDT complex/kg lipid; range 56–193 μg DDT complex/kg lipid and thus two orders of magnitude above LOQ [0.5 μg/kg lipid]). Figure 12 illustrates this dependence of the decrease over 10 years in 44 countries on the concentration range with repeated participation between 2000 and 2019.

Table 7 Overall decrease (%) of DDT concentrations (expressed as μg DDT complex/kg lipid) in human milk over 10 years calculated by the Theil–Sen method and their dependence on the concentration range. Negative decreases are to be read as increase

	<200 μg/kg lipid	200–500 μg/kg lipid	500–2000 μg/kg lipid	>2000 μg/kg lipid
N	13	13	15	3
min	10	−96	−13	79
median	49	63	77	82
max	71	84	97	91

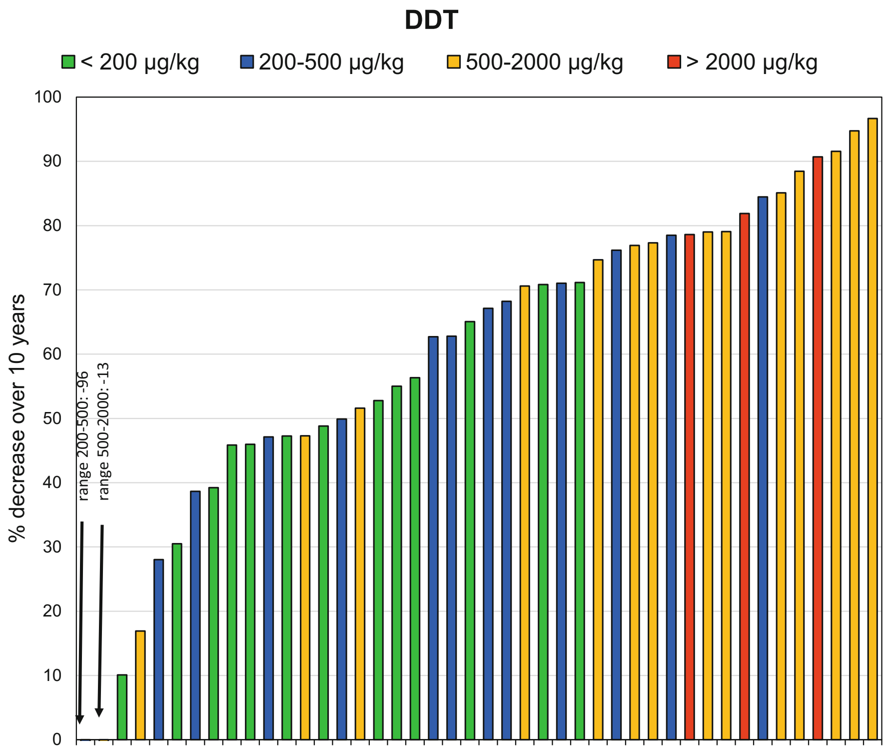

Fig. 12 Dependence of the decrease over 10 years (calculated by the Theil–Sen method) for DDT on concentrations in human milk in 44 countries with repeated participation between 2000 and 2019, with differentiation into four ranges of concentration (<200; 200–500; 500–2000; >2000 μg DDT complex/kg lipid)

4 Beta-Hexachlorocyclohexane (beta-HCH)

4.1 Global Level and Comparison between UN Regional Groups

Large differences of beta-HCH concentrations in 119 pooled human milk samples from 44 countries with repeated participation between 2000 and 2019 were found, with a minimum of <0.5 μg beta-HCH/kg lipid found in few countries and a maximum of 1020 μg beta-HCH/kg lipid found in 2002 (median: 6.0 μg/kg). This is the same range as found in 134 country results (based on aggregated data) of 82 countries (as total number regardless the number of participations between 2000 and 2019), with a median of 5.9 μg beta-HCH/kg lipid (Malisch et al. 2023b).

An overall decrease within a 10-year period between 50% and 98% was achieved for beta-HCH levels in all the UN regions and at a global level. The overall reduction rates per 1 year and 10 years calculated by the Theil–Sen method for exponential trends and the additionally applied median method to derive time trends were quite comparable in nearly all UN regions and at the global level (Table 8). On a global level and in all UN regions except Latin America and the Caribbean, all trends were significant (*p*-value <0.001 in three regions and globally, 0.037 in Eastern Europe).

The variation of concentration ranges and reduction rates between countries in the five UN Regional Groups is shown in the following subsections.

The exponential trends of beta-HCH concentrations in human milk derived by the Theil–Sen method in the five UN regions and worldwide by combination of data from countries are illustrated in Fig. 13a–f. These figures are normalized according to the maximum concentration found in the respective UN Regional Groups. Thus, the different scales illustrate the different ranges between and within the UN Regional Groups. For a detailed discussion of the regional data, see the following subsections 4.2–4.6.

Table 8 Overall decrease (%) of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid) in the five UN Regional Groups and worldwide (computed using all samples from countries with repeated participation)

UN Regional Group	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall[a]
		Theil–Sen method	Median method	Theil–Sen method	Median method	
Africa	13	16.0	14.2	82.5	78.4	<0.001
Asia-Pacific	8	31.0	13.3	97.5	76.1	<0.001
Latin America and Caribbean	9	7.0	6.3	51.9	48.0	0.105
Eastern Europe	6	9.6	7.8	63.4	55.5	0.037
Western Europe and Others	8	9.5	10.2	62.9	65.8	<0.001
Global	44	16.5	9.6	83.5	63.4	<0.001

[a] for Theil–Sen method

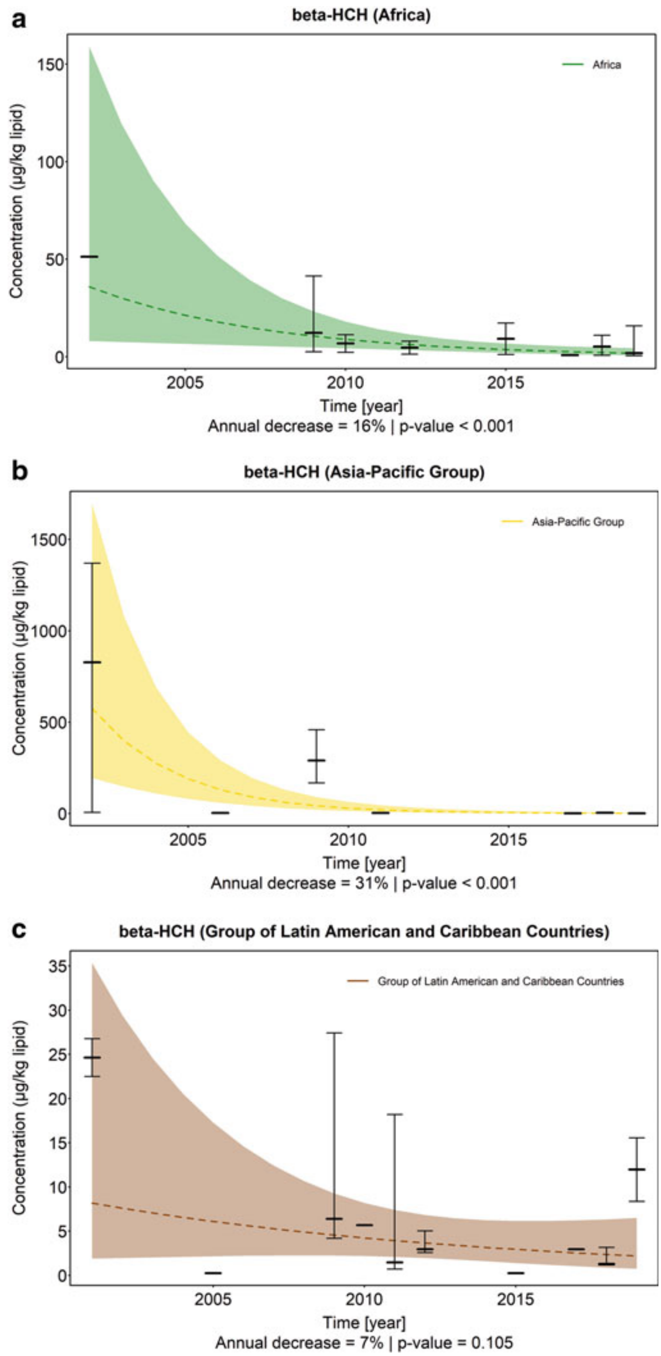

Fig. 13 (**a**–**f**) Theil–Sen exponential trends of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid) worldwide and in the five UN regions. The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, whiskers show ranges between fifth and 95th percentiles

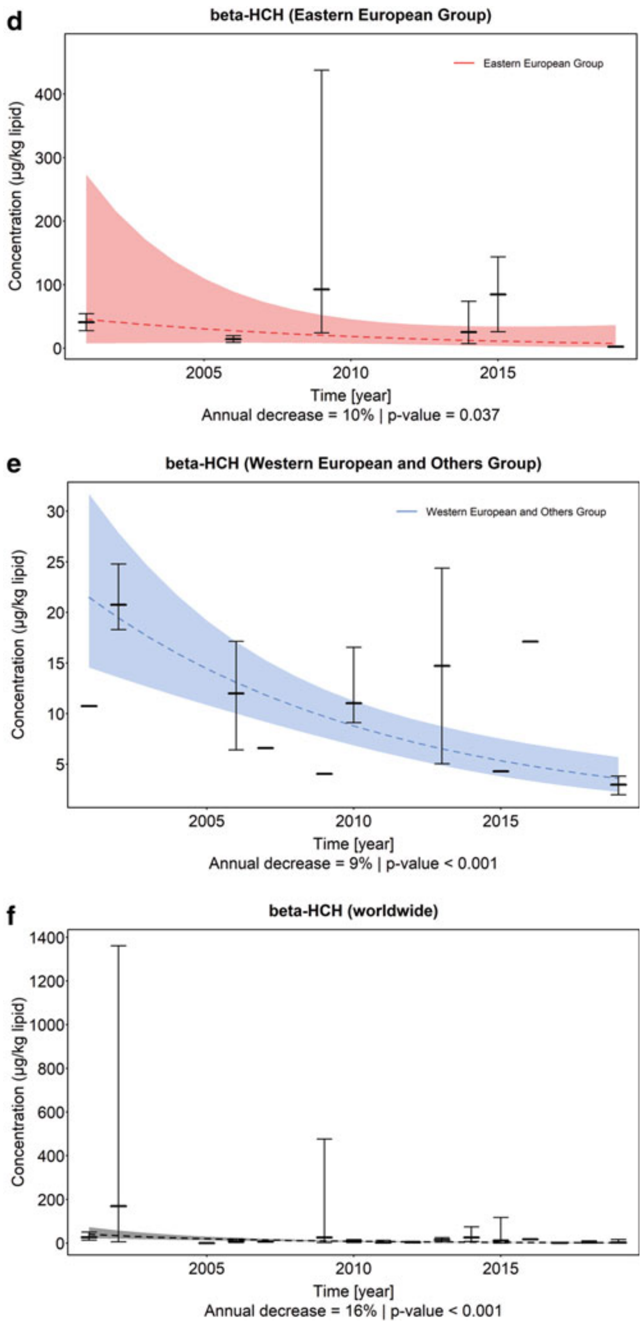

Fig. 13 (continued)

4.2 African Group

Figures 14 (for aggregated data) and 15 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of beta-HCH concentrations in 13 countries from Africa with repeated participation between 2000 and 2019. In Egypt, a reduction of 62% was observed from 51.3 μg/kg lipid in the 2000–2003 period to the 2016–2019 period. Most countries participated for the first time in the 2008–2011 period (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 on average by about 63% until the period 2016–2019 (decreases in the range between 43% and 92%). In Côte d'Ivoire, which was in the middle of the frequency distribution of beta-HCH concentrations in Africa, levels increased from 11.8 μg/kg lipid in 2010 to 2015 by 54%.

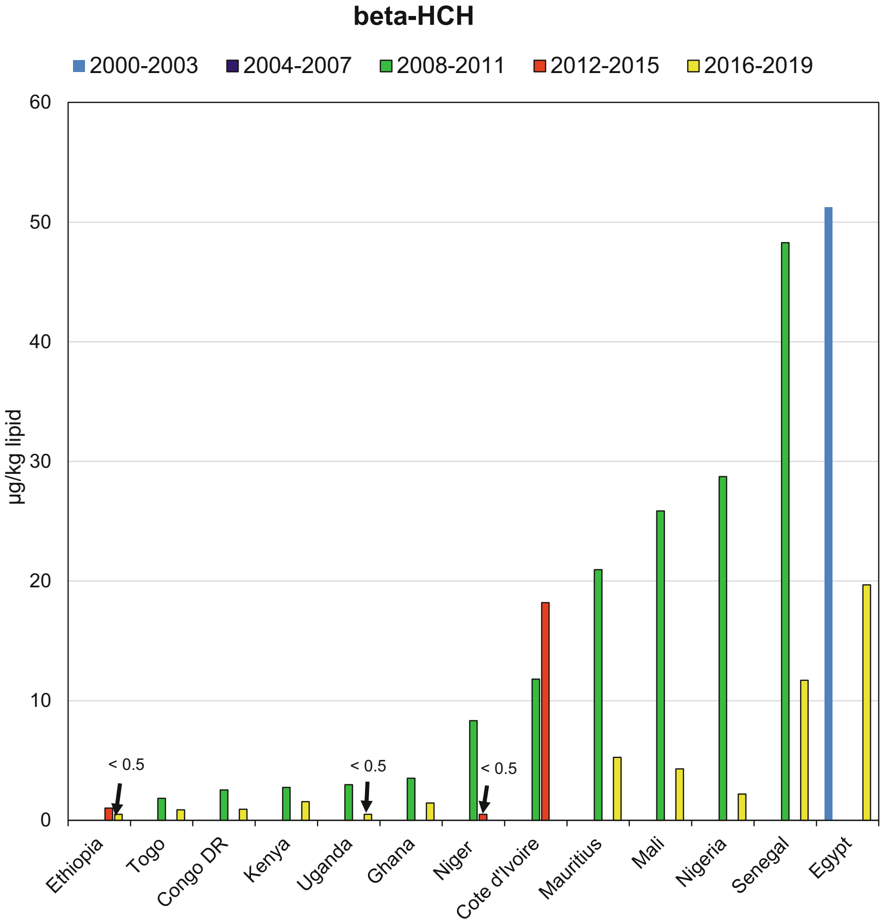

Fig. 14 Overview of the development of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid; aggregated data) over time in African countries with repeated participation between 2000 and 2019

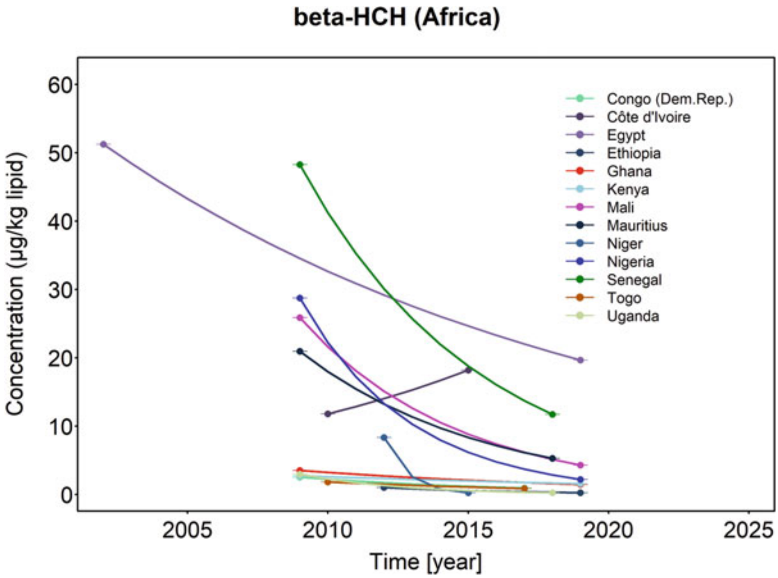

Fig. 15 Temporal tendencies of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) in African countries with repeated participation between 2000 and 2019 using the Theil–Sen method

Table 9 Overall decrease (%) of beta-HCH concentrations in human milk per 1 year, 5 years 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 5 years	Overall decrease (%) per 10 years	Trend *p*-value overall
Congo (DR)	11.8	46.6	71.5	1.000
Côte d'Ivoire	−9.0	−54.2	−137.8	1.000
Egypt	5.5	24.5	43.1	1.000
Ethiopia	18.2	63.4	86.6	1.000
Ghana	8.5	35.7	58.7	1.000
Kenya	5.5	24.5	43.1	1.000
Mali	16.4	59.2	83.4	1.000
Mauritius	14.2	53.6	78.4	1.000
Niger	68.9	99.7	100.0	1.000
Nigeria	22.6	72.3	92.3	1.000
Senegal	14.6	54.5	79.3	1.000
Togo	9.9	40.7	64.9	1.000
Uganda	24.1	74.8	93.6	1.000
Median	14.2	53.6	78.4	

The overall decreases per 1 year, 5 years and 10 years are given in Table 9. The limited number of samples did not allow to determine statistically significant decreases. In all African countries except Côte d'Ivoire, the decreases in the levels of beta-HCH within a 10-year period were in the range between 43% and 100%. Similar decreases were found as well at the higher end of the frequency distribution of concentrations as at the lower end in the range of background contamination. The median of 78% is in line with the statistically significant ($p < 0.001$) decrease over 10 years for all African countries of 83% calculated by the Theil–Sen method (see Sect. 4.1).

4.3 Asia-Pacific Group

Figures 16 (for aggregated data) and 17a–b (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of beta-HCH concentrations in 8 countries from the Asia-Pacific Group with repeated participation between 2000 and 2019. Overall decrease rates per 1 year, 5 years and 10 years are given in Table 10.

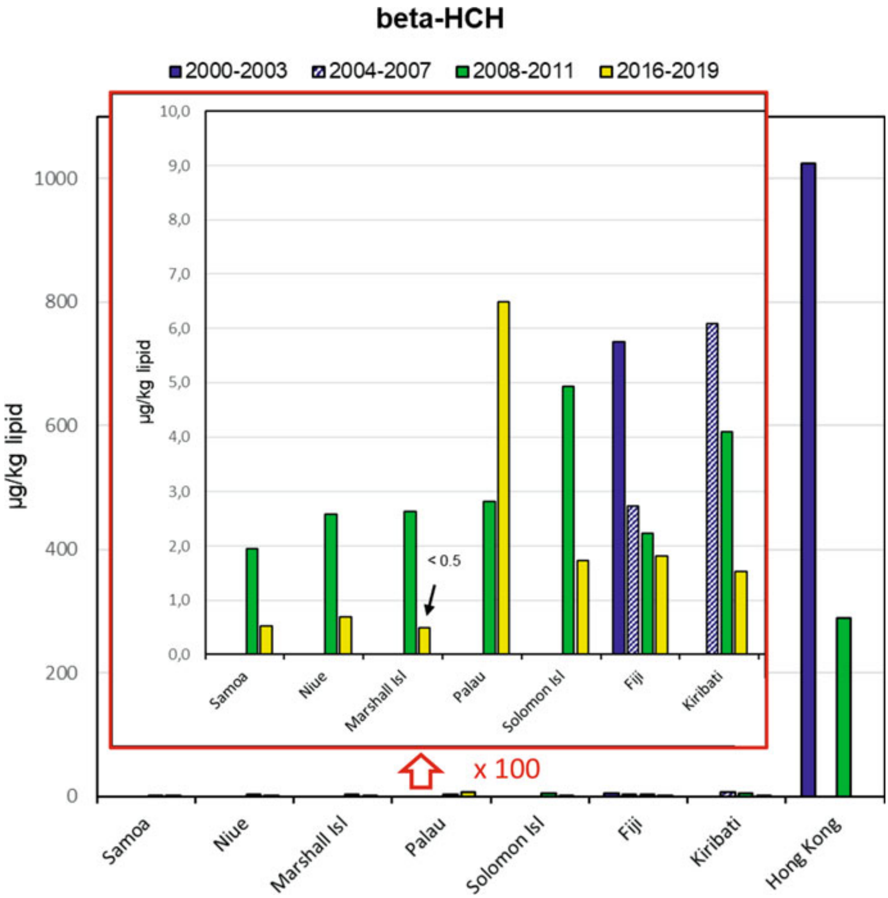

Fig. 16 Overview of the development of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid; aggregated data) over time in countries of the Asia-Pacific Group with repeated participation between 2000 and 2019

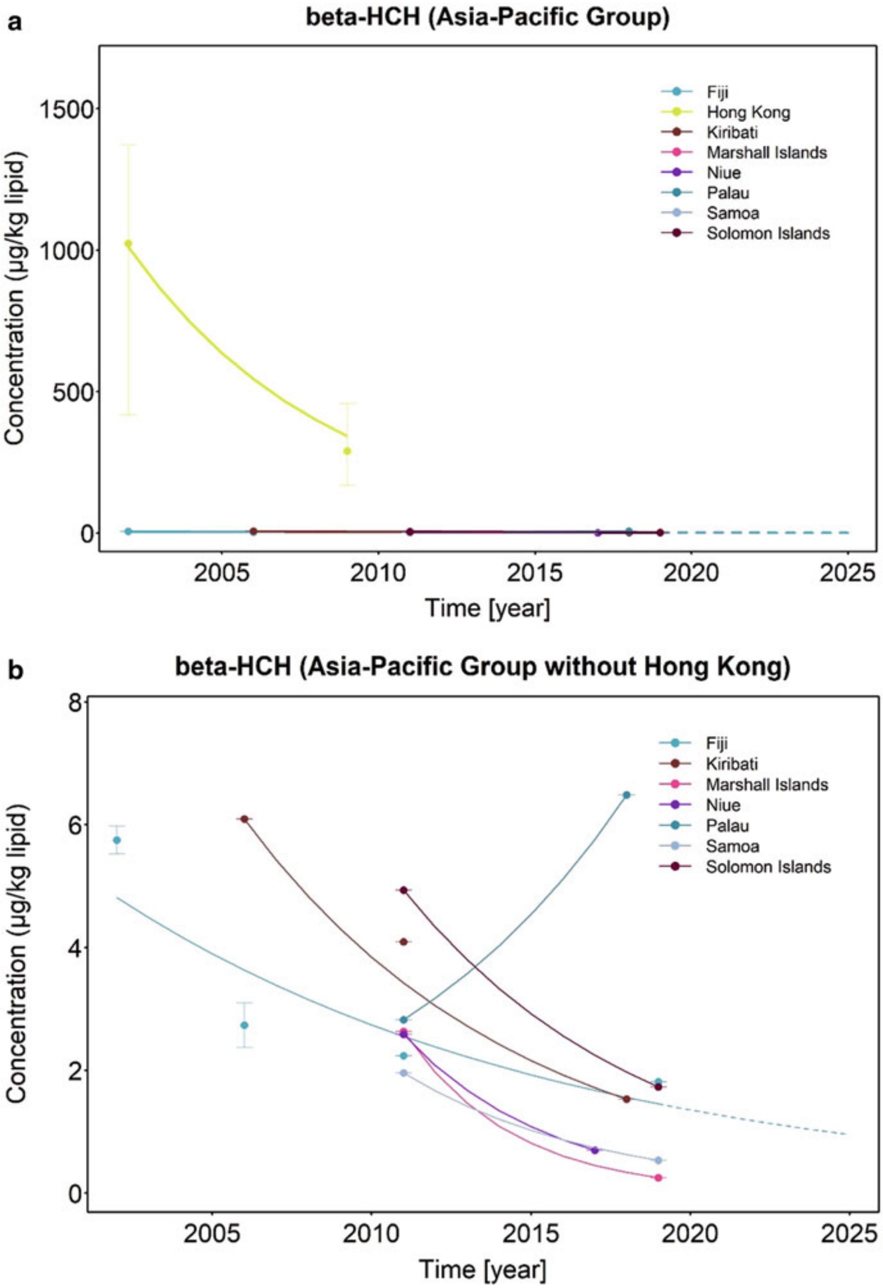

Fig. 17 (**a–b**) Temporal tendencies of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) in (**a**) all countries of the Asia-Pacific Group and (**b**) Pacific Islands countries with repeated participation between 2000 and 2019 using the Theil–Sen method (with statistically significant time trends in Hong Kong SAR of China and Fiji)

Table 10 Overall decrease (%) of beta-HCH concentrations in human milk in countries of the Asia-Pacific Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025[μg/kg lipid]	Trend *p*-value overall
Fiji	6.8	29.7	50.6	0.95	< 0.001
Hong Kong	14.4	53.9	78.8		< 0.001
Kiribati	10.9	43.8	68.4		0.25
Marshall Isl	25.5	77.0	94.7		1.000
Niue	19.6	66.4	88.7		1.000
Palau	−12.6	−81.2	−228.4		1.000
Samoa	15.0	55.6	80.3		1.000
Solomon Isl	12.3	48.1	73.1		1.000
Median	13.3	51.1	76.1		

The highest concentration found in Hong Kong SAR of China in 2002 (1020 μg beta-HCH/kg lipid as the median of 10 pooled samples [Hedley et al. 2010]) decreased until 2009 by 78% to 290 μg/kg as median of 4 samples from different population groups (two age groups [above or below 30 years] which had been living in Hong Kong for either more or less than 10 years).

In comparison to these concentration ranges, beta-HCH levels in human milk from 7 Pacific Islands countries were lower by about a factor of 100 and more and in the range of low background contamination between <0.5 μg/kg lipid and 6 μg/kg lipid over the whole period between 2000 and 2019. With this, all countries of the Pacific region were among the countries with the lowest beta-HCH concentrations in human milk on a global level. As in the African countries, downtrends were also seen at these comparably low concentration ranges. In Fiji, a reduction of nearly 70% was observed from 5.8 μg beta-HCH/kg lipid in 2002 until 2019. Most other countries participated the first time in the period 2008–2011 and Kiribati in 2004–2007. In nearly all of these countries, a decrease in the range of 65–81% was observed in the following years, whereas in Palau, beta-HCH concentrations increased from the relatively low level of 2.8 μg/kg in 2002 to 6.5 μg/kg in 2019.

A statistically significant decrease in the levels of beta-HCH within a 10-year period of 79% was achieved for the initially high levels by Hong Kong SAR of China. For Fiji with a statistically significant decrease of 51% over 10 years and participation also in the 2016–2019 round, in addition the concentration in 2025 was estimated.

High reduction rates were also found in the range of lower background contamination of other Pacific Islands countries, with exception of Palau. However, due to the limited number of pooled samples per country, these decreases are not statistically significant (see subsection 2.3). Furthermore, the variation should be seen in context with the advised differentiation of levels above or—as in these cases—in the range of background contamination (approximately <5 μg beta-HCH/kg lipid), as explained in subsection 3.7.

4.4 Group of Latin American and Caribbean Countries (GRULAC)

Figures 18 (for aggregated data) and 19 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of beta-HCH concentrations in 9 Latin American and Caribbean countries with repeated participation between 2000 and 2019. In Brazil, a decrease of median concentrations by 88% was found from 2001 to 2012. In Haiti, beta-HCH concentrations in human milk remained constantly low in the range of the limit of quantification (0.5 μg/kg) from 2004 to 2015. The other countries participated for the first time in the period 2008–2011. In nearly all of them, a decrease was observed in the following years (range 41–67%). In Jamaica and Mexico with beta-HCH concentrations in the lower background range below 3 μg/kg lipid, the levels remained quite constant.

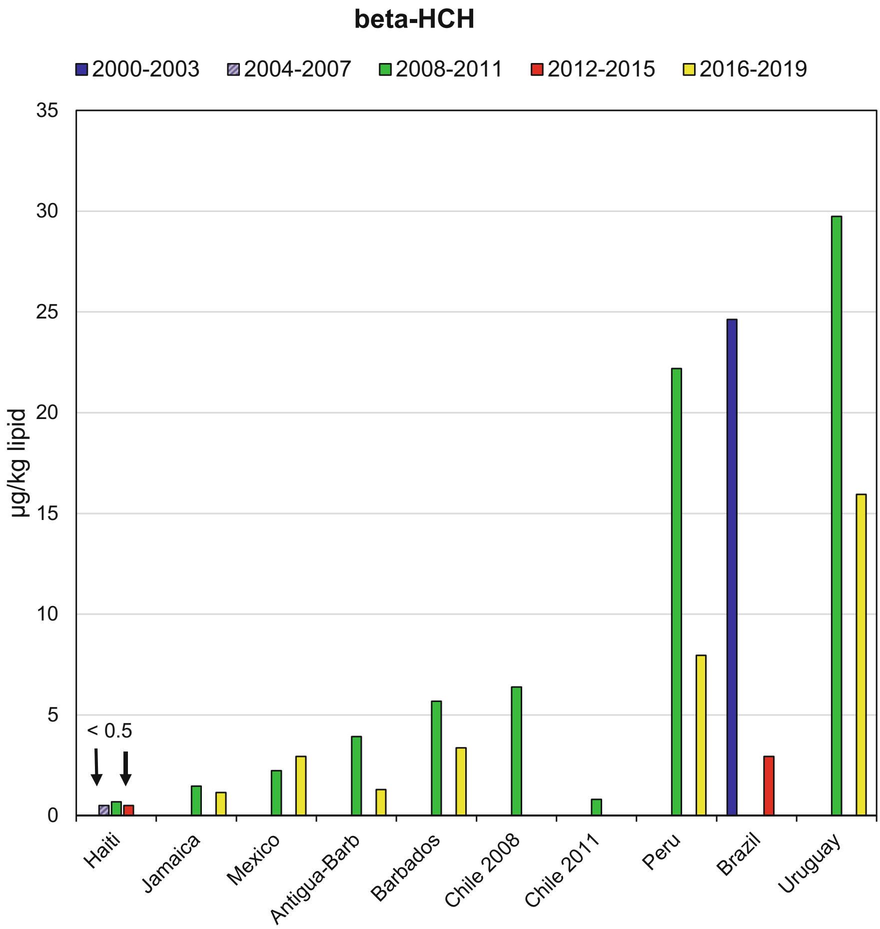

Fig. 18 Overview of the development of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid; aggregated data) over time in Latin American and Caribbean Countries with repeated participation between 2000 and 2019

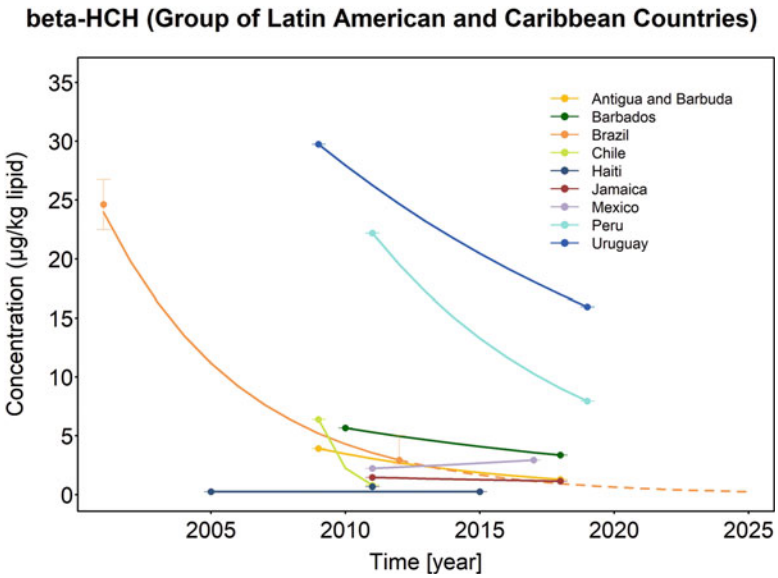

Fig. 19 Temporal tendencies of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) in Latin American and Caribbean Countries with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Brazil)

These findings are also reflected in the overall decreases per 1 year, 5 years and 10 years (Table 11). In all countries except Brazil, the limited number of pooled samples did not allow to determine statistically significant reduction rates. A decrease in the beta-HCH levels within a 10-year period between 46% and 85% was seen in countries at the upper end of the frequency distribution (Brazil, Peru and

Table 11 Overall decrease (%) of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) per 1 year, 5 years and 10 years in Latin American and Caribbean Countries and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [µg/kg lipid]	Trend *p*-value overall
Antigua-Barb.	11.6	46.1	71.0		1.000
Barbados	6.3	27.9	48.0		1.000
Brazil	17.4	61.5	85.2	0.25	0.031
Chile	64.6	99.4	100.0		1.000
Haiti	0.0	0.0	0.0		1.000
Jamaica	3.3	15.7	28.9		1.000
Mexico	−4.7	−25.9	−58.6		1.000
Peru	12.0	47.3	72.2		1.000
Uruguay	6.0	26.8	46.4		1.000
Median	6.3	27.9	48.0		

Uruguay), whereas in the other countries with beta-HCH concentrations mostly in the range of the background contamination (<5 μg/kg), a wide range of reduction rates was found. Therefore, as explained above for the Asia-Pacific countries, a differentiation of levels above or in the range of background contamination seems to be advised. The calculated tendencies for Haiti, Mexico and Jamaica show the variation at low background levels over time.

4.5 Eastern European Group

Figures 20 (for aggregated data) and 21 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of beta-HCH concentrations in 6 countries of the Eastern European Group with

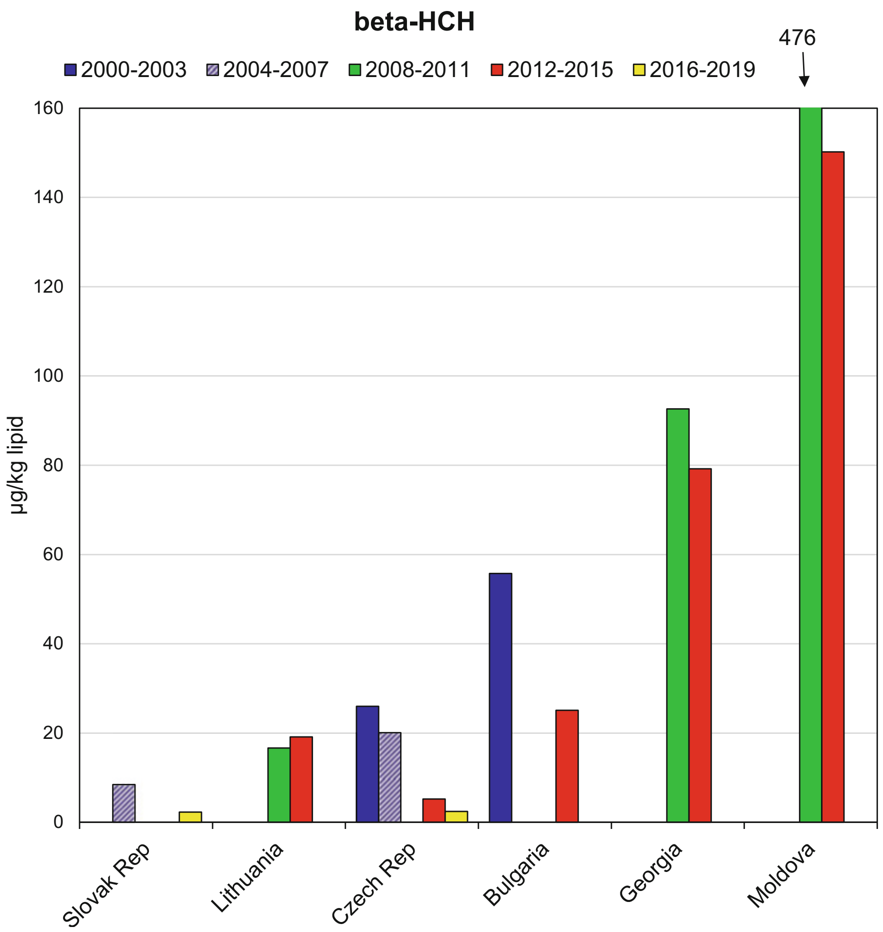

Fig. 20 Overview of the development of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid; aggregated data) over time in countries of the Eastern European Group with repeated participation between 2000 and 2019

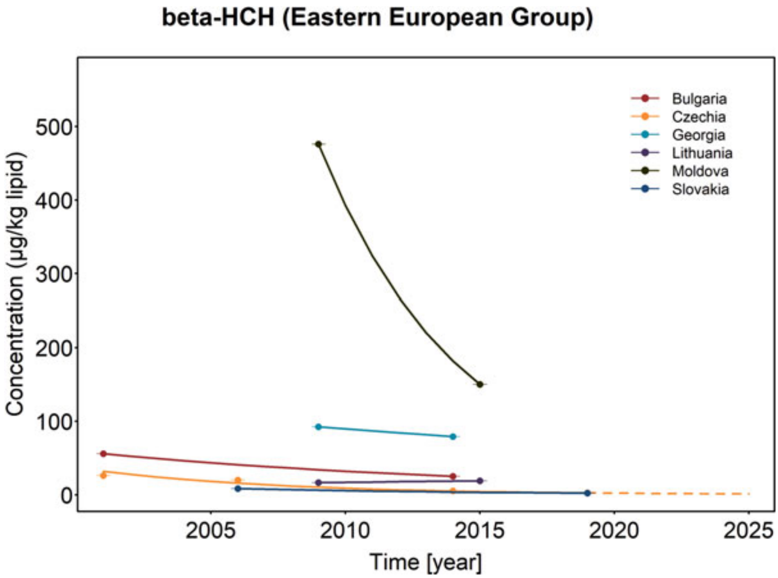

Fig. 21 Temporal tendencies of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid) in countries of the Eastern European Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Czechia)

Table 12 Overall decrease (%) of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid) in countries of the Eastern European Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [ng/g lipid]	Trend *p*-value overall
Bulgaria	5.9	26.4	45.8		1.000
Czechia	13.2	50.6	75.6	1.08	0.031
Georgia	3.1	14.5	26.8		1.000
Lithuania	−2.4	−12.4	−26.4		1.000
Moldova	17.5	61.8	85.4		1.000
Slovakia	9.6	39.5	63.4		1.000
Median	7.8	33.3	55.5		

repeated participation between 2000 and 2019. A continuous decrease is observed in nearly all countries over all periods. Most data points are available for the Czech Republic with four participations between the 2001 and 2019. Here, beta-HCH concentrations in human milk fell from 26 μg/kg lipid by 91% until 2019. The highest concentration found in Moldova in 2009 (476 μg beta-HCH/kg lipid) decreased by 68% until 2015.

The overall decreases per 1 year, 5 years and 10 years are given in Table 12. A statistically significant decrease over 10 years of 76% was observed in Czechia; also for this country, a prognosis of the estimated beta-HCH concentration in 2025 was calculated. In all other countries, the limited number of samples did not allow to determine statistically significant reduction rates. For most Eastern European countries, the decrease over 10 years was between 27% and 85%. In Lithuania, beta-HCH concentrations slightly increased from 16.6 μg/kg lipid in 2009 by 15% until 2014.

4.6 Western European and Others Group (WEOG)

Figures 22 (for aggregated data) and 23 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes for beta-HCH in 8 countries of the Western European and Others Group with repeated participation between 2000 and 2019. In Germany, which covers the entire period between 2000 and 2019, a decrease of beta-HCH concentrations in human milk from 19.4 μg/kg lipid in 2002 by 84% to 2019 was achieved, with a statistically significant decrease over 10 years of 67%. Based on this, a prognosis of the estimated concentration in 2025 was derived for Germany (Table 13).

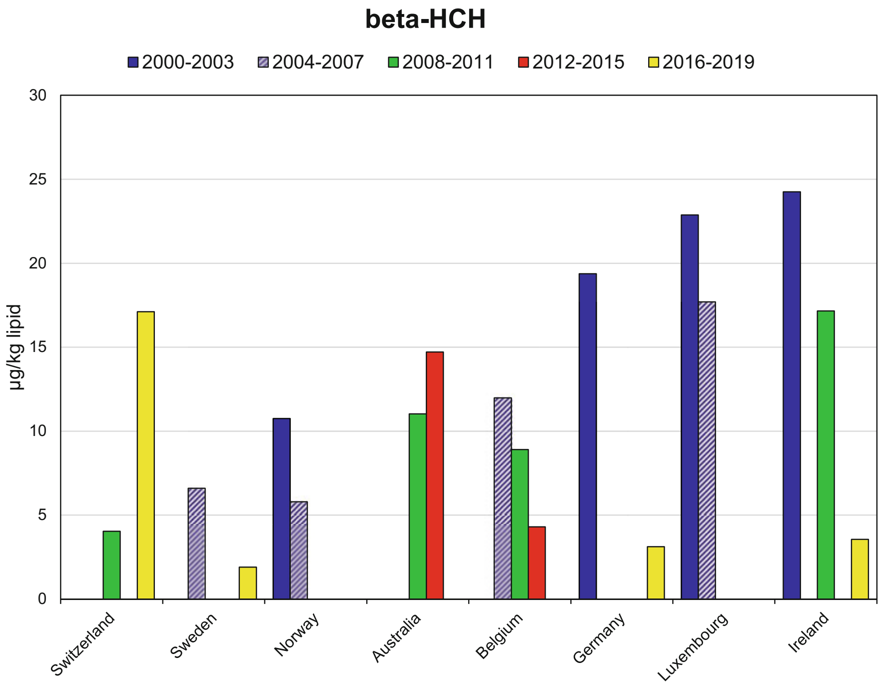

Fig. 22 Overview of the development of beta-HCH concentrations in human milk (expressed as μg beta-HCH/kg lipid; aggregated data) over time in countries of the Western European and Others Group with repeated participation between 2000 and 2019

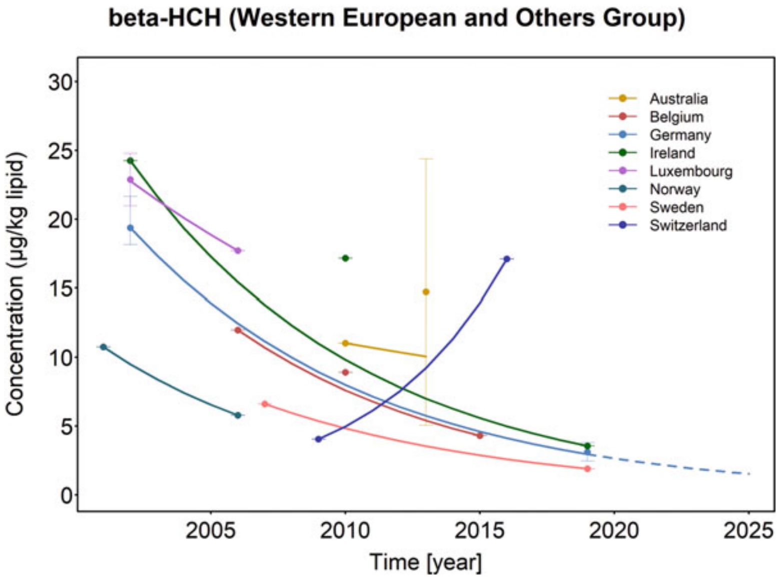

Fig. 23 Temporal tendencies of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) in countries of the Western European and Others Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Germany)

Table 13 Overall decrease (%) of beta-HCH concentrations in human milk (expressed as µg beta-HCH/kg lipid) in countries of the Western European and Others Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [ng/g lipid]	Trend *p*-value overall
Australia	3.0	14.2	26.5		1.000
Belgium	10.8	43.4	68.0		0.250
Germany	10.5	42.5	66.9	1.52	0.008
Ireland	10.7	43.2	67.7		0.250
Luxembourg	6.1	27.0	46.7		0.500
Norway	11.6	46.1	71.0		1.000
Sweden	9.9	40.5	64.6		1.000
Switzerland	−22.9	−180.5	−686.6		1.000
Median	10.2	41.5	65.8		

In all other countries, the limited number of pooled samples did not allow to determine statistically significant decreases. The median of the reduction rates per 10 years for five Western European countries (Belgium, Ireland, Luxembourg, Norway and Sweden) was 68% (range 47–71%). In Switzerland, beta-HCH concentrations increased from 4.0 µg/kg in 2009 to 17.1 µg/kg in 2016.

For Australia, data are available for 2010 (one pooled sample with 11 μg beta-HCH/kg lipid) and 2013 (two pooled samples with 25.5 μg beta-HCH/kg lipid and 4.0 μg beta-HCH/kg lipid, respectively). Therefore, the median of the two pooled samples from 2013 shows an increase, whereas based on the Theil–Sen method an overall decrease is calculated with regard to the two very different levels found in 2013 (Table 13; Fig. 23).

4.7 Dependence of Decrease (Decrease Rate Constants) on Concentration

Beta-HCH concentrations in human milk in the 44 countries with repeated participation between 2000 and 2019 comprise a wide range between <0.5 μg beta-HCH/kg lipid and 1020 μg beta-HCH/kg lipid. This was grouped into five ranges (<5; 5–10; 10–20; 20–100; >100 μg beta-HCH/kg lipid) to check the dependence of the decrease (*decrease rate constants*) on the "initial" (the first measured) concentration in a country. The lower end of the frequency distribution below 5 μg beta-HCH/kg lipid is considered as (low) background contamination. The upper end with the two highest levels (476 μg/kg; 2010 μg/kg) is related to a more recent use.

Table 14 shows, that the reduction rates over 10 year for beta-HCH concentrations above 20 μg/kg lipid vary less than in the lower part: For all samples above 20 μg/kg, only downward trends were observed, with a decrease of about 80% over 10 years for the two samples with highest concentrations (>100 μg/kg) and a wide variation in the range 20–100 μg/kg. In particular, samples in the low background range below 5 μg beta-HCH/kg lipid show a high variation of the reduction rates (range between −690% and 95%, median 68%, for 15 of the 16 pooled samples in this range below 5 μg beta-HCH/kg lipid, but above LOQ [0.5 μg/kg lipid] and one pooled sample below LOQ). At lower background levels, many factors, e.g. contamination of feed and food by air via long-range transport and subsequent bioaccumulation, cannot be influenced locally. It might be concluded that the large variation of reduction rates for beta-HCH concentrations in this lower background range limits the applicability of this parameter for assessments at the country-level, but allows a more general assessment of the temporal trends of background contamination.

Figure 24 illustrates this dependence of decrease rates over 10 years in 44 countries on the concentration range with repeated participation between 2000 and 2019.

Table 14 Overall decrease (%) of beta-HCH concentrations in human milk (μg/kg lipid) over 10 years calculated by the Theil–Sen method and their dependence on the concentration range. Negative decreases are to be read as increase

	<5 μg/kg	5–10 μg/kg	10–20 μg/kg	20–100 μg/kg	>100 μg/kg
N	16	7	6	13	2
min	−687	48	−138	27	79
median	68	65	47	72	82
max	95	100	71	92	85

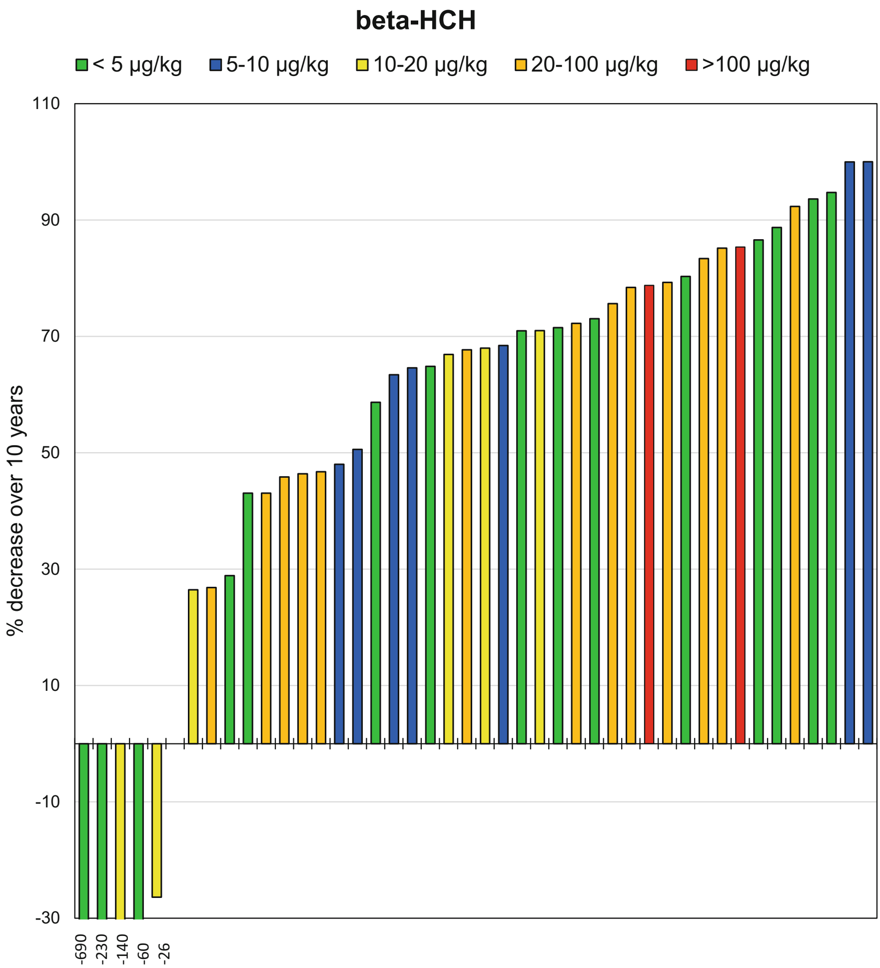

Fig. 24 Dependence of the decrease over 10 years (calculated by the Theil–Sen method) for beta-HCH on concentrations in human milk in 44 countries with repeated participation between 2000 and 2019, with differentiation into five ranges of concentration (<5; 5–10; 10–20; 20–100; >100 µg/kg lipid)

5 Hexachlorobenzene (HCB)

5.1 Global Level and Comparison Between UN Regional Groups

The maximum levels and therefore the ranges of HCB were much lower than found for DDT and beta-HCH, with a minimum of about 1–2 µg/kg lipid found in some countries and a maximum of 154 µg/kg lipid found in 2009 in an Eastern European

Table 15 Overall decrease (%) of the HCB concentration in human milk (expressed as μg/kg lipid) in the five UN Regional Groups and worldwide (computed using all samples from countries with repeated participation)

UN Regional Groups	*N* of countries	Overall decrease (%) per 1 year		Overall decrease (%) per 10 years		Trend *p*-value overall[a]
		Theil–Sen method	Median method	Theil–Sen method	Median method	
Africa	13	1.2	1.1	11.3	10.2	0.002
Asia-Pacific	8	10.5	6.5	67.2	49.1	<0.001
Latin America and Caribbean	9	3.6	6.6	30.7	49.6	<0.001
Eastern Europe	6	6.1	6.9	46.6	51.2	0.037
Western Europe and Others	8	5.8	1.9	44.8	17.3	<0.001
Global	44	8.0	5.8	56.7	44.8	<0.001

[a]For Theil–Sen method

country. The median of 119 country results from 44 countries with repeated participation was 6.1 μg/kg lipid. This is the same range as found in 134 country results (based on aggregated data) of 82 countries (as total number regardless the number of participations between 2000 and 2019), with a similar median of 5.1 μg/kg. Many results were in the lower background range, such as the results of all 19 countries from Africa over the whole period (Malisch et al. 2023b). As explained above (subsections 3.7 and 4.7), some fluctuation of decrease rates within a 10-year period might be expected when concentrations tend to level out.

In the African Group with all pooled samples collected over time being in the background range of 5 μg HCB/kg lipid or below, the overall decrease within 10 years was 11% calculated by the Theil–Sen method and 10% by the median method. Overall decreases between 30% and 67% were calculated by the Theil–Sen method for the other UN Regional Groups, and 57% on a global level. Overall decreases calculated by the median method are in the range between 17% and 50% in these UN Regional Groups, with 48% as median of the five UN regions and 26% as median of 44 countries (Table 15). The variation between countries in the UN Regional Groups is shown in the following subsections.

The exponential trends of HCB concentrations derived by the Theil–Sen method in the five UN regional groups and worldwide are illustrated in Fig. 25a–f. Again, these figures are normalized according to the maximum concentration found in the respective UN Regional Groups. Thus, the different scales illustrate the different ranges between and within the UN Regional Groups. For a detailed discussion of the regional data, see the following subsections 5.2–5.6.

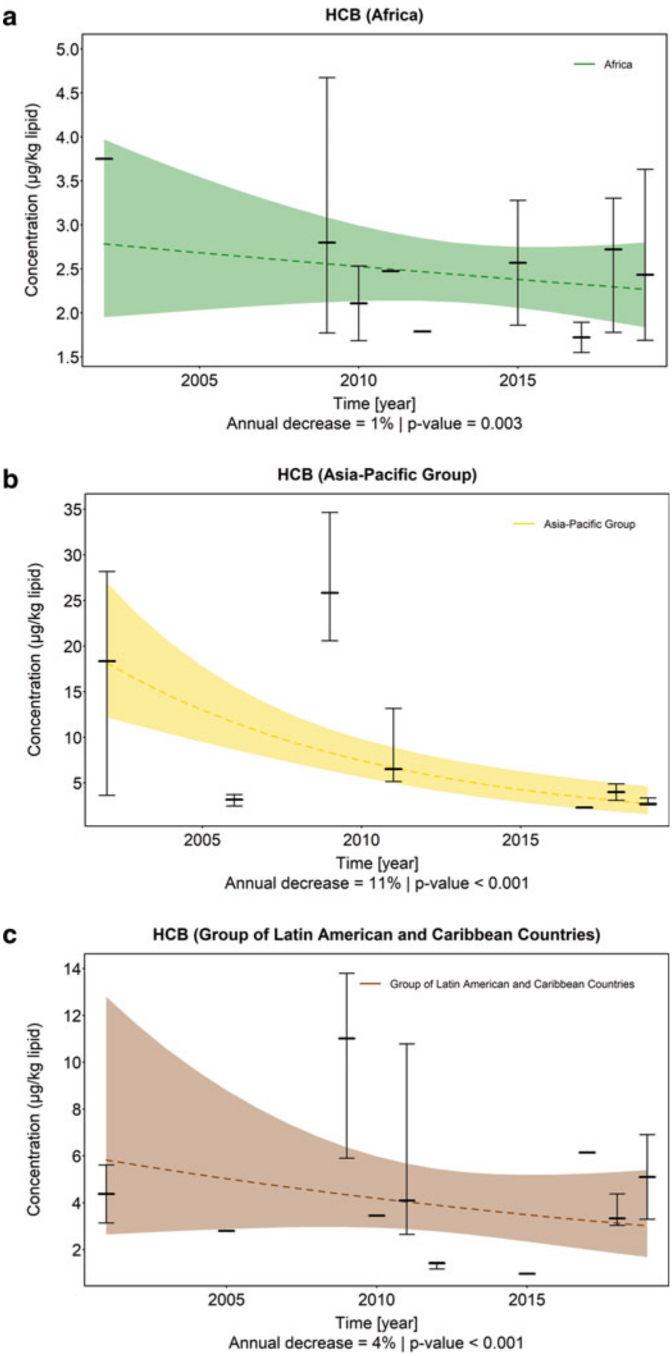

Fig. 25 (**a**–**f**) Theil–Sen exponential trends of the HCB concentration in human milk (expressed as µg/kg lipid) worldwide and in the five UN regions. The shaded area shows the 95% confidence interval of the trend; the thick black lines in the middle of the frequency distribution in a certain year show median concentrations in individual years, whiskers show ranges between fifth and 95th percentiles

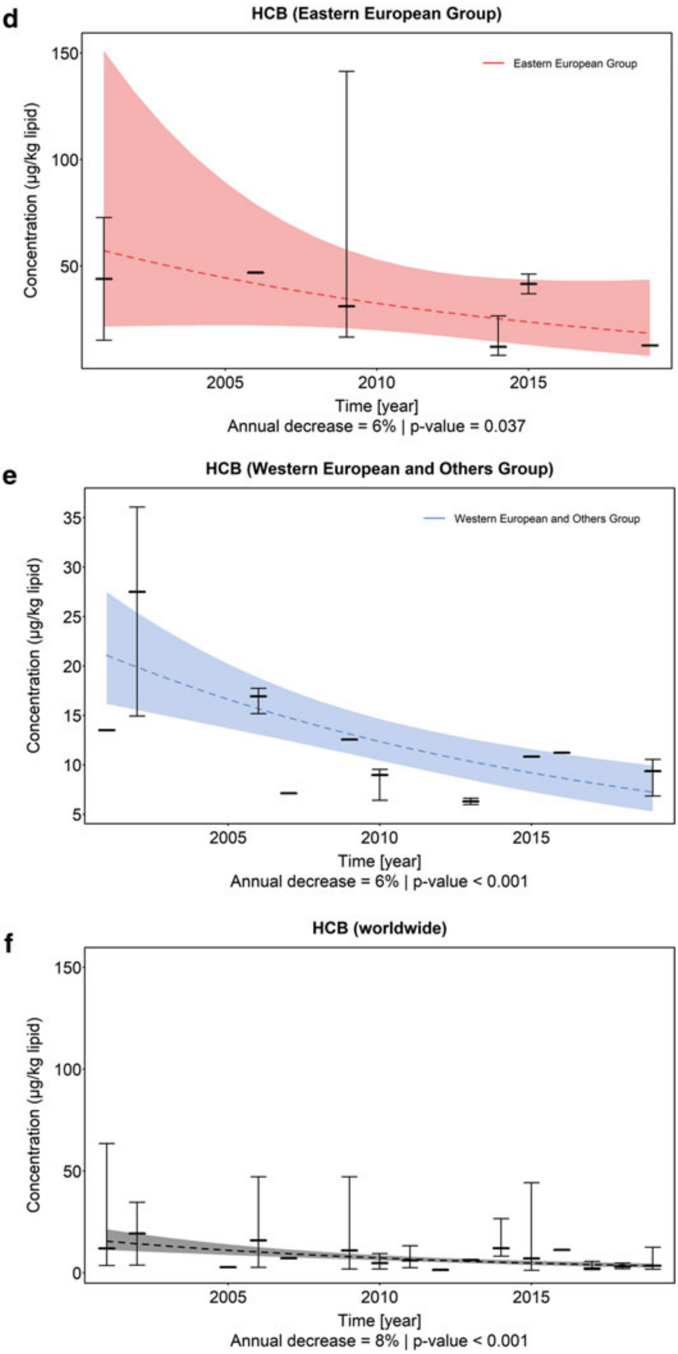

Fig. 25 (continued)

5.2 African Group

All 13 countries from Africa with repeated participation were at all times in the low background range below 5 μg HCB/kg lipid. In most countries, even these comparably low levels seemed to decrease over time, whereas in few countries, an increase seemed to be observed. However, it is more likely that these findings reflect the HCB variation in the lower background range over time, which might be levelling out and quite stable at low concentrations of approximately 2–3 μg HCB/kg lipid.

Figures 26 (for aggregated data) and 27 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of HCB. Table 16 compiles the overall decreases calculated by the Theil–

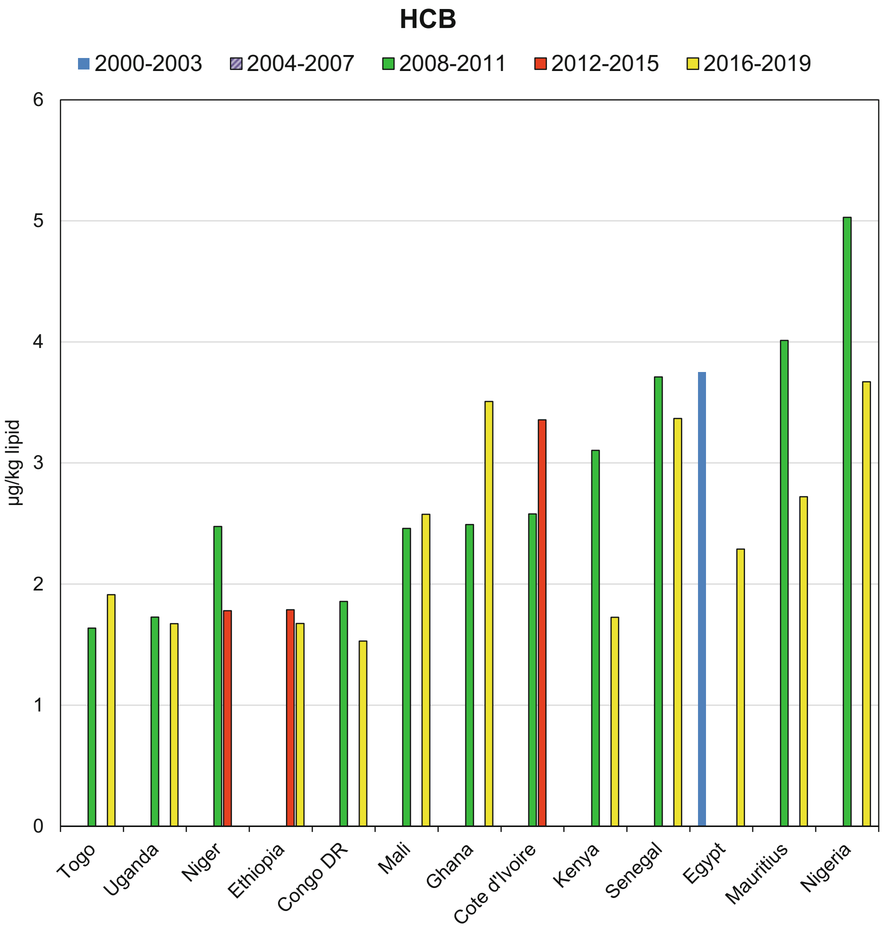

Fig. 26 Overview of the development of the HCB concentration in human milk (expressed as μg/kg lipid; aggregated data) over time for African countries with repeated participation between 2000 and 2019

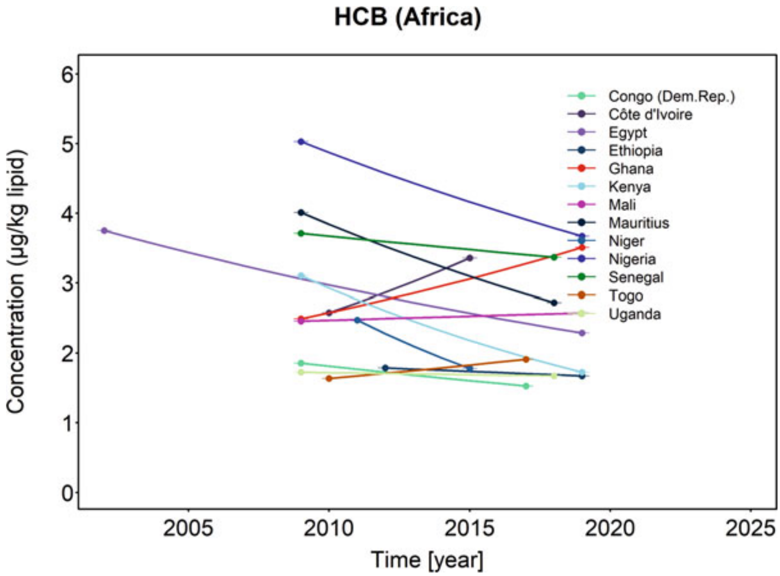

Fig. 27 Temporal tendencies of the HCB concentration in human milk (expressed as µg/kg lipid) in African countries with repeated participation between 2000 and 2019 using the Theil–Sen method

Table 16 Overall decrease (%) of the HCB concentration in human milk in African countries per 1 year, 5 years and 10 years (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Trend *p*-value overall
Congo (DR)	2.4	11.4	21.5	1.000
Côte d'Ivoire	−5.4	−30.1	−69.3	1.000
Egypt	2.9	13.5	25.2	1.000
Ethiopia	0.9	4.6	9.0	1.000
Ghana	−3.5	−18.7	−40.8	1.000
Kenya	5.7	25.4	44.4	1.000
Mali	−0.5	−2.3	−4.7	1.000
Mauritius	4.2	19.4	35.0	1.000
Niger	7.9	33.8	56.1	1.000
Nigeria	3.1	14.6	27.0	1.000
Senegal	1.1	5.2	10.2	1.000
Togo	−2.2	−11.7	−24.9	1.000
Uganda	0.4	1.7	3.5	1.000
Median	1.1	5.2	10.2	

Sen method. The limited number of samples did not allow to determine statistically significant decrease rates.

5.3 Asia-Pacific Group

Figures 28 (for aggregated data) and 29 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of HCB concentrations in 8 countries from the Asia-Pacific Group with repeated participation between 2000 and 2019. Table 17 compiles the overall decreases

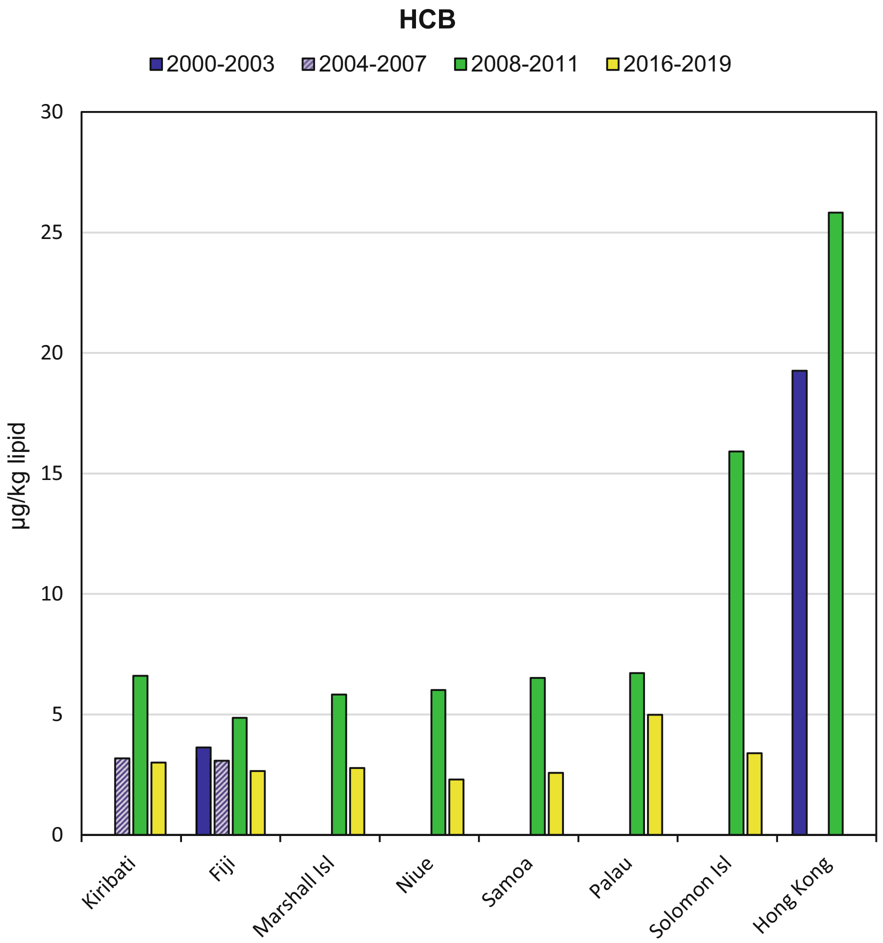

Fig. 28 Overview of the development of the HCB concentration in human milk (expressed as µg/kg lipid; aggregated data) over time in countries of the Asia-Pacific Group with repeated participation between 2000 and 2019

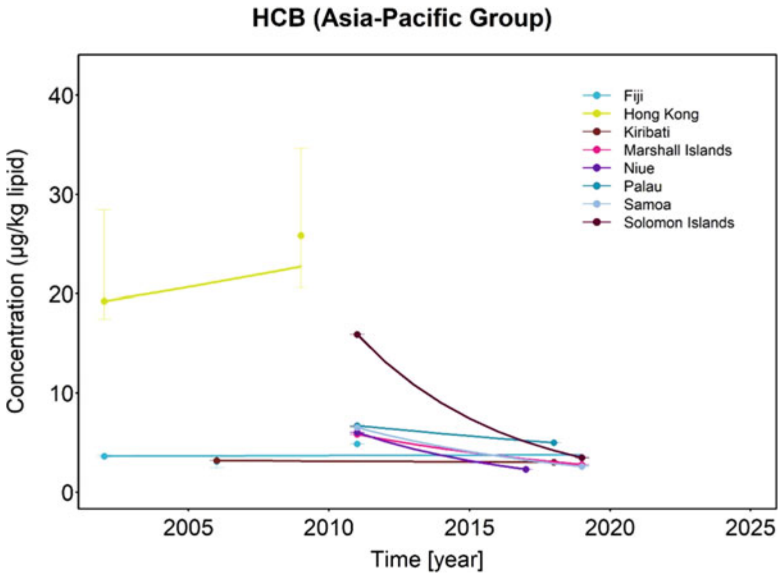

Fig. 29 Temporal tendencies of the HCB concentration in human milk (expressed as µg/kg lipid) over time in countries of the Asia-Pacific Group with repeated participation between 2000 and 2019 using the Theil–Sen method

Table 17 Overall decrease (%) of the HCB concentration in human milk in countries of the Asia-Pacific Group per 1 year, 5 years and 10 years (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Trend *p*-value overall
Fiji	−0.2	−1.1	−2.2	0.950
Hong Kong	−2.4	−12.6	−26.8	< 0.001
Kiribati	0.5	2.3	4.6	1.000
Marshall Islands	8.8	37.0	60.3	1.000
Niue	14.8	55.1	79.9	1.000
Palau	4.2	19.2	34.8	1.000
Samoa	11.0	44.0	68.7	1.000
Solomon Islands	17.4	61.5	85.2	1.000
Median	6.5	28.1	47.6	

calculated by the Theil–Sen method. In nearly all countries, the limited number of samples did not allow to determine statistically significant decreases.

From the Asian part of this UN regional group, Hong Kong SAR of China participated twice, with an increase of the median concentrations from 2002 (19.3 μg HCB/kg lipid as median of 10 pooled samples [Hedley et al. 2010]) to the 2009 level of 25.8 μg/kg (median of 4 samples from different population subgroups). These summarizing temporal trends could be further differentiated: Two subgroups of 2009 with residents who had been living in Hong Kong for 10 years or more had comparable HCB concentrations (20.5–21.0 μg/kg) to 2002, whereas two subgroups of 2009 who had been living in Hong Kong for less than 10 years had HCB concentrations of 30–35 μg/kg.

Fiji participated four times between 2002 and 2019 and Kiribati three times between 2006 and 2018. Over the entire period, the HCB concentrations remained quite stable in the range of background contamination, i.e. around 3 μg/kg in Fiji, 2002 and 2019, and Kiribati, 2006 and 2018. The highest HCB concentration found in Solomon Islands in 2011 (15.9 μg/kg) was reduced by 79% until 2018. The other four countries of the Pacific Islands had HCB concentrations in human milk in the 2008–2011 period around 6–7 μg/kg and had overall decrease rates per 10 years between 35% and 80%.

5.4 Group of Latin American and Caribbean Countries (GRULAC)

Figures 30 (for aggregated data) and 31 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of HCB concentrations in 9 Latin American and Caribbean countries with repeated participation between 2000 and 2019. Table 18 compiles the overall decreases calculated by the Theil–Sen method. In all countries except Brazil, the limited number of samples did not allow to determine statistically significant decrease rates.

In all countries, HCB concentrations in human milk decreased over time. In Haiti, the level of 2.8 μg HCB/kg lipid in 2004 was reduced by 65% until 2015. In Brazil, a decrease of the median concentration of 4.4 μg/kg in 2001 by 68% was found until 2012. Most other countries participated for the first time in the period 2008–2011. In all of these countries, a decrease was observed until the 2016–2019 period (range 5–50%).

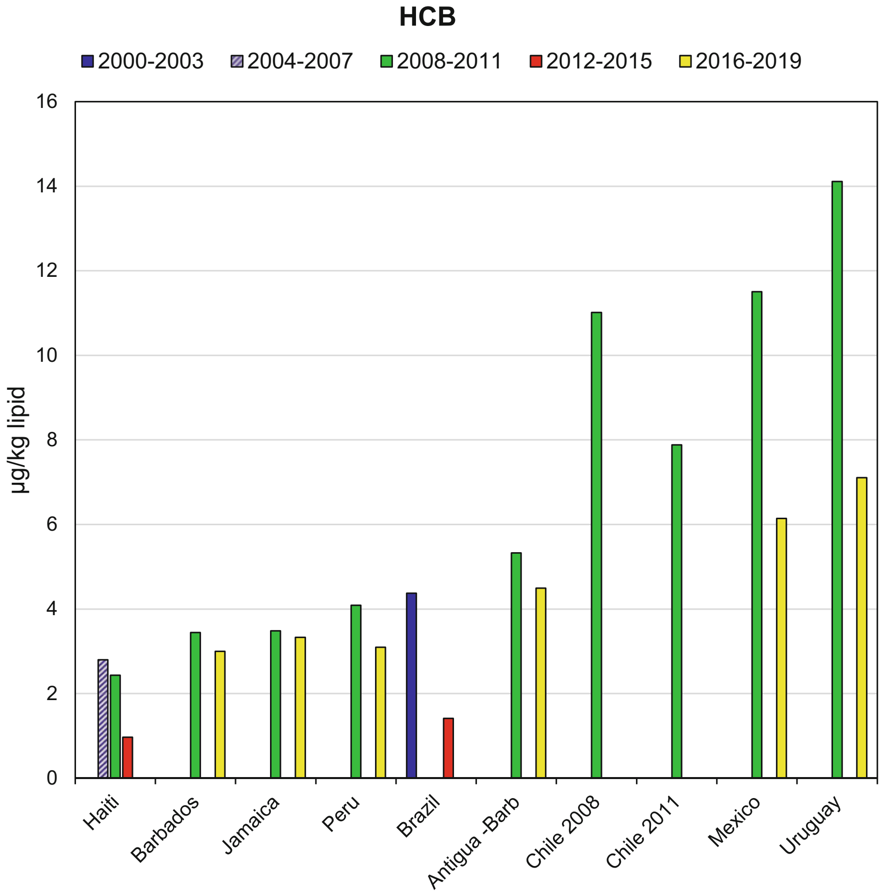

Fig. 30 Overview of the development of the HCB concentration in human milk (expressed as µg/kg lipid; aggregated data) over time in countries of the Group of Latin American and Caribbean Countries with repeated participation between 2000 and 2019

For countries with first participation in the 2008–2011 period and HCB concentrations in human milk in the range of the background contamination around or below 5 µg/kg (Antigua-Barbuda, Barbados, Jamaica and Peru), the overall decrease per 10 years was lower (range 6.1% to 29.5%) than for the countries with higher HCB levels in this period (range of HCB concentrations in Chile, Mexico and Uruguay: 8–14 µg/kg; decrease: range 50–81%).

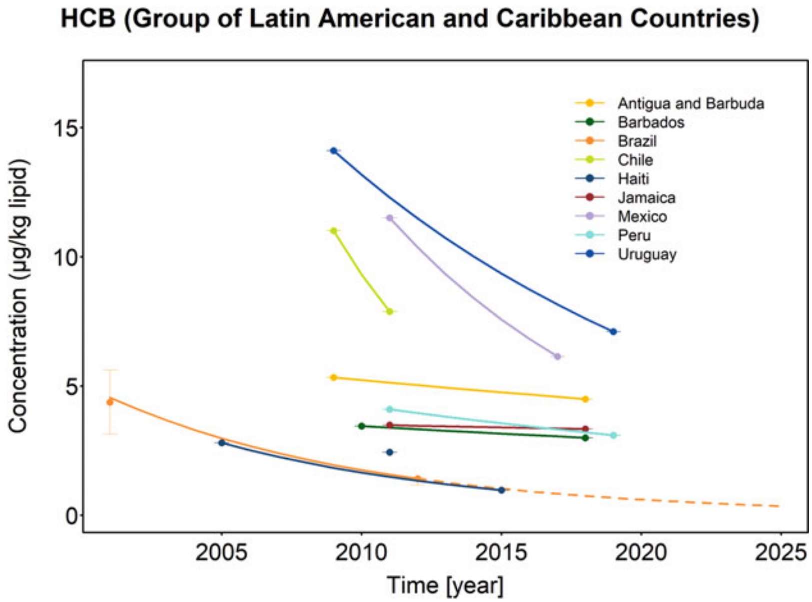

Fig. 31 Temporal tendencies of the HCB concentration in human milk (expressed as µg/kg lipid) in countries of the Group of Latin American and Caribbean Countries with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Brazil)

Table 18 Overall decreases (%) of the HCB concentration in human milk in countries of the Group of Latin American and Caribbean Countries per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method)

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [ng/g lipid]	Trend *p*-value overall
Antigua-Barb.	1.9	9.0	17.2		1.000
Barbados	1.7	8.4	16.0		1.000
Brazil	10.1	41.3	65.5	0.35	0.031
Chile	15.4	56.7	81.3		1.000
Haiti	10.1	41.1	65.4		0.250
Jamaica	0.6	3.1	6.1		1.000
Mexico	9.9	40.7	64.9		1.000
Peru	3.4	16.0	29.5		1.000
Uruguay	6.6	29.0	49.6		1.000
Median	6.6	29.0	49.6		

5.5 Eastern European Group

Figures 32 (for aggregated data) and 33 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of HCB concentrations in 6 countries of the Eastern European Group with repeated participation between 2000 and 2019. Table 19 compiles the overall decreases calculated by the Theil–Sen method. In all countries except Czechia, the limited number of samples did not allow to determine statistically significant decreases.

A wide range of concentrations was observed with a maximum of 154 μg/kg found in Moldova, 2009. These concentrations decreased considerably by 70% until 2015. Czechia participated four times between 2000 and 2019; here a continuous downtrend was found from 76 μg HCB/kg lipid in 2001 with a reduction by 84% until 2019.

The overall reduction rates were lower in countries with initially lower HCB levels (Bulgaria, 2001: 12 μg/kg; Georgia, 2009: 15 μg/kg; decrease over 10 years:

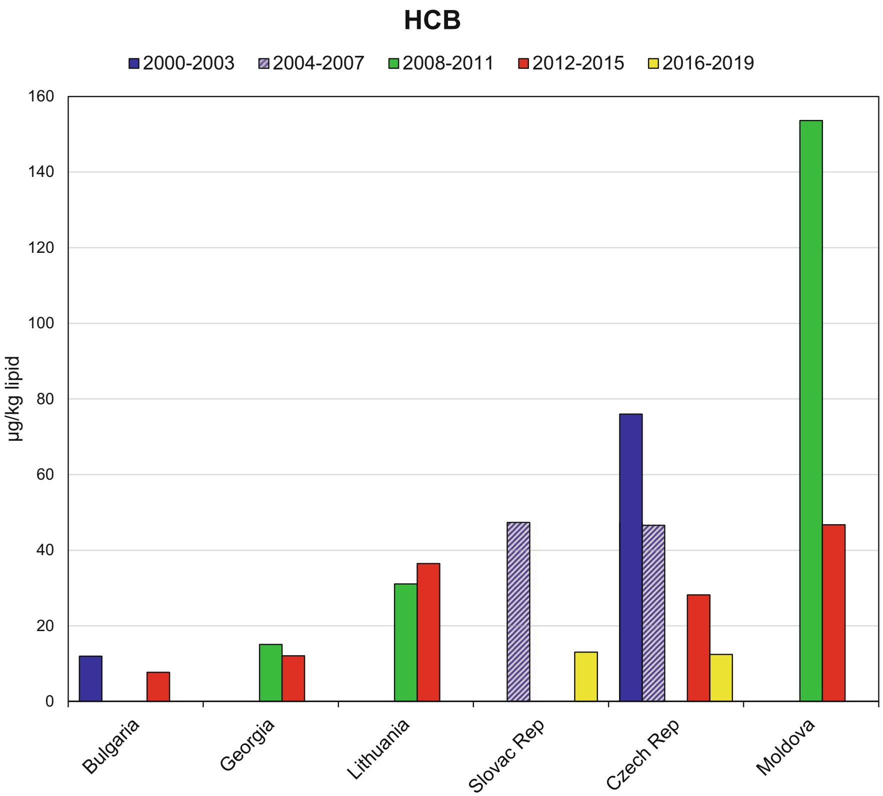

Fig. 32 Overview of the development of the HCB concentration in human milk (expressed as μg/kg lipid; aggregated data) over time in countries of the Eastern European Group with repeated participation between 2000 and 2019

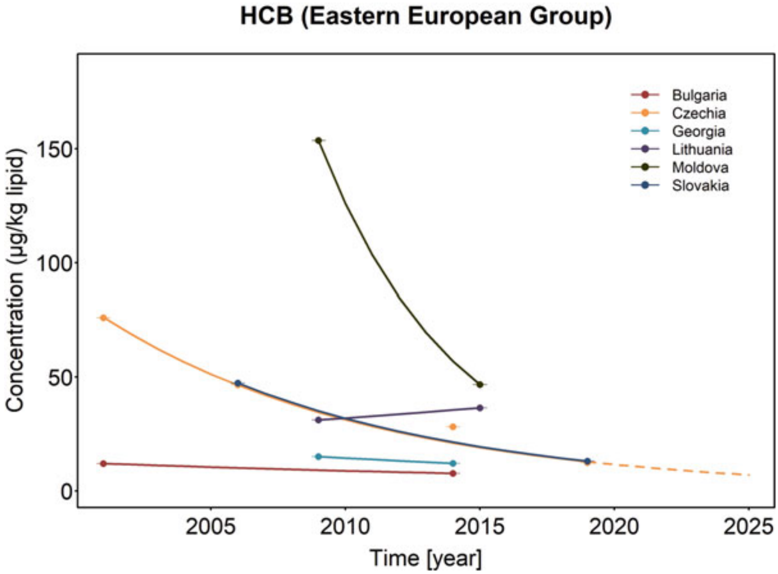

Fig. 33 Temporal tendencies of the HCB concentration in human milk in countries of the Eastern European Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Czechia)

Table 19 Overall decrease (%) of the HCB concentration in human milk (expressed as μg/kg lipid) in countries of the Eastern European Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [ng/g lipid]	Trend *p*-value overall
Bulgaria	3.3	15.7	28.9		1.000
Czechia	9.4	39.1	62.9	7.06	0.031
Georgia	4.3	19.9	35.8		1.000
Lithuania	−2.7	−14.2	−30.5		1.000
Moldova	18.0	62.9	86.2		1.000
Slovakia	9.4	39.1	62.9		1.000
Median	6.9	30.1	51.2		

29% and 36%, respectively) than in countries with higher HCB levels (Czechia, Moldova, Slovakia; range of HCB concentrations: 47–154 μg/kg; decrease over 10 years: 63–86%). Only in Lithuania were the HCB concentrations increased slightly from 31.1 μg/kg in 2009 by 17% until 2015.

5.6 Western European and Others Group (WEOG)

Figures 34 (for aggregated data) and 35 (comprising all individual pooled samples and assuming exponential trends, see subsection 2.3) illustrate the temporal changes of HCB concentrations in 8 countries of the Western European and Others Group with repeated participation between 2000 and 2019. Table 20 compiles the overall decreases calculated by the Theil–Sen method. In all countries except Germany, the limited number of samples did not allow to determine statistically significant decreases.

The highest HCB concentrations (range 13.5 μg/kg to 31.6 μg/kg) were found in countries participating in the 2000–2003 period. The highest overall decrease over 10 years of 48% was observed in Germany, with a decrease of the median of four pooled samples collected in 2002 (31.6 μg HCB/kg lipid as median, range 28 μg/kg to 37 μg/kg) by 67% until 2019 (10.4 μg/kg as median, range 10.2 μg/kg to 10.6 μg/kg). In most other countries, in particular with lower initial HCB concentrations, the decreases were lower. Over shorter periods, the levels remained quite constant in Australia (from 2010 to 2013) or increased slightly in Norway (from 2001 to 2006).

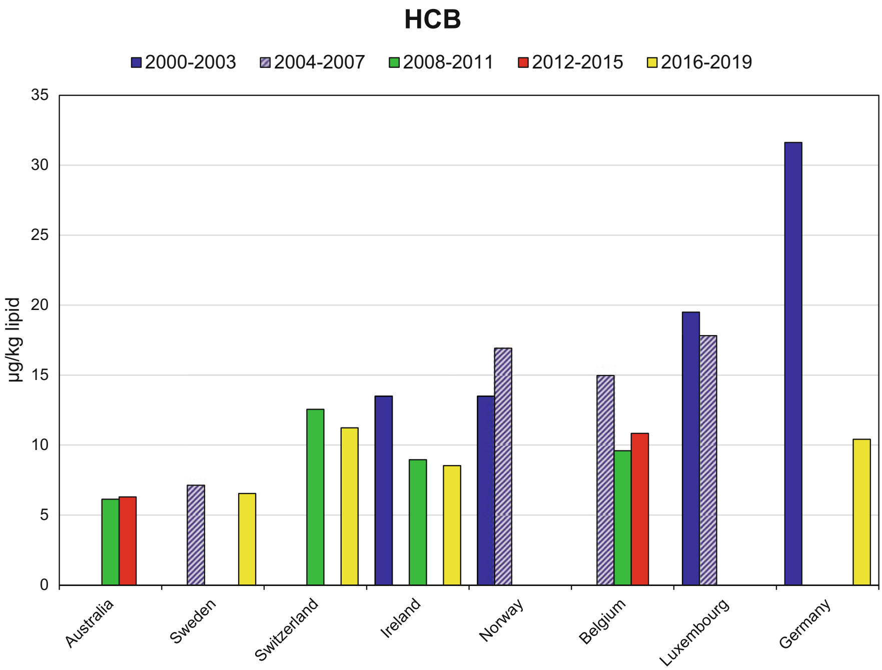

Fig. 34 Overview of the development of the HCB concentration in human milk (expressed as μg/kg lipid; aggregated data) over time in countries of the Western European and Others Group with repeated participation between 2000 and 2019

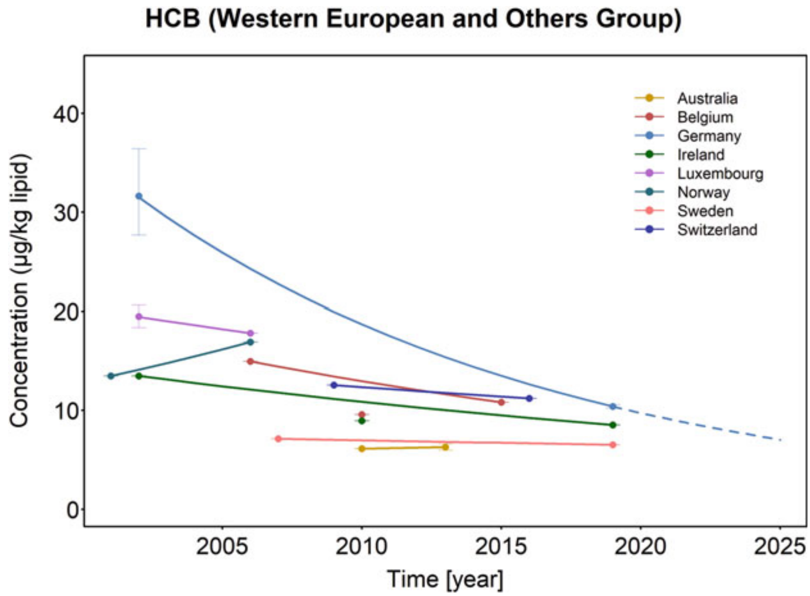

Fig. 35 Temporal tendencies of the HCB concentration in human milk (expressed as µg/kg lipid) in countries of the Western European and Others Group with repeated participation between 2000 and 2019 using the Theil–Sen method (with a statistically significant time trend in Germany)

Table 20 Overall decrease (%) of the HCB concentration in human milk (expressed as µg/kg lipid) in countries of the Western European and Others Group per 1 year, 5 years and 10 years and for one country, an estimated concentration in 2025 (calculated by the Theil–Sen method). Negative decreases are to be read as increase

Country	Overall decrease (%) per 1 year	Overall decrease (%) per 5 years	Overall decrease (%) per 10 years	Estimated concentration in 2025 [ng/g lipid]	Trend *p*-value overall
Australia	−0.9	−4.4	−9.0		1.000
Belgium	3.5	16.5	30.2		0.500
Germany	6.3	27.8	47.9	7.04	0.008
Ireland	2.7	12.6	23.7		0.250
Luxembourg	2.2	10.4	19.8		0.500
Norway	−4.6	−25.3	−57.1		1.000
Sweden	0.7	3.5	6.9		1.000
Switzerland	1.6	7.7	14.7		1.000
Median	1.9	9.0	17.3		

5.7 Dependence of Decrease (Decrease Rate Constants) on Concentration

As explained in subsection 3.7, the decrease (*decrease rate constants*) within a 10-year period have to be seen also in context with the concentration range: A differentiation of levels above or in the range of background contamination seems to be advisable.

HCB concentrations in human milk in the 44 countries with repeated participation between 2000 and 2019 comprise a wide range between 1.6 μg/kg and 154 μg/kg. This was grouped into four ranges (<5; 5–10; 10–20; >20 μg HCB/kg lipid) to check the dependence of the decrease on the "initial" (the first measured) concentration in a country. The lower end of the frequency distribution below 5 μg HCB/kg lipid is considered as the background contamination.

Table 21 shows, that the decrease over 10 year for HCB concentrations vary in all ranges. However, for samples above 20 μg/kg, the highest median decrease over 10 years (63%) for five samples was observed, whereas the median decrease in the low background range was 13%. Note that all samples in the low background range had concentrations above LOQ (0.5 μg/kg lipid) with a range between 1.6 and 5.0 μg HCB/kg lipid. Figure 36 illustrates this variation of the decreases over 10 years in 44 countries with repeated participation between 2000 and 2019 in all concentration ranges, with a tendency to higher decreases at higher levels. It might be concluded that the large variation of the decrease for beta-HCH concentrations in the low background range (< 5 μg/kg) limits the applicability of this parameter for assessments at the country-level at this low level, but allows a more general assessment of the temporal trends of the background contamination.

Table 21 Overall decrease (%) of HCB concentrations in human milk (μg/kg lipid) over 10 years calculated by the Theil–Sen method and their dependence on the concentration range

	<5 μg/kg	5–10 μg/kg	10–20 μg/kg	>20 μg/kg
N	20	7	12	5
min	−69	−9	−57	−30
median	13	35	30	63
max	65	80	85	86

Negative decreases are to be read as increase

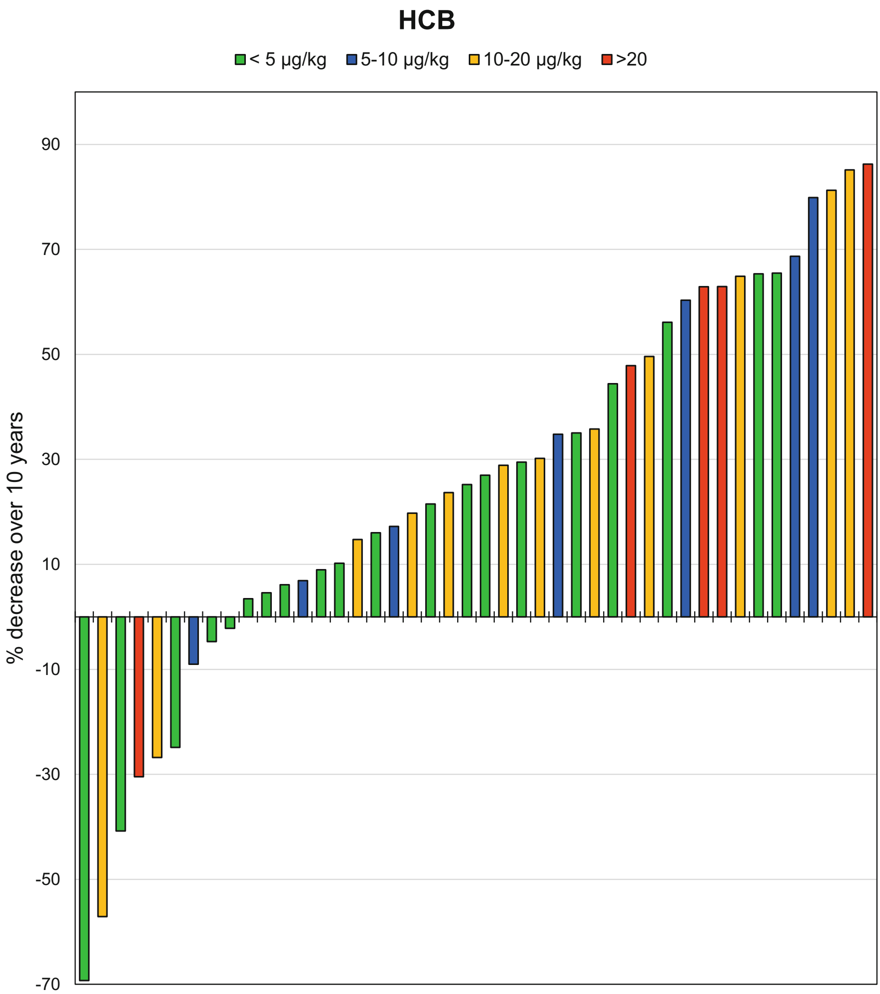

Fig. 36 Dependence of the decrease over 10 years (calculated by the Theil–Sen method) for HCB on concentrations in human milk in 44 countries with repeated participation between 2000 and 2019, with differentiation into four ranges of concentration (<5; 5–10; 10–20; >20 μg/kg lipid)

6 Summary and Conclusions

The assessment of temporal trends is an important objective of the WHO/UNEP-coordinated studies for the evaluation of the effectiveness of the Stockholm Convention on POPs to eliminate or reduce emissions of listed POPs.

The presentation and discussion of the 2000–2019 results for chlorinated pesticides and industrial chemicals in Part III of this compendium includes a *general* estimation of time trends during the five rounds for *all* participating countries over these 20 years. However, a more precise approach is the evaluation of results from *only countries with repeated participation* in these studies: This allows more certainty in drawing of conclusions on temporal trends, which are not potentially influenced by single results of a countries submitted for a single round. The time trends of 119 pooled samples from 44 countries with repeated participation seemed optimal for the evaluation of the effectiveness for the purpose of Article 16. With regard to the found concentrations, most relevant were DDT (as sum parameter “DDT complex”, comprising the p,p′-and o,p′-isomers of DDT and the metabolites p,p′-DDE and p,p′-DDD), beta-HCH and HCB, which are presented in this chapter.

For statistically significant *trends*, a minimum of five data points has to be available. However, for most countries only less than five data points are available. This prevents deriving statistically significant temporal trends in these cases. Yet, the existing data can indicate decreasing or increasing *tendencies* in POP concentrations. Furthermore, pooling of data in regions allows to derive statistically significant time trends for the UN regional groups. To minimize possible sources of variation for time trend analysis of POPs, the concept of the WHO/UNEP-coordinated exposure studies has two basic elements (preparation of pooled samples from a number of individual samples considered to be representative for a country or region/subgroup; analysis by a reference laboratory).

To provide reliable monitoring information for the Parties to the Stockholm Convention, as quantitative objective for temporal studies *The Guidance Document on the Global Monitoring Plan* (GMP) proposed the ability to detect a 50% decrease in the levels of POPs within a 10-year period. However, there is no stipulation that this is a quantitative goal for the rate of decrease in the levels of listed POPs. 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 the change is nowhere specified or required.

Decreases (*decrease rate constants*) per 1 year, 5 years (about 20% higher than for 4 years as average lengths of the WHO/UNEP-coordinated studies) and 10 years were computed using the Theil–Sen trend estimator including values of all individual samples for the trend analysis. For confirmation, a method of deriving the regional trend as a median of trends in countries within the region was used (“median method”).

DDT

For DDT concentrations in human milk, large differences were found comprising a range between a minimum of 17 μg DDT complex/kg lipid found in 2019 and a maximum of 23,500 μg DDT complex/kg lipid found in 2012 (median 283/kg).

An overall decrease within a 10-year period between 50% and 80% was achieved for DDT complex levels in Africa, the Asia-Pacific Group and the Group of Latin American and Caribbean Countries, and at a global level. Lower decreases were observed in the Eastern European Group and the Western European and Others Group. Generally, the highest DDT concentrations in the five periods between 2000 and 2019 were found in the Africa Group, the Asia-Pacific Group or the Group Latin America and Caribbean Countries, whereas Western European countries had the lowest median and lowest maximum of DDT concentrations. This is an indication that the decrease might be faster in regions with higher concentration, compared to a slower decrease in less contaminated regions, which banned DDT decades ago. This is supported by the assessment of the reduction rates based on the frequency distribution of DDT concentrations in 44 countries showing that the decrease over 10 year in the upper part of the frequency distribution for the three samples with concentrations above 2000 μg DDT complex/kg lipid is considerably higher than in the lower part (<200 μg DDT complex/kg lipid).

Beta-HCH

Large differences were also found of beta-HCH concentrations, with a minimum of <0.5 μg/kg in few countries and a maximum of 1020 μg/kg lipid in a sample of 2002 (median: 6.0 μg/kg). An overall decrease within a 10-year period between 50% and 98% was achieved for beta-HCH levels in the UN regional groups and at the global level.

On the country level, the reduction rates should also be seen in context with the concentration range (differentiation of levels above or in the range of the background contamination). If high levels are found, sources might be detected which could be eliminated. However, at low background levels (< 5 μg beta-HCH/kg lipid), other factors, e.g. contamination of feed and food by air via long-range transport and subsequent bioaccumulation, cannot be influenced locally.

HCB

For HCB, the maximum levels and therefore the ranges were much lower than found for DDT and beta-HCH, with a minimum of about 1–2 μg/kg found in some countries and a maximum of 154 μg/kg found in 2009 in an Eastern European country (median: 6.1 μg/kg).

In the African Group with all pooled samples and all collection periods, the low background range at or below 5 μg HCB/kg lipid resulted the overall decrease within 10 years of only about 11% calculated by the Theil–Sen method. Overall decreases between 30% and 67% were calculated by the Theil–Sen method for the other UN Regional Groups, and 57% on a global level.

However, the high variation of the reduction rates in African countries with all samples at all times in the lower background range at or below 5 μg HCB/kg lipid shows that use of these calculated reduction rates is questionable in this low range. As concluded for beta-HCH, the large variation of decrease rates for HCB concentrations in the low background range (<5 μg/kg lipid) limits the applicability of this parameter for assessments at the country-level at these low levels, but allows a more general assessment of the temporal trends of the low background contamination.

Overall Conclusions

The concept of WHO/UNEP-coordinated exposure studies with standardized protocols for preparation of pooled samples considered to be representative for a country or subgroup within a country and analysis in a reference laboratory with long-term quality control provides a cost-effective method to obtain reliable data on POPs in human milk samples over time. The use of only those countries with repeated participation provides the best possible data base for assessment of temporal trends. Statistically significant decreasing trends for DDT, beta-HCH and HCB were observed for all parameters in the five UN regional groups and at a global level. However, for the majority of individual countries the limited available data did not allow for the statistically significant assessment of time trends, but decreasing tendencies were observed for many of them and constant or increasing levels for few of them. It is highly recommended to continue this monitoring effort to secure enough data for a proper time trend assessment in the future.

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