

Midterm Workshop of the UN Environment/GEF project
'Continuing Regional Support for the POPs Global
Monitoring Plan under the Stockholm Convention'
in the Asia Region
Ulaanbaatar, Mongolia, 8-10 August 2018

**2016-2019 round of UNEP-coordinated
exposure studies on human milk
in the Asia region**

Rainer Malisch

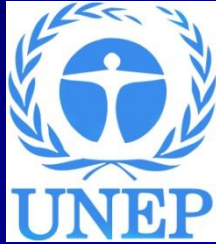


CVUA Freiburg, Germany

(State Institute for Chemical and Veterinary Analysis of Food)



- ✓ State Institute for Chemical and Veterinary Analysis of Food
- ✓ WHO / UNEP Reference Laboratory
- ✓ EU Reference Laboratory (EURL) for Halogenated POPs in Feed and Food
- ✓ EURL for Pesticides in Food of Animal Origin



WHO/UNEP-coordinated exposure studies on levels of POPs in human milk

Round	Years	Organized by	No of countries	Parameters
1	1987-1988	WHO-EURO	12	Dioxins and PCBs
2	1992-1993	WHO-EURO	19	Dioxins and PCBs
3	2000-2003	WHO-EURO	26	Dioxins and PCBs
				later Stockholm Convention Initial POPs
4	2004-2007	WHO/UNEP	13	Stockholm Convention POPs
5	2008-2011	WHO/UNEP	45	Stockholm Convention POPs
6	2012-2015	UNEP	17	Stockholm Convention POPs
7	since 2016	UNEP	42	Stockholm Convention POPs

Participants 2000 - 2015

Africa					America					Asia					Australia, NZ, Pacific Islands					Europe				
	2000-2003	2004-2007	2008-2011	2012-2015		2000-2003	2004-2007	2008-2011	2012-2015		2000-2003	2004-2007	2008-2011	2012-2015		2000-2003	2004-2007	2008-2011	2012-2015		2000-2003	2004-2007	2008-2011	2012-2015
Congo (DR)			x		Antigua-Barb.			x		Hong Kong SAR	x		x		Australia	x		x	x	Belgium	x	x	x	x
Côte d'Ivoire			x	x	Barbados			x		India			x		Fiji	x	x	x		Bulgaria	x			x
Djibouti			x		Brazil	x			x	Indonesia			x		Kiribati		x	x		Croatia	x			x
Egypt	x				Chile			2 x		Israel				x	Marshall Islands			x		Cyprus		x		
Ethiopia				x	Cuba			x		Korea (Rep)			x		New Zealand	x		x		Czech Rep	x	x		x
Ghana			x		Haiti		x	x	x	Philippines	x				Niue			x		Finland	x	x		
Kenya			x		Jamaica			x		Syria			x		Palau			x		Georgia			x	x
Mali			x		Mexico			x		Tajikistan			x		Samoa			x		Germany	x			
Mauritius			x		Peru			x		total no: 8					Solomon Islands			x		Hungary	x	x		
Niger			x	x	Suriname				x					Tonga			x		Ireland	x		x		
Nigeria			x		Uruguay			x						Tuvalu			x		Italy	x				
Senegal			x		USA	x								total no: 11					Lithuania			x	x	
Sudan		x			total no: 12														Luxembourg	x	x			
Togo			x																Moldova			x	x	
Uganda			x																Netherlands	x				x
total no: 15																			Norway	x	x			
																			Romania	x				x
																			Russia	x				
																			Slovak Rep	x	x			
																			Spain	x				
																			Sweden	x	x			
																			Switzerland			x		
																			Ukraine	x				
																			total no: 23					

total number of countries:	69
from these participating	
- once	43
- twice	21
- three times	4
- four times	1
resulting number of country/year-data:	101
total number of pooled samples:	188

Asia					
	2000- 2003	2004- 2007	2008- 2011	2012- 2015	2016- 2019
Hong Kong SAR	x		x		
Cambodia					x
India			x		
Indonesia			x		(x)
Israel				x	
Korea (Rep)			x		
Lao PDR					x
Mongolia					x
Philippines	x				x
Syria			x		
Tajikistan			x		
Thailand					x
Vietnam					x
<i>total no: 13 (2016-2019:6)</i>					

Participants from Asia 2000 - 2019

Key aspects for human milk monitoring

- Effectiveness evaluation (time trends)
- Inclusion of new POPs
- Cost-effective approach to evaluate relevance of individual POPs
- Support of capacity building (quality control in labs)

Human tissues as indicators of human exposure to POPs

Human samples as suitable indicators for bioaccumulation of POPs :

- ✓ **Breast milk**
- ✓ **Blood**
- ✓ **Adipose tissues**

Comparable results on fat basis

Advantage of breast milk samples

- ✓ non-invasive mean to estimate the exposure
- ✓ Less toxicological concern (relatively high risk of contacts with infectious agents: AIDS virus, hepatitis) than for human blood
- ✓ human milk has higher fat content than blood
 - **For analysis: available amount of lipids important factor with regard to number of analytically covered POPs and LOQs**
 - Lipid amount of type of sample
 - Mixing (pooling)

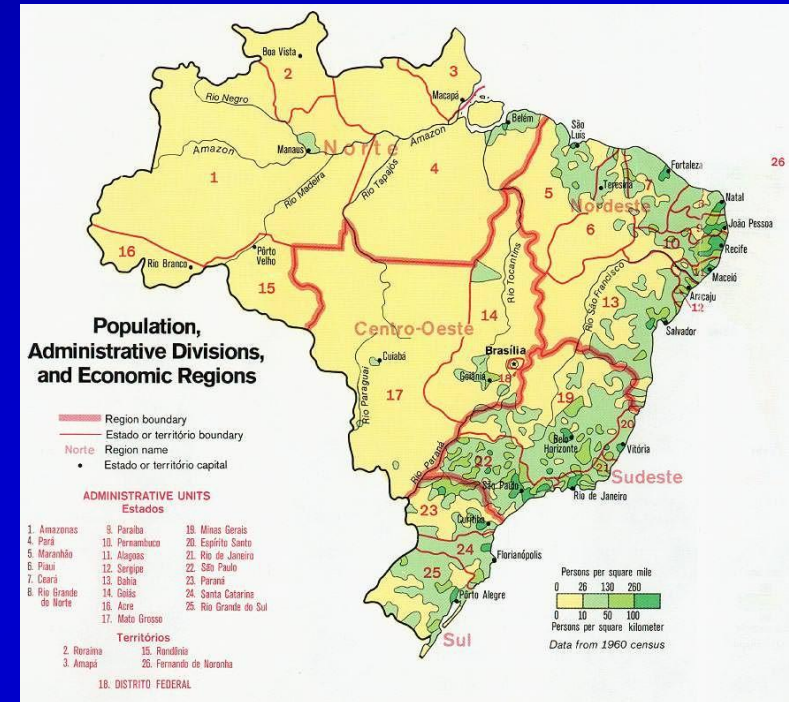
Standardized protocol



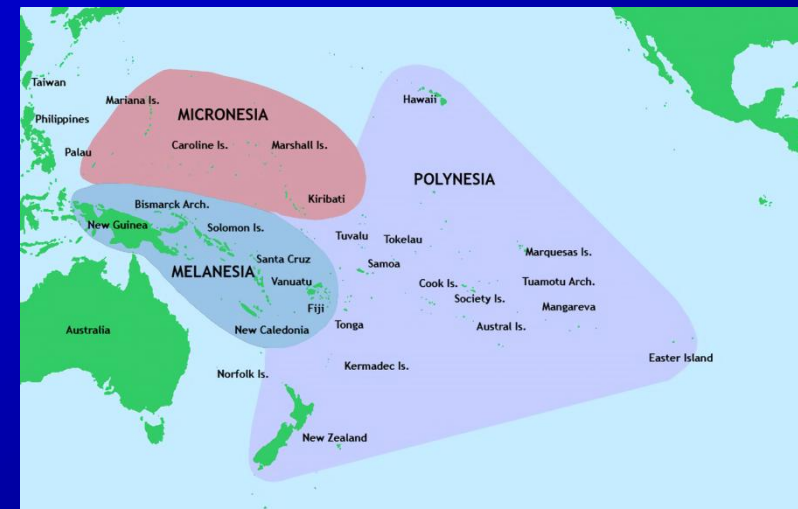
- **Collection of human milk from representative individuals** (*since 2007: n = 50*)
- **Preparation of one pooled (=mixed) sample representative** for a country / region
- **Analysis by Reference Laboratory for reliable and comparable data**
 - (+) **Cost-effective** and useful non-invasive mean to estimate the overall exposure of a local population
 - Possible to get a rough estimate on the exposure in different regions of the world and time trends with only very few samples

Standardized protocol

- **Collection of human milk from representative individuals** (*since 2007: n = 50*)
- **Preparation of one pooled (=mixed) sample representative for a country / region**
- **Some flexibility:**
 - Countries with populations greater than 50 million should include at least one additional participant per one million population over 50 million. Countries with populations well over 50 million (or with sufficient resources) are encouraged to prepare a second pooled sample (or more) if feasible.
 - Small countries: less?



Brazil 210,000,000



e.g. Fiji 880,000; Tuvalu 10,000

Support of capacity building

Sample preparation scheme: Preparation of individual samples for analysis of basic POPs by country and of pooled (mixed) samples

(Before taking an aliquot, shake intensely at room temperature and then take the aliquot immediately.
Storage and shipment of all samples deep-frozen.)



50 ml



take 25 ml into 2000 ml bottle (50 * 25 ml ea = 1250 ml)



Send the 2000 ml bottle with frozen pooled samples to WHO/UNEP Reference Lab



25 ml



Individual samples for analysis by country for basic POPs

For (deep-frozen) storage in and analysis by country. If no analysis in country possible and no storage capacity available, contact UNEP.



Mean of individual samples should be comparable to concentration found by reference lab



UNEP-coordinated Survey of Human Milk for Persistent Organic Pollutants

Guidelines for Organization, Sampling and Analysis

January 2017

Page 1 of 38

Guidelines

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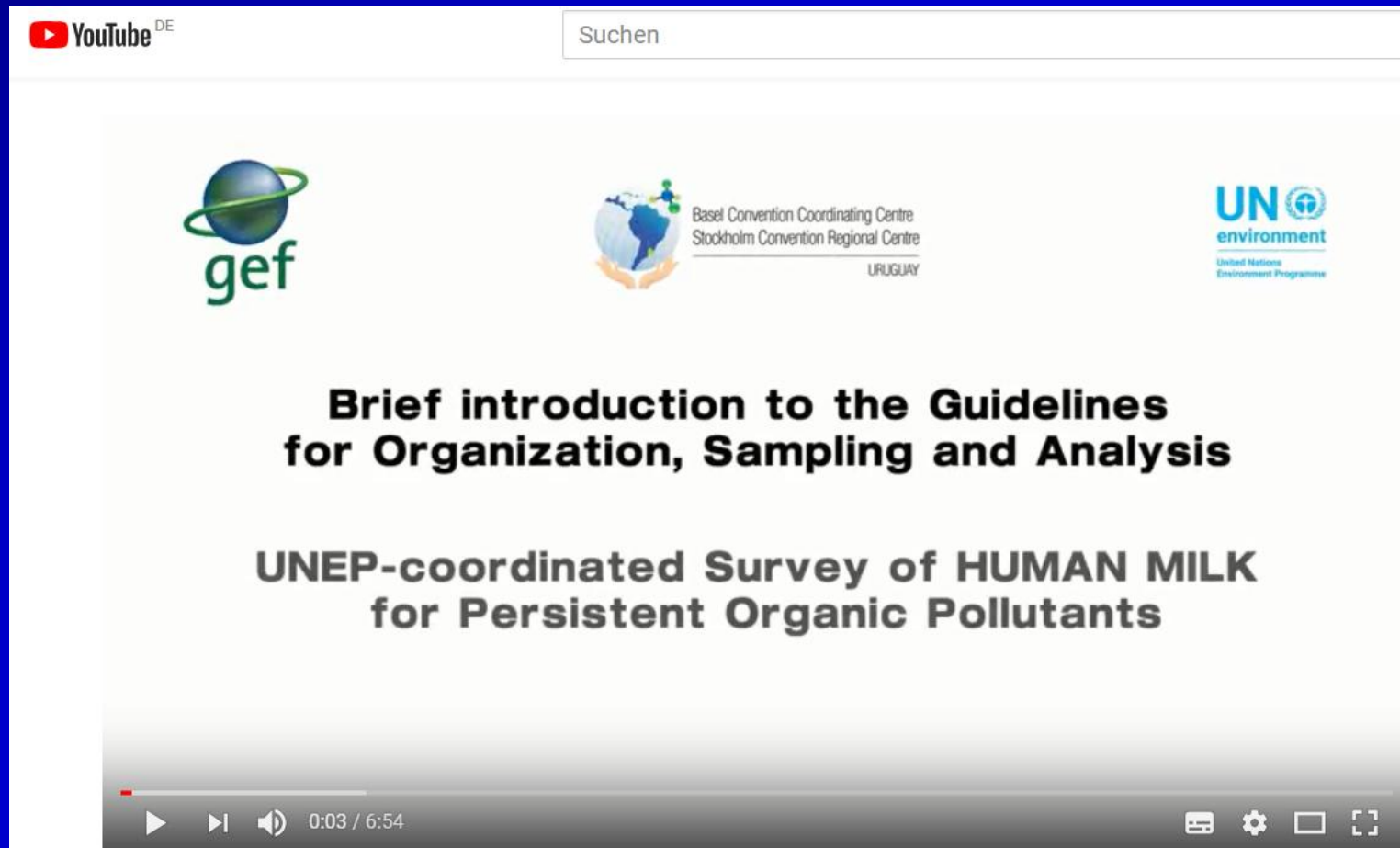
Chemicals Branch
Division of Technology, Industry and Economics (DTIE)
United Nations Environment Programme (UNEP)

Video on guidelines

https://youtu.be/7LwJ0x2_PXQ

Preparing the video:

Alejandra Torre
Gabriela Medina
Virginia Santana



The screenshot shows a YouTube video player interface. At the top left is the YouTube logo with 'DE' next to it. To its right is a search bar containing the text 'Suchen'. The video content area features three logos at the top: 'gef' (Global Environment Fund) on the left, the 'Basel Convention Coordinating Centre Stockholm Convention Regional Centre URUGUAY' logo in the center, and the 'UN environment United Nations Environment Programme' logo on the right. The main title of the video is 'Brief introduction to the Guidelines for Organization, Sampling and Analysis' in bold black text. Below the title is the subtitle 'UNEP-coordinated Survey of HUMAN MILK for Persistent Organic Pollutants' in bold black text. At the bottom of the player, there is a progress bar showing '0:03 / 6:54' and several control icons including play, volume, and settings.

Project Cooperation Agreement (PCA), part I: shipment of glassware containers (60 x 100 ml; 1 x 2000 ml) (here: 7 countries from Asia)

Activity	Comments for 2016	Comments for 2017
Activity 1		
Supply of samplers for human milk for 9 countries of Pacific Islands	Purchase and cleaning of glassware for 9 countries, shipment to 8 countries	Shipment to 1 country; with 1 country (Fiji) not accepting the first shipment; therefore a second shipment necessary
Supply of samplers for human milk for 7 countries of Asia-Pacific	Purchase and cleaning of glassware for 7 countries, no shipment in 2016	Shipment to 7 countries, with 1 country (Philippines) not accepting the first shipment; therefore a second shipment necessary
Supply of samplers for human milk for 15 countries in Africa	Purchase and cleaning of glassware for 15 countries, shipment to 13 countries	Shipment to 2 countries
Supply of samplers for human milk for 12 countries of GRULAC	Purchase and cleaning of glassware for 12 countries, no shipment in 2016 (one country not selected, so far)	Shipment to 11 countries (1 country not responding)

- **Finally, 6 countries received glassware**
- **1 country sent glassware back (non-participation of Indonesia)**



Participation of Asian countries 2016-2019

No	Country	Shipment of glassware	Date of shipment of glassware	Receipt of glassware	Comments
1	Cambodia	yes	10.01.2017	25.04.2017	1st shipment lost
2	Indonesia	yes	14.02.2017		glassware was sent back after a waiting time of 28 days returned to Freiburg on 05 06 2017
3	Lao PDR	yes	10.01.2017	23.01.2017	according to DHL tracking
4	Mongolia	yes	10.01.2017	30.01.2016	2 bottles broken, others opened by custom
5	Philippines	yes	14.02.2017; 15.12.2017	13.03.2018	1. glassware lost, was sent back to Freiburg April 18
6	Thailand	yes	15.12.2017	15.01.2018	12.1.2018 delivery not possible
7	Vietnam	yes	10.01.2017	04.04.2017	according to DHL

- ✓ 6 participants received glassware
- ✓ 2 countries ready to send human milk sample

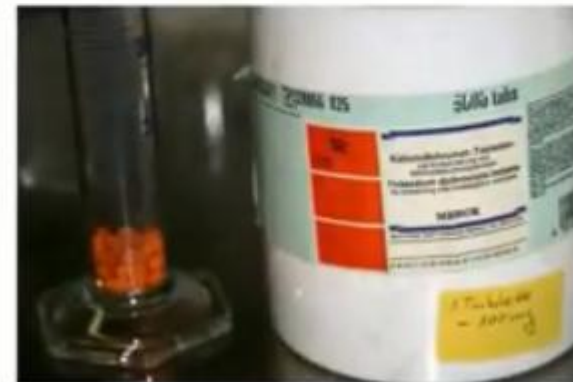


STEP 3 - PRESERVATION

Refrigeration:

$-4^{\circ}\text{C} \leq 72\text{h}$

$-20^{\circ}\text{C} > 72\text{h}$



Supply of dichromate tablets via distributor of chemicals in Germany?



STEP 5 - TRANSPORT by express shipment

The shipment of the pooled samples should be done in close cooperation with Karin Malisch who should be informed about date of shipment and tracking number for follow-up of the shipment and in particular solving possible problems at customs in Germany and to avoid any problems of possible delays due to shipment at inappropriate times

Contact e-mail:

pops@cvuafr.bwl.de

For questions:

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Germany



- Shipment of samples in close contact with Dr Karin Malisch
- Coordination before shipment – preferably on Monday; no holidays
- Express; frozen; cooling elements
- Tracking number allowing to contact to customs in Germany !!!

Project Cooperation Agreement (PCA), part II: analysis – compounds to be analyzed in pooled national mother's milk samples

2003

Compounds to be analysed in pooled national mothers milk samples by CVUA under this Agreement	
Initial POPs	
Aldrin	Aldrin
Chlordane	<i>cis</i> - and <i>trans</i> -chlordane; and <i>cis</i> - and <i>trans</i> -nonachlor, oxychlordane
DDT	4,4'-DDT, 2,4'-DDT and 4,4'-DDE, 2,4'-DDE, 4,4'-DDD, 2,4'-DDD
Dieldrin	Dieldrin
Endrin	Endrin
HCB	HCB
Heptachlor	Heptachlor and heptachlorepoxide
Mirex	Mirex
PCB	∑PCB ₆ (6 congeners): 28, 52, 101, 138, 153, and 180 PCB with TEFs* (12 congeners): 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189
PCDD/PCDF	2,3,7,8-substituted PCD/PCDF (17 congeners)
Toxaphene	Congeners P26, P50, P62

* PCB with TEFs (Toxic Equivalency Factors) assigned by WHO in 1998

2009

POPs listed at COP-4	
Chlordecone	Chlordecone
α-HCH	α-HCH
β-HCH	β-HCH
γ-HCH	γ-HCH
Hexabromobiphenyl	PBB 153
Pentachlorobenzene	PeCBz
c-penta BDE	BDE 47, 99, 153, 154, 175/183 (co-eluting)
c-octa BDE	Optional: BDE 100

2011

POPs listed at COP-5	
Endosulfan	α-, β-endosulfan; and endosulfan sulfate

2013

POPs listed at COP-6	
HBCD	α-HBCD, β-HBCD, γ-HBCD



UNITED NATIONS ENVIRONMENT PROGRAMME

Stockholm Convention on Persistent Organic Pollutants

اتفاقية استكهولم بشأن الملوثات العضوية الثابتة • 关于持久性有机污染物的斯德哥尔摩公约 • Convention de Stockholm sur les polluants organiques persistants
Convenio de Estocolmo sobre Contaminantes Orgánicos Persistentes • Стокгольмская конвенция о стойких органических загрязнителях



Expert meeting to update the Global Monitoring Plan guidance document

Brno, Czech Republic, 7-9 November 2017

2015
2017

2. Introduction and context:
 - (a) Outcomes of COP-7 and COP-8 relevant to the update of the global monitoring plan (GMP) guidance document;
 - (b) Mandate and process for updating the GMP guidance document;
3. Experiences from monitoring programmes in sampling and analyzing the newly listed POPs in core matrices and other media:
 - (a) Hexachlorobutadiene;
 - (b) Pentachlorophenol and its salts and esters;
 - (c) Polychlorinated naphthalenes;
 - (d) Decabromodiphenyl ether (BDE-209);
 - (e) Short-chain chlorinated paraffins;

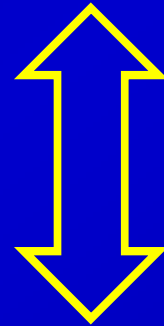


**Aim of CVUA Freiburg: inclusion also of voluntary POPs
(COP 7, COP 8)
(except PFAS, analysed at University Örebro)**

mandatory (according to PCA)	
2003	1) Initial POPs: aldrin, chlordane, DDT, dieldrin, endrin, HCB, heptachlor, mirex, toxaphene, PCB, PCDD, PCDF
2009	2) POPs listed at COP-4: chlordecone, HCH (alpha, beta, gamma), hexabromobiphenyl (PBB 153), Pentachlorobenzene, PBDE (47, 99, 153, 175/183-co-eluting); optional: BDE 100; PFOS
2011	3) POPs listed at COP-5: endosulfan
2013	4) POPs listed at COP-6: HBCDD (alpha, beta, gamma)
voluntary:	
2015	5) POPs listed at COP-7: Hexachlorobutadiene (Annex A), pentachlorophenol + salts + esters, polychlorinated naphthalenes
2017	6) POPs listed at COP-8: Decabromodiphenyl ether, SCCP, hexachlorobutadiene (Annex C)
2019	7) possible candidates at COP-8: dicofol, pentadecafluorooctanoic acid (PFOA) and salts, perfluorohexane sulfonic acid (PFHxS)

**Selected results and discussion –
examples for a complex picture**

Global WHO/UNEP-Studies 2000 - 2019



Asian countries 2016-2019

Aspects for differentiation

- ✓ **Parameters**
23 parameters
(without congeners, metabolites ...)
- ✓ **Regions**
 - **Continents**
 - **Countries**
- ✓ **Time trends**

**UNITED
NATIONS**



SC

UNEP/POPS/COP.6/INF/33

Distr.: General
26 March 2013

English only



**Stockholm Convention
on Persistent Organic
Pollutants**

**Conference of the Parties to the Stockholm
Convention on Persistent Organic Pollutants
Sixth meeting**

Geneva, 28 April–10 May 2013

Item 5 (i) of the provisional agenda*

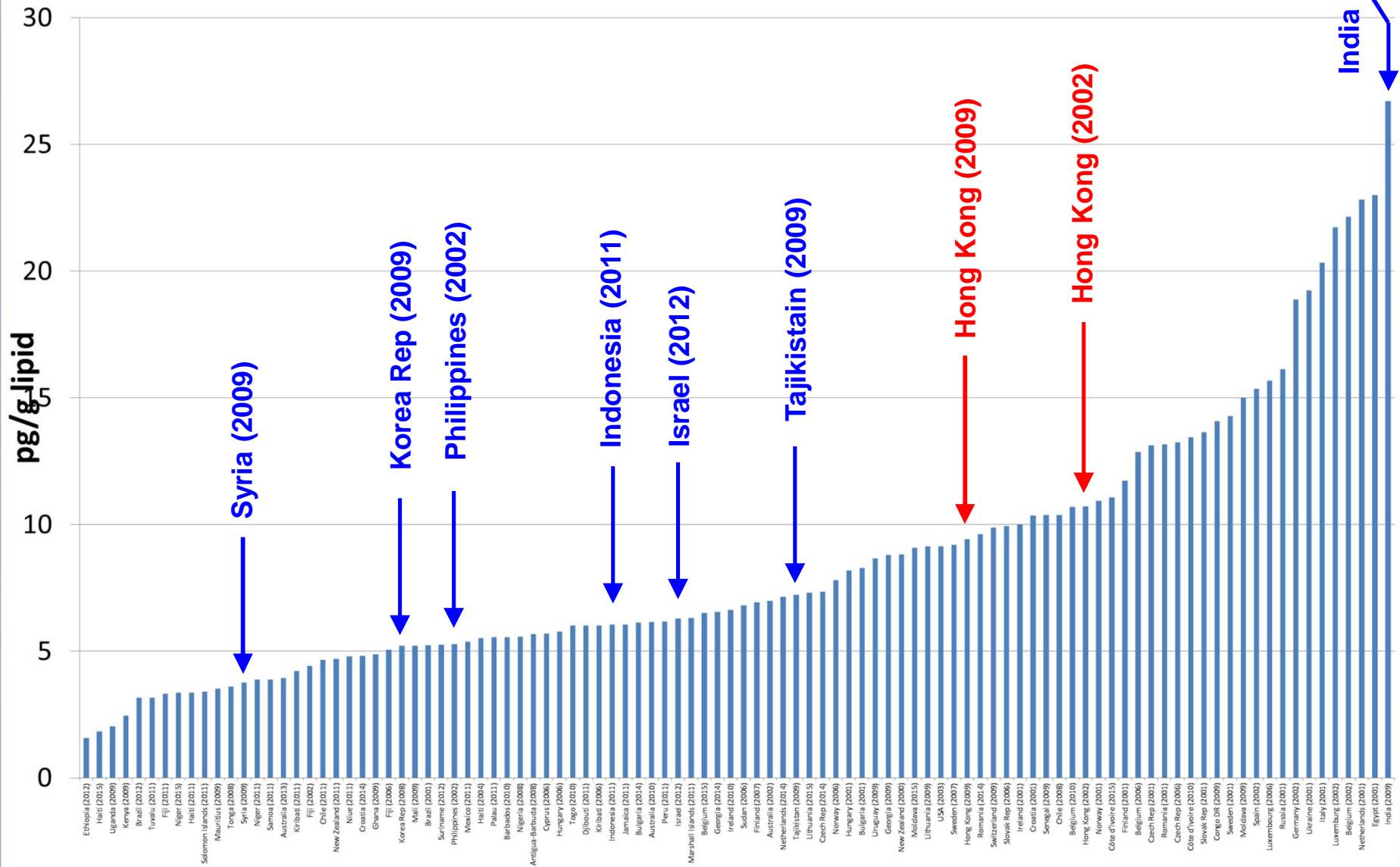
**Matters related to the implementation of the Convention:
effectiveness evaluation**

**Results of the global survey on concentrations in human milk of
persistent organic pollutants by the United Nations Environment
Programme and the World Health Organization**

Comparison of levels between countries

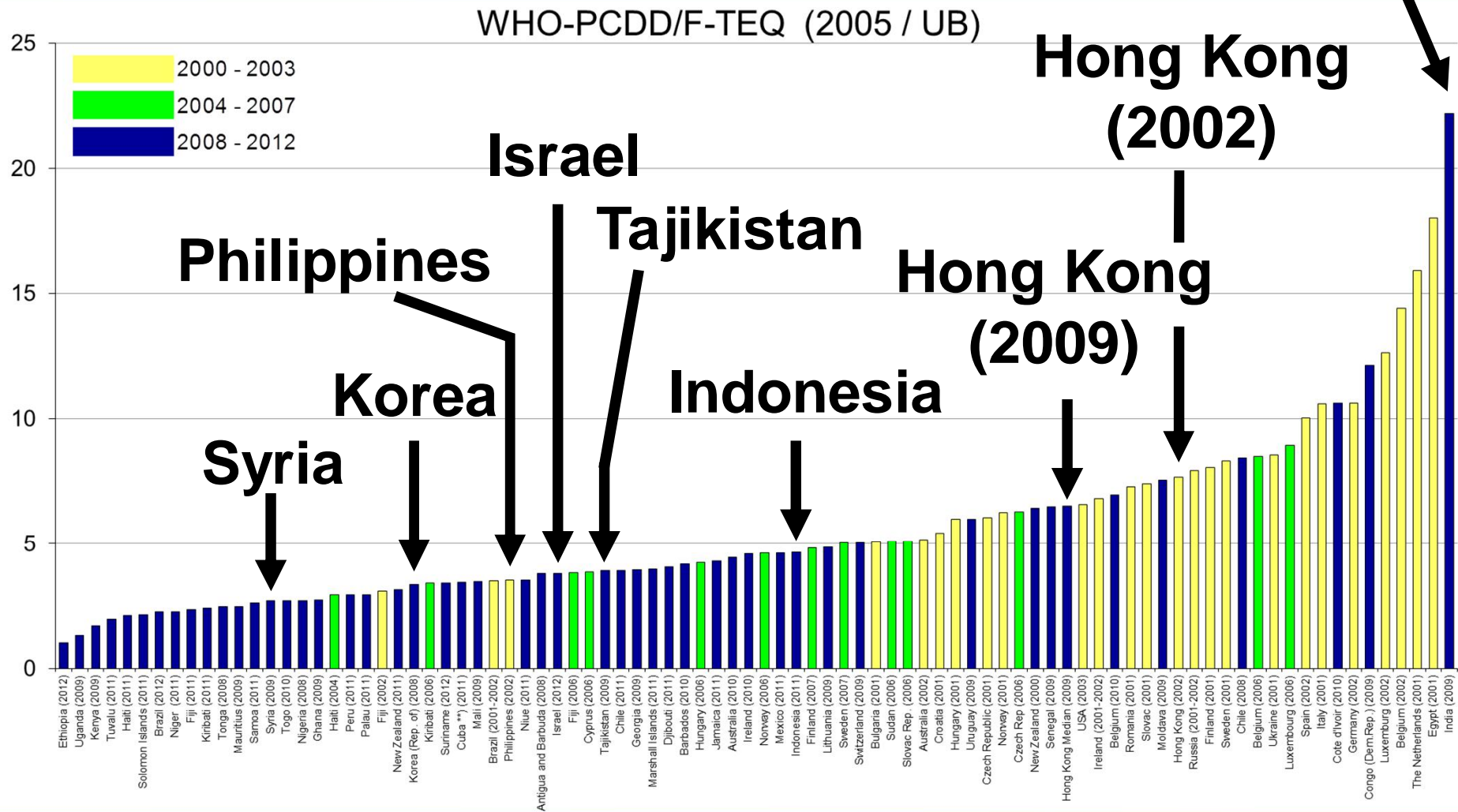
- NO „ranking“ between countries
- But identification of lower / middle / upper ranges
 - ✓ **Goal: findings allow setting priorities in different regions and countries**

WHO-PCDD/F-PCB-TEQ (2005 / UB)



Asia

India



pg WHO-PCDD/F-TEQ (2005) / g lipid

PCP and dioxins in guar gum from India

RASFF, July 2007, notification from Switzerland:

Very high contamination levels of dioxins and pentachlorophenol (PCP) found in certain batches of guar gum from India:

- about 1000 times the level of what can be considered as normal background contamination
- 9 EU Member States affected

Guar gum

- extracted from guar bean
- use as thickening, emulsifying, binding, gelling additive
- India produces about 80 % of world market
- **Food grade** guar gum: authorised as food additive
- **Industrial grade** guar gum: for non-food uses, e.g. in printing and textile industry
 - ✓ Technical note (2004): guar gum used as printing thickener for printing inks on textile (in particular in textiles made from polyester). Frequently/often preserved with pentachlorophenol (PCP).

Two EU missions to India (2007, 2009)

Na-PCP extensively used in India for industrial grade guar gum

- either sold as food grade
- or cross-contamination from industrial to food grade gums

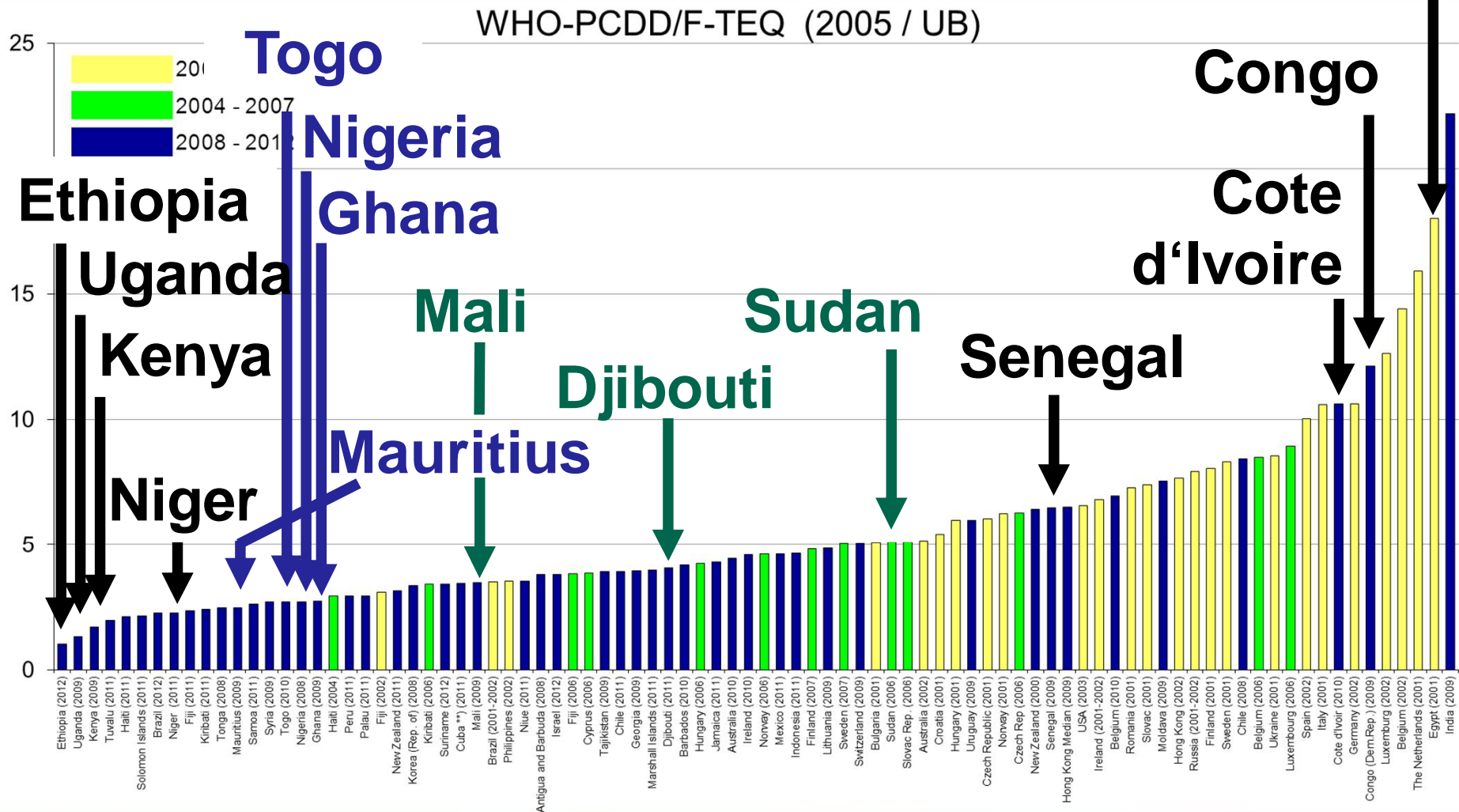
Detection in food grade guar gum points to use for textiles –

- **elimination of sources is quite complex (environment, food, ...)**

Africa

(3 rounds 2000 -2011)

Egypt



pg WHO-PCDD/F-TEQ (2005) / g lipid

Egypt

Ministry of State for Environmental Affairs
Egyptian Environmental Affairs Agency

EEAA-EIMP



➤ Main source of dioxins is waste incineration



Geophagia

- **Consumption of clay** quite common among ethnic minorities in the Netherlands, UK and certain parts of the population in Africa
- **Consumption of clay by pregnant women**
 - use against morning sickness, but also source of minerals



Clays collected from Africa (n=20)



- Increased dioxin levels in some of the clays with a highest observed level of 103 ng TEQ/kg



Ball clay / caolinitic clay I

Food and Drug Administration (USA), 1997:

- **Ball clay (bentonite) as source of dioxin contamination in poultry, commercial catfish and eggs**
- **Used as feed additive (to soybean meal, as flowing or anticaking agent)**
- **Origin: mine in Mississippi**

Ball clay / caolinitic clay II

EU, 1999:

- ✓ Caolinitic clay as source of dioxin contamination
- ✓ Feed additive (anticaking agent)
- ✓ Origin: mine in Germany
- ✓ Same PCDD/F pattern as in clay from Mississippi (OCDD-dominated; no furans; similar to PCP)
- ✓ Range of contamination:
> 100,000 to > 500,000 pg WHO-TEQ/kg

Ball clay / caolinitic clay III

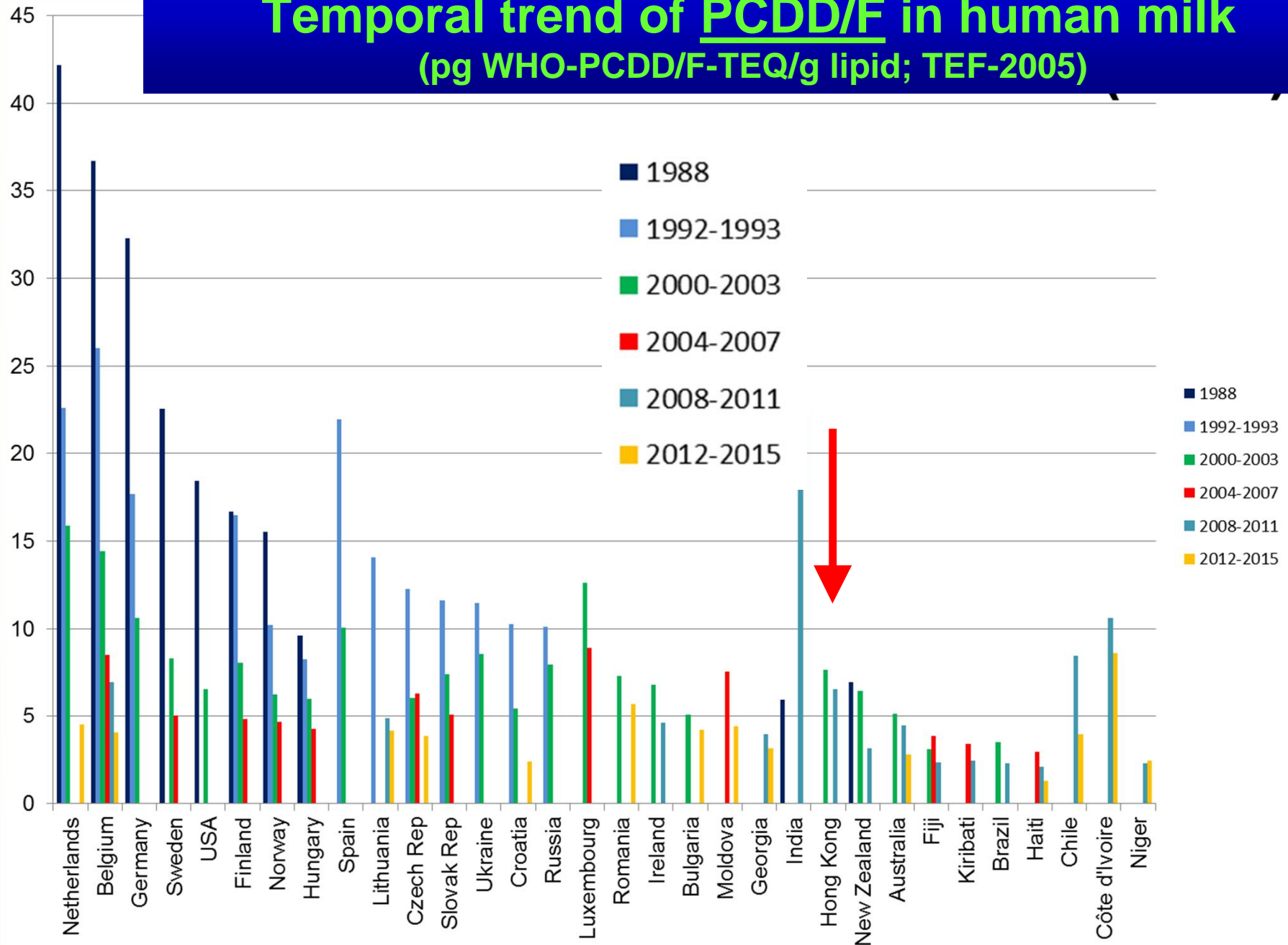
Obvious: natural source

- Possibly, geological processes formed this unique pattern of dioxins over time from organic material and chlorine.

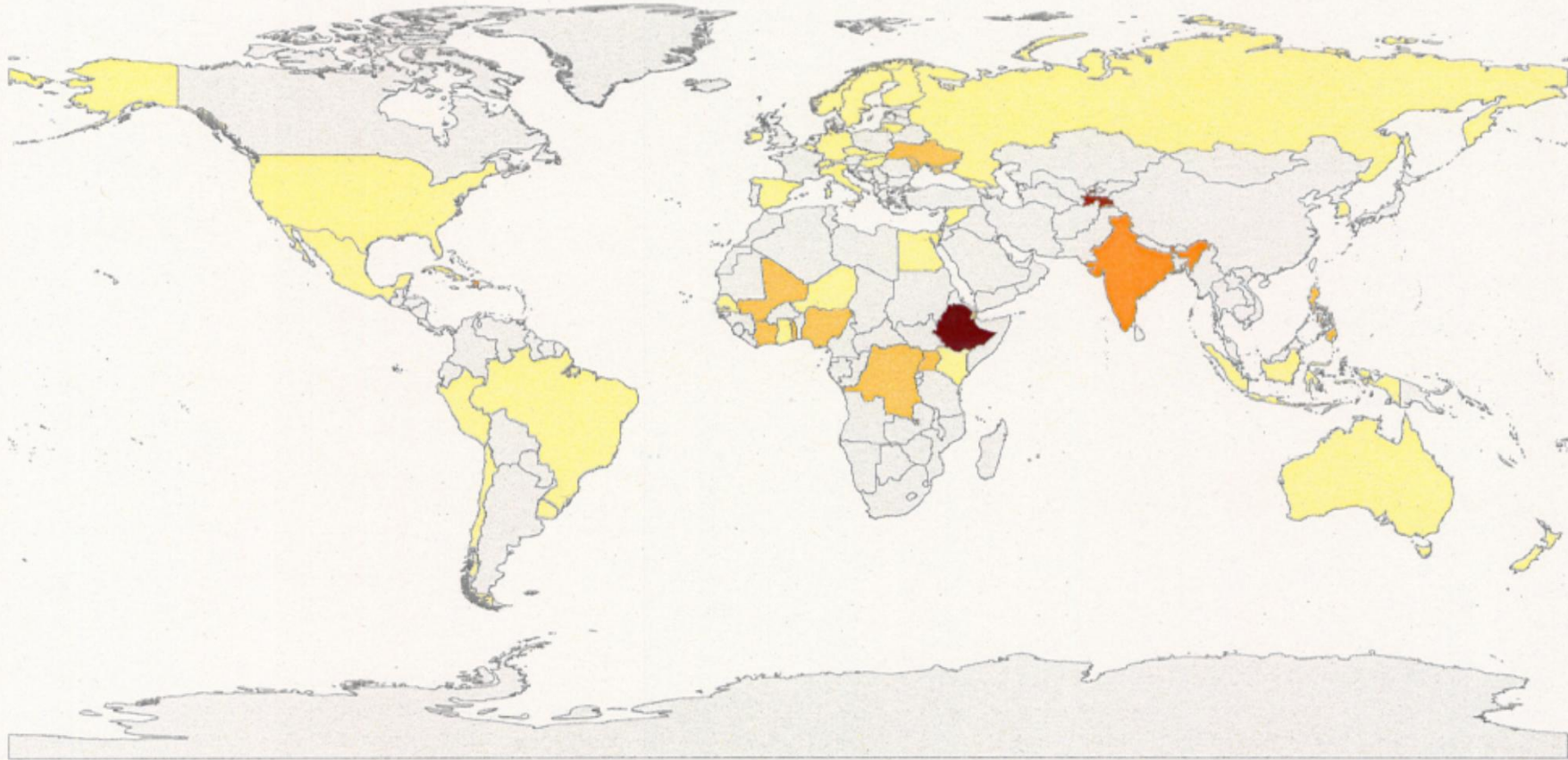
Conclusions for breast milk from Ivory Coast and Congo

- Dioxin pattern in clays can explain pattern in human milk
- Use of clay likely to be responsible for elevated dioxin levels in breast milk from some African countries
- Potential risk

Temporal trend of PCDD/F in human milk (pg WHO-PCDD/F-TEQ/g lipid; TEF-2005)



SUM DDTs



Legend



26 – 725



725 – 1740



1740 – 4320



4320 – 7640

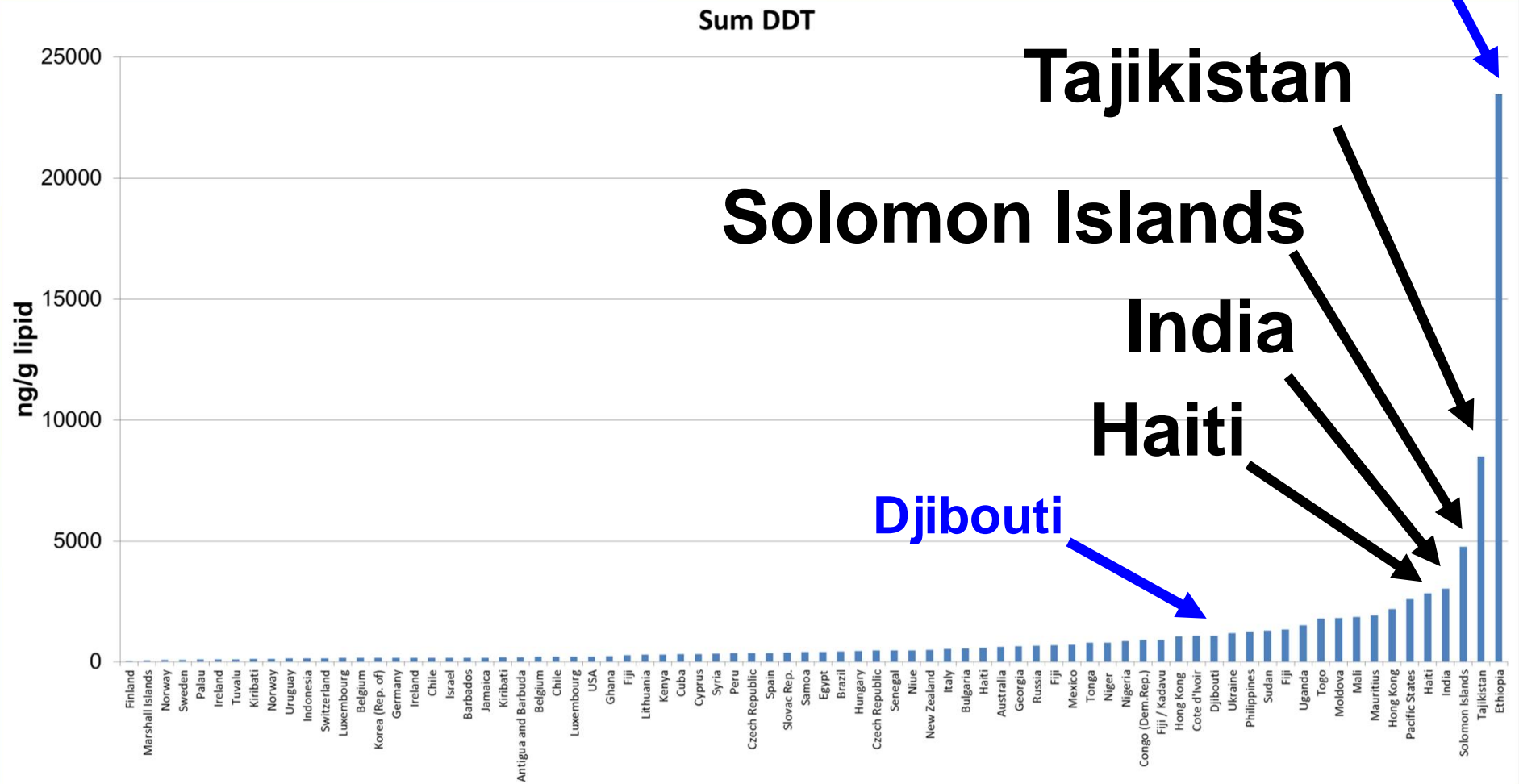


7640 – 22300

ng Sum DDT/g lipid

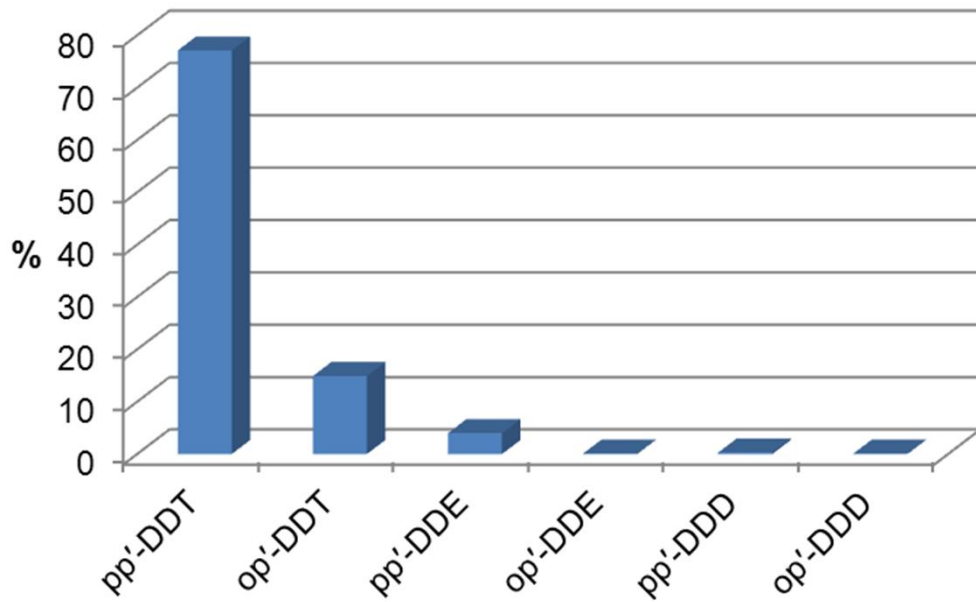
Median levels of DDT (ng/g lipid)

Ethiopia

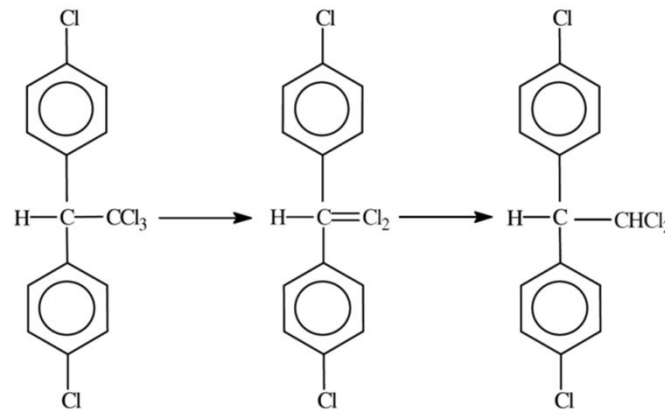


Max: 23500 ng sum DDT/g lipid (= 23.5 mg/kg)

DDT



Composition of technical DDT (%)



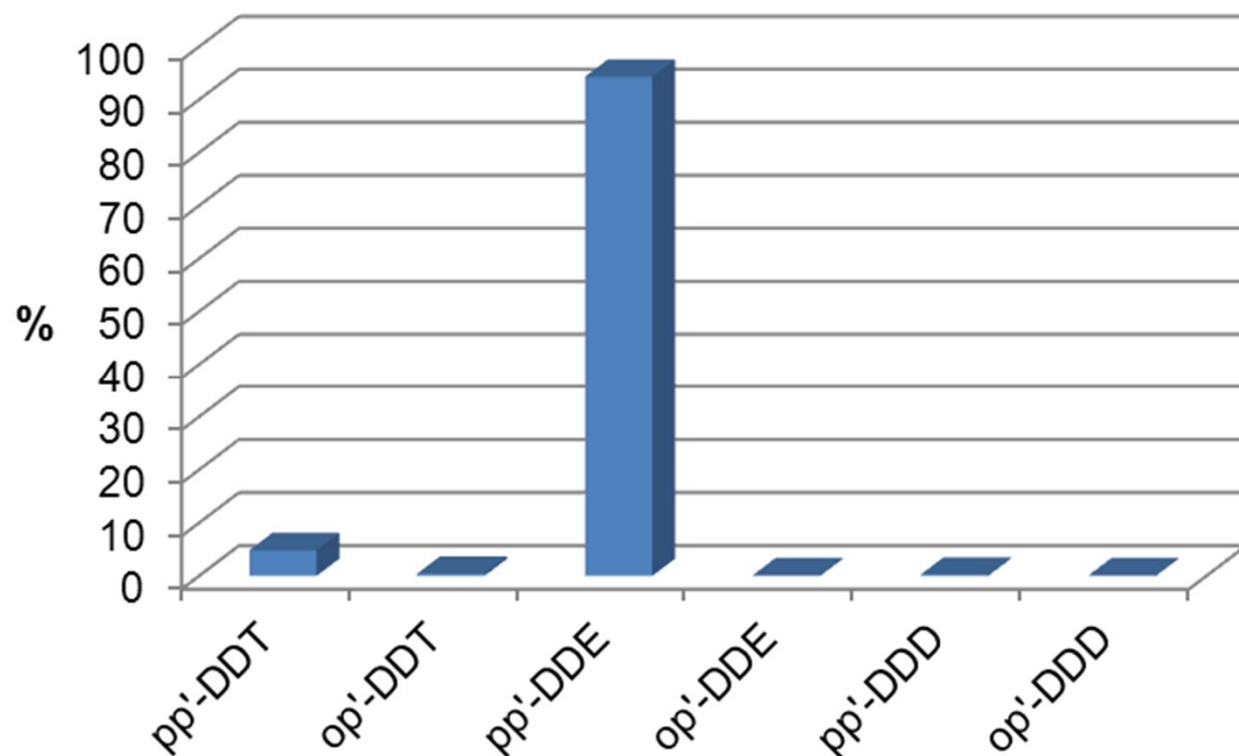
DDT

DDE

DDD

Contribution (%) to Sum DDT in humans

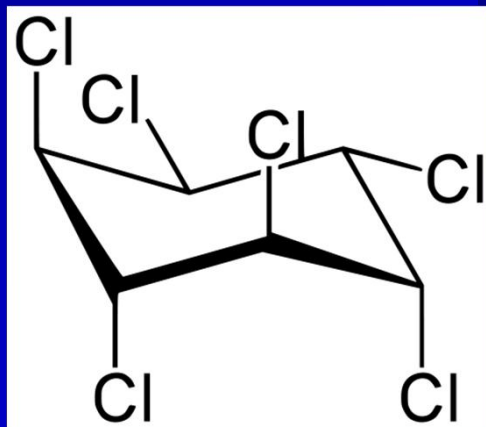
(all samples except from Ethiopia and Djibouti, median of 97 samples)



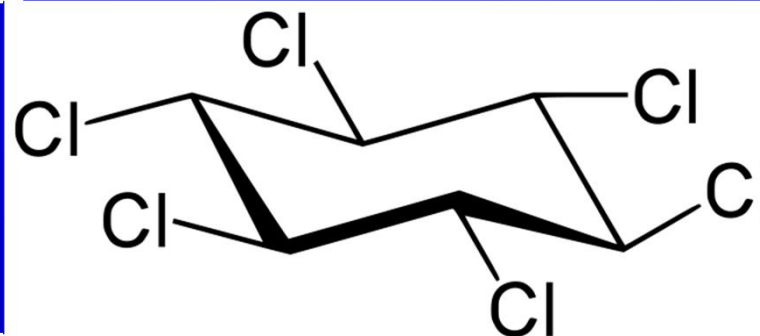
Technical HCH:

HCH

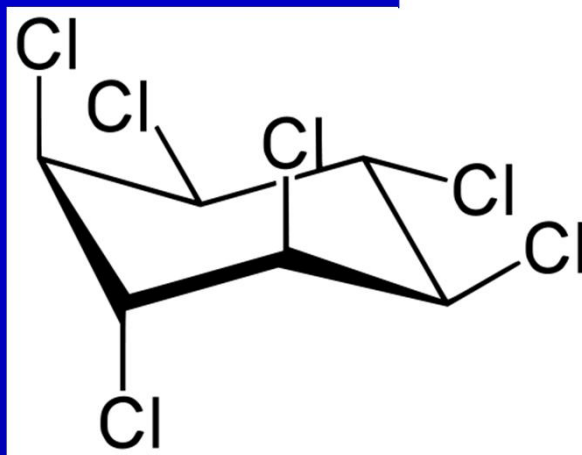
✓ **Alpha-HCH**
(65 – 70 %)



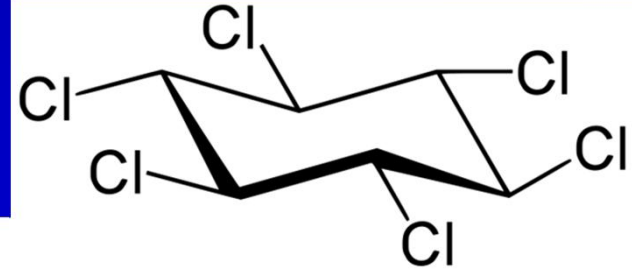
✓ **Beta-HCH**
(7 – 20 %)



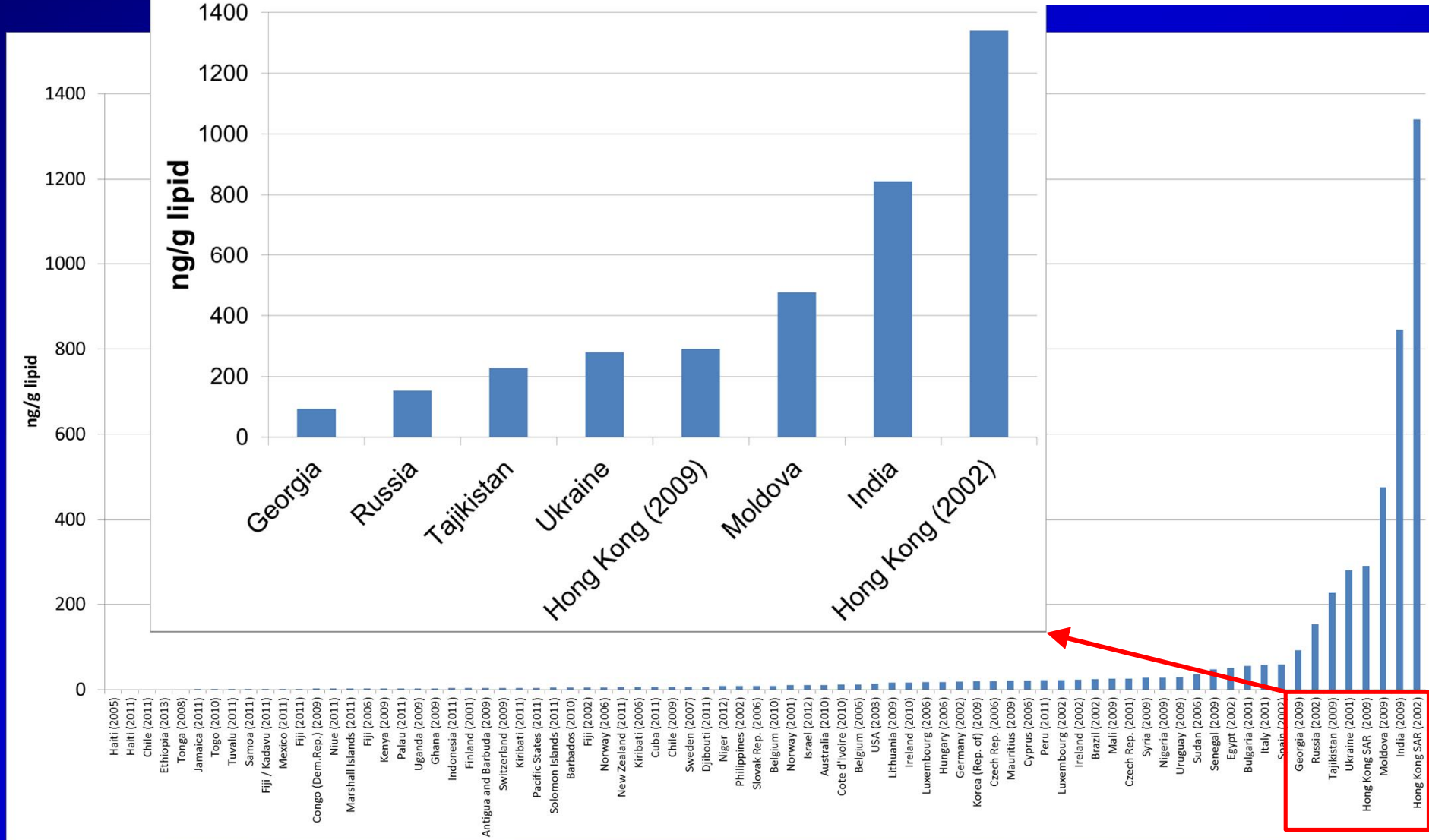
✓ **Gamma-HCH**
(14 – 15 %)



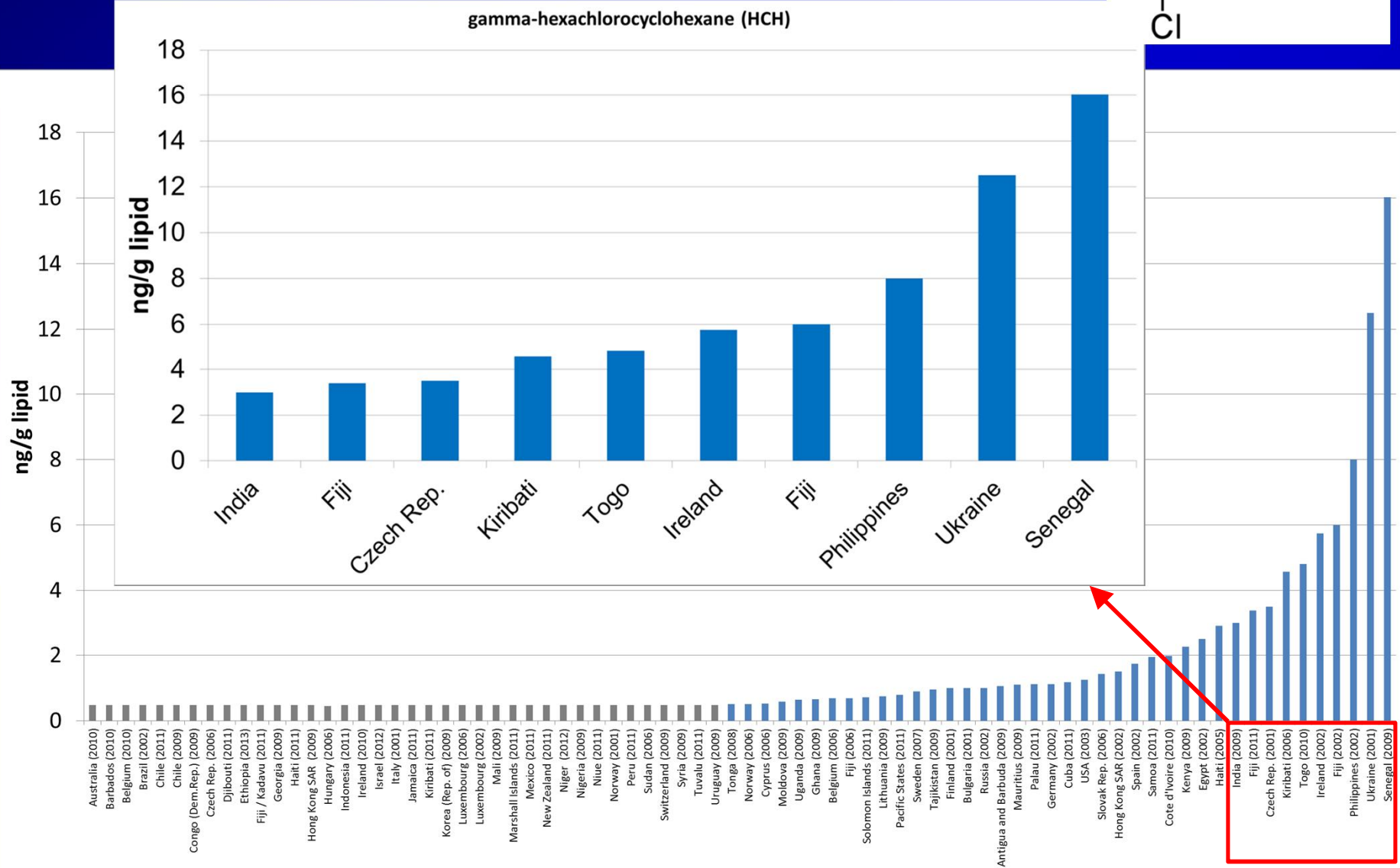
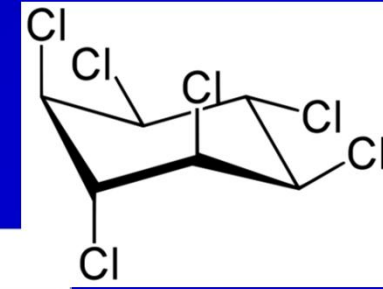
Beta-HCH



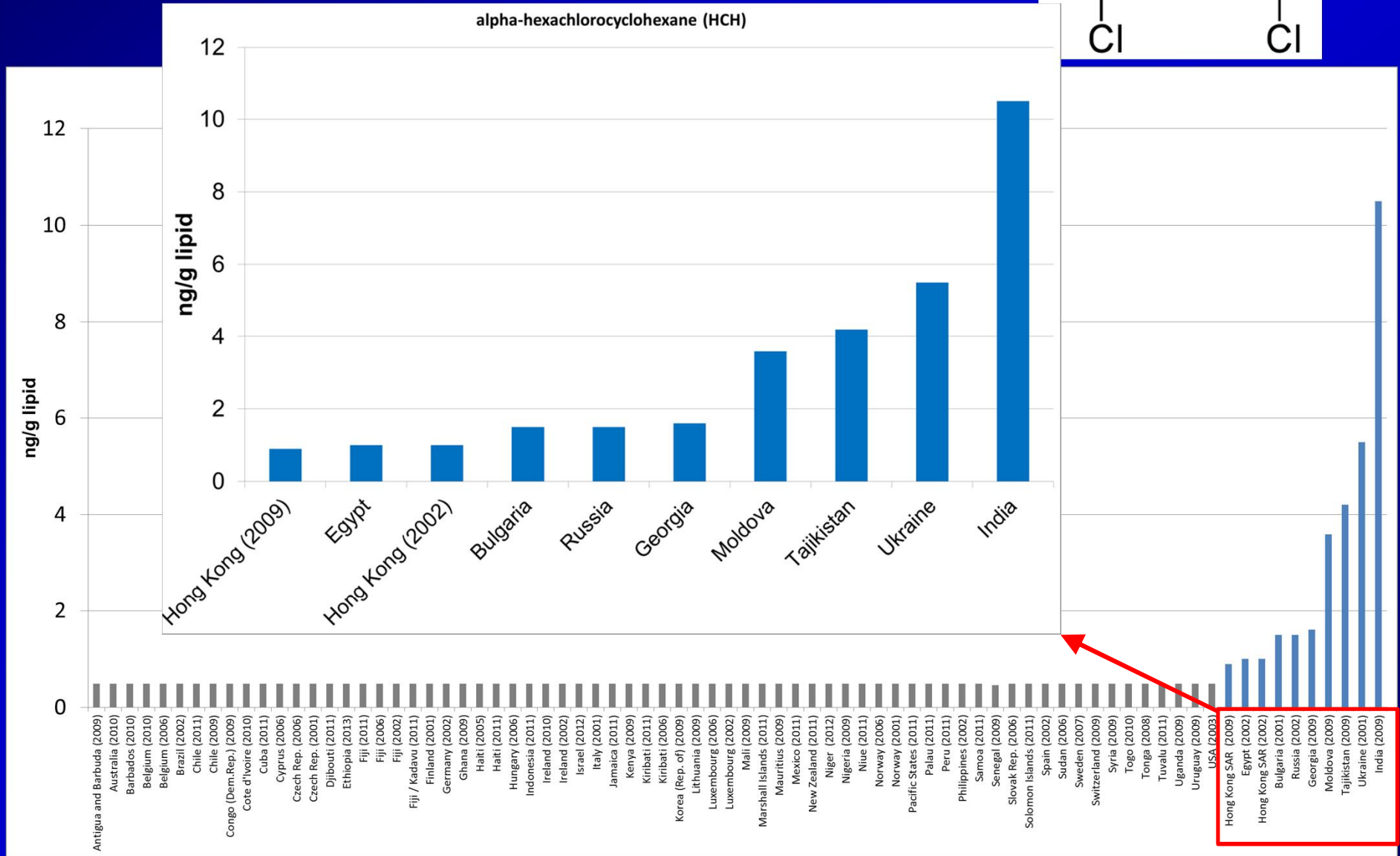
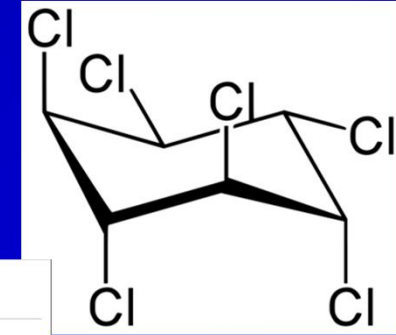
beta-hexachlorocyclohexane (HCH)



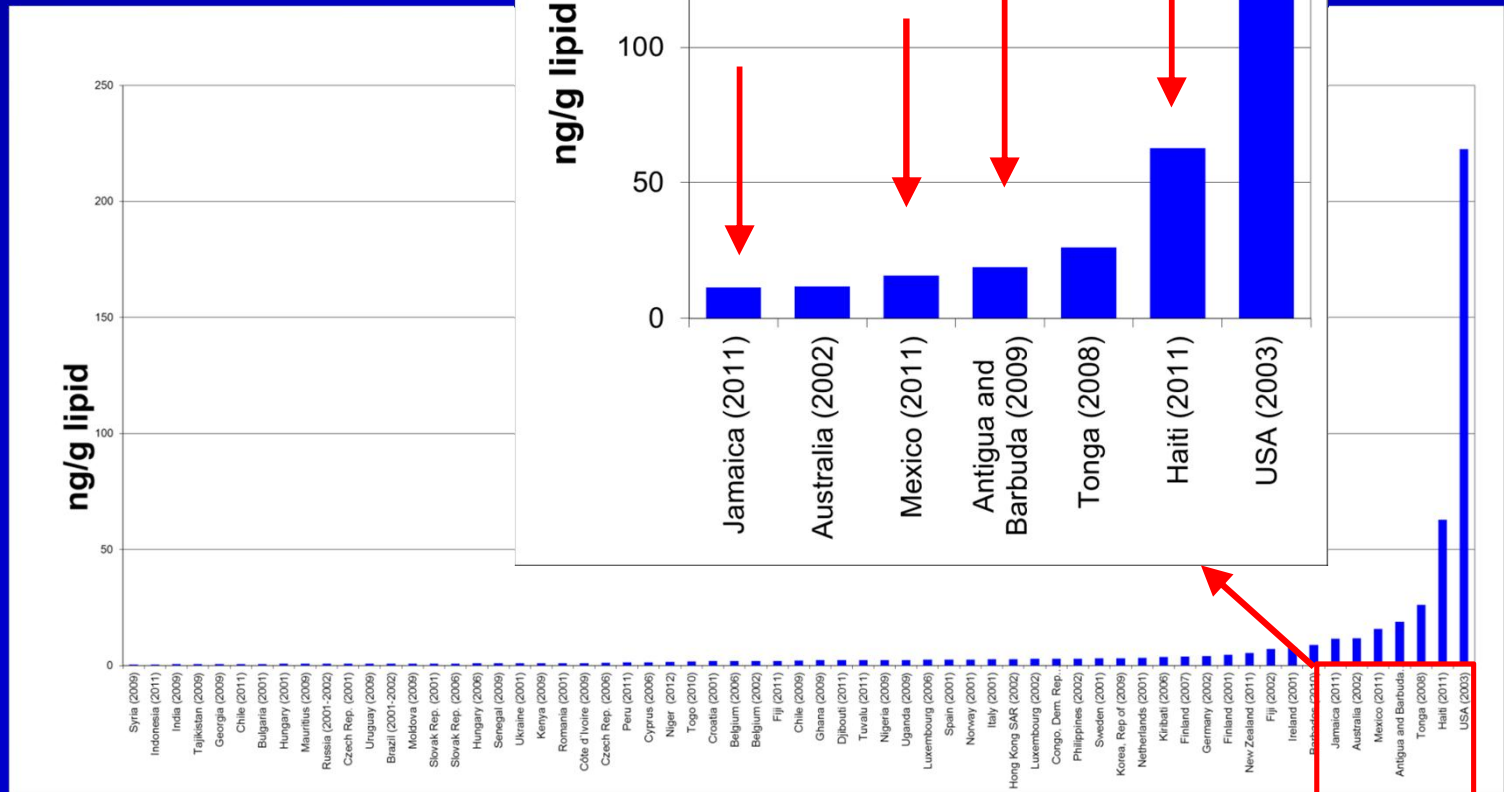
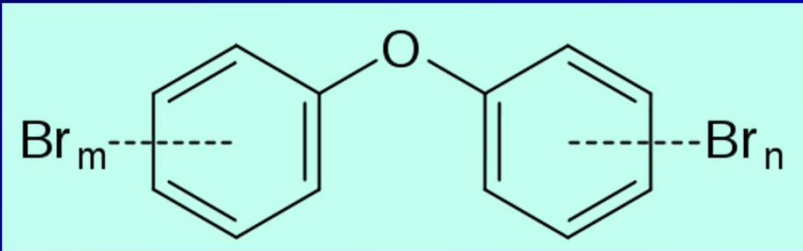
Gamma-HCH



Alpha-HCH



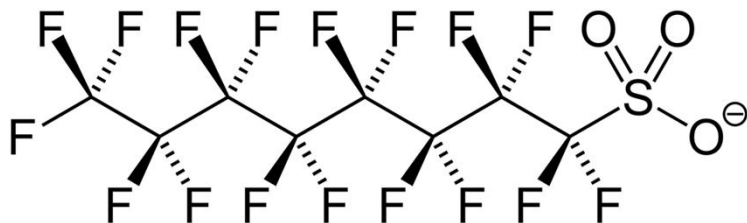
Sum PBDE



No increase of PBDD/F-levels with increasing PBDE levels

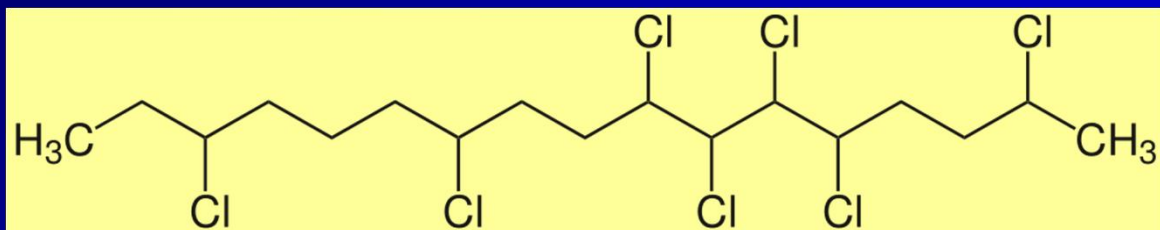
Perfluorinated compounds (PFCs)

- Lack of lipophilicity results in relatively low levels in milk compared to serum/blood (distribution milk/serum ~1:100).
- Distribution varies for different PFCs.
- More reliable data would be generated using blood/serum.



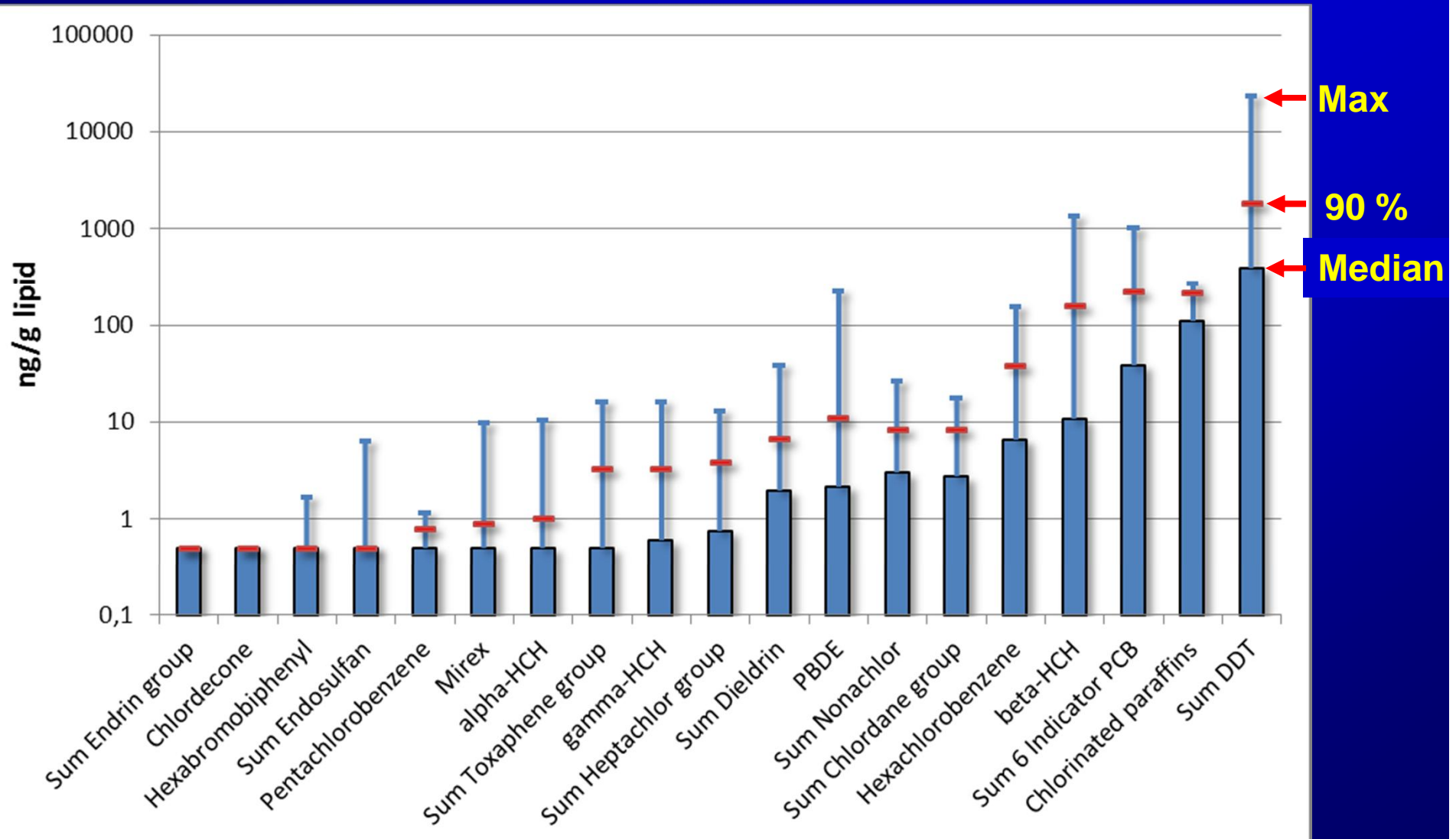
PFOS

Chlorinated Paraffins (CPs)

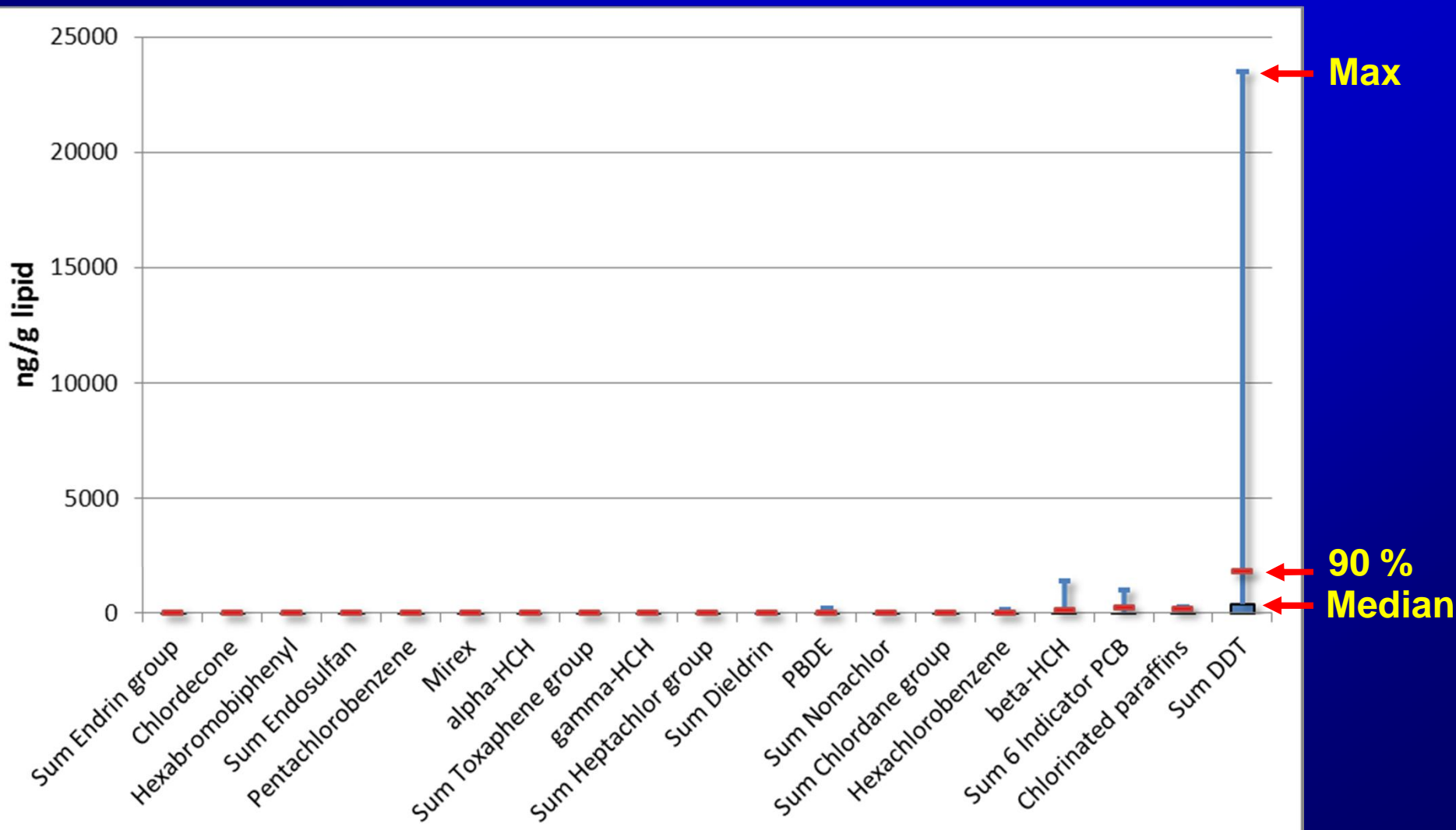


- ✓ general formula $C_nH_{2n+2-z}Cl_z$
- ✓ variation of chain length, number and position of chlorines
 - short chain CPs (SCCP; C10 – C13)
 - medium chain CPs (MCCP; C14 – C17)
 - long chain CPs (LCCP; > C17)
- ✓ **complex mixtures: > 10.000 compounds**
- ✓ **chlorine content of commercially available mixtures between 30 – 70 %**

Stockholm Convention POPs in breast milk (ng/g lipid)

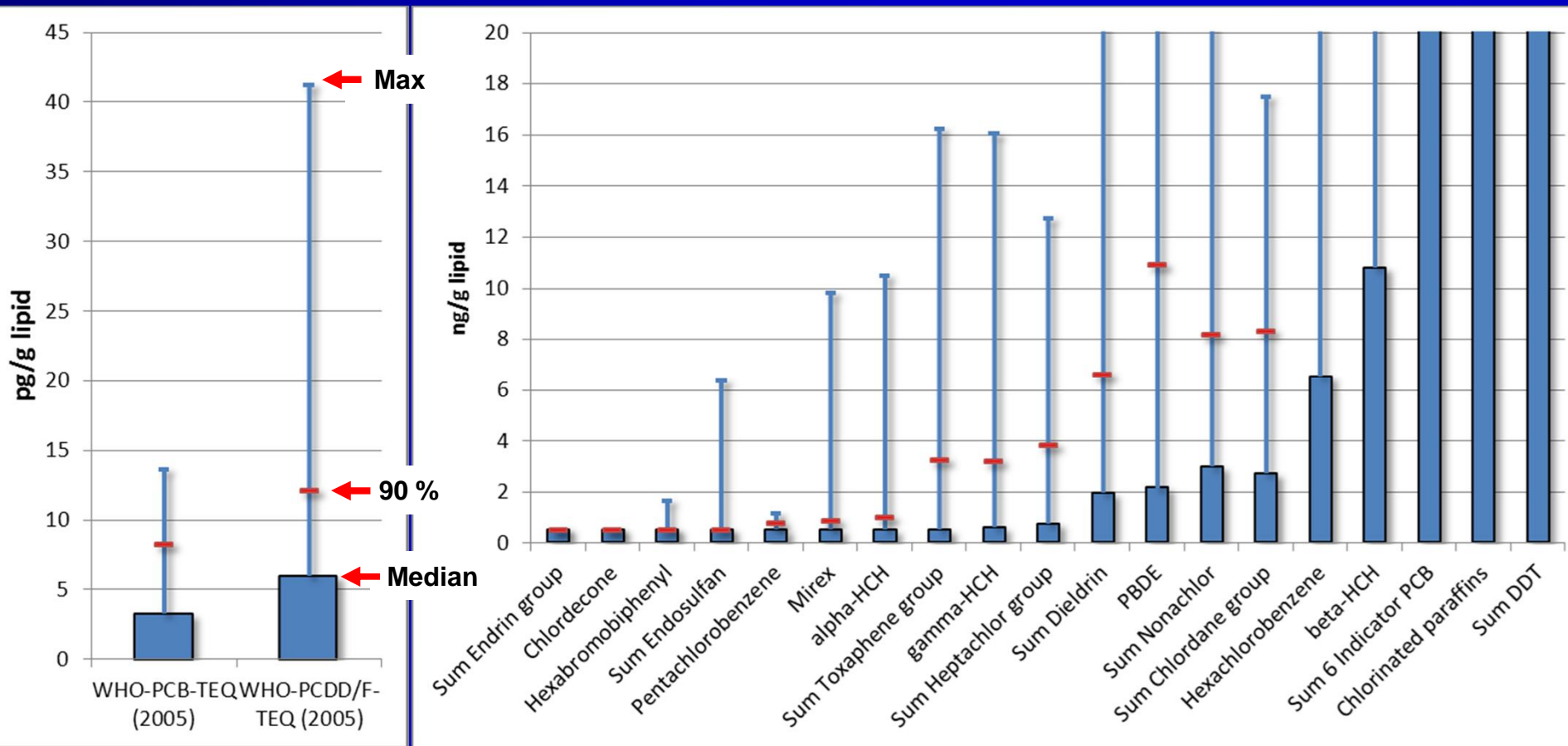


Stockholm Convention POPs in breast milk (ng/g lipid)



Stockholm Convention POPs in breast milk

(pg/g lipid resp. ng/g lipid)



WHO recommendation over last decades

- **Support and promotion of exclusive breastfeeding for the first six months**


Risk evaluation of breast feeding with regard to concentrations of PCDDs, PCDFs, PCBs and DDTs

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REVIEW ARTICLE

WHO/UNEP global surveys of PCDDs, PCDFs, PCBs and DDTs in human milk and benefit–risk evaluation of breastfeeding

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Risk evaluation of breast feeding with regard to concentrations of PCDDs, PCDFs, PCBs and DDTs

Conclusions from several studies:

- Prenatal exposure to these compounds is more important for effects than breastfeeding itself

Safety standards for human milk

Recommended intake for (PCDD/F + dl-PCB)-TEQ

- European Commission, Scientific Committee on Food (2001): tolerable weekly intake (TWI) of 14 pg WHO-TEQ/kg bw
- Joint FAO/WHO Expert Committee on Food Additives (JECFA) (2001): provisional tolerable monthly intake (PTMI) of 70 pg WHO-TEQ/kg bw/month
- US EPA (2010): oral reference dosis (RfD) of 0.7 pg TCDD/kg bw/day

Recommended intake for DDT

- WHO (2001): provisional tolerable daily intake (TDI) of 10 µg/kg bw
- US EPA and ATSDR (2011): oral reference dosis (RfD) of 0.5 µg/kg bw/day

Recommended daily intake

- **TDI, TWI, PTMI, RfD:**
meant for **chronic life time exposure**
- **Not applicable to breastfeeding situation**
(covering a much shorter period of life –
with exceedance of TWI/PTMI with one or two
order of magnitudes)

Comparison of results for human milk from WHO/UNEP studies (2000 – 2012) with „safe“ levels

	unit	safety standards as "Equivalent milk level"	Ranges in human milk (pooled samples)						
			Min	25th perc.	Median	75th perc.	90th perc.	95th perc.	Max
WHO-PCDD/F-PCB-TEQ (2005 / UB)	pg/g lipid	0.2 – 0.9	1,5	5,6	9,4	14,3	20,3	23,7	49,0
Total PCBs *)	ng/g lipid	7	2	18	38	121	223	347	1009
Sum DDT **)	ng/g lipid	2300	23	171	396	1015	1849	2616	23472

*) in human milk as sum of 6 indicator PCBs

***) in human milk calculated after correction of metabolites for molecular weight

Risk-benefit assessment for PCDDs, PCDFs, PCBs and DDTs

CONCLUSIONS (1) :

- Human milk levels of PCDDs, PCDFs and PCBs are still significantly above those considered safe
- Σ DDTs are below or around those considered safe in most countries.
- In comparison to pooled samples, individual samples will show some variation.
- Picture gets more complex, if other POPs included.

Risk-benefit assessment for PCDDs, PCDFs, PCBs and DDTs

CONCLUSIONS (2) :

- With respect to potential adverse health effects, in utero exposure is more important than lactational exposure.
- If potential adverse effects are balanced against positive health aspects for (breastfed) infants, the advantages of breastfeeding far outweigh the possible disadvantages.
- In view of the importance of *in utero* exposure due to maternal body burdens, all efforts should still be directed to further reducing human dietary and environmental exposure to these POPs.

Outlook

Complex evaluation possible after performance of 7th round based on cost-effective study **with pooled human milk samples** (as end-point of bioaccumulation)

- ✓ Regional differentiation allowing identification of priorities for follow-up with regard to wide range of POPs (including new POPs)
- ✓ Effectiveness Evaluation: Time trends for countries with repeated participation

Thank you !

