Summary

Mother's milk is healthy and of high psychological meaning to the newborn. Otherwise, we know mother milk may contain toxic residues. Since almost 30 years, the State-Laboratory of Basel-City is investigating mother's milk from women living in Basel-City and the surrounding villages. These periodical monitoring of human milk should give information about contaminants. Potential risks for man can be estimated based on such data. The present data, based on the periodic investigations of the State-Laboratory of Basel-City from 1970 to 2018, shall give an overview of the development of contaminants in human milk in Basel and surrounding villages[1] [2] [3] [4] [5] [6].

Introduction

Mother's milk is the first nutrition in our life but also an important indicator for studying bioaccumulation of contaminants in the human body. Since 1970, the State-Laboratory of Basel-City is analysing human milk. Many substances are enriched in fat tissue (including milk-producing cells) and can result in undesirable concentrations in the body. The focus in those years was on residues of organochlorine pesticides and polychlorinated diphenyls (PCB). Later on, other pesticide classes, such as insecticides or fragrances were monitored. In addition, the contamination with radioactive substances was of interest.

Materials and Methods

Collection of the human milk

All samples were collected with the help of the local University hospital of Basel-City, the Basle childbirth Trust and Mutter- und Vater-Beratungsstelle Basel. All samples were anonymized in the laboratory. When mothers were interested in the results of their own

[1] Schüpbach M R and Egli H. Rückstandsuntersuchung in Muttermilch. Kurzbericht aus der Insektizid-Abteilung des Kantonalen Laboratoriums Basel-Stadt. Internal report of the State-Laboratory of Basel-City 1971.
[2] Schüpbach M R and Egli H. Organochlorpestizide und polychlorierte Biphenyle in Muttermilch. Mitt. Gebiete Lebensm. Hyg. 70, 451-463 (1979).
[3] Reichert A, Durrer H. Egli H und Schüpbach M R. Muttermilchuntersuchungen in Basel, 1984/85: die Rückstände an Organochlorpestiziden und polychlorierten Biphenylen im Vergleich zur Situation von 1978. Mitt. Gebiete Lebensm. Hyg. 77, 554-564 (1986).
[4] Ramseier C, Raggini S, Eymann W (1998) Mitt Geb Lebensm Hyg. 89: 741-757 (1998).
[5] Zehringer M, Herrmann A. Analysis of polychlorinated biphenyls, pyrethroid insecticides and fragrances in human milk using a laminar cup liner in the GC injector. Eur Food Res Technol (2001) 212:247-251.
[6] Zehringer M. Radio-contamination of Swiss human milk. In: annual report of the State-Laboratory of Basel-City 2018: 41-45.

milk sample, the laboratory send them a standard report. The majority of the women were not interested in getting any personal information.

Analytical techniques

Organochlorine pesticides, polychlorinated biphenyls, pyrethroid and pyrethrine insecticides and nitro-musk fragrances were analysed with capillary gas chromatography and electron capture detection (ECD). The ECD-analysis were performed using two capillary columns of different polarity. Polycyclic musk fragrances were detected with a quadrupole mass-spectrometric detector.

Radio contaminants were analysed with gamma ray spectrometry with high-resolution Ge-detectors (radiocesium) and α,β-spectrometry with a gas-proportional counter (radiostrontium).

Details of the methods are described in the specific publications (foot-notes of page 1).

Results

Organochlorine pesticides and PCB

Some prominent insecticides were found in human milk:

p,p'-Dichlorotriphenyldichloroethane (DDT), banned in Switzerland in 1972. Hexachlorocyclohexane (BHC, the γ-isomere is called lindane), which was produced in the 1950ies and 1960ies near the border to France. BHC-dust was emitted into the town of Basel over years. After 1969, local production was stopped. Heptachlor epoxide is a degradation product of the insecticide heptachlor. Dieldrin is a powerful insecticide against ants. The fungicide hexachlorobenzene (HCB), also a chemical used for the synthesis of pentachlorophenol (PCP), was emitted from chemical industry in Badisch-Rheinfelden, 25 km above the town of Basel. Since 1993, emissions into the river Rhine were stopped. Polychlorinated diphenyls (PCB) were widely used in transformators, in oils, as plasticiser etc. In 2001, DDT, Heptachlor, Dieldrin, HCB and PCB were banned worldwide (Stockholm convention).

Year of investigation pesticides	1970 Basel	1978 Basel	1981[7] Zurich	1984/85 Basel	1993/95 Basel	2001 Basel
Lindane and isomeres (BCH)*	9.2 ± 13.9 N=50	13 ± 17 N=50	5.3 1 – 24, n=30	6.9 ± 5.8 N=70	3.9 ± 4.6 N=39	---
Hexachloro benzene (HCB)	53 ± 72 N=50	17 ± 17 N=50	3.7 1 – 13, n=30	5.5 ± 9.5 N=70	2.5 ± 3.2 N=41	---
Heptachloro epoxide	3.9 ± 4.4 N=50	1.0 ± 0.5 2.0 N=50	0.8 0.1 – 1.4, n=8	0.9 ± 0.4 N=69	<1 N=0	---
DDT and metabolites **	253 ± 175 N=50	98 ± 94 N=50	25 1 – 76, n=30	48 ± 29 N=70	15 ± 20 N=73	---
PCB ***	---	66 ± 45 N=50	34 10 – 85, n=30	73 ± 63 N=63	25 ± 17 N=59	16 ± 19 N=45
Dieldrin	---	---	0.5 0.1 – 1, n=6	0.8 ± 0.3 N=69	<1 N=0	

Table 1 Organochlorine pesticides and PCB in human milk of Basel, Zurich and surrounding villages.*total of the isomers α–,β– γ- and δ-HCH. **included the metabolites p,p'-DDD and p,p'-DDE. ***Sum of the detected PCBs is calculated as Aroclor 1260.

[7] Disler R.L., Glatt V., Meier W. Rückstände von chlorierten Insektiziden und polychlorierten Biphenylen in Humanmilch. Mitt Gebiete Lebensm. Hyg. 75, 205-213 (1984).

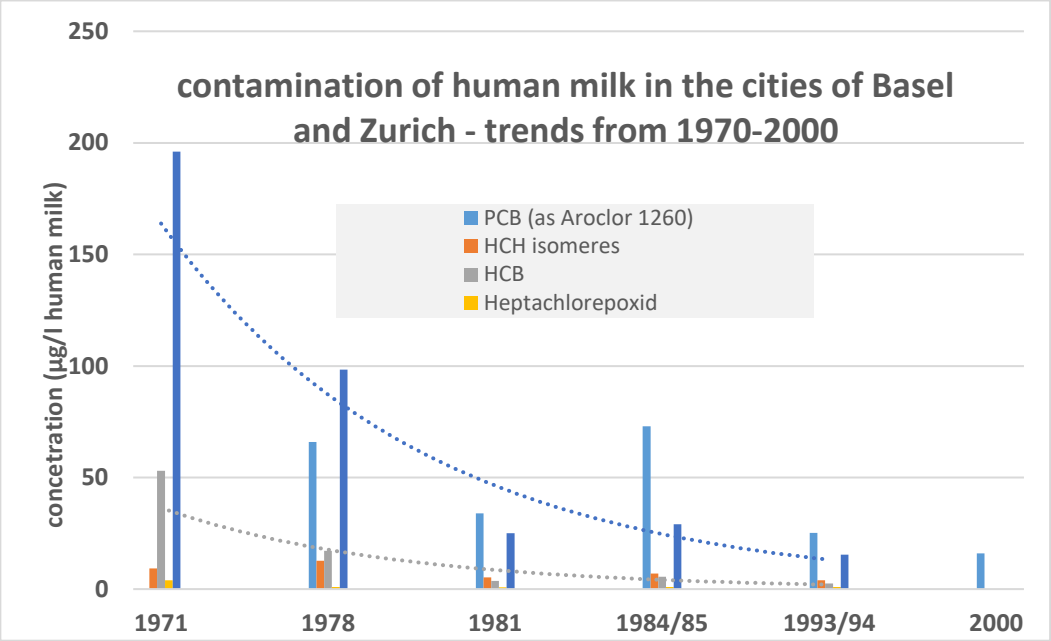


Fig 1 Organochlorine pesticides in Swiss human milk

In the 1970ies, DDT was the most prominent insecticide in human milk in Switzerland and other countries. The Swiss ban in 1972, led to a powerful reduction of the residues of over 90%. A similar, but smaller trend show HCB and lindane. PCB were banned from the Swiss market later than the insecticides, its trend showed a maximum in 1984/85, followed by a decrease to 25% of the level in 1978. The values for DDT in 1971 and 1984 are the median values instead of the arithmetic means.

Pyrethrins and pyrethroid insecticides

These groups of insecticides show a rapid degradation in the body and it seems that they do not accumulate in human milk. However, the database is poor: they are investigated rarely. Later investigations show, that these insecticides may be enriched in human fat tissues[8] when people are exposed to them (e.g. in agriculture).

Year of investigation Pyrethrins/Pyrethroids	Switzerland mean ± SD	Spain 2012[8] mean ± SD
Allethrin	38 ± 16, n=3	
Bifenthrin	82 ± 39, n=11	6 ± 7, n=67
Cyfluthrin	89 ± 59, n=2	
Fenpropathrin	54 ± 21, n=8	
Fluvalinate	60, n=1	
Permethrin	103, n=1	69 ± 23, n=100
Pyrethrins *	172 ± 93, n=14	

Table 2 Residues of pyrethrins and pyrethroids in human milk from women living in Basel and surrounding villages (values in µg/L human milk). A total of 14 substances was analysed for. *sum of six main constituents in pyrethrins. SD: standard deviation, n: number of samples with concentrations bellow LOD.

Fragrances

fragrance	1998/99, Basel	2004, Massachusetts[9]
Musk mosken	3.8 ± 1.3, n=4	---
Musk tibeten	25, n=1	---
Musk xylene	35, n=1	100 ± 120, n=39
Galaxolide	73 ± 46, n=55	703 ± 707, n=39
Tonalide	44 ±30, n=37	156 ± 121, n=39
Traseolide	38 ±10, n=2	---

Table 3 Summary of fragrances found in human milk. Investigations in 1998/99 (Basel) and 2004 (USA). From five analysed musk compounds, musk ambrette and musk ketone were not detectable. The polycyclic musk compounds muscon, phantolide, thibetolide, versalilde, cashmeran and phantolide were not detectable.

[8] Corcellas C et al. Pyrethroids in human breast milk: Occurrence and nursing daily intake estimation. Environment International 47 (2012) 17–22.
[9] Reiner J et al. Synthetic musc fragrances in human milk from the United States. Environ Sci Technol 2007, 41:3815-3820.

Fragrances are spread ubiquitously in the environment. According to their apolar structure, they accumulate mainly in fat tissues. Since some of the toxic nitro musk fragrances were banned, they can no more be detected in human milk. On the other side, polycyclic musk fragrances, such as galaxolide or tonalide, are enriched in higher concentration levels and can be detected more frequently in human milk. Chronic toxicity of these substance classes is still under discussion.

Radio contaminants

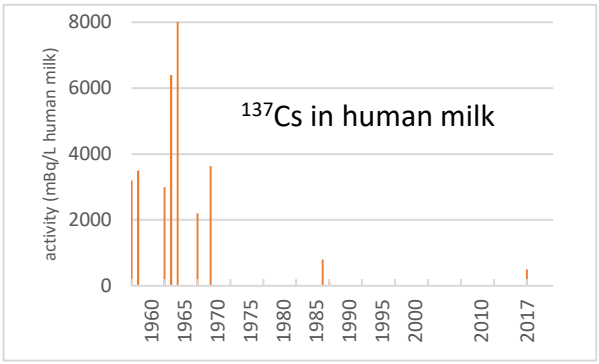
In 1986, only a few samples of collected human milk were analysed for radio-contamination. Unfortunately, only for ^{131}I data are available. In 2016/2017, human milk was collected in the city of Basel and the surrounding villages. The University-hospital of Basel could deliver us with 53 samples of 50 - 100 mL each. The samples had to be pooled. Otherwise, the detection would have been not sensitive enough for quantitative results. The pooling resulted in 25 samples of at 500 mL each.

Radionuclide	unit	1986	2017
^{40}K	Bq/L	---	17 ± 7, n=25
^{90}Sr	mBq/L	---	32 ± 12, n=19
^{131}I	Bq/L	17 ± 8, n=9	<20
^{134}Cs	mBq/L	---	<20
^{137}Cs	mBq/L	<5*	51 ± 37, n=13

Table 4 Summary of investigations of human milk from women living in Basel.*In all samples analysed after June 13 1986 the sum of the activities of ^{134}Cs and ^{137}Cs was below 5 Bq/L[10].

In 1986/87, the database for human milk was very poor. In Basel, only few investigations were realised in a non-systematic way. In those years, focus was on the radio-monitoring of food, and the capacity of the radiological laboratory in Basel was limited. The German laboratories investigated a series of 70 human milk samples (radiocesium and radioiodine). These results were reported only as a general summary with the comment that these values were not of great relevance. ^{131}I data was not published[10]. However, this radionuclide is of high importance for health of infants and foetuses. ^{131}I will be deposited in the glands. This can induce cancer (glands, leukaemia).

[10] Miethke H, Heffter A. , Hörtig W. Humanmilch-Untersuchungen 1980-1986. Deutsche Lebensm.-Runds. 84 (1988), 137-143.



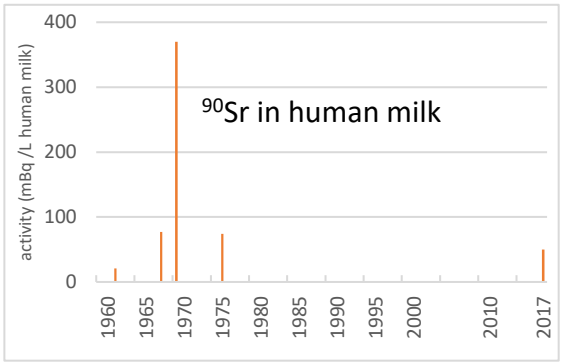


Fig 2 and 3 Radiocesium and radiostrontium levels in human milk from different countries.

After the maximal bomb fallout in 1962, a significant rise of the contamination in human milk was observed. In 2017, residues, due to global fallout and fallout from the reactor-fire at Chernobyl, are at a low level again.

Conclusions

The summarized investigations of human milk at the State-Laboratory of Basel-City throw spotlights on the contamination level of man for the years of sample collection. The following statements summarise these investigations.

- The ban of some persistent organochlorine pesticides and PCB resulted in a respectable decrease of these compounds in mother's milk in Basel and surroundings during the last 25 years. Today, concentrations are noticeable low, sometimes no more detectable.
- Residues of fragrances in human milk have to be monitored in the future. Their widespread use results in the accumulation in fat tissues.
- New substance classes should be monitored in human milk: That could be plasticizers, phosphorous pesticides, whitening agents and other substances, which may accumulate in fat tissues.
- Glyphosate, a polar herbicide, seems not to be accumulated in human milk[11,12].
- After emissions of artificial radioactivity, e.g. after a release from a nuclear power plant, a monitoring of human samples (human milk, urine, blood, hair, baby teeth) is essential for the assessment of doses received by the exposition to fallout. This monitoring cannot be replaced by a whole-body counting, which do not detect the incorporation of beta-nuclides, such as ^{90}Sr.

- [11]Steinborn A et al. Determination of Glyphosate Levels in Breast Milk Samples from Germany by LC-MS/MS and GC-MS/MS. *J. Agric. Food Chem.* 2016, 64, 6, 1414–1421.
[12] McGuire M et al. Glyphosate and aminomethylphosphonic acid are not detectable in human milk. *The American Journal of Clinical Nutrition*, Volume 103, Issue 5, May 2016, Pages 1285–1290,