

PCDD/PCDF in Butter Samples from Egypt

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1. Introduction

Food is considered to be the major source for human exposure to polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF). Thus, in 1994 a joint project as part of the cooperation between the German Research Society (DFG) of Germany and the Academy of Scientific Research and Technology (ASRT) of Egypt was used for a preliminary study to determine the PCDD/PCDF contamination of Egyptian food (1). To the best of our knowledge, this was the first survey of these contaminants in Egypt.

It was found

- that meat products sold in Cairo and its surrounding area had a low PCDD/PCDF-contamination;
- that two margarine samples sold in Cairo were low contaminated with PCDD/PCDF, also;
- that butter samples from different areas of Egypt had a wide range of PCDD/PCDF-contamination, however. Whereas samples from some areas met reference values proposed in Germany for milk and milk products, other exceeded these reference values considerably.

Further investigations were necessary to confirm the butter results. Now, new data are available to present an actual overview on the PCDD/PCDF contamination in butter samples from different Egyptian provinces.

2. Materials and Methods

8 samples of butter were collected from different locations in October 1995. Additionally, 9 butter samples from lower Egypt and 8 butter samples from upper Egypt were collected and shipped to Freiburg for dioxin analysis in February 1996.

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The basic steps of the analytical procedure for determination of PCDD/PCDF were presented elsewhere (2-3). The pure butter fat was spiked with all $^{13}\text{C}_{12}$ -labelled PCDD/PCDF. Clean up includes gelchromatography, silica gel/sulfuric acid and silica gel/NaOH column chromatography, florisil column chromatography and Carbopack C column chromatography. As recovery standard, $^{13}\text{C}_{12}$ -labelled 1,2,3,4-TCDD was used. GC/MS-detection was performed on a VG Autospec at 10,000 resolution using a 60 m DB5-MS-column. The AS 200 autosampler injected 5 μl into the Multinjector of a Carlo Erba Mega GC. With every sequence, a 5 point calibration curve was acquired in duplicate.

3. Results and Discussion

The general idea of these confirmation tests was to focus on 7 of the 26 Egyptian provinces and on the Cairo area. With these locations, more than 70 % of the Egyptian population is represented (the Cairo area involves 22 % of the total population, alone). The selected provinces include rural and urban areas.

In each of these provinces, 2 or 3 butter samples were collected. Thus, only a limited number of samples had to be analyzed to give a reliable overview on the situation in the most important parts of Egypt.

The results of the I-TEQ-contents of all butter samples collected in 1994, 1995 and 1996 are given in table 1. The congener pattern is shown in table 2. Samples of the same region were listed together.

For comparison, the following reference values are recommended in Germany for milk and milk products (in pg I-TEQ/g fat) (4, 5):

- < 0.9 Desirable target value to be met
- > 3 Investigate sources and start measures to minimize releases. Recommendation to stop distribution of milk and dairy products directly to consumer.
- > 5 Ban on trade of milk and dairy products

Although Egypt has not fixed any PCDD/PCDF-tolerance for food samples up to now, these German reference values can be used to find out whether increased levels of food contamination occur.

The results of the 33 butter samples show a wide range of PCDD/PCDF-contamination: Whereas all butter samples from El-Fayoum district and Kom Ombo have been below 3 pg I-TEQ/g fat, so far, nearly all samples from lower Egypt have exceeded even 5 pg I-TEQ/g fat. 7 samples from the El Monofia district had 10.3 pg I-TEQ/g fat in the mean. The mean of all samples was 7.69 pg I-TEQ/g fat.

| No. | year | origin | pg I-TEQ/g fat |
|-----|------|----------------------------|----------------|
| 1 | 1994 | Lower Egypt (H,) | 10.2 |
| 2 | 1994 | Lower Egypt (M,) | 28.9 |
| | | mean | 19.5 |
| 3 | 1994 | EL-Monofia Gov, (H,) | 8.55 |
| 4 | 1994 | EL-Monofia Gov, (M,) | 3.13 |
| 5 | 1995 | East Cairo, Monofia Dist, | 11.5 |
| 6 | 1995 | North Cairo, Monofia Dist | 5.71 |
| 7 | 1996 | Lower Egypt, Monofia | 15.3 |
| 8 | 1996 | Lower Egypt, Monofia | 13.8 |
| 9 | 1996 | Lower Egypt, Monofia | 13.8 |
| | | mean | 10.3 |
| 10 | 1995 | North Cairo, Sharkia Dist | 2.37 |
| 11 | 1996 | Lower Egypt, Al-Sharkia | 9.03 |
| 12 | 1996 | Lower Egypt, Al-Sharkia | 9.02 |
| 13 | 1996 | Lower Egypt, Al-Sharkia | 8.92 |
| | | mean | 7.34 |
| 14 | 1996 | Lower Egypt, Alesmaelia | 10.1 |
| 15 | 1996 | Lower Egypt, Alinsweira | 12.2 |
| 16 | 1996 | Lower Egypt, Al-Kasasin | 12.2 |
| 17 | 1994 | EL-Fayoum Gov,(H,) | 1.84 |
| 18 | 1994 | EL-Fayoum Gov,(M,) | 1.83 |
| 19 | 1995 | El-Fayoum Dist, | 1.80 |
| | | mean | 1.82 |
| 20 | 1995 | West-Cairo, El-Menya Dist | 1.72 |
| 21 | 1995 | South Cairo, Danta Dist, | 12.1 |
| 22 | 1995 | East Cairo, Khaliobia Dis | 10.9 |
| 23 | 1995 | South Cairo, Cairo Dist, | 0.85 |
| 24 | 1994 | Upper Egypt (H,) | 2.42 |
| 25 | 1994 | Bany Swif Gov,(M,) | 0.41 |
| 26 | 1996 | Upper Egypt, Bany Swif | 8.84 |
| 27 | 1996 | Upper Egypt, Bany Swif | 7.48 |
| 28 | 1996 | Upper Egypt, Bany Swif | 17.6 |
| | | mean | 8.57 |
| 29 | 1996 | Upper Egypt, Komombo | 0.81 |
| 30 | 1996 | Upper Egypt, Komombo | 0.64 |
| 31 | 1996 | Upper Egypt, Komombo | 1.04 |
| | | mean | 0.83 |
| 32 | 1996 | Upper Egypt, Menia (Malaw) | 4.22 |
| 33 | 1996 | Upper Egypt, Menia (Malaw) | 4.72 |
| | | mean | 4.47 |
| | | mean of all samples: | 7.69 |

Table 1. Results of PCDD/PCDF-contamination of butter samples of different regions of Egypt

Table 2. PCDD/PCDF pattern of butter samples of different regions of Egypt (in pg/g fat)

| No. | 2378- TCDD | 12378- PeCDD | 123478- HxCDD | 123678- HxCDD | 123789- HxCDD | 1234678- HpCDD | OCDD | 2378- TCDF | 12378- PeCDF | 23478- PeCDF | 123478- HxCDF | 123678- HxCDF | 234678- HxCDF | 123789- HxCDF | 1234678- HpCDF | 1234789- HpCDF | OCDF |
|------|---------------|-----------------|------------------|------------------|------------------|-------------------|-------|---------------|-----------------|-----------------|------------------|------------------|------------------|------------------|-------------------|-------------------|--------|
| 1 | 1,59 | 2,89 | 0,73 | 0,99 | 0,47 | 1,63 | 30,09 | 0,86 | 1,31 | 11,10 | 4,75 | 4,66 | 1,91 | 0,09 | 0,72 | 0,16 | 0,88 |
| 2 | 4,85 | 8,10 | 1,99 | 2,63 | 0,97 | 1,06 | 4,19 | 2,58 | 4,10 | 31,92 | 12,78 | 12,18 | 4,98 | 0,18 | 1,74 | 0,39 | 0,31 |
| mean | 3,22 | 5,50 | 1,36 | 1,81 | 0,72 | 1,35 | 17,14 | 1,72 | 2,71 | 21,51 | 8,77 | 8,42 | 3,45 | 0,14 | 1,23 | 0,28 | 0,60 |
| 3 | 1,18 | 2,51 | 0,69 | 1,01 | 0,30 | 0,57 | 1,62 | 1,55 | 1,82 | 9,45 | 3,98 | 3,70 | 1,58 | 0,04 | 0,65 | 0,09 | 0,10 |
| 4 | 0,44 | 0,82 | 0,25 | 0,42 | 0,16 | 0,54 | 2,03 | 0,65 | 0,43 | 3,51 | 1,40 | 1,32 | 0,67 | 0,00 | 0,37 | 0,06 | 0,15 |
| 5 | 1,33 | 3,00 | 0,78 | 1,03 | 0,57 | 1,75 | 3,29 | 0,69 | 1,54 | 13,31 | 6,38 | 6,46 | 2,50 | 0,00 | 1,43 | 0,16 | 0,20 |
| 6 | 0,69 | 1,41 | 0,42 | 0,68 | 0,28 | 1,32 | 4,41 | 0,78 | 1,28 | 6,63 | 2,91 | 2,58 | 1,24 | 0,00 | 0,58 | 0,09 | 0,23 |
| 7 | 1,78 | 4,69 | 1,23 | 1,40 | 0,45 | 0,61 | 1,51 | 1,93 | 2,93 | 17,51 | 7,35 | 7,29 | 2,90 | < 0,23 | 0,84 | 0,23 | < 0,19 |
| 8 | 1,95 | 3,98 | 1,00 | 1,15 | 0,37 | 0,52 | 1,10 | 1,85 | 2,49 | 15,65 | 6,58 | 5,46 | 2,39 | < 0,12 | 0,78 | 0,13 | 0,08 |
| 9 | 1,74 | 3,67 | 1,02 | 1,24 | 0,52 | 0,54 | 1,38 | 1,63 | 2,60 | 16,21 | 7,07 | 5,91 | 2,53 | 0,09 | 0,81 | 0,17 | 0,08 |
| mean | 1,30 | 2,87 | 0,77 | 0,99 | 0,38 | 0,84 | 2,19 | 1,30 | 1,87 | 11,75 | 5,10 | 4,68 | 1,97 | 0,02 | 0,78 | 0,13 | 0,12 |
| 10 | 0,28 | 0,59 | 0,20 | 0,29 | 0,16 | 0,96 | 22,52 | 0,26 | 0,51 | 2,70 | 1,23 | 1,21 | 0,48 | 0,00 | 0,27 | 0,04 | 0,14 |
| 11 | 1,40 | 2,48 | 0,47 | 0,61 | 0,21 | 0,33 | 1,15 | 0,62 | 1,29 | 10,54 | 3,92 | 3,32 | 1,31 | < 0,05 | 0,42 | 0,09 | < 0,05 |
| 12 | 1,46 | 2,38 | 0,51 | 0,63 | 0,21 | 0,45 | 1,45 | 0,73 | 1,43 | 10,49 | 3,98 | 3,07 | 1,27 | 0,05 | 0,46 | 0,07 | 0,07 |
| 13 | 1,42 | 2,54 | 0,55 | 0,59 | 0,22 | 0,53 | 1,89 | 0,70 | 1,35 | 10,21 | 3,75 | 3,20 | 1,38 | 0,04 | 0,45 | 0,10 | 0,13 |
| mean | 1,14 | 2,00 | 0,43 | 0,53 | 0,20 | 0,57 | 6,75 | 0,58 | 1,15 | 8,49 | 3,22 | 2,70 | 1,11 | 0,02 | 0,40 | 0,08 | 0,08 |
| 14 | 1,46 | 2,57 | 0,62 | 0,97 | 0,36 | 0,53 | 1,89 | 1,41 | 1,81 | 11,68 | 4,61 | 4,45 | 1,66 | < 0,05 | 0,54 | 0,10 | 0,06 |
| 15 | 1,67 | 3,31 | 0,82 | 1,26 | 0,37 | 0,98 | 4,43 | 1,39 | 1,80 | 14,11 | 5,81 | 5,32 | 2,14 | 0,04 | 0,74 | 0,12 | 0,08 |
| 16 | 1,57 | 3,47 | 0,62 | 1,18 | 0,33 | 0,50 | 1,62 | 1,44 | 1,84 | 14,27 | 5,39 | 5,15 | 1,99 | 0,05 | 0,62 | 0,09 | 0,11 |
| 17 | 0,22 | 0,51 | 0,15 | 0,26 | 0,17 | 1,05 | 32,72 | 0,13 | 0,19 | 2,05 | 0,81 | 0,84 | 0,42 | 0,04 | 0,35 | 0,04 | 0,16 |
| 18 | 0,29 | 0,48 | 0,16 | 0,26 | 0,16 | 0,91 | 33,21 | 0,22 | 0,18 | 1,90 | 0,84 | 0,88 | 0,40 | 0,04 | 0,20 | 0,03 | 0,08 |
| 19 | 0,23 | 0,43 | 0,12 | 0,24 | 0,14 | 1,10 | 26,68 | 0,31 | 0,39 | 2,01 | 0,86 | 0,83 | 0,37 | 0,00 | 0,26 | 0,05 | 0,22 |
| mean | 0,25 | 0,47 | 0,14 | 0,25 | 0,16 | 1,02 | 30,87 | 0,22 | 0,25 | 1,99 | 0,84 | 0,85 | 0,40 | 0,03 | 0,27 | 0,04 | 0,15 |
| 20 | 0,18 | 0,41 | 0,12 | 0,22 | 0,13 | 0,96 | 23,40 | 0,24 | 0,39 | 1,98 | 0,86 | 0,80 | 0,36 | 0,00 | 0,21 | 0,03 | 0,14 |
| 21 | 1,73 | 3,34 | 0,95 | 1,35 | 0,51 | 1,36 | 3,28 | 1,29 | 2,28 | 13,27 | 6,57 | 6,02 | 2,28 | 0,00 | 1,37 | 0,17 | 0,23 |
| 22 | 1,74 | 3,09 | 0,60 | 1,12 | 0,36 | 0,58 | 1,89 | 1,16 | 1,75 | 12,13 | 4,35 | 4,56 | 1,81 | 0,00 | 0,59 | 0,08 | 0,09 |
| 23 | 0,17 | 0,35 | 0,45 | 1,01 | 0,54 | 4,35 | 7,59 | 0,07 | 0,05 | 0,29 | 0,31 | 0,26 | 0,23 | 0,00 | 0,57 | 0,06 | 0,27 |
| 24 | 0,47 | 0,61 | 0,12 | 0,38 | 0,15 | 2,54 | 15,41 | 0,82 | 0,54 | 2,45 | 0,70 | 0,79 | 0,39 | 0,00 | 1,56 | 0,15 | 4,98 |
| 25 | 0,00 | 0,07 | 0,11 | 0,40 | 0,34 | 8,04 | 34,05 | 0,61 | 0,07 | 0,17 | 0,07 | 0,07 | 0,06 | 0,00 | 0,14 | 0,00 | 0,14 |
| 26 | 1,04 | 2,24 | 0,48 | 0,89 | 0,24 | 0,43 | 1,61 | 1,29 | 1,43 | 10,55 | 4,30 | 4,10 | 1,90 | < 0,06 | 0,67 | 0,17 | 0,06 |
| 27 | 0,81 | 1,92 | 0,54 | 0,87 | 0,33 | 0,51 | 2,19 | 1,01 | 1,08 | 8,99 | 3,46 | 3,61 | 1,62 | 0,05 | 0,68 | 0,14 | 0,17 |
| 28 | 1,87 | 4,65 | 1,14 | 1,69 | 0,71 | 0,83 | 1,30 | 1,61 | 2,47 | 20,93 | 9,68 | 8,18 | 4,31 | 0,09 | 1,84 | 0,31 | 0,11 |
| mean | 0,93 | 2,22 | 0,57 | 0,96 | 0,40 | 2,45 | 9,79 | 1,13 | 1,26 | 10,16 | 4,38 | 3,99 | 1,97 | 0,03 | 0,83 | 0,16 | 0,12 |
| 29 | 0,13 | 0,17 | 0,05 | 0,13 | 0,05 | 0,30 | 1,51 | 0,29 | 0,17 | 0,89 | 0,33 | 0,29 | 0,12 | < 0,04 | 0,09 | < 0,02 | < 0,05 |
| 30 | 0,07 | 0,14 | < 0,04 | 0,10 | 0,05 | 0,22 | 1,12 | 0,30 | 0,13 | 0,76 | 0,22 | 0,21 | 0,14 | < 0,01 | 0,07 | < 0,01 | 0,05 |
| 31 | 0,14 | 0,21 | 0,08 | 0,17 | 0,08 | 0,35 | 1,23 | 0,32 | 0,14 | 1,17 | 0,49 | 0,43 | 0,39 | < 0,02 | 0,10 | 0,02 | 0,04 |
| mean | 0,12 | 0,18 | 0,04 | 0,13 | 0,06 | 0,29 | 1,29 | 0,31 | 0,15 | 0,94 | 0,35 | 0,31 | 0,21 | 0,00 | 0,09 | 0,01 | 0,03 |
| 32 | 0,50 | 1,08 | 0,19 | 0,54 | 0,16 | 0,70 | 3,06 | 0,43 | 0,53 | 5,13 | 1,66 | 1,82 | 0,92 | < 0,05 | 0,33 | 0,05 | 0,13 |
| 33 | 0,59 | 1,21 | 0,23 | 0,59 | 0,21 | 0,53 | 1,22 | 0,75 | 0,68 | 5,58 | 1,94 | 2,16 | 1,03 | < 0,02 | 0,49 | 0,07 | 0,07 |
| mean | 0,54 | 1,15 | 0,21 | 0,56 | 0,19 | 0,62 | 2,14 | 0,59 | 0,61 | 5,35 | 1,80 | 1,99 | 0,98 | 0,00 | 0,41 | 0,06 | 0,10 |
| all: | 1,06 | 2,10 | 0,53 | 0,80 | 0,31 | 1,14 | 8,36 | 0,91 | 1,24 | 8,77 | 3,62 | 3,37 | 1,44 | 0,02 | 0,63 | 0,11 | 0,29 |

Obviously, there can be a wide range of contamination in the same province (above all, Bany Swif: minimum 0.41 pg I-TEQ/g fat, maximum 17.6 pg I-TEQ/g fat).

Generally, PCDF contribute about 70 % of the I-TEQ-burden. 2,3,4,7,8-PeCDF is toxicologically the predominant congener (Table 2).

The relatively high contamination of butter samples of large areas (as in the most of lower Egypt and of the northern parts of upper Egypt) is surprising. These amounts indicate local sources of contamination. However, further investigations are necessary to detect these sources.

4. Summary

33 Butter samples from different areas of Egypt were analyzed for PCDD/PCDF contamination. These areas were rural as well as urban; they include about 70 % of the Egyptian population. A wide range of PCDD/PCDF-contamination was found. Whereas samples from some areas meet reference values proposed in Germany for milk and milk products, others exceed these tolerances, many samples considerably. The mean of all samples was 7.7 pg I-TEQ/g fat, with a range from 0.41 to 28.9 pg I-TEQ/g fat. Further investigations are necessary to detect the sources of the PCDD/PCDF contamination of the environment.

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