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## DIOXINS AND PCBs IN EGGS AND EGG PRODUCTS – A STATISTICAL EVALUATION OF DATA RECEIVED BY AN ANALYTICAL SERVICE PROVIDER

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### Introduction

Findings of elevated levels of polychlorinated dibenzo-p-dioxins and -furans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in eggs above the EU-maximum levels for food raised ongoing attention in the public over the last years. For this reason a statistical evaluation of data received from more than 8400 samples analysed within the routine analysis of eggs and egg products on behalf of companies resp. authorities from the food sector was performed in order to gain an overview of levels of dioxins and PCBs in these types of food products.

### Materials and methods

**Samples.** Sampling was done resp. organized by the customers of the analytical service provider Eurofins GfA Lab Service. Samples were not selected following a specific study design, but were therefore received due to the customers' own strategies. As a side-effect, informative content on samples is sometimes restricted when customers were not generous in sharing information. The statistical data analysis described in the following was therefore done on basis of the information that is available. More than 8400 samples - from various, mainly European sources in 2008 to 2016, being clearly recognizable as egg and egg products - were selected for this study, only taking into account those products that might be used as food products.

**Analysis of PCDD/Fs, DL-PCBs and NDL-PCBs.** Analyses of 2,3,7,8-substituted congeners of PCDD/Fs, dioxin-like PCBs (DL-PCBs) and non-dioxin-like PCBs (NDL-PCBs, ICES6-PCBs) were performed on samples of eggs and egg products like whole egg, egg yolk or albumin according to the analytical procedures of Eurofins GfA Lab Service using high resolution gas chromatography and high resolution mass spectrometry (HRGC/HRMS) or Quadrupole GC/MS/MS analogue to methods described before [1, 2]. The applied analytical procedures - being accredited by DAkkS (German Accreditation Body) according to DIN EN ISO 17025:2005 - fulfill the requirements of the European Union (EU) concerning confirmatory methods of sampling and analysis for the control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs [3]. In brief, for each native PCDD/F-, DL-PCB- and NDL-PCB-congener to be determined the corresponding <sup>13</sup>C<sub>12</sub>-UL-labeled internal standard was added to the samples before the extraction procedure. After spiking the samples were Soxhlet extracted overnight with a mixture of appropriate polar and non-polar solvents for ultratrace-analyses (e.g. nanograde). Lipid determination was performed gravimetrically. Subsequently the crude extract samples were subjected to a clean-up procedure using a multicolumn system (involving carbon-on-glasfibre or carbon-on-celite for PCDD/Fs and PCBs). Further <sup>13</sup>C<sub>12</sub>-UL-labeled internal standards were added to the extracts for the determination of the recovery of the internal standards added before. Analyses were performed by HRGC/HRMS on Waters Autospec or DFS mass spectrometers or GC-MS/MS. For each substance 2 isotope masses were measured. The quantification was carried out by means of the isotope dilution method with the use of internal and external standards including a multi-point calibration. TEQ values are calculated by using WHO(2005)-TEFs according to the upper bound procedure. Results are related to the lipid content or fresh weight of the samples depending on the reference base of the corresponding EU-maximum value or action level [4, 5].

### Results and discussion

8439 different data sets were statistically evaluated. The whole database includes samples being clearly recognizable as hen eggs (80%), hen egg products (18%), preparation of hen eggs like custard or boiled eggs (<2%) as well as some eggs or egg products from other poultry than hens (duck or quail, <0,3%). For hen egg samples the method of rearing is known for 85% of the samples consisting of 26% organic eggs, 40% eggs from free ranging hens (free-range eggs), 32% barn eggs, remaining samples originate

from caged hens. The origin is clearly recognizable for nearly 80% of the samples. Roughly 75% of all hen egg samples were reared in Germany, the Netherlands or Belgium.

Each about 46% of the hen egg product samples could be recognized as being whole egg or egg yolk products. About 6% of the analysed egg product samples belong to the group of albumen products. All hen egg products are either fresh or dried, with or without additional ingredients. The country of origin was not taken into account because this information could not be clearly identified due to unknown international chains of distribution within the commercial sale and resale of such products.

Results for PCDD/Fs, DL-PCBs and NDL-PCBs including percentiles and rates of results above EU-maximum levels [4] and EU-action levels [5] are presented as an overview for the different groups of samples as described before in the attached table.

For calculation of the rates of samples being free of doubt below or above the corresponding maximum values, a general uncertainty of measurement of 20% was taken into account for PCDD/Fs and NDL-PCBs, 25% were considered for DL-PCBs. A result was regarded as being free of doubt below the maximum value in case the concentration of the sample plus the uncertainty of measurement was below or equal to the corresponding maximum value. In contrast, a result was regarded as being free of doubt above the maximum value in case the concentration of the sample minus the uncertainty of measurement was above the corresponding maximum value.

Looking on hen eggs, concentrations of PCDD/Fs, DL-PCBs as well as NDL-PCBs seems to be highest for organic eggs, followed by free-range eggs, then barn eggs and eggs from caged hens, when taking into account the 25th percentile (P25) to the 75th percentile (P75) excluding potential contamination situations via percentiles P95 and P99.

Egg products excluding albumin products seem to be higher contaminated than free range or barn eggs within this set of data. Because samples of hen eggs include also samples from customers applying a strict QA/QC-system when rearing the hens by analyzing eggs for PCDD/Fs and PCBs very frequently and initiating measures at early stages in the laying period of hen eggs, this situation could imply a shift towards lower concentrations of hen eggs included in this set of data evaluated here. This conclusion should be therefore confirmed by other / additional studies.

The EU developed a strategy [6] to support protecting the environmental and human health by reducing the concentration of dioxins and PCBs in both - the environment and food and feeding stuff. This strategy includes setting of limit values on a low, but practicable level, which will be gradually reduced, so that unacceptable high contaminated products will then no longer be used. Rates below the corresponding EU-maximum values on lipid basis determined in this study are clear above 95% for most groups of hen eggs and hen egg products. Despite the limitations of the set of data evaluated in this study, this seems to a proof of the effectiveness of the EU-strategy.

### **Acknowledgements**

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### **References**

1. Neugebauer F, Pöpke O, Opel M, Arkudas R. (2011); *Organohalogen Compounds* 73: 1223 - 1226
2. Focke F., Meyer A. (2014); Comparison of HRMS and MS/MS in Quantitative Dioxin and PCB Analysis. Oral Presentation 844, Dioxin 2014, 31. Aug to 05. Sept 2014, Madrid, Spain
3. Commission Regulation (EU) No 589/2014 of 2 June 2014 laying down methods of sampling and analysis for the control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs and repealing Regulation (EU) No 252/2012 (OJ L 164, 3.6.2014, p. 18)
4. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs (OJ L 364, 20.12.2006, p. 5), as amended by Commission Regulation (EU) 2015/704 of 30 April 2015 (OJ L 113, 1.5.2015, p. 27)
5. Commission Recommendation of 3 December 2013 on the reduction of the presence of dioxins, furans and PCBs in feed and food (2013/711/EU) (OJ L 323, 4.12.2013, p. 37), as amended by Commission Recommendation of 11 September 2014 (2014/663/EU) (OJ L 272, 13.9.2014, p. 17)

6. Communication from the Commission to the Council, the Social Committee of 24 October 2001. Community strategy for dioxins, furans and polychlorinated biphenyls COM (2001) 593 final (OJ C 322, 17.11.2001, p. 2)

Table 1: Results for sums of PCDD/Fs, DL-PCBs and NDL-PCBs including percentiles and rates of results above EU-maximum levels [4] and EU-action levels [5]

	Type of egg or egg product	n	Unit	P5	P25	MED	AM	P75	P95	P99	MV	AL	Rate below MV - free of doubt -	Rate below MV - not free of doubt -	Rate above MV - not free of doubt -	Rate above MV - free of doubt -	Rate below AL - no additional action necessary -	Rate above AL - additional action necessary -	
WHO(2005)-PCDD/F-TEQ (upperbound)	Hen eggs, all samples	6770	pg/g fat	0.181	0.234	0.302	0.456	0.541	1.10	2.38	2.5	1.75	98.5%	0.59%	0.27%	0.64%	97.8%	2.2%	
	- Method of rearing not known	945	pg/g fat	0.179	0.223	0.285	0.561	0.545	1.95	3.90	2.5	1.75	95.4%	1.8%	0.85%	1.9%	93.9%	6.1%	
	- Organic eggs	1510	pg/g fat	0.214	0.295	0.486	0.559	0.646	1.28	1.97	2.5	1.75	99.0%	0.40%	0.20%	0.40%	98.2%	1.8%	
	- Free range eggs	2315	pg/g fat	0.199	0.252	0.319	0.450	0.542	0.861	2.28	2.5	1.75	98.7%	0.43%	0.22%	0.60%	98.4%	1.6%	
	- Barn eggs	1857	pg/g fat	0.171	0.202	0.245	0.336	0.339	0.607	1.82	2.5	1.75	99.3%	0.38%	0.05%	0.27%	98.8%	1.2%	
	- Eggs from caged hens	143	pg/g fat	0.169	0.199	0.238	0.329	0.303	0.589	1.68	2.5	1.75	99.3%	0%	0%	0.70%	99.3%	0.70%	
	Egg products (> 2 % fat)	1496	pg/g fat	0.172	0.229	0.316	0.445	0.413	0.991	2.78	2.5	1.75	98.0%	0.80%	0.53%	0.67%	97.8%	2.2%	
	- Whole egg products (> 2 % fat)	687	pg/g fat	0.176	0.220	0.276	0.402	0.417	0.736	2.33	2.5	1.75	98.4%	0.87%	0.44%	0.29%	98.3%	1.7%	
	- Egg yolk products (> 2 % fat)	689	pg/g fat	0.163	0.261	0.340	0.487	0.411	1.05	3.09	2.5	1.75	97.8%	0.58%	0.44%	1.2%	97.7%	2.3%	
	- Egg white products (< 2 % fat)	86	pg/g	0.009	0.010	0.012	0.030	0.0177	0.028	0.644	0.05	N/A	97.7%	0%	0%	2.3%	---	---	
	Egg preparation of hen eggs	160	pg/g fat	0.167	0.203	0.246	0.262	0.285	0.415	0.581	2.5	1.75	100%	0%	0%	0%	100%	0%	
	Eggs and egg products of other poultry	24	pg/g fat	0.147	0.167	0.226	0.464	0.473	0.638	3.47	N/A	N/A	---	---	---	---	---	---	
	Hen eggs, all samples	6402	pg/g fat	0.126	0.148	0.213	0.391	0.339	0.721	2.42	N/A	1.75	---	---	---	---	---	98.4%	1.6%
	- No further information available	863	pg/g fat	0.123	0.143	0.174	0.705	0.320	1.92	10.5	N/A	1.75	---	---	---	---	---	94.7%	5.3%
- Organic eggs	1422	pg/g fat	0.161	0.266	0.353	0.521	0.494	1.14	4.27	N/A	1.75	---	---	---	---	---	97.5%	2.5%	
- Free range eggs	2230	pg/g fat	0.126	0.148	0.203	0.292	0.323	0.533	1.37	N/A	1.75	---	---	---	---	---	99.4%	0.58%	
- Barn eggs	1766	pg/g fat	0.122	0.141	0.158	0.270	0.262	0.362	0.509	N/A	1.75	---	---	---	---	---	99.7%	0.34%	
- Eggs from caged hens	121	pg/g fat	0.122	0.140	0.157	0.198	0.224	0.354	0.555	N/A	1.75	---	---	---	---	---	100%	0%	
Egg products (> 2 % fat)	1376	pg/g fat	0.119	0.151	0.206	1.03	0.382	3.91	9.79	N/A	1.75	---	---	---	---	---	90.0%	10.0%	
- Whole egg products (> 2 % fat)	606	pg/g fat	0.123	0.142	0.179	0.537	0.284	0.902	7.45	N/A	1.75	---	---	---	---	---	96.5%	3.5%	
- Egg yolk products (> 2 % fat)	654	pg/g fat	0.109	0.179	0.262	1.62	1.15	5.77	12.0	N/A	1.75	---	---	---	---	---	82.4%	17.6%	
- Egg white products (< 2 % fat)	87	pg/g	0.006	0.006	0.0065	0.010	0.011	0.019	0.033	N/A	N/A	---	---	---	---	---	---	---	
Egg preparation of hen eggs	146	pg/g fat	0.124	0.134	0.147	0.198	0.217	0.455	0.602	N/A	1.75	---	---	---	---	---	100%	0%	
Eggs and egg products of other poultry	19	pg/g fat	0.103	0.113	0.124	0.167	0.190	0.341	0.347	N/A	N/A	---	---	---	---	---	---	---	
Hen eggs, all samples	6344	pg/g fat	0.323	0.390	0.526	0.831	0.888	1.76	4.57	5.0	N/A	98.7%	0.43%	0.16%	0.71%	---	---		
- No further information available	820	pg/g fat	0.318	0.369	0.461	1.22	0.874	3.17	12.2	5.0	N/A	96.0%	0.61%	0.49%	2.9%	---	---		
- Organic eggs	1421	pg/g fat	0.411	0.606	0.881	1.09	1.19	2.28	4.88	5.0	N/A	98.2%	0.84%	0.21%	0.70%	---	---		
- Free range eggs	2226	pg/g fat	0.340	0.407	0.532	0.741	0.873	1.41	3.18	5.0	N/A	99.1%	0.45%	0.13%	0.31%	---	---		
- Barn eggs	1757	pg/g fat	0.311	0.349	0.406	0.579	0.586	0.949	1.30	5.0	N/A	99.8%	0%	0%	0.23%	---	---		
- Eggs from caged hens	120	pg/g fat	0.305	0.341	0.384	0.461	0.508	0.872	0.943	5.0	N/A	100%	0%	0%	0%	---	---		
Egg products (> 2 % fat)	1358	pg/g fat	0.307	0.397	0.547	1.49	0.984	4.51	10.4	5.0	N/A	93.7%	2.1%	1.10%	3.1%	---	---		
- Whole egg products (> 2 % fat)	595	pg/g fat	0.316	0.376	0.481	0.958	0.722	2.34	8.77	5.0	N/A	97.3%	1.0%	0.17%	1.5%	---	---		
- Egg yolk products (> 2 % fat)	650	pg/g fat	0.293	0.463	0.672	2.12	1.76	6.37	12.6	5.0	N/A	89.4%	3.4%	2.2%	5.1%	---	---		
- Egg white products (< 2 % fat)	89	pg/g	0.013	0.016	0.018	0.030	0.029	0.041	0.134	0.1	N/A	98.9%	0%	0%	1.1%	---	---		
Egg preparation of hen eggs	146	pg/g fat	0.301	0.343	0.402	0.462	0.516	0.789	1.05	5.0	N/A	100%	0%	0%	0%	---	---		
Eggs and egg products of other poultry	19	pg/g fat	0.261	0.291	0.335	0.459	0.637	0.800	0.847	N/A	N/A	---	---	---	---	---	---		
Hen eggs, all samples	4356	ng/g fat	1.18	1.41	2.17	3.33	3.24	6.41	24.5	40	N/A	99.3%	0.14%	0.16%	0.39%	---	---		
- No further information available	577	ng/g fat	1.18	1.36	1.85	6.28	3.31	13.3	129	40	N/A	97.1%	0.69%	0.17%	2.1%	---	---		
- Organic eggs	1014	ng/g fat	1.43	2.37	3.14	3.96	3.97	7.96	25.4	40	N/A	99.5%	0%	0.39%	0.10%	---	---		
- Free range eggs	1712	ng/g fat	1.19	1.37	1.77	2.56	3.11	4.53	11.3	40	N/A	99.7%	0.12%	0.06%	0.12%	---	---		
- Barn eggs	978	ng/g fat	1.14	1.32	1.60	2.38	3.08	3.53	6.44	40	N/A	99.7%	0%	0.10%	0.20%	---	---		
- Eggs from caged hens	75	ng/g fat	0.997	1.27	1.45	1.85	2.65	3.40	3.47	40	N/A	100%	0%	0%	0%	---	---		
Egg products (> 2 % fat)	1007	ng/g fat	0.834	1.44	2.21	11.3	6.27	60.4	148	40	N/A	90.6%	2.0%	1.2%	6.3%	---	---		
- Whole egg products (> 2 % fat)	391	ng/g fat	0.870	1.30	1.94	5.86	2.96	21.0	101	40	N/A	96.7%	0.26%	0.26%	2.8%	---	---		
- Egg yolk products (> 2 % fat)	560	ng/g fat	0.820	1.69	2.54	16.0	15.6	77.4	164	40	N/A	85.4%	3.4%	2.0%	9.3%	---	---		
- Egg white products (< 2 % fat)	70	ng/g	0.060	0.070	0.101	0.118	0.147	0.253	0.355	0.8	N/A	100%	0%	0%	0%	---	---		
Egg preparation of hen eggs	25	ng/g fat	0.928	1.23	1.38	2.01	3.05	4.12	4.41	40	N/A	100%	0%	0%	0%	---	---		
Eggs and egg products of other poultry	8	ng/g fat	0.571	1.01	1.04	0.971	1.09	1.14	1.16	N/A	N/A	---	---	---	---	---	---		

n = number of data sets, P5 = 5th percentile, P25 = 25th percentile, MED = Median, AM = Arithmetic mean, P75 = 75th percentile, P95 = 95th percentile, N/A = not applicable, MV = Maximum value according to [4], AL = Action level according to [5]