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Levels of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans in Food of Animal Origin. The Swiss Dioxin Monitoring Program

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Between 1997 and 1999, several cases of dioxin contaminations in foodstuffs of animal origin occurred in Europe due to feed contaminated by several independent sources: citrus pulp pellets, fat containing polychlorinated biphenyls (PCB), and kaolinitic clay as anti-caking agent in feedingstuffs. As a consequence of the latter, a survey on polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F) in food of animal origin was initiated by the Swiss authorities to assess the extent of PCDD/F contamination and to document the efficiency of the measures taken to ensure the decontamination of the food supply. Investigation of a total of 128 samples of cow's milk, poultry, eggs, and meat revealed several cases of residue levels distinctly above the background exposure limit of approximately 2.5 ng of I-TEQ/kg (fat basis). Particularly, elevated concentrations were found in eggs (maximum 13 ng of I-TEQ/kg), poultry (maximum 3.9 ng of I-TEQ/kg), and pork (maximum 7.5 ng of I-TEQ/kg). On the basis of the observed PCDD/F congener pattern, the contamination could be attributed to PCDD/F-contaminated kaolin that was used as an anti-caking agent in particular feedstuffs.

KEYWORDS: PCDD; PCDF; food; egg; meat; poultry; chicken; cow's milk; dioxins; dioxin monitoring program Switzerland

INTRODUCTION

The levels of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F) in food of animal origin are mainly due to bioaccumulation and biomagnification along the food chain. As a rule, feedstuffs of animal origin (e.g., animal fat, fish meal, and fish oil) therefore contain higher PCDD/F levels than vegetable feed materials (*1*). The diet of terrestrial species of farm animals such as pigs, ruminants, or chicken is mainly based on feed of plant origin such as grass or seeds. The PCDD/F contamination of these material results from direct deposition of airborne pollutants after atmospheric transport (*2*−*4*). The major part of PCDD/F originates from combustion and other thermal processes, such as waste incineration or metal production and recycling. Various studies show that environmental levels have decreased during the last 20 years due to specific measures to reduce the emission of PCDD/F from combustion processes (*5*): In a study on sediment cores from Lake Constance, situated on the northeastern border of Switzerland, Hagenmaier et al. report a strong decrease of the PCDD/F levels after 1974 (*6*). For Switzerland, decreased total emissions of PCDD/F after 1980 have been documented (*7*). At the same time, PCDD/F levels in food have been decreasing continuously: Between 1984 and 2001, the average PCDD/F content in Swiss cow's milk dropped from 2.3 to 0.51 ng of I-TEQ/kg (milk fat basis) (*8*). The same trend has been observed for human exposure: Average PCDD/F levels in German mother's milk were 31 (1991) and 13 ng of I-TEQ/kg (1998), respectively (*9*).

In contrast to this trend, several cases of specific contamination have caused high PCDD/F levels in foodstuffs. In 1997, exceptional PCDD/F levels in dairy products from Baden-Württemberg (Germany) were discovered, which could be attributed to the use of a pellet feed consisting of Brazilian citrus pulp (*10*). The levels of up to 10 ng of I-TEQ/kg in the citrus pulp product far exceeded the maximum content of 0.75 ng of WHO-TEQ/kg established by the Council of the EU (*11*). Citrus pulp is obtained as a byproduct from the production of juice from citrus fruits. To make it applicable as animal feed, the fruit acids are partly neutralized by adding 2% lime (calcium hydroxide). The use of lime contaminated with PCDD/F originating from industrial production processes involving chlorine led to the contaminated citrus pulp (*12*). A survey of feedstuffs and feed components in Switzerland revealed levels

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Table 1. PCDD/F Levels in Food of Animal Origin and in the Feedstuff Used in the Breed as Determined in a Preliminary Survey (July 1999)[a]

foodstuff			corresponding feedstuff	
type	no. of samples	ng of I-TEQ/kg (fat basis)	type	ng of I-TEQ/kg (dry weight)
cow's milk	1	0.88 (2.1)	supplementary feed	0.044
eggs	1	3.7 (2.7)	supplementary feed	0.022
			supplementary feed	1.4
eggs	1	6.0 (2.7)	supplementary feed	0.94
			supplementary feed	2.2
eggs	1	15 (2.7)	sole feed	0.93
eggs	1	21 (2.7)	sole feed	1.8
eggs	1	15 (2.7)	sole feed	2.1
fattened chicken	1	2.9 (1.0)	fattening feed	0.084
pork	1	3.8 (0.4)	fattening feed	2.6
lard (pork)	3	2.2–2.5 (0.4)	fattening feed	0.69

[a] Values in parentheses represent the average background PCDD/F contamination of the respective foodstuff in countries of the EU (*28*). In the cases where several types of feed with different PCDD/F contents were used simultaneously, the respective quantitative compositions are not known.

of up to 20 ng of I-TEQ/kg in citrus pulp pellets (data not published). Thereafter, imports, delivery, and feeding of the contaminated material were stopped throughout Europe, and more than 100 000 metric tons of feedstuff had to be disposed off.

Only two years later, in spring 1999, high PCDD/F levels detected in Belgian eggs and poultry could be traced back to feed containing waste vegetable oil contaminated with residues of polychlorinated biphenyls (PCB) and PCDD/F (*13*). Spot checks of suspicious foodstuffs in Switzerland revealed no indication of food contamination caused by this incident.

As far back as 1994, farm-raised catfish in Mississippi, USA (*14*), were found to contain high levels of PCDD/F with a previously unknown congener pattern. This same congener pattern was later found in chickens (*15*). The source of the contamination was found to be soybean meal (*16*). The PCDD/F in the soybean meal could be traced back to ball clay from a particular mine in Mississippi (*17*). In 1999, similar PCDD/F containing materials were discovered in sediments from Queensland, Australia (*18*), and in kaolin from Germany (*19*). Contaminated kaolin from deposits in the German State ("Bundesland") Rhineland-Palatinate was used as an anti-caking aid in feedstuff causing PCDD/F levels up to 14 ng of I-TEQ/kg in the feed (*20*). The PCDD/F composition of kaolin was similar to the PCDD/F observed in the materials from Mississippi (*21*) and from Queensland (*22*). The congener profiles were dominated by hepta- and octachlorodibenzo-*p*-dioxins (HeptaCDD and OctaCDD) with typical levels in kaolin of 1500 and 25 000 ng/kg, respectively, whereas polychlorinated dibenzofurans (PCDF) were almost completely absent. As in the case of ball clay and marine sediment from Queensland, the formation pathways of polychlorinated dibenzo-*p*-dioxins (PCDD) in kaolin are still unknown. However, no indication for anthropogenic influences could be found (*19*). As a consequence of the high PCDD levels, a tolerance limit of 500 pg of WHO-TEQ/kg was defined for PCDD/F in kaolinitic clays to be used for feedstuffs (*23*).

In a response to the European kaolin contamination incident, a preliminary survey was initiated in July 1999 by the Swiss authorities to measure PCDD/F levels in feedstuffs and foodstuffs of animal origin (cow's milk, eggs, poultry, and pork) produced in farms where feed with polluted kaolin had been used. The results of this survey provided the starting point of a systematic investigation of food of animal origin that was ordered in July 1999 by the Swiss Federal Department of Home Affairs. In the present work, the results of this monitoring program are compiled and discussed.

MATERIALS AND METHODS

Samples. The samples investigated in the preliminary survey were collected from farms to which feedstuffs containing contaminated kaolin had been delivered (sampling in July 1999). The food samples for the monitoring program (cow's milk, eggs, chicken meat (broilers), pork, veal, and beef) were collected from September 1999 to March 2000 by the official food control and veterinary authorities of several Swiss cantons. The samples were kept frozen (−20 °C) until analysis.

Analytical Methods. Fat extraction of milk and eggs was based on an extraction method for human milk described by Fürst et al. (*24*). The fatty parts of meat were mixed with anhydrous sodium sulfate and extracted in a Soxhlet apparatus using cyclohexane (recovery of lipids >95%). After gravimetric determination of the fat content, the internal standard (mixture of the 17 $^{13}C_{12}$-labeled 2,3,7,8-chlorosubstituted PCDD/F, EDF-4067, Cambridge Isotope Laboratories, Andover, MA) was added. Further treatment of the fat extract followed a method described by Smith et al. (*25*). $^{13}C_{12}$-1,2,7,8-TetraCDF (EF-1438, Cambridge Isotope Laboratories) was added as a recovery standard.

Gas chromatography/high-resolution mass spectrometry (GC/HRMS) analysis was carried out on a MAT 95 (Thermo Finnigan MAT, Bremen, Germany) mass spectrometer coupled to a HRGC Mega 2 series (Fisons Instruments, Rodano, Italy) gas chromatograph equipped with an A200S (CTC Analytics, Zwingen, Switzerland) autosampler. Samples were injected in splitless mode (splitless time 20 s) at an injector temperature of 260 °C and at an initial oven temperature of 110 °C. After 1 min, the temperature was ramped at 20 °C/min to 240 °C and at 4 °C/min up to 300 °C. The latter temperature was held for 5 min. A glass capillary column of 20 m × 0.30 mm, coated with a DB-5 analogue stationary phase (film thickness 0.15 μm) was used. The ion source was operated at 180 °C and an electron energy of 70 eV, and the mass spectrometer was tuned to a mass resolution of 10 000. The two most abundant signals of the molecular ion cluster of the tetra- to octachlorodibenzo-*p*-dioxins and -dibenzofurans were recorded in single ion monitoring (SIM) mode. Quantification was performed using signal heights in the respective mass chromatograms. Calculation of concentrations was based on internal standard levels and comparison with a reference mixture of the 17 2,3,7,8-chlorosubstituted PCDD/F (EDF-7999, Cambridge Isotope Laboratories). For calculation of the detection limits, a signal-to-noise ratio of 3 was applied.

Data Processing. Calculation of I-TEQ was based on I-TEF (*26*); for congeners below the detection limits the respective detection limits were used. The detection limits of these congeners contributed less than 5% to the total I-TEQ of any sample.

RESULTS AND DISCUSSION

Table 1 shows PCDD/F levels in selected foodstuffs of animal origin determined within the scope of a preliminary survey after the detection of the PCDD/F contamination in kaolin. The corresponding PCDD/F levels in the feed are also given. The data show that most of the concentrations in the

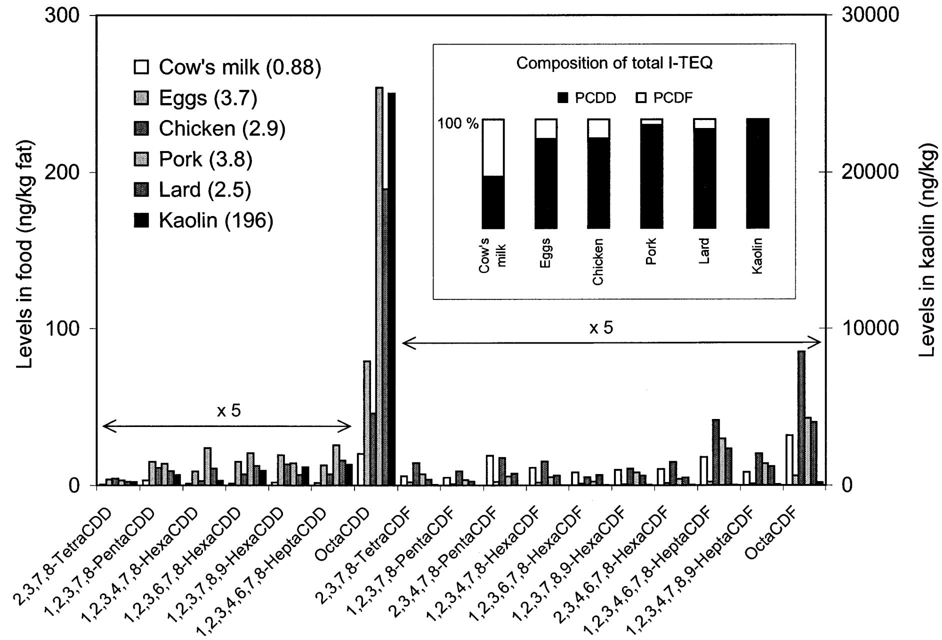

Figure 1. Typical PCDD/F congener profiles of samples of cow's milk, eggs, chicken, pork, lard, and kaolin (sequence within each congener from left to right as indicated in the legend). Corresponding total I-TEQ levels are given in parentheses (ng of I-TEQ/kg fat for food samples, ng of I-TEQ/kg dry weight for kaolin). The small chart shows the percentages of PCDD and PCDF of the total I-TEQ, respectively.

feedstuffs were distinctly higher than the levels usually occurring in feed of vegetable origin, being in the range of 0.2 ng of I-TEQ/kg (dry weight) for roughage (*1*). Those foodstuffs that were found to have higher than usual background concentrations were consistently found at sites with feedstuffs having higher PCDD/F concentrations. These results support the conclusion that PCDD/F were transferred from the contaminated feed, through the animal, to the foodstuff. Moreover, the congener patterns observed in the contaminated foodstuffs and in the feed match with the PCDD/F pattern of contaminated kaolin, substantiating again the transfer of PCDD/F from the feed into the animal and its products, respectively. **Figure 1** shows the PCDD/F congener profiles of some of the food samples from the preliminary survey together with the pattern of contaminated kaolin. The contaminant in kaolin consists exclusively of PCDD with particularly high levels of OCDD and is further characterized by complete absence of PCDF. Whereas the egg, chicken, pork, and lard samples with 3.7, 2.9, 3.8, and 2.5 ng of I-TEQ/kg (fat basis), respectively, reflect the profile of kaolin, the levels and profile of the cow's milk sample (0.88 ng of I-TEQ/kg) are consistent with the general input of airborne particle-associated PCDD/F from the ubiquitous background contamination in Switzerland (*4*) with percentages of the PCDD and PCDF of the total I-TEQ of ca. 50% each. In the case of PCDD/F contamination from kaolin, the share of the total I-TEQ resulting from PCDD rises distinctly (>80%), depending on the degree of contamination.

On the basis of the results of the preliminary survey (reported in **Table 1**), a systematic survey of food of animal origin was ordered by the Swiss Federal Department of Home Affairs in July 1999. The objectives of this survey were data collection on PCDD/F levels in cow's milk, eggs, poultry, pork, and ruminant's meat produced in Switzerland. On the basis of these data, the compliance of the affected farms concerning the ordered exchange of contaminated feedstuffs was verified and the effectiveness of the measures introduced was investigated.

Table 2 shows a summary of the food samples analyzed within the scope of the monitoring program and the ranges of

Table 2. Overview of the Outcome of the Swiss Dioxin Monitoring Program: Food Items Analyzed, Number of Samples, and Levels (ng of I-TEQ/kg, Fat Basis)

product category	no. of samples	av	std dev	median	min	max.
cow's milk	44	0.63	0.17	0.60	0.35	1.1
eggs	18	2.9	3.0	1.9	0.41	13
chicken (broilers)	24	1.1	0.90	0.84	0.32	3.9
pork	31	0.96	1.8	0.37	0.052	7.5
veal	5	0.83	0.14	0.79	0.66	1.0
beef	5	0.61	0.10	0.62	0.49	0.73
cow	1	0.85				

Table 3. Temporary Tolerance Limits (TTL) and Maximum Residue Limits (MRL) Applied in the Monitoring Program in ng of I-TEQ/kg (Fat Basis) with Relative Number of Samples Exceeding the TTL

category	TTL	MRL	range (min – max.)	exceeding of TTL
cow's milk	3	20	0.35–1.1	0/44
eggs	5 (barn-reared hens)	60	0.41–13	2/18
	10 (free range hens)			1/18
poultry	5	80	0.32–3.9	0/24
pork	2	40	0.052–7.5	3/31
veal	6	40	0.66–1.0	0/5
beef	6	40	0.49–0.73	0/5
cow's meat	6	40	0.85	0/1

[a] For eggs, separate TTL were defined for extensive indoor keeping and keeping of the hens (see text).

the PCDD/F levels measured. For classification of the detected levels, temporary tolerance limits (TTL) and maximum residue limits (MRL) were defined by the Swiss Federal Office of Public Health for each food category (see **Table 3**): Exceeding the TTL indicates contamination and/or impaired quality of a foodstuff; exceeding the MRL qualifies a foodstuff unsuitable for human feeding (*27*). The table indicates the cases where TTL were exceeded. In contrast, no MRL was exceeded by any of the samples. A graphical representation of the data is given in **Figure 2**. The levels are classified into foodstuff categories

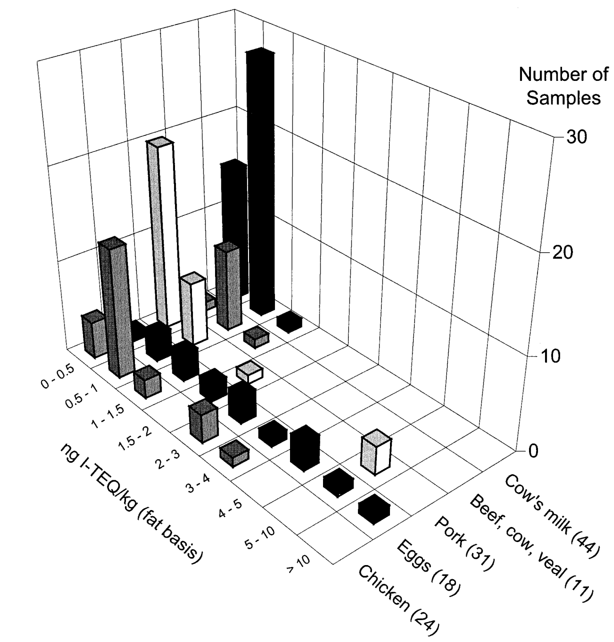

Figure 2. Distribution of the PCDD/F levels detected in food by categories (in parentheses: number of samples).

and concentration ranges demonstrating the frequency distribution of the levels within food categories. Although the collection of the samples was carried on for 6 months (September 1999 to March 2000), no distinct temporal trend regarding a decrease of the PCDD/F levels within the sample collection period could be observed. On one hand, there was no scheduled collection of the samples enabling a continuous monitoring of the levels, on the other hand, the slow elimination pharmacokinetics of PCDD/F in part of the animals may have delayed the effects of withdrawal of contaminated feedstuffs in July 1999. However, the various food categories exhibit characteristics that are closely related to the specific feeding and keeping conditions of the respective animals, as discussed below.

Cow's Milk. All PCDD/F levels of the 44 cow's milk samples are within a narrow range far below the TTL of 3 ng of I-TEQ/kg (fat basis). The average level (0.63 ± 0.17 ng of I-TEQ/kg) is not significantly different from the average concentration of approximately 0.5 ng of I-TEQ/kg usually observed in not specifically exposed cow's milk in Switzerland and Western Europe (*8*, *28*). Although all samples were collected in farms where feedstuffs containing kaolin were used, no elevated PCDD/F milk levels indicating the feeding of large amounts of contaminated material to cows could be found. This is consistent with data reported by Malisch (*20*). However, the relative amount of the contaminated feed in the diet of the cows is unknown. In Switzerland, the feed of cows mainly consists of locally grown roughage (grass, hay, and silage) (*29*). During the vegetation period, cows are fed by outdoor foraging. Therefore, the common levels occurring in cow's milk are mainly due to transfer of PCDD/F from the grass, which is contaminated by deposition of airborne PCDD/F originating from atmospheric transport. In absence of PCDD/F point sources in the vicinity of a farm, the particle-associated PCDD/F are mainly due to long-range transport. Therefore, the resulting PCDD/F milk levels reflect the general input of PCDD/F, and the concentrations are similar to the values usually detected in Swiss cow's milk. Comparison of the results with older data from 1984 (*30*) and 1990/1991 (*31*) indicates a decrease of the general background contamination that generally applies to cow's milk from Western Europe (*32*, *33*).

Poultry (Eggs and Meat). PCDD/F concentrations in eggs ranged from 0.41 to 13 ng of I-TEQ/kg (fat basis) with maximum levels exceeding the TTL. In fact, the five egg samples taken before exchange of contaminated feed exhibited levels between 3.7 and 21 ng of I-TEQ/kg (see **Table 1**). All samples with concentrations above approximately 2 ng of I-TEQ/kg showed the characteristic congener pattern of kaolin (>80% of the total I-TEQ due to PCDD, see **Figure 1**), clearly indicating kaolin as the source of contamination; similar data were reported by Malisch (*20*). Accordingly, the PCDD/F levels detected in chicken meat ranged from 0.32 to 3.9 ng of I-TEQ/kg, and the samples with concentrations above 2 ng of I-TEQ/kg could be attributed to contaminated kaolin due to their congener patterns in this case as well.

Besides the feed, the type of keeping can be an important factor influencing the PCDD/F exposure of poultry. Whereas the feed is the major contributor in the case of housing in cages, the PCDD/F content of the soil can play an important role in free range keeping with outdoor foraging. As absorption of soil contributes to about 10% of the feed, transfer from contaminated soil can significantly increase the PCDD/F levels in chicken and in eggs (*4*). As free range keeping is more frequently used in egg production than in chicken-rearing, a higher TTL was fixed for eggs from free range keeping of laying hens (see **Table 3**). Although the records of the egg samples did not allow an unambiguous classification to one of the two categories and the data had to be merged to one category, this situation is reflected in the distribution of the levels observed in eggs and in chicken (see **Figure 2**): Some of the egg samples exhibit relatively high concentrations (1.5−3 ng of I-TEQ/kg) not being accompanied by the typical kaolin pattern but suggesting the influence of soil as mentioned above. In contrast, the PCDD/F levels in chicken meat distinctly show a bimodal distribution with levels below approximately 1.5 ng of I-TEQ/kg and congener profiles due to transfer from feed with the usual background contamination and high levels with congener profiles, indicating kaolin-contaminated feed (similar to profiles shown in **Figure 1**), respectively.

Pork. Similar to the results obtained for the poultry products, the levels observed in pork can be classified into two categories: Low levels (<0.1−0.60 ng of I-TEQ/kg) indicating a low and nonspecific exposure, due to the use of feedstuff without considerable PCDD/F content, were found in 27 samples. High levels (1.8−7.5 ng of I-TEQ/kg) were detected in 4 samples taken on the same date (November 1999), and the corresponding animals were from the same farm. These high levels were accompanied by the same characteristic congener pattern of contaminated kaolin (similar to profiles of the pork and lard samples shown in **Figure 1**). It is well-known that PCDD/F have half-lives in the order of years in mammals (*34*). Thus, it is supposed that the high PCDD/F levels in these animals resulted from the use of contaminated feed before the ordered exchange of contaminated feed materials 4 months before (July 1999).

In the keeping of cows, the feed is mainly based on locally grown roughage containing fairly constant PCDD/F levels originating from the atmospheric deposition and leading to small variations of the PCDD/F milk concentrations only (see **Table 2**). In contrast, pigs are usually fed with diverse materials such as commercial feed, leftovers of foodstuffs, or fodder soups. It is therefore expected that PCDD/F levels in pork may vary, accordingly. These considerations are substantiated by the observed levels which vary from very low levels up to levels clearly indicating contamination. As no levels in the intervening range were observed, it can be concluded that the usual feed for pigs is relatively low in PCDD/F, resulting in a generally low exposure. Therefore, it is expected that the low level of PCDD/F exposure can be reconstituted by eliminating the contaminated feedstuffs.

Meat of Ruminants (Beef, Cow's Meat, Veal). Due to the small number of samples (11), no general conclusions can be drawn from these results. Nevertheless, no samples exhibited increased levels or conspicuous congener patterns indicating consumption of contaminated kaolin. The distribution of the levels and the congener profiles are similar to the data of cow's milk, which is dominated by the general PCDD/F exposure by atmospheric deposition, suggesting a similar feeding of these animals.

CONCLUSIONS

The survey revealed that poultry, eggs, and pork were the most affected products, whereas no samples of cow's milk and ruminant meat (cow, veal, and beef) exhibited increased PCDD/F levels due to PCDD/F contaminated kaolin. On the basis of the levels determined in all samples of cow's milk, eggs, poultry, pork, and meat of ruminants, a PCDD/F exposure above average of the majority of the consumers due to transfer from polluted feedstuffs can be excluded, as the average PCDD/F levels for all samples fell below the TTL for the respective foodstuff, presumably a reflection of the enforced feedstuff exchange program. Data from the foodstuffs without the kaolin PCDD/F contamination indicate that dietary PCDD/F exposure of the Swiss population is similar to that in other Western European countries (*35*). As the dietary exposure of a considerable proportion of the European population still exceeds the recommendations on tolerable intake (TWI 14 pg of WHO-TEQ/kg of bodyweight, provisional TMI 70 pg of WHO-TEQ/kg of bodyweight, range TDI 1−4 pg of WHO-TEQ/kg of bodyweight (*36*, *37*)), there is need for continuing efforts to identify and eliminate sources of contamination.

ABBREVIATIONS USED

EU, European Union; GC/HRMS, gas chromatography/high-resolution mass spectrometry; HeptaCDD, heptachlorodibenzo-*p*-dioxin; I-TEF, international toxic equivalency factor; I-TEQ, international total toxic equivalency; MRL, maximum residue limit; OctaCDD, octachlorodibenzo-*p*-dioxin; PCB, polychlorinated biphenyls; PCDD, polychlorinated dibenzo-*p*-dioxins; PCDD/F, polychlorinated dibenzo-*p*-dioxins and dibenzofurans; PCDF, polychlorinated dibenzofurans; SIM, single ion monitoring; TDI, tolerable daily intake; TMI, tolerable monthly intake; TTL, temporary tolerance limit; TWI, tolerable weekly intake.

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Supporting Information Available: Extensive version of **Table 2** (single congener data, WHO-TEQ). This material is available free of charge via the Internet at http://pubs.acs.org.

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