

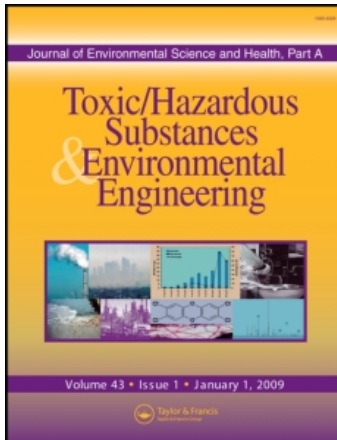
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Uptake of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in laying ducks

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Uptake of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in laying ducks was determined at different degree of feed contamination. To observe the extent of the transfer of 17 PCDD/Fs from feed to the duck eggs and duck meat, 18 ducks were divided into 3 groups (6 in each group) and fed feed with two different levels of PCDD/Fs. As a control, one group of ducks was fed with the non-contaminated feed for comparison, while the other 2 groups were exposed to the feed doped with EAF dusts (fly ash). The experiment lasted for 60 days, with an exposure duration of 41 days and the subsequent non-contaminated feed being given for an additional 19 days. PCDD/F levels in the eggs of the all 3 groups were observed to increase significantly on the 15th day. For the low contaminated group, PCDD/F levels reached 2.61 pg WHO-TEQ/g lipid at day 41, whereas those of the high contaminated group accounted exceeded 3 pg/g lipid on the 15th day. Furthermore, PCDD/Fs levels in the duck meat were analyzed before and after exposure duration, and at the end of the experiment. The results showed that the level of PCDD/F in the duck eggs and the duck meat may reach unacceptable levels due to the effect of accumulation, although the PCDD/Fs in the duck feed were at acceptable levels.

Keywords: Dioxins, PCDD/Fs, ducks, eggs, feed.

Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) are well known anthropogenic toxicants which could cause serious diseases such as cancer, immune deficiency, reproductive and developmental abnormality, and central and peripheral nervous system pathology.^[1] In 1998, the tolerable daily intake of 1 to 4 pg toxic equivalent (TEQ)/kg body weight was established by the World Health Organization. Among various routes of human intake PCDD/Fs, i.e., diet, dermal absorption and soil ingestion,^[2] more than 90% of PCDD/F uptake is contributed by dietary intake.^[3] Since an incident involving PCDD/F contaminated feeds in Belgium in 1998, many studies have been performed to evaluate PCDD/F concentrations in food,^[4] dietary intake,^[2,3,5–8] and the health

risk of exposure to PCDD/Fs through food.^[5] The main sources of PCDD/F intake from food have been found to be fish and fishery products, meat and meat products, and milk and dairy products.^[3,6]

PCDD/Fs are accumulated and distributed among animal tissues. Since the metabolism of PCDD/Fs by different animals is varied, the PCDD/F distribution in tissues also varies among animals and is influenced by the lipid content and metabolic ability.^[9] Thus, livers and eggs are expected to accumulate high levels of PCDD/Fs for fish and birds, respectively.^[10] While in beef cattle the PCDD/Fs were predominantly found in association with circulating blood lipid rather than in equilibrium with fat deposits.^[11]

Contaminated feed is the most common cause of PCDD/F dietary intake. Thus, many studies have been performed to observe the biotransfer, bioaccumulation and bioavailability of PCDD/Fs in animals through contaminated feed. These include cows fed grass silage from a field that had a history of sewage sludge applications,^[12,13] chickens exposed to low and high contaminated feed blended with two types of contaminated soil, with an analysis of 17 PCDD/F congeners in liver, adipose, meat and

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eggs;^[14] chickens exposed to fly ash and steam cleaning oil, with PCDD/F analysis of the liver, eggs, muscles, and excreta;^[15,16] and pigs and broilers from the contaminated feed incident in Belgium.^[17] In short, the bioavailability of PCDD/Fs obtained in the results of the above-mentioned studies all decreased according to the number of chlorine atoms in the congeners. Moreover, the residues of PCDD/Fs could be predicted according to the PCDD/F levels in the feed, and various cooking processes had no influence on the PCDD/F residues in the meat.^[18]

In June 2004, a high level of PCDD/Fs in duck eggs (30.0–45.0 pg WHO/g-fat) was discovered in Changhua County, located in the middle of Taiwan. Those PCDD/F I-TEQ concentrations were approximately 10–15 times higher than the European Union limits of 3 pg WHO/g-fat. Several previous reports suspected that a specific electric arc furnace dust treatment plant (EAFDT plant) located near the duck farm was responsible for the event. However, the influence of the EAFDT plant has been proven very minor, and EAF dust (fly ash) added into feed was probably the major contributor of PCDD/Fs in this case.^[19]

In Asia, both duck tissue and eggs are major food sources. This study is the first to undertake an investigation of the transfer of PCDD/Fs from feed to duck eggs. The bioconcentration factors and the fraction of PCDD/Fs ex-

creted from the duck eggs were determined and compared for both the non-contaminated and contaminated feed, in which the extent of contamination was adjusted by blending different amounts of EAF dust. In addition, the decrease of PCDD/Fs in eggs and tissues after the exposure stopped was also observed to supply data on PCDD/Fs excreted from ducks. The results can provide useful information as to whether illegal open-dumping sites for electric arc furnace (EAF) dust lead to an unacceptable PCDD/F level in duck eggs.

Materials and methods

Duck feed

The non-contaminated feed (NCF) was purchased from Taiwan Sugar Corporation. The contaminated feed was then made by mixing the NCF with a certain amount of EAF dust, resulting in low-contaminated feed (LCF, 0.3 wt% EAF dust) and high-contaminated feed (HCF, 0.6 wt% EAF dust). The PCDD/F concentrations of the NCF and the EAF dust are given in Table 1. Note that the PCDD/Fs in the contaminated feed were calculated based on the mass balance of those present in the non-contaminated feed and EAF dust.

Table 1. PCDD/F concentrations (pg/g) in non-contaminated feed, EAF dust and contaminated feed (mixed with non-contaminated feed and EAF dust).

PCDD/Fs (pg/g)	Feed		EAF dust		Contaminated feed	
	Mean (n = 4)	RSD (%)	Mean (n = 4)	RSD (%)	0.3 wt% fly ash	0.6 wt% fly ash
2,3,7,8-TeCDD	0.0045	28	5.55	18	0.0211	0.0378
1,2,3,7,8-PeCDD	0.0095	26	47.9	8.8	0.153	0.297
1,2,3,4,7,8-HxCDD	0.0047	23	42.2	12	0.131	0.258
1,2,3,6,7,8-HxCDD	0.0104	17	78.5	12	0.246	0.481
1,2,3,7,8,9-HxCDD	0.0089	44	96.1	7.9	0.297	0.585
1,2,3,4,6,7,8-HpCDD	0.0944	34	797	27	2.48	4.88
OCDD	1.03	43	1520	43	5.59	10.2
Total PCDDs	1.16	—	2589	—	8.92	16.7
2,3,7,8-TeCDF	0.0260	19	34.4	24	0.129	0.232
1,2,3,7,8-PeCDF	0.0563	10	93.3	4.3	0.336	0.616
2,3,4,7,8-PeCDF	0.0693	22	91.6	12	0.344	0.619
1,2,3,4,7,8-HxCDF	0.154	29	167	6.4	0.654	1.15
1,2,3,6,7,8-HxCDF	0.0456	19	190	7.4	0.615	1.18
1,2,3,7,8,9-HxCDF	0.0410	59	151	9.4	0.494	0.947
2,3,4,6,7,8-HxCDF	0.0794	29	48.4	4.9	0.224	0.370
1,2,3,4,6,7,8-HpCDF	0.125	34	722	7.0	2.29	4.45
1,2,3,4,7,8,9-HpCDF	0.0407	32	125	8.3	0.416	0.792
OCDF	0.127	48	687	14	2.19	4.25
Total PCDFs	0.76	—	2310	—	7.69	14.6
Total PCDD/Fs	1.92	—	4900	—	16.6	31.3
Total WHO-TEQ	0.0912	—	201	—	0.695	1.30

RSD: (standard deviation/mean) × 100%.

Study design

Eighteen 6- to 7-month-old ducks were purchased from a farm located in a rural region of Taiwan. These ducks were randomly assigned to three groups (six each) and housed in a yard, with the cement ground washed daily. Each group of ducks was separated by a net, and the feed and water were refreshed and the related containers cleaned daily. Before exposure to the PCDD/F contaminated feed, all the ducks were fed for 10 days with NCF under the same environmental conditions. Baseline duck eggs and duck meats were analyzed before PCDD/F exposure. Exposure of the low-contaminated group (LCG) and the high-contaminated group (HCG) lasted for 41 days. Thereafter, these ducks were then fed with NCF from days 42 to 60, while the non-contaminated group (NCG) was provided with NCF for the entire duration of the experiment.

The eggs of all groups were analyzed 7 times during the exposure period (days 1, 8, 15, 21, 27, 33, and 41), and 3 times after exposure (days 47, 54, and 60). Moreover, the duck meat was collected and analyzed at days 41 and 60.

Sample pretreatment and PCDD/F analyses

Egg samples were unshelled and the egg yolks were homogenized. Duck feathers were removed with hot water. All edible parts of duck meat with skin were taken and homogenized, while the metacarpus was excluded.

PCDD/Fs were quantified by following EPA method 1613B^[20] in the Super Micro Mass Research and Technology Center of Cheng Shiu University. Egg yolks and meat samples were dried with Sodium Sulfate and Soxhlet extracted with mixture of n-Hexane and dichloromethane (1:1, v/v). The extracts were concentrated to obtain the lipid. These lipid samples were spiked with ¹³C₁₂ internal standards and cleaned up with silica gels (acid and basic), alumina, and activated carbon columns.^[20] The final extracts were concentrated to about 1 mL in rotary vacuum concentrators, further concentrated to near dryness by evaporation with nitrogen blowing, and spiked with the internal standards prior to being analyzed.

The recoveries of PCDD/F internal standards for the tetra- through hexa-chlorinated homologues were between 67% and 99%, and met the criteria within 40%–130%, while that for the hepta- and octa-chlorinated homologues were between 53% and 102%, and met the criteria within 25%–130%. Laboratory (method) blank samples were also sampled and analyzed for quality assurance purposes. The PCDD/F mass of method blank samples for the tetra- through hexa-chlorinated homologues were much lower than 20 pg, while that for the hepta- and octa-chlorinated homologues were much lower than 150 pg.

A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PCDD/F analyses. The HRGC (Hewlett Packard 6970 Series gas, CA, USA) was equipped with a DB-5MS fused

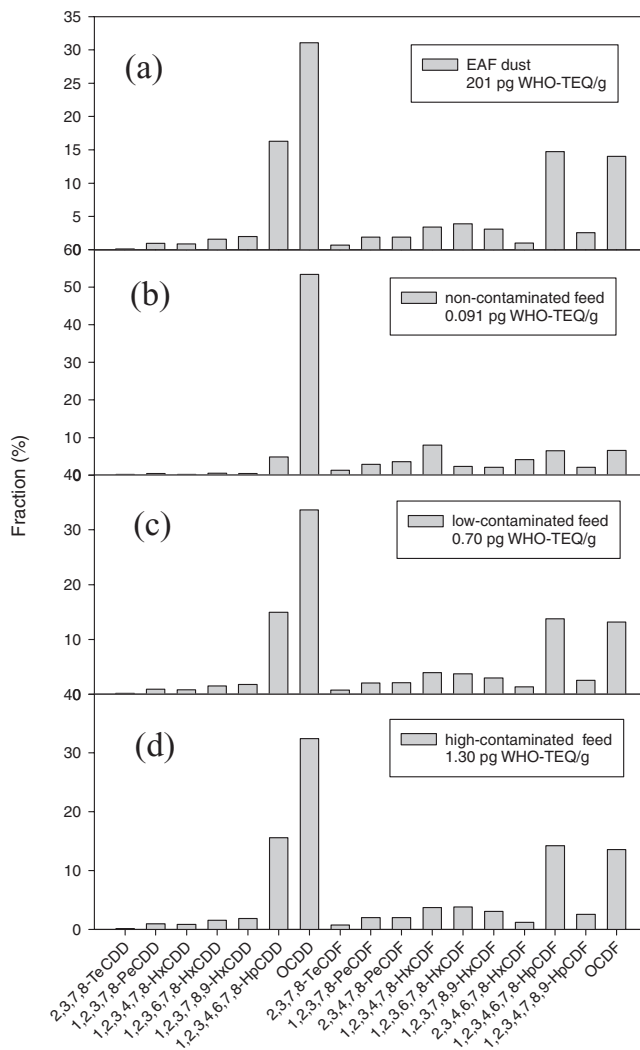


Fig. 1. Congener profiles of PCDD/Fs in (a) EAF dust, (b) NCF, (c) LCF, and (d) HCF.

silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μ m) (J&W Scientific, CA, USA), and with a splitless injection. Helium was used as the carrier gas. The oven temperature program was set according to follows: begin at 150°C (held for 1 min), then increased at 30°C min⁻¹ to 220°C (held for 12 min), then increased at 1.5°C min⁻¹ to 240°C (held for 5 min), and finally increased at 1.5°C min⁻¹ to 310°C (held for 20 min). The HRMS (Micromass Autospec Ultima, Manchester, UK) was equipped with a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring (SIM) was used with resolving power at 10,000. The electron energy and source temperature were specified at 35 eV and 250°C, respectively.

Results and discussion

Feed and EAF dust

Table 1 shows that the PCDD/F concentration was 0.091 pg WHO-TEQ/g in NCF, while the EAF dust

Table 2. PCDD/F concentrations (pg/g lipid) and lipid content (%) in duck eggs of the NCG.

PCDD/PCDFs	Period of feeding ducks with feed									
	1st day	8th day	15th day	21st day	27th day	33rd day	41st day	47th day	54th day	60th day
2,3,7,8-TeCDD	0.10	0.11	0.05	0.08	0.08	0.027	0.051	0.06	0.10	0.11
1,2,3,7,8-PeCDD	0.38	0.41	0.24	0.34	0.36	0.309	0.284	0.31	0.39	0.34
1,2,3,4,7,8-HxCDD	0.20	0.22	0.22	0.29	0.278	0.160	0.145	0.16	0.23	0.21
1,2,3,6,7,8-HxCDD	0.63	0.69	0.65	0.79	0.697	0.420	0.379	0.42	0.55	0.49
1,2,3,7,8,9-HxCDD	0.25	0.27	0.27	0.33	0.295	0.183	0.173	0.19	0.26	0.24
1,2,3,4,6,7,8-HpCDD	0.95	1.02	2.38	2.79	2.31	2.06	1.80	1.90	2.01	2.14
OCDD	2.91	3.15	3.62	4.32	4.40	4.57	4.96	4.43	4.35	3.86
Total PCDDs	5.42	5.87	7.43	8.92	8.42	7.73	7.79	7.46	7.88	7.39
2,3,7,8-TeCDF	1.18	1.27	1.12	0.75	0.72	0.83	0.90	0.99	1.24	1.08
1,2,3,7,8-PeCDF	0.82	0.75	0.92	0.98	0.79	0.68	0.57	0.62	1.04	1.10
2,3,4,7,8-PeCDF	1.03	0.93	1.32	1.42	1.12	0.78	0.72	0.80	0.92	0.73
1,2,3,4,7,8-HxCDF	0.54	0.59	0.67	0.78	0.61	0.52	0.47	0.51	0.75	0.73
1,2,3,6,7,8-HxCDF	0.53	0.57	0.71	0.81	0.61	0.44	0.40	0.44	0.71	0.74
1,2,3,7,8,9-HxCDF	0.39	0.42	0.48	0.56	0.42	0.34	0.30	0.33	0.45	0.42
2,3,4,6,7,8-HxCDF	0.03	0.03	0.04	0.05	0.03	0.03	0.05	0.06	0.05	0.03
1,2,3,4,6,7,8-HpCDF	0.27	0.30	0.52	0.58	0.54	0.36	0.30	0.32	0.54	0.57
1,2,3,4,7,8,9-HpCDF	0.02	0.02	0.12	0.13	0.10	0.08	0.06	0.07	0.09	0.09
OCDF	0.17	0.19	0.46	0.51	0.39	0.34	0.26	0.29	0.28	0.17
Total PCDFs	4.98	5.07	6.36	6.57	5.33	4.40	4.03	4.43	6.07	5.67
Total PCDD/Fs	10.4	10.9	13.8	15.5	13.7	12.1	11.8	11.9	14.0	13.1
WHO-TEQ	1.42	1.44	1.44	1.64	1.44	1.08	1.03	1.13	1.45	1.29
Lipid content	11.7	12.0	12.5	11.8	13.3	12.7	12.8	9.7	11.8	11.2

exhibited a concentration of 201 pg WHO-TEQ/g. The PCDD/F concentrations of duck feed in this study are close to those of the chicken feed (0.08–0.2 pg WHO TEQ/g dry w) in an Egyptian study,^[21] and that of the EAF dust is lower than that from hazardous waste incineration.^[22]

As shown in Figure 1(b), for total PCDD/F concentration in the NCF, more than 50% was contributed by OCDD and the fractions of the remaining 16 congeners were all below 10%. As for the PCDD/F congener profiles of the EAF dust (Fig. 1a), in addition to OCDD, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDF were also the main contributors. According to the mass ratio of the EAF dust and the NCF, concentrations of the LCF and HCF were calculated to be 0.70 and 1.30 pg WHO-TEQ/g. The LCF was slightly lower and the HCF was much higher than both the maximum threshold level of 0.75 pg WHO-TEQ/g for feed^[23] and the chicken feed (0.78 pg WHO-TEQ/g) in the Belgian dioxin incident.^[24]

PCDD/F concentrations in eggs and duck meat

The PCDD/F contents of eggs over the entire period of the experiments for the NCG are given in Table 2. First, the lipid contents present in duck eggs ranged from 9.7–13.3%, which was slightly higher than in Hsu et al. (9.7%).^[6] Second, in the NCG (Table 2), there was no clear increasing/decreasing trend related to the PCDD/F content of

eggs over time. It ranged from 1.03 to 1.64 pg WHO-TEQ/g lipid, which was well below the maximum threshold level of 3 pg WHO-TEQ/g lipid.^[25] As shown in Table 3, the PCDD/F concentrations (Total PCDD/Fs or WHO-TEQ) of eggs in the LCG reached a steady state at day 15. After the ducks had ingested the NCF for an additional 19 days, the PCDD/F content of eggs for the LCG did not decrease to the baseline level, although the concentrations during the period were below acceptable levels (3 pg WHO-TEQ/g lipid). For eggs in the HCG, shown in Table 4, PCDD/F concentrations kept increasing until day 21. Moreover, the concentrations started to be higher than 3pg WHO-TEQ/g lipid from day 15, and then dropped promptly after exposure stopped (on day 41). On day 60, the concentration was still about 1.9 times (2.59/1.34) higher than that before exposure, although it had dropped below the threshold level. Compared with previous studies for chicken,^[14,16] the PCDD/F content of eggs reached a steady state at 30–60 days and 8 weeks, while the eggs from ducks that were fed 35.8 and 9 pg WHO-TEQ/g contained more fraction of lipid than eggs from chickens fed less contaminated feed.

Figures 2, 3, and 4 show the variations of different chlorinated congeners during the exposure. In the NCG (Fig. 2), only Hepta-CDD and OCDD had very slight variation, while the others were all at a steady state. For the eggs of the LCG (Fig. 3) and HCG (Fig. 4), the distribution

Table 3. PCDD/F concentrations (pg/g lipid) and lipid content (%) in duck eggs of the LCG.

PCDD/PCDFs	<i>The period of feeding ducks contaminated feed</i>							<i>The period after stopping feeding contaminated feed</i>		
	<i>1st day</i>	<i>8th day</i>	<i>15th day</i>	<i>21st day</i>	<i>27th day</i>	<i>33rd day</i>	<i>41st day</i>	<i>47th day</i>	<i>54th day</i>	<i>60th day</i>
2,3,7,8-TeCDD	0.07	0.06	0.08	0.07	0.05	0.08	0.10	0.13	0.15	0.13
1,2,3,7,8-PeCDD	0.33	0.28	0.47	0.40	0.45	0.39	0.57	0.97	0.82	0.61
1,2,3,4,7,8-HxCDD	0.15	0.19	0.34	0.34	0.45	0.36	0.41	0.37	0.41	0.36
1,2,3,6,7,8-HxCDD	0.36	0.59	0.97	0.90	0.99	0.82	0.96	0.88	0.96	0.85
1,2,3,7,8,9-HxCDD	0.16	0.27	0.51	0.45	0.56	0.41	0.50	0.43	0.41	0.39
1,2,3,4,6,7,8-HpCDD	0.70	2.32	3.70	3.96	3.94	3.53	3.66	3.34	3.05	1.87
OCDD	2.51	4.71	6.09	6.15	5.95	5.86	6.12	5.95	4.94	4.00
Total PCDDs	4.28	8.42	12.2	12.3	12.4	11.5	12.3	12.1	10.7	8.21
2,3,7,8-TeCDF	0.80	0.84	0.91	0.90	1.01	0.80	1.16	1.15	1.67	1.32
1,2,3,7,8-PeCDF	0.67	0.73	1.08	1.01	1.27	1.20	1.38	1.41	1.50	1.51
2,3,4,7,8-PeCDF	0.83	1.01	1.41	1.36	1.39	1.45	1.40	1.50	1.32	1.10
1,2,3,4,7,8-HxCDF	0.41	0.69	1.27	1.42	1.45	1.42	1.35	1.27	1.41	1.25
1,2,3,6,7,8-HxCDF	0.40	0.67	1.33	1.49	1.46	1.45	1.30	1.32	1.37	1.27
1,2,3,7,8,9-HxCDF	0.26	0.52	1.08	1.04	1.05	0.99	1.00	0.80	0.91	0.75
2,3,4,6,7,8-HxCDF	0.02	0.03	0.06	0.05	0.07	0.07	0.13	0.15	0.12	0.08
1,2,3,4,6,7,8-HpCDF	0.21	1.08	1.97	1.89	2.15	2.08	1.94	1.59	0.95	0.76
1,2,3,4,7,8,9-HpCDF	0.04	0.21	0.36	0.47	0.54	0.44	0.50	0.37	0.21	0.15
OCDF	0.15	0.57	0.88	0.86	0.85	0.71	0.58	0.33	0.28	0.38
Total PCDFs	3.79	6.34	10.4	10.5	11.2	10.6	10.7	9.89	9.73	8.57
Total PCDD/Fs	8.07	14.8	22.5	22.7	23.6	22.1	23.1	21.9	20.5	16.8
WHO-TEQ	1.11	1.30	2.01	1.92	2.03	1.95	2.18	2.61	2.47	2.03
Lipid content	11.4	10.4	11.5	12.8	11.2	11.5	12.5	11.3	12.9	12.4

Table 4. PCDD/F concentrations (pg/g lipid) and lipid content (%) in duck eggs of the HCG.

PCDD/PCDFs	<i>The period of feeding ducks contaminated feed</i>							<i>The period after stopping feeding contaminated feed</i>		
	<i>1st day</i>	<i>8th day</i>	<i>15th day</i>	<i>21st day</i>	<i>27th day</i>	<i>33rd day</i>	<i>41st day</i>	<i>47th day</i>	<i>54th day</i>	<i>60th day</i>
2,3,7,8-TeCDD	0.06	0.10	0.12	0.14	0.14	0.11	0.06	0.13	0.15	0.17
1,2,3,7,8-PeCDD	0.33	0.39	0.71	0.94	0.99	0.77	0.85	0.85	0.85	0.81
1,2,3,4,7,8-HxCDD	0.14	0.21	0.63	0.72	0.68	0.73	0.50	0.54	0.46	0.39
1,2,3,6,7,8-HxCDD	0.87	0.65	1.52	1.68	1.50	1.55	1.23	1.23	1.01	1.04
1,2,3,7,8,9-HxCDD	0.28	0.26	0.80	0.89	0.80	0.83	0.62	0.63	0.49	0.51
1,2,3,4,6,7,8-HpCDD	1.98	1.60	4.82	5.20	4.86	4.74	4.34	4.04	3.00	2.42
OCDD	3.24	3.62	6.29	6.94	7.29	7.95	8.16	7.44	5.26	4.21
Total PCDDs	6.90	6.83	14.9	16.5	16.3	16.7	15.8	14.9	11.2	9.53
2,3,7,8-TeCDF	0.92	1.11	1.33	1.65	1.77	1.12	1.20	1.27	1.34	1.44
1,2,3,7,8-PeCDF	0.85	0.76	1.80	2.25	2.27	2.28	2.25	1.75	1.55	1.72
2,3,4,7,8-PeCDF	1.08	1.01	2.02	2.51	2.51	2.47	2.39	1.69	1.37	1.48
1,2,3,4,7,8-HxCDF	0.47	0.57	2.11	2.45	2.44	2.50	2.30	1.75	1.44	1.53
1,2,3,6,7,8-HxCDF	0.44	0.54	2.31	2.56	2.43	2.56	2.40	1.85	1.50	1.61
1,2,3,7,8,9-HxCDF	0.28	0.41	1.57	1.78	1.73	1.78	1.51	1.11	0.89	0.92
2,3,4,6,7,8-HxCDF	0.02	0.02	0.06	0.08	0.09	0.09	0.22	0.20	0.09	0.10
1,2,3,4,6,7,8-HpCDF	0.25	0.59	3.13	3.09	2.98	2.99	2.65	1.60	0.79	0.99
1,2,3,4,7,8,9-HpCDF	0.06	0.09	0.67	0.68	0.68	0.73	0.75	0.34	0.16	0.18
OCDF	0.21	0.39	1.22	1.15	0.85	1.14	0.37	0.57	0.35	0.33
Total PCDFs	4.59	5.48	16.2	18.2	17.7	17.7	16.0	12.1	9.47	10.3
Total PCDD/Fs	11.5	12.3	31.1	34.7	34.0	34.3	31.8	27.0	20.7	19.8
WHO-TEQ	1.34	1.43	3.04	3.72	3.73	3.43	3.29	2.83	2.52	2.59
Lipid content	12.6	12.7	11.5	11.7	11.6	11.0	11.9	12.3	11.2	11.5

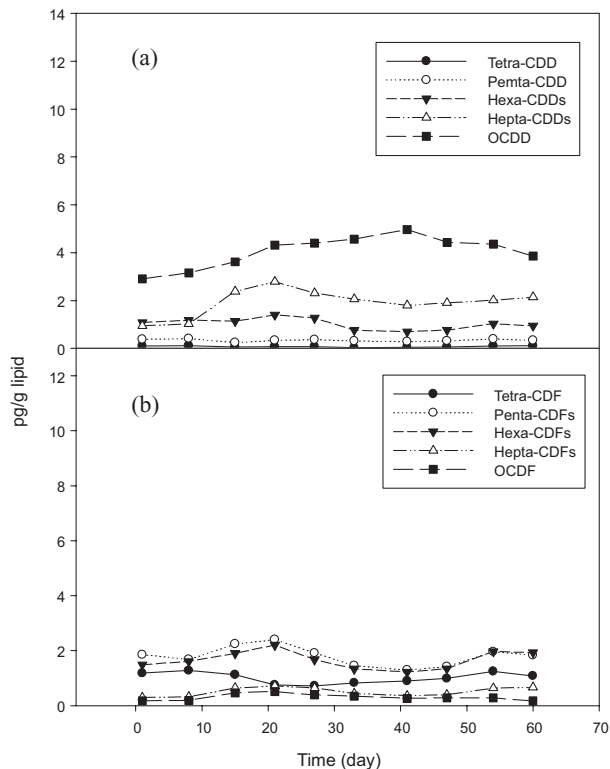


Fig. 2. Concentration profiles of (a) PCDDs and (b) PCDFs in duck eggs of the NCG for the duration of this study.

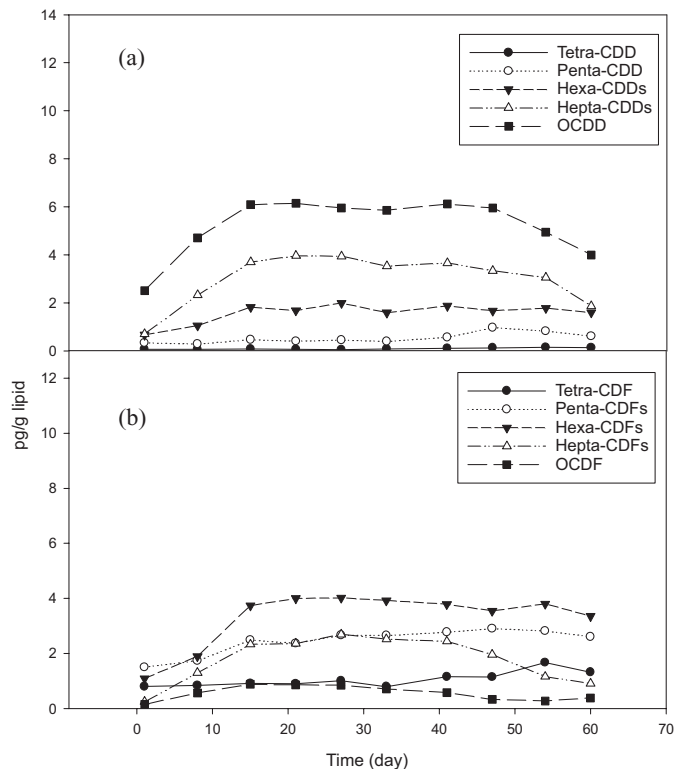


Fig. 3. Concentration profiles of (a) PCDDs and (b) PCDFs in duck eggs of the LCG for the duration of this study.

and variation of all chlorinated congeners were similar for the duration of the experiment, and the only the difference in the PCDD/F levels was observed due to the exposure levels. Little variation was shown with all congeners during the first week of exposure, which may suggest that most of the PCDD/Fs absorbed accumulated in the duck meat.^[16] OCDD, Hepta-CDD and Hexa-CDF were detected to increase significantly after the sample taken at day 15. This is expected, due to the large fraction of OCDD, Hepta-CDD, and Hexa-CDF in the fly ash. However, OCDF was at a steady state over the 60 days, even though it was at almost the same level as Hexa-CDF in the feed.

As for the PCDD/F concentration in duck meat shown in Table 5, after 41 days of exposure, the NCG was at a level of 0.92 pg WHO-TEQ/g lipid, indicating that the feed contaminated at the level below 0.1 pg WHO-TEQ/g did not cause any further accumulation. However, the duck meat in the LCG that was exposed to the contaminated feed slightly below the maximum level achieved a concentration of 2.43 pg WHO-TEQ/g lipid at day 41, and those in the HCG after exposure were at an even high level of 9.50 pg WHO-TEQ/g lipid. For both groups, the PCDD/F concentrations of muscles remained over 2 pg WHO-TEQ/g lipid after 19 days of ingesting NCF, which shows that even though the PCDD/Fs in feed are under an acceptable level, they may accumulate in the duck meat to an unacceptable level.

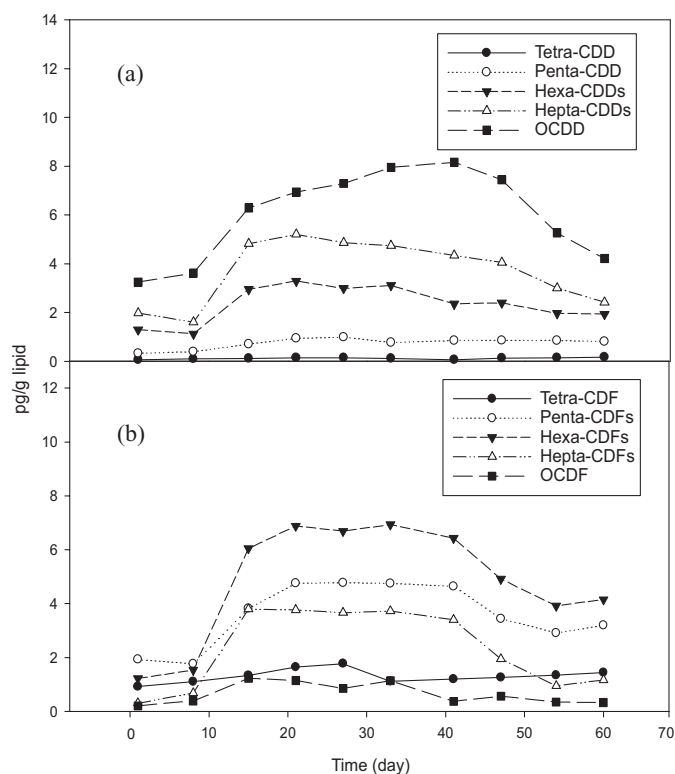


Fig. 4. Concentration profiles of (a) PCDDs and (b) PCDFs in duck eggs of the HCG for the duration of this study.

Table 5. PCDD/F concentrations (pg/g lipid) and lipid content (%) in duck meat (pg/g lipid) before dosing the ducks (0 day), after dosing (41th day) and after stopping dosing for 19 days (60th day).

PCDD/PCDFs	0 day	41st day			60th day		
		NCG	LCG	HCG	NCG	LCG	HCG
2,3,7,8-TeCDD	0.081	0.076	0.11	0.73	0.24	0.19	0.17
1,2,3,7,8-PeCDD	0.41	0.24	0.68	3.40	0.32	0.86	0.60
1,2,3,4,7,8-HxCDD	0.25	0.06	0.46	1.34	0.10	0.40	0.43
1,2,3,6,7,8-HxCDD	0.60	0.41	0.93	2.69	0.27	0.97	0.97
1,2,3,7,8,9-HxCDD	0.25	0.10	0.4616	0.74	0.27	0.33	0.44
1,2,3,4,6,7,8-HpCDD	0.92	0.45	1.67	1.71	1.17	0.82	1.26
OCDD	4.74	1.18	15.2	4.55	3.99	1.62	2.08
Total PCDDs	7.26	2.51	19.5	15.2	6.35	5.18	5.95
2,3,7,8-TeCDF	1.02	0.93	1.43	6.91	0.45	2.33	1.33
1,2,3,7,8-PeCDF	0.92	0.56	1.56	4.17	0.56	2.21	1.45
2,3,4,7,8-PeCDF	1.17	0.69	1.72	6.80	0.92	2.39	1.77
1,2,3,4,7,8-HxCDF	0.77	0.34	1.4	2.13	0.69	1.34	1.27
1,2,3,6,7,8-HxCDF	0.74	0.25	1.2	2.08	0.26	1.23	1.24
1,2,3,7,8,9-HxCDF	0.52	0.20	0.82	1.17	0.31	0.66	0.78
2,3,4,6,7,8-HxCDF	0.13	0.029	0.09	0.23	0.07	0.06	0.12
1,2,3,4,6,7,8-HpCDF	0.73	0.17	0.76	0.87	0.80	0.37	0.69
1,2,3,4,7,8,9-HpCDF	0.15	0.028	0.18	0.41	0.14	0.14	0.19
OCDF	1.36	0.16	1.77	0.65	0.83	0.19	0.25
Total PCDFs	7.51	3.36	10.9	25.4	5.01	10.9	9.10
Total PCDD/Fs	14.8	5.87	30.5	40.6	11.4	16.1	15.1
WHO-TEQ	1.57	0.92	2.43	9.50	1.31	3.10	2.41
Lipid content	14.6	19.7	20.5	7.1	7.6	22.6	33.7

Bioconcentration factors

Bioconcentration factors (BCFs) were calculated for each PCDD/F congener as the ratio between the concentrations of the duck meats (lipid weight) and those of the feed (dry weight) which the ducks ingested. The BCFs of the eggs and duck meat in each group are shown in Table 6. Among all congeners, 2,3,7,8-TeCDF showed significant higher BCFs than the others. As observed with chicken,^[14–16] the BCFs decreased with the increasing degree of PCDD/F chlorination, this study is consistent with this trend. Since the higher chlorinated PCDD/Fs were less absorbed by the gastrointestinal tract,^[16] they were less present in the eggs. Additionally, for BCFs of duck meat from the LCG and the HCG, only 4–6 chlorinated congeners increased significantly with the increase of PCDD/F exposure levels, and little difference was observed for more chlorinated congeners. Since the BCFs of eggs do not show significant differences between the LCG and HCG, as those of duck meat do, it may suggest that either the absorption rate increases with the uptake of PCDD/Fs levels or the steady state had not been reached.

Output fractions

The variations of the PCDD/F output fractions of the eggs in each group are given in Figures 5–7. In the LCG

Table 6. Individual BCFs in the duck eggs and the duck meat.

Congeners	Duck eggs		Duck meat	
	LCG	HCG	LCG	HCG
2,3,7,8-TeCDD	4.3	2.3	5.0	19.2
1,2,3,7,8-PeCDD	3.1	2.7	4.4	11.5
1,2,3,4,7,8-HxCDD	2.9	2.4	3.5	5.2
1,2,3,6,7,8-HxCDD	3.6	2.9	3.8	5.6
1,2,3,7,8,9-HxCDD	1.5	1.2	1.6	1.3
1,2,3,4,6,7,8-HpCDD	1.5	0.9	0.7	0.4
OCDD	1.1	0.8	2.7	0.4
Total PCDD	1.3	1.0	2.2	0.9
2,3,7,8-TeCDF	7.6	5.0	11.1	29.8
1,2,3,7,8-PeCDF	3.8	3.7	4.6	6.8
2,3,4,7,8-PeCDF	4.1	3.9	5.0	11.0
1,2,3,4,7,8-HxCDF	2.1	2.1	2.1	1.9
1,2,3,6,7,8-HxCDF	2.2	2.1	2.0	1.8
1,2,3,7,8,9-HxCDF	2.0	1.7	1.7	1.2
2,3,4,6,7,8-HxCDF	0.5	0.4	0.4	0.6
1,2,3,4,6,7,8-HpCDF	0.9	0.6	0.3	0.2
1,2,3,4,7,8,9-HpCDF	1.1	0.9	0.4	0.5
OCDF	0.3	0.2	0.8	0.2
Total PCDF	1.4	1.2	1.4	1.7

BCFs of eggs were calculated by dividing the average concentration at days 33 and 41 in eggs by the concentration in the feed of each group. BCFs of duck meat were calculated by dividing the concentrations at day 41 by the feed concentration.

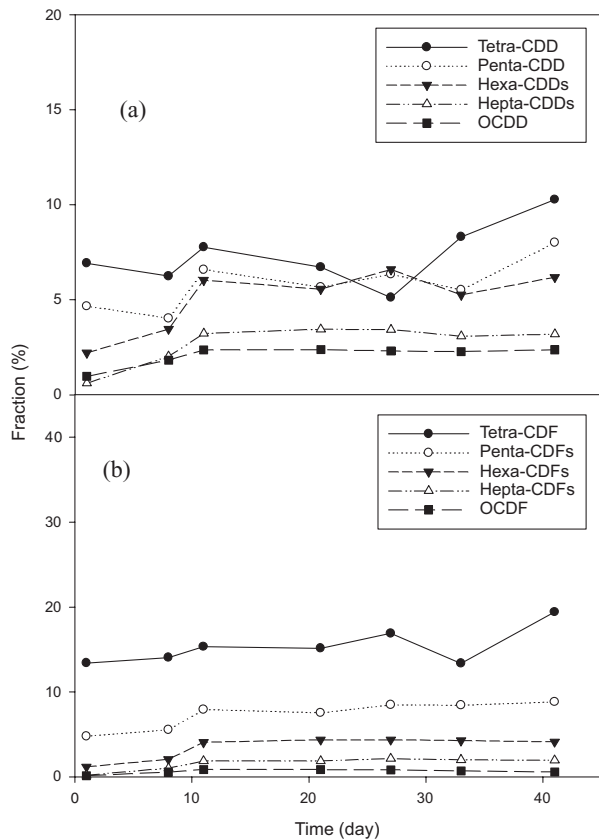


Fig. 5. Output fractions of (a) PCDDs and (b) PCDFs in duck eggs of the LCG for the duration of this study.

(Fig. 5), with feed concentration lower than the limit of 0.75 pg WHO-TEQ/g lipid, the output fraction only slightly increased. For tetra-CDD it increased from 6.9% on the first day to 10.3% on the last day of exposure, while it rose from 0.97% to 2.37% for OCDD. Tetra-CDF, which was output the most from the eggs, was about 13.4% at the beginning and about 19.4% at the end of the exposure. The output fraction of the other chlorinated congeners decreased regularly from low to high chlorinated congeners.

The output fractions from the HCG eggs were higher than those of the LCG in 4 to 6 chlorinated congeners. However, 7 to 8 chlorinated congeners did not show much difference, because of the low absorption of the high degree of chlorination for both PCDDs and PCDFs. This is consistent with the results found in previous studies.^[14–16] The output fractions of Tetra-CDD and Tetra-CDF showed instability after 4 weeks of exposure. As found by McLachlan and Richter,^[12] in cows, 2,3,7,8-TCDF is more easily metabolized than other congeners, which may explain the instability.

Furthermore, 9 to 10% of total TEQ on the lipid weight basis were stably released after three weeks of exposure to LCF, while 6 to 7% of total TEQ were released with exposure to HCF. The released fractions observed in this study are lower than those (17%) of the chickens exposed

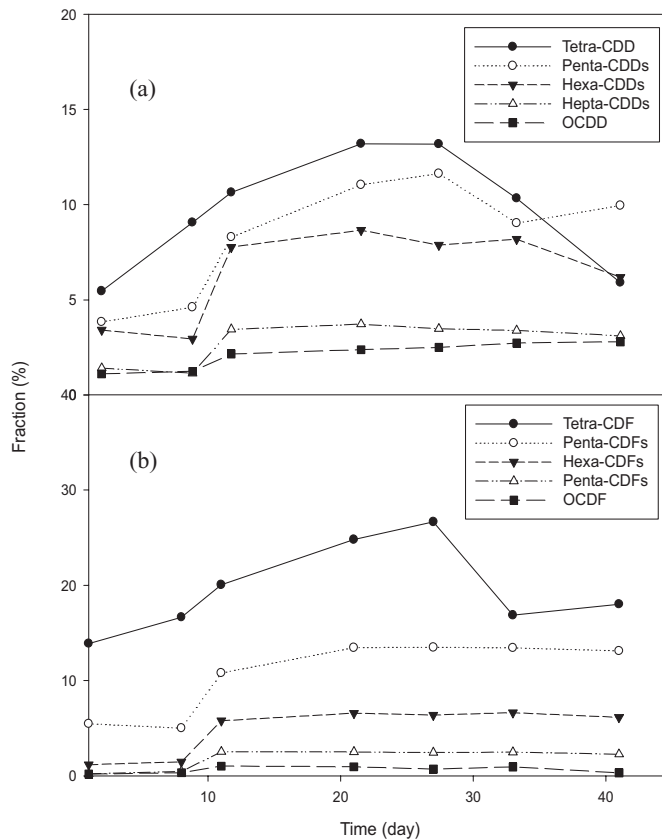


Fig. 6. Output fractions of (a) PCDDs and (b) PCDFs in duck eggs of the HCG for the duration of this study.

to feeds of 10 pg WHO-TEQ/g in earlier research.^[16] We thus suggest that the absorption of PCDD/Fs is dependent on the concentration in the feed. One can expect that about 6.7% of PCDD/Fs were excreted through the eggs by ducks fed with 0.75 pg WHO-TEQ/g feed and assuming that

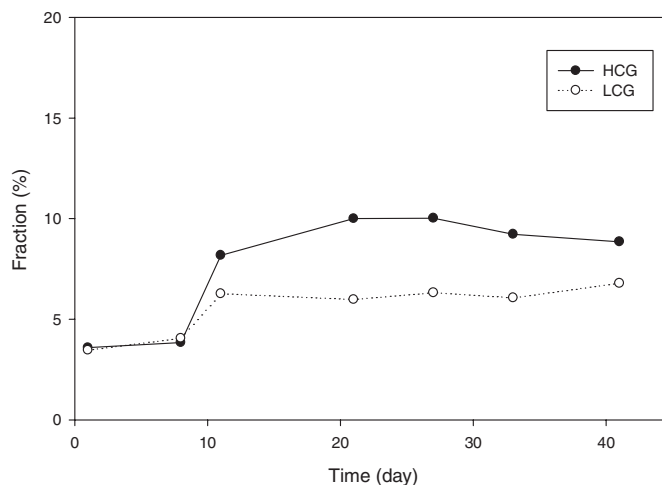


Fig. 7. Output fractions of toxicity (TEQ) of (a) PCDDs and (b) PCDFs in duck eggs for the duration of this study.

200 g of feed per day per duck was ingested, the ducks would lay eggs with less than 1 pg WHO-TEQ/g lipid.

Conclusions

Since the BCFs and released fraction of duck eggs decrease with the increase of the degree of chlorination of PCDD/Fs, we can suggest that higher chlorinated congeners were absorbed less. As laying eggs is an important route to excrete PCDD/Fs, PCDD/Fs may keep accumulating to an unacceptable level in meat from ducks who do not lay. A longer period of exposure and recovery may be needed to ensure the steady state of PCDD/F levels in duck meat. Nevertheless, a lower concentration in feed would be needed to produce duck meat less contaminated than 2 pg TEQ/g lipid.

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