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Assessment of polychlorinated dibenzo-*p*-dioxins and dibenzofurans contribution from different media to surrounding duck farms

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ABSTRACT

Since the "Toxic Egg Event" broke out in central Taiwan, the possible sources of the high content of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in eggs have been a serious concern. In this study, the PCDD/F contents in different media (feed, soil and ambient air) were measured. Evaluation of the impact from electric arc furnace dust treatment plant (abbreviated as EAFDT plant), which is site-specific to the "Toxic Egg Event", on the duck total-PCDD/F daily intake was conducted by both Industrial Source Complex Short Term model (ISCST) and dry and wet deposition models.

After different scenario simulations, the worst case was at farm A and at 200 g feed and 5 g soil for duck intake, and the highest PCDD/F contributions from the feed, original soil and stack flue gas were 44.92, 47.81, and 6.58%, respectively. Considering different uncertainty factors, such as the flow rate variation of stack flue gas and errors from modelling and measurement, the PCDD/F contribution fraction from the stack flue gas of EAFDT plant may increase up to twice as that for the worst case (6.58%) and become 13.2%, which was still much lower than that from the total contribution fraction (86.8%) of both feed and original soil. Fly ashes contained purposely in duck feed by the farmers was a potential major source for the duck daily intake. While the impact from EAFDT plant has been proven very minor, the PCDD/F content in the feed and soil, which was contaminated by illegal fly ash landfills, requires more attention.

1. Introduction

In June 2004, a high level of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in duck eggs (30.0–45.0 pg WHO/g-fat) was discovered in Changhua County, located in the middle Taiwan. The PCDD/F I-TEQ concentration was approximately 10–15 times higher than the EU limits (3 pg WHO/g-fat) [1]. This issue was called "Toxic Egg Event of Changhua" and abbreviated as "Toxic Egg Event". The possible sources and major transport routes that caused those eggs to contain so high an amount of PCDD/Fs are obvious of public concern. In the environment, various media and exposure routes, such as inhalation and ingestion of food, and water, could result in different possible levels of PCDD/F accumulation [2]. Therefore, the measurement of PCDD/F contents in different media and the establishment of a methodology for the assessment of major PCDD/F routes for the ducks' intake is a critical issue for clearer understanding of the "Toxic Egg Event".

Several reports were suspecting a specific electric arc furnace dust treatment plant (EAFDT plant), locating near the duck farm, to be responsible for the "Toxic Egg Event". During the period before the "Toxic Egg Event", the mean PCDD/F concentration in the stack flue gases of EAFDT plant was 181 ng I-TEQ/Nm^3 [3]. In 2004, there was no stack emission standard regulated for EAFDT plant in Taiwan. Comparing with other kind of pollutant sources, 181 ng I-TEQ/Nm^3 was much higher than the regulated standard of large-scale municipal solid wastes incinerators (0.1 ng I-TEQ/Nm^3) and sintering plants (2.0 ng I-TEQ/Nm^3). Indeed, a high PCDD/F concentration of stack flue gas from the EAFDT plant was released into the atmosphere and was subject to atmospheric dispersion and deposition [4]. However, without solid scientific evidence, the effect of the EAFDT plant on the surrounding duck farms was still unclear.

Human or animals intake PCDD/Fs through various routes, such as dermal absorption, respiration, and diet [5], particularly more than 90% of PCDD/F intake is contributed by the last route [6]. In

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this study, PCDD/F contents in the feed, soil, or concentrations in the ambient air were measured and the result of PCDD/F concentrations in stack flue gas from the EAFDT plant was cited from a previous study [3]. Then, by using the dispersion, dry deposition and wet deposition modelling, the impact of PCDD/F emission from the stack flue gas of the EAFDT plant on the surrounding duck farms was assessed. Consequently, the major contribution fractions on resulting in the PCDD/F “Toxic Egg Event” are presented and discussed.

2. Experimental method

2.1. EAFDT plant

The EAFDT plant was established in 1999 to recover zinc oxide via the Waelz process [7,8]. The EAFDT capacity is approximately 70,000 t/year and the input materials include EAF dust (9.5 t/h), coke (1.71 t/h), and sand (1.71 t/h). The ignition is done by a burner that is fed with diesel oil and ambient air [3]. The treatment process is conducted in a high temperature rotary-kiln that is 40 m long and has a 3.6 m outer diameter. It is equipped with various air pollution control devices, including a dust setting chamber (DSC), venturi tower, cyclone, and bag filters. The height of the stack is 35 m and the flow rate of flue gas is 52,200 Nm^3/h (Table 1) [3].

2.2. Sampling and analysis of PCDD/Fs

The stack flue gas sampling procedures followed those of the US EPA Modified Method 23 [9], and each stack gas sampling lasted for 2–3 h. The PCDD/Fs in the ambient air were collected by a standard semi-volatile sampling train (General Metal works PS-1) according to the revised US EPA Reference Method TO9A [10].

Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards pre-labelled with isotopes. The analyses for stack flue gas and ambient air samples were performed according to the US EPA Modified Method 23 [9] and US EPA Reference Method TO9A [10], respectively. Soil, feed and egg samples were analyzed with the US EPA Method 1613B [11]. Essentially, samples were extracted with toluene for 24 h and this was then followed by a series of sample cleanup procedures. The extract was transferred to a vial, and finally further concentrated by a N_2 gas stream.

Two high-resolution gas chromatographs/high-resolution mass spectrometers (HRGC/HRMS) were used for PCDD/F analysis. The HRGC (Hewlett-Packard 6970 Series gas, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, ID = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA) with a splitless injection, while the HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact (EI+) source. The analyzer mode of the selected ion monitoring was used with the resolving power at 10,000. The electron energy and source temperature were specified at 35 eV and 250 °C, respectively. The oven temperature program was set according to the following: initially at 150 °C (held for 1 min), then increased by 30 °C/min to 220 °C (held for 12 min), and finally increased by 1.5 °C/min to 310 °C (held for 20 min). Helium was used as the carrier gas.

Table 1
Basic information of the EAFDT plant

Feed materials	EAF fly ash, coke, sand
Furnace type	Waelz rotary kiln
Air pollution control devices	Dust settling chamber, venturi tower, cyclones, bag filters
Stack height (m)	35
Flue gas rate (Nm^3/h)	52,200
Treatment capacity (t/year)	Approximately 70,000

2.3. Gas/particle partitioning

The issue of gas/particle partitioning has particular significance for PCDD/Fs, because it will affect the deposition processes (e.g., the relative importance of gaseous and particulate dry/wet depositions, respectively) which can transfer PCDD/Fs to terrestrial and aquatic food chains and compartments [12].

The distribution of PCDD/Fs between the gas phase and particle phase depends on the particle concentration, particle size distribution, phase related properties, ambient temperature, relative humidity and the compounds themselves [12]. An equation that has been used with success to describe gas–particle partitioning is

$$K_p = \frac{F/\mathrm{TSP}}{A}$$

where K_p ($m^3/\mu g$) is a temperature-dependent partitioning constant, TSP ($\mu g/m^3$) is the concentration of total suspended particulate materials, F (pg/m^3) is the concentration of the compound bound to particles, and A (pg/m^3) is the gaseous concentration of the compound of interest [13–16]. Plotting log K_p against the logarithm of the subcooled liquid vapor pressure, P_L^o, gives

$$\log K_p = m_r \times \log P_L^o + b_r$$

where m_r is the slope and b_r is the y-intercept of the trend line.

Eitzer and Hites [17] have correlated P_L^o of PCDD/Fs with gas chromatographic retention indexes (GC-RI) on a non-polar (DB-5) GC-column using p,p'-DDT as a reference standard. The correlation has been redeveloped by Hung et al. [18] as

$$\log {P_L}^o = \frac{-1.34(\mathrm{RI})}{T} + 1.67 \times 10^{-3}(\mathrm{RI}) - \frac{1320}{T} + 8.087$$

where P_L^o is the subcooled liquid vapor pressure, RI is the gas chromatographic retention indexes derived by Donnelly et al. [19] and Hale et al. [20], and T is ambient temperature (K). In this study, the RIs derived by Donnelly et al. [19] and Hale et al. [20] and the equation redeveloped by Hung et al. [18] were taken to generate the P_L^o values. A complete dataset on the gas–particle partitioning of PCDD/Fs in Taiwan has been reported [21] with the data giving values for $m_r = -1.29$ and $b_r = -7.2$ with $R^2 = 0.94$. In this study, the trend line proposed by Chao et al. [21] was taken to estimate the partitioning constant, K_p.

2.4. Dry and wet deposition processes of PCDD/Fs

The dry deposition flux of PCDD/Fs in the atmosphere is a combination of both gas-phase and the particle-phase fluxes, which is given by

$$F_T = F_g + F_p,$$

F_T is the summation of PCDD/F deposition fluxes from both gas and the particle phases; F_g is the PCDD/F deposition flux from the gas phase, F_p is the PCDD/F deposition flux from the particle phase, where $F_T = C_T \times V_{d,T}$, $F_g = C_g \times V_{d,g}$, and $F_p = C_p \times V_{d,p}$.

Therefore,

$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p}$$

where C_T is the measured concentration of total PCDD/Fs in the ambient air, $V_{d,T}$ is the dry deposition velocity of total PCDD/Fs, C_g is the calculated concentration of PCDD/Fs in the gas phase, $V_{d,g}$ is the dry deposition velocity of the gas-phase PCDD/Fs, C_p is the calculated concentration of PCDD/Fs in the particle phase, $V_{d,p}$ is the dry deposition velocity of the particle-phase PCDD/Fs.

The mean dry deposition velocity of total PCDD/Fs (0.42 cm/s) was proposed by Shih et al. [22]. This value (V_d = 0.42 cm/s) is also used for the approximate calculation of total PCDD/F dry deposition flux. Dry deposition of gas-phase PCDD/Fs is mainly by diffusion, and because of a lack of measured data for PCDD/Fs, a selected value (0.010 cm/s) of gas-phase PAH dry deposition velocity, $V_{d,g}$, proposed by Sheu et al. [23] and used by Lee et al. [24], is also used here to calculate the PCDD/F dry deposition flux contributed by its gas phase.

The wet deposition flux of PCDD/Fs is a combination of both vapor dissolution into rain and the removal of suspended particulate by precipitation. The gas scavenging ratio, S_g, can be estimated by

$$S_g = \frac{RT}{H},$$

S_g is the gas scavenging ratio of PCDD/Fs (dimensionless), R is the universal gas constant (82.06×10^{-6} m^3 atm/mol K), T is the ambient temperature (K), H is the Henry constant (m^3 atm/mol).

$$S_g = \frac{C_{rain,dis}}{C_g}$$

$C_{rain,dis}$ is the dissolved-phase concentration of PCDD/Fs in the raindrop, C_g is the concentration of PCDD/Fs in the gas phase.

The particle scavenging ratio, S_p, on the other hand, can be calculated by

$$S_p = \frac{C_{rain,particle}}{C_p}$$

S_p is the particle scavenging ratio of PCDD/Fs (dimensionless), $C_{rain,particle}$ is the particle-phase concentration of PCDD/Fs in the raindrop, C_p is the concentration of PCDD/Fs in the particle phase.

Total scavenging of precipitation (S_{tot}) is the sum of gas and particle scavenging, it can be calculated by

$$S_{tot} = S_g(1 - \Phi) + S_p \times \Phi \quad (9)$$

S_{tot} is the total scavenging ratio of PCDD/Fs (dimensionless), Φ is the fraction of the total air concentration bound to particles.

The particle scavenging ratio of PCDD/Fs based on average Bloomington air and rain concentrations were measured by Eitzer and Hites [25]. Because of a lack of measured data, the S_p values of OCDD and OCDF measured by Eitzer and Hites [25] were averaged and used in this study.

2.5. PCDD/F dispersion from the stack of EAFDT plant to eight surrounding duck farms

The concentrations of PCDD/Fs emitted from the stack of the EAFDT plant to the eight surrounding duck farms were estimated by using the Industrial Source Complex Short Term model (ISCST). Table 2 summarizes the dispersion parameters used in this study. The model has been widely used to simulate ambient air concentrations at specified receptor points for various sources [26–28]. The coordinates and location of the EAFDT plant and eight duck farms are shown in Table 3 and Fig. 1.

The operating period of ISCST, which began on 1 October 1999 and ended on 16 June 2005, on EAFDT plant was totally 2084 days. The meteorological information was obtained from both Wuchi and Banciao weather stations.

3. Results and discussion

3.1. PCDD/F concentration in different media

The mean of total PCDD/F concentration (total concentration of seventeen 2,3,7,8-substituted PCDD/F congeners) of the stack flue gas from the EAFDT plant was 1877 ng/Nm^3, and the mean of total PCDD/F I-TEQ concentration was 181 ng I-TEQ/Nm^3 [3]. These results were used to calculate or simulate the dry and wet depositions from stack flue gas for eight duck farms. Fig. 2 shows the congener profiles of the seventeen 2,3,7,8-substituted PCDD/Fs in different media. The y-coordinate was normalized by dividing the concentration or content of each congener by that of total PCDD/Fs. For stack flue gas, the most abundant congeners were 1,2,3,4,6,7,8-HpCDF, 1,2,3,6,7,8-HxCDF and 1,2,3,4,7,8-HxCDF (Fig. 2a) [3]. Table 4 lists the PCDD/F contents or concentrations in the soil, feed and ambient air. The mean total PCDD/F and total PCDD/F I-TEQ contents

Table 2
Parameters used in the ISCST model for simulating PCDD/F concentration transported to eight sampling sites

Item	Parameters
Program control	Algorithm: RURAL MODE Influence of buoyancy: ignored
Receptor	Sea level: 0 m Elevation from ground: 0 m Location: downwind-side
Emission source (stack)	Emission source: single The base of the stack (sea level): 0 m Stack height: 35 m Emission rate: fixed Decay of pollutant: no decay during the transportation process Smoke height: as the parameter of wash down distances Effective smoke height: calculated Pollutant concentration: constant at steady state Flow trace effect: ignored
Meteoric parameter	Wind direction: perpendicular to smoke direction Air temperature: 12–33 °C Mixing-layer height: 500 m Temperature inclined: varies with the stability

Table 3
The coordinates of the EAFDT plant and eight duck farms

Symbol	Location
EAF plant	193387,2672519
Duck farm A	193379,2671510
Duck farm B	193563,2671500
Duck farm C	193541,2670854
Duck farm D	193543,2670554
Duck farm E	193215,2669696
Duck farm F	192723,2669786
Duck farm G	194926,2672953
Duck farm H	199405,2676558

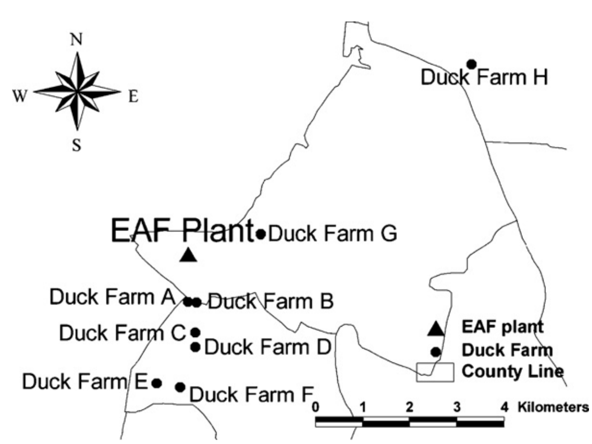

Fig. 1. The location of an EAFDT plant and eight duck farms.

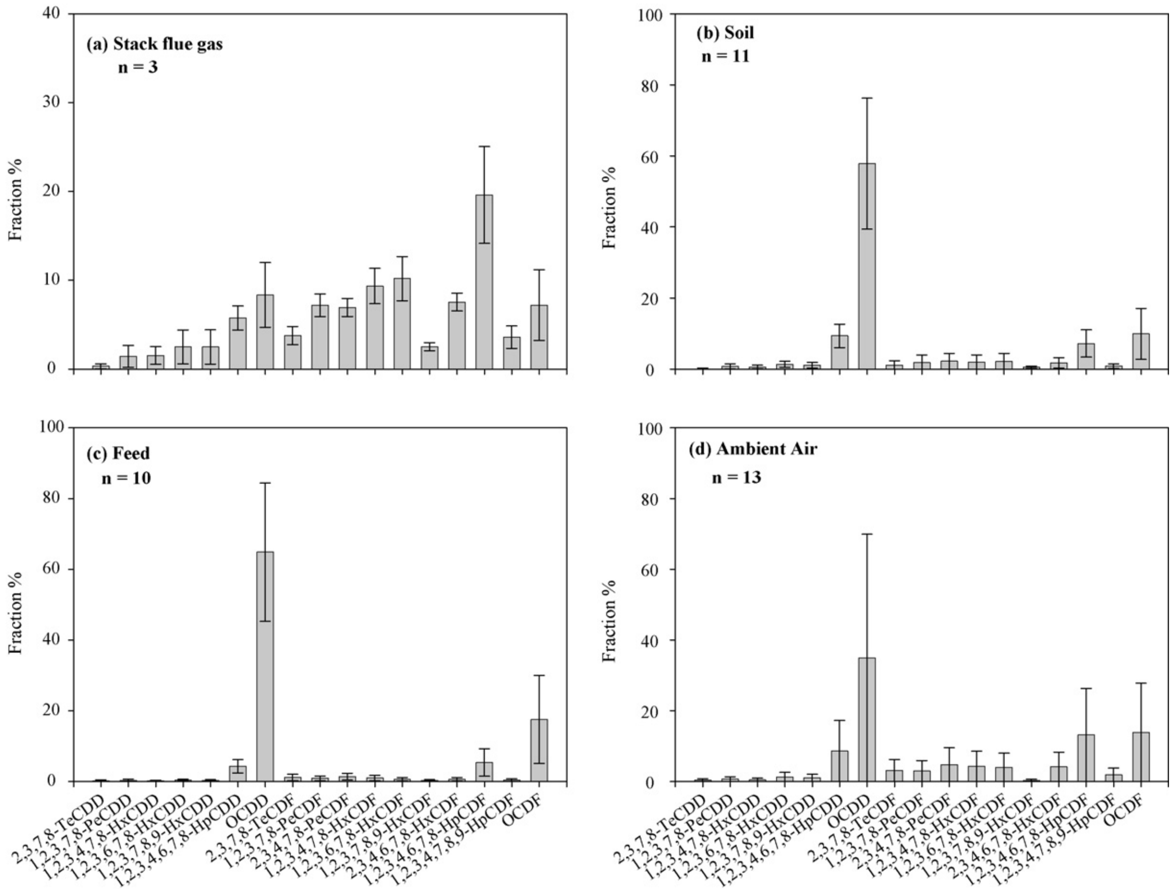

Fig. 2. The congener profiles of 17 2,3,7,8-substituted PCDD/Fs in different media.

Table 4
PCDD/F contents or concentrations in the soil, feed and ambient air

PCDD/Fs (ng/kg)	Soil		Feed		Ambient air	
	Mean (n = 11)	R.S.D. (%)	Mean (n = 10)	R.S.D. (%)	Mean[a] (n = 13)	R.S.D. (%)
2,3,7,8-TeCDD	0.362	94.5	0.0305	156	0.00429	105
1,2,3,7,8-PeCDD	2.29	105	0.0343	113	0.00592	79.2
1,2,3,4,7,8-HxCDD	2.02	119	0.0243	86.9	0.00496	88.5
1,2,3,6,7,8-HxCDD	5.06	115	0.0479	92.9	0.0112	78.3
1,2,3,7,8,9-HxCDD	4.17	121	0.0415	80.5	0.00881	79.1
1,2,3,4,6,7,8-HpCDD	40.8	115	0.603	89.6	0.0772	83.8
OCDD	221	97.5	7.54	72.9	0.312	122
2,3,7,8-TeCDF	2.79	103	0.131	84.0	0.0268	79.9
1,2,3,7,8-PeCDF	5.38	122	0.106	86.6	0.0246	75.8
2,3,4,7,8-PeCDF	7.15	119	0.159	90.7	0.0387	72.5
1,2,3,4,7,8-HxCDF	7.38	130	0.121	84.9	0.0384	81.3
1,2,3,6,7,8-HxCDF	7.95	130	0.0814	82.7	0.0350	79.4
1,2,3,7,8,9-HxCDF	2.07	137	0.0319	84.2	0.00379	117
2,3,4,6,7,8-HxCDF	7.46	135	0.0693	81.1	0.0391	85.6
1,2,3,4,6,7,8-HpCDF	36.9	126	0.651	88.7	0.133	91.3
1,2,3,4,7,8,9-HpCDF	4.63	160	0.0417	79.7	0.0185	92.8
OCDF	55.1	131	2.75	114	0.152	119
PCDDs	275	97.3	8.32	72.3	0.424	107
PCDFs	137	116	4.14	101	0.510	88.6
PCDDs/PCDFs ratio	2.01	–	2.01	–	0.832	–
Total PCDD/Fs	412	99.5	12.5	78.2	0.934	93.8
PCDDs I-TEQ	3.26	101	0.0726	104	0.0108[b]	88.6
PCDFs I-TEQ	7.08	120	0.138	85.9	0.0365[b]	76.8
PCDDs/PCDFs I-TEQ ratio	0.461	–	0.527	–	0.296	–
Total PCDD/Fs I-TEQ	10.3	114	0.210	90.1	0.0474[b]	79.2

[a] The unit is pg/Nm^3.
[b] The unit is pg I-TEQ/Nm^3.

in the soil were 412 ng/kg and 10.3 ng I-TEQ/kg, respectively. By comparing the above total PCDD/Fs I-TEQ with those of the municipal solid waste incinerators in Taiwan (ranged from 1.19 ng-TEQ/kg to 3.03 ng-TEQ/kg) [29], the content levels indicate that the soil in the present study was heavily contaminated. This result could be attributed to the fact that an illegal fly ash landfill was discovered near duck farms that caused the “Toxic Egg Event” The major congeners in the soil were OCDD, CODF and 1,2,3,4,6,7,8-HpCDD (Fig. 2b) and the mean total I-TEQ content in the feed and ambient air were 0.210 ng I-TEQ/kg (R.S.D. = 90.1%) and 0.0474 pg I-TEQ/Nm^3 (R.S.D. = 79.2%) (Table 4), respectively. For feed and ambient air, the dominant congeners were OCDD, CODF and 1,2,3,4,6,7,8-HpCDF (Fig. 2c and d).

At duck farm A, which is located at the downwind direction of EAFDT plant (Fig. 1), the PCDD/F concentrations in the ambient air were measured on various time. They were 0.0704, 0.0945, 0.174, and 0.121 pg I-TEQ/Nm^3, respectively. Via inputting the results of PCDD/F emission from the stack of EAFDT plant and the dispersion parameters (as shown in Table 2) into the ISCST model, the simulated concentrations of PCDD/Fs were 0.00027, 0.00010, 0.00026, and 0.00055 pg I-TEQ/Nm^3, respectively. Thus, the contributions of stack emission from EAFDT plant were minor and were 0.384% (0.00027/0.0704), 0.106% (0.00010/0.0945), 0.149% (0.00026/0.174), and 0.455% (0.00055/0.121), respectively. These results also indicated that during the “Toxic Egg Event” the impact of PCDD/Fs from EAFDT plant on the duck daily intake was insignificant and could be ignored.

Based on the experimental results of our laboratory, the mean and/or range of PCDD/F contents in the toxic eggs from each farm (A to H) were 19.63 (9.6–32.6), 3.46, 3.31, 3.00, 8.58 (7.34–9.81), 5.13, 2.50, and 7.83 pg WHO–TEQ $_{DF}$/g fat. Comparing the above results with total dioxin toxicity equivalency concentration guidelines (3 pg WHO–TEQ $_{DF}$/g fat) regulated for chicken egg in European Union, it was found that most of the PCDD/F contents in the toxic eggs from duck farms were higher than this guidance except that from duck farm G. The results in the experiment disclose that duck eggs were heavily contaminated and it is crucial to pinpoint the possible sources of these PCDD/Fs.

3.2. Dry/wet depositions of PCDD/Fs

The dry/wet depositions of PCDD/Fs from ambient air and stack flue gas for eight duck farms are shown in Table 5. The total dry deposition and wet depositions of PCDD/Fs from the ambient air were 16.18 ng I-TEQ/m^2 (1533 days) and 8.61 ng I-TEQ/m^2 (533 days), respectively. According to the weather and operation information, there were 1357 days for PCDD/F dry deposition and 458 days for PCDD/F wet deposition from the stack flue gas from EAFDT plant to model. Of all duck farms, duck farms A and B are the closest to EAFDT plant and are also located at downwind of the stack flue gas. The dry deposition (183.31–193.15 ng I-TEQ/m^2) and wet deposition (45.16–52.75 ng I-TEQ/m^2) of PCDD/Fs were much higher than farms G and H (3.15–16.04 ng I-TEQ/m^2, 0.94–3.61 ng I-TEQ/m^2), located at upwind of the stack flue gas. Therefore, the stack flue gas from the EAFDT plant had a higher impact on duck farms A and B than the other locations.

The questionnaire surveying and interviewing made among the duck farmers has stated a fact that was significantly associated with the “Toxic Egg Event”. In order to have a better market and more favorable prices for the duck eggs, some additives were added into the eggs. Among them fly ashes were effective in turning the duck egg yolk from the yellow to the red. As a result, these duck eggs will have a good sale and prices. Nevertheless, these fly ashes contributed critical level of PCDD/Fs to the duck eggs.

With respect to the factors that could be responsible for the high PCDD/F contents of duck eggs, the impact of dry/wet PCDD/F depositions from the ambient air and EAFDT plant on the soils has been detailed in previous discussion. In addition, the comparison between the PCDD/F contents in the original soils in the present study and other soils that was not contaminated should be provided. Based on the results obtained from our laboratory, the PCDD/F contents in the soils near certain incinerator was 1.48 ng I-TEQ/kg (n = 8). Comparing the original PCDD/F contents in the soils at duck farm A and H in this study with the above result, it is found that they were approximately 6 (8.95/1.48) and 7 (10.25/1.48) times higher. This result indicated that the original soils of the duck farms in this study had been heavily polluted by PCDD/Fs. In addition to the dry/wet depositions from the ambient air and EAFDT plant, the probable reason maybe the contribution of feces excreted by the ducks in the farms. Ducks in the farms intake the contaminated feed and soils, then feces was excreted to the soils and increased their PCDD/F contents.

3.3. Different scenario simulation

Humans and animals intake PCDD/Fs through various routes, e.g., dermal absorption, respiration, and diet [5], and more than 90% of PCDD/F intake is contributed by the last route [6]. Since ducks always walk or take a rest on the ground, the soil is probably accidentally ingested. In this study, the duck intake was simulated for diet based on the feed and soil, while both dermal absorption and respiration were ignored. In addition, the PCDD/Fs were deposited on the surface of the feed and soil through dry/wet deposition of ambient air and stack flue gas of EAFDT plant.

After field investigation, each duck was found to ingest approximately 200 g of feed per day. The duck feed area was 3 m × 3 m, and the feed exposure time was 4 h/day. There were six scenarios that contained from 0 to 5 g of soil with 200 g of feed to simulate what the duck ingested as shown in the following equation:

$$E_{total} = E_{feed} + E_{soil} \quad (1)$$

where E_{total} is the total PCDD/F mass of duck daily intake (pg); E_{feed} is the PCDD/F mass of duck daily intake contributed by feed (pg); E_{soil} is the PCDD/F mass of duck daily intake contributed by soil (pg).

Table 5
Dry deposition and wet deposition from ambient air and stack flue gas for eight duck farms

Location	Ambient air (ng I-TEQ/m^2)		Stack flue gas (ng I-TEQ/m^2)	
	Dry deposition (1533 days)	Wet deposition (551 days)	Dry deposition (1357 days)	Wet deposition (458 days)
Duck farm A	16.18	8.61	193.15	45.16
Duck farm B	16.18	8.61	183.31	52.75
Duck farm C	16.18	8.61	168.54	47.69
Duck farm D	16.18	8.61	155.84	43.57
Duck farm E	16.18	8.61	109.23	25.66
Duck farm F	16.18	8.61	96.71	21.32
Duck farm G	16.18	8.61	16.04	3.61
Duck farm H	16.18	8.61	3.15	0.94

Table 6
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 0 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	99.878	0.00	0.009	0.00	0.00	0.112	0.00	0.00	0.112
Duck farm B	Contribution I-TEQ	99.884	0.00	0.009	0.00	0.00	0.106	0.00	0.00	0.106
Duck farm C	Contribution I-TEQ	99.893	0.00	0.009	0.00	0.00	0.098	0.00	0.00	0.098
Duck farm D	Contribution I-TEQ	99.899	0.00	0.009	0.00	0.00	0.091	0.00	0.00	0.091
Duck farm E	Contribution I-TEQ	99.927	0.00	0.009	0.00	0.00	0.063	0.00	0.00	0.064
Duck farm F	Contribution I-TEQ	99.934	0.00	0.009	0.00	0.00	0.056	0.00	0.00	0.056
Duck farm G	Contribution I-TEQ	99.981	0.00	0.009	0.00	0.00	0.009	0.00	0.00	0.009
Duck farm H	Contribution I-TEQ	99.989	0.00	0.009	0.00	0.00	0.001	0.00	0.00	0.001

[a] The PCDD/F concentration from Li et al. [3].

In addition, the PCDD/F mass of duck daily intake contributed by feed (E_{feed}, pg) includes the PCDD/F mass in the original daily feed ($F_{original\ feed}$, pg), the daily PCDD/F mass entering the feed due to dry deposition from original ambient air PCDD/Fs ($D_{ambient\ air}$, pg) and the daily PCDD/F mass entering the feed due to dry deposition from the plume of stack flue gas ($D_{stack\ flue\ gas}$, pg). This can be shown as the following equation:

$$E_{feed} = F_{original\ feed} + D_{ambient\ air} + D_{stack\ flue\ gas} \quad (2)$$

Moreover, the PCDD/F mass of duck daily intake contributed by soil (E_{soil}, pg), includes the PCDD/F mass of duck daily intake contributed by the original soil ($S_{original\ soil}$, pg), the daily PCDD/F mass deposited on the soil by the dry ($D_{ambient\ air}$) or wet ($W_{ambient\ air}$) depositions from original ambient air PCDD/Fs (pg), and the daily PCDD/F mass deposited on the soil by the dry ($D_{stack\ flue\ gas}$) or wet ($W_{stack\ flue\ gas}$) depositions from the plume of stack flue gas (pg). This can be shown as following equation:

$$E_{soil} = S_{original\ soil} + D_{ambient\ air} + W_{ambient\ air} + D_{stack\ flue\ gas} + W_{stack\ flue\ gas} \quad (3)$$

In the first scenario, as shown in Table 6, the duck only ingested 200 g of feed with no soil. In the eight duck farms (A–H), more than 99.87% of the PCDD/Fs came from the feed, and the stack flue gas contributed 0.001–0.112%. In the second scenario, as shown in Table 7, 200 g of feed and 1 g of soil was ingested by the duck. For these duck farms, the PCDD/F contributions of feed, original soil, and stack flue gas were 80.23–80.30, 17.08–19.41, and 0.04–2.42%, respectively. From the third to sixth scenarios (as shown in Tables 8–11), the amount of soil ingested increased from 2 to 5 g. In each duck farm, the PCDD/F contributions from

Table 7
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 1 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	80.25	17.08	0.008	0.158	0.084	0.090	1.89	0.442	2.42
Duck farm B	Contribution I-TEQ	80.23	17.12	0.008	0.158	0.084	0.086	1.80	0.517	2.40
Duck farm C	Contribution I-TEQ	80.24	17.32	0.008	0.159	0.084	0.079	1.65	0.467	2.20
Duck farm D	Contribution I-TEQ	80.24	17.48	0.008	0.159	0.084	0.073	1.53	0.427	2.03
Duck farm E	Contribution I-TEQ	80.26	18.12	0.008	0.159	0.084	0.051	1.07	0.251	1.37
Duck farm F	Contribution I-TEQ	80.26	18.28	0.008	0.159	0.084	0.045	0.95	0.209	1.20
Duck farm G	Contribution I-TEQ	80.29	19.26	0.008	0.159	0.084	0.008	0.16	0.035	0.20
Duck farm H	Contribution I-TEQ	80.30	19.41	0.007	0.159	0.084	0.001	0.03	0.009	0.04

[a] The PCDD/F concentration from Li et al. [3].

Table 8
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 2 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	67.06	28.55	0.006	0.265	0.141	0.075	3.16	0.74	3.98
Duck farm B	Contribution I-TEQ	67.04	28.61	0.006	0.265	0.141	0.072	3.00	0.86	3.93
Duck farm C	Contribution I-TEQ	67.04	28.94	0.006	0.265	0.141	0.066	2.76	0.78	3.61
Duck farm D	Contribution I-TEQ	67.05	29.21	0.006	0.265	0.141	0.061	2.55	0.71	3.32
Duck farm E	Contribution I-TEQ	67.06	30.28	0.006	0.265	0.141	0.043	1.79	0.42	2.25
Duck farm F	Contribution I-TEQ	67.06	30.55	0.006	0.265	0.141	0.038	1.58	0.35	1.97
Duck farm G	Contribution I-TEQ	67.08	32.18	0.006	0.265	0.141	0.006	0.26	0.06	0.33
Duck farm H	Contribution I-TEQ	67.09	32.43	0.006	0.265	0.141	0.001	0.05	0.02	0.07

[a] The PCDD/F concentration from Li et al. [3].

Table 9
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 3 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	57.60	36.78	0.005	0.341	0.181	0.065	4.07	0.95	5.08
Duck farm B	Contribution I-TEQ	57.57	36.86	0.005	0.341	0.181	0.061	3.87	1.11	5.04
Duck farm C	Contribution I-TEQ	57.58	37.28	0.005	0.341	0.181	0.057	3.55	1.01	4.62
Duck farm D	Contribution I-TEQ	57.58	37.63	0.005	0.341	0.181	0.052	3.29	0.92	4.26
Duck farm E	Contribution I-TEQ	57.59	39.00	0.005	0.341	0.181	0.037	2.30	0.54	2.88
Duck farm F	Contribution I-TEQ	57.59	39.36	0.005	0.341	0.181	0.032	2.04	0.45	2.52
Duck farm G	Contribution I-TEQ	57.61	41.44	0.005	0.341	0.181	0.005	0.34	0.08	0.42
Duck farm H	Contribution I-TEQ	57.61	41.77	0.005	0.341	0.181	0.001	0.07	0.02	0.09

[a] The PCDD/F concentration from Li et al. [3].

Table 10
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 4 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	50.48	42.98	0.005	0.399	0.212	0.057	4.76	1.11	5.93
Duck farm B	Contribution I-TEQ	50.45	43.06	0.005	0.399	0.212	0.054	4.52	1.30	5.87
Duck farm C	Contribution I-TEQ	50.45	43.55	0.005	0.399	0.212	0.050	4.15	1.18	5.38
Duck farm D	Contribution I-TEQ	50.46	43.97	0.005	0.399	0.212	0.046	3.84	1.07	4.96
Duck farm E	Contribution I-TEQ	50.46	45.57	0.005	0.399	0.212	0.032	2.69	0.63	3.35
Duck farm F	Contribution I-TEQ	50.46	45.98	0.005	0.399	0.212	0.028	2.38	0.53	2.94
Duck farm G	Contribution I-TEQ	50.48	48.42	0.005	0.399	0.212	0.005	0.39	0.09	0.49
Duck farm H	Contribution I-TEQ	50.48	48.80	0.005	0.399	0.212	0.001	0.08	0.02	0.10

[a] The PCDD/F concentration from Li et al. [3].

feed decreased, while those from original soil and stack flue gas increased. The worst case occurred at duck farm A in the sixth scenario (Table 11), where 47.81% of the PCDD/Fs came from the original soil, 44.92% from feed and 6.58% from stack flue gas. Notably, because duck farms G and H were upstream of the stack flue gas, the PCDD/F contributions from it were lower than 1.00% in all scenarios. In contrast, duck farms A and B were downstream of the stack flue gas, the PCDD/F contributions were higher than duck farms G and H. Particularly, the highest PCDD/F contributions were 6.58 and 6.52% for duck farms A and B in sixth scenario (Table 11), which were close to 10.5% [30]. If we considered the different uncertainty factors, such as flow rate variation of stack flue gas, errors from modelling and measurement, the PCDD/F contribution fraction from the stack flue gas may increase up to twice as that for the worst case (duck farm A in Table 11, 6.58%), 13.2%, which was still much lower than that from the total contribution (86.8%) of both feed and original soil. This result indicated that the PCDD/F contribution from the stack flue gas of EAFDT plant was minor, and the major source of PCDD/F intake for ducks is attributed to the feed and the original soil.

As can be seen in Tables 6–11, there were trends occurred in these tables. The PCDD/F contributions of EAFDT plant decreased from A to H for each scenario. For instance, at the scenario of 200 g feed and 5 g soil (Table 11), they were 6.58, 6.52, 5.97, 5.50, 3.72, 3.26, 0.54, and 0.11%. On the other hand, they increased with increasing soil fed for specified duck farm. For instance, for duck farm A, they were 0.112, 2.42, 3.98, 5.08, 5.93, and 6.58% when the soil varied from 0 to 5 g. According to these results, the maximum contribution of EAFDT plant on the duck total PCDD/F daily intake occurred at farm A with 5 g soil (200 g feed). Notably, there

Table 11
The PCDD/F contribution from feed, soil, ambient air and stack flue gas for eight duck farms (200 g feed and 5 g soil)

Media Exposure		Feed	Soil	Ambient air			Stack flue gas[a]			Contribution fraction by stack flue gas (%)
				Dry deposition		Wet deposition	Dry deposition		Wet deposition	
		Feed (%)	Original soil (%)	Feed farm (%)	Soil (%)	Soil (%)	Feed farm (%)	Soil (%)	Soil (%)	
Duck farm A	Contribution I-TEQ	44.92	47.81	0.004	0.443	0.236	0.051	5.29	1.24	6.58
Duck farm B	Contribution I-TEQ	44.90	47.90	0.004	0.444	0.236	0.048	5.02	1.45	6.52
Duck farm C	Contribution I-TEQ	44.90	48.45	0.004	0.444	0.236	0.044	4.62	1.31	5.97
Duck farm D	Contribution I-TEQ	44.90	48.91	0.004	0.444	0.236	0.041	4.27	1.19	5.50
Duck farm E	Contribution I-TEQ	44.91	50.68	0.004	0.444	0.236	0.029	2.99	0.70	3.72
Duck farm F	Contribution I-TEQ	44.91	51.15	0.004	0.444	0.236	0.025	2.65	0.58	3.26
Duck farm G	Contribution I-TEQ	44.92	53.86	0.004	0.444	0.236	0.004	0.44	0.10	0.54
Duck farm H	Contribution I-TEQ	44.92	54.29	0.004	0.444	0.236	0.001	0.08	0.03	0.11

[a] The PCDD/F concentration from Li et al. [3].

was a shelter constructed over the feed area, so no rain with PCDD/Fs reached the feed. Therefore, no data about wet deposition of PCDD/Fs to the feed appeared in Tables 6–11.

As previously described, the contribution of PCDD/Fs from the EAFDT plant on each farm was quite different. For each scenario, they decreased from A to H owing to $D_{\text{stack flue gas}}$ (feed), $D_{\text{stack flue gas}}$ (soil) and $W_{\text{stack flue gas}}$ (soil) were all decreased from duck farms A–H. The above results also claim the contribution of PCDD/Fs from the EAFDT plant on each farm and distance between EAFDT plant and each farm are adversely correlated. As shown in Fig. 1, the distance increased from duck farms A–H and the contribution decreased accordingly. As mentioned previously, duck farms G and H were in the upwind direction of EAFDT plant and then had much lower contributions.

Regarding the impacts of PCDD/Fs from EAFDT plant and ambient air on the soils, duck farms A and H were selected to compare the PCDD/F contents in the soils before and after 1815-days operation of EAFDT plant owing to the maximum and minimum dry and wet depositions occurred at duck farms A and H, respectively (Table 5). Additionally, discussion was focused on the scenario of 200 g feed and 1 g soil (Table 7). Via sampling and analyzing, both of the PCDD/F contents in the soils at duck farms A and H were 10.3 ng I-TEQ/kg after 1815-days operation of EAFDT plant. By subtracting the PCDD/F masses contributed from dry and wet depositions, the original PCDD/F contents in the soils at duck farms A and H were then be calculated as 8.95 and 10.15 ng I-TEQ/kg, respectively. The ratios of PCDD/F contents in the original soil and the soil after 1815-days operation of EAFDT plant were represented by duck farms A and H with the results 1.15 (10.3/8.95) and 1.01 (10.3/10.15), respectively. These results indicated that the increase of PCDD/F contents in the soil from EAFDT plant and ambient air were limited, meaning that the impacts of PCDD/Fs of EAFDT plant and ambient air on the soils were minor. Furthermore, EAFDT plant could not be the main PCDD/F source of the "Toxic Egg Event".

As shown in Table 11, the PCDD/F contributions of EAFDT plant on duck farms E and H were 3.72 and 0.11%, respectively, and they were much lower among these eight farms. Nevertheless, the mean PCDD/F contents in the toxic eggs from duck farms E and H were 8.58 and 7.83 pg WHO–TEQ $_{\text{DF}}$/g fat, respectively. Compared with other duck farms (except duck farm A), they were significantly higher. These results indicated that higher PCDD/F contributions of EAFDT plant were not necessarily resulting in the higher PCDD/F contents in eggs for these duck farms. Fly ashes contained purposely in duck feed by the farmers was a potential major source for the duck daily intake. Furthermore, more attention should be paid to the PCDD/F content in both feed and soil, which was contaminated by illegal fly ash landfills.

4. Conclusions

The mean PCDD/F I-TEQ content in the soil, feed, and ambient air were 10.3 ng I-TEQ/kg, 0.210 ng I-TEQ/kg and 0.0474 pg I-TEQ/Nm^3, respectively. The PCDD/F deposition from ambient air was 16.18 ng I-TEQ/m^2 (dry deposition) and 8.61 ng I-TEQ/m^2 (wet deposition). Of all the duck farms, A and B were the closest sites to the EAFDT plant and also located at downstream of the stack flue gas. The dry deposition (183.31–193.15 ng I-TEQ/m^2) and wet deposition (45.16–52.75 ng I-TEQ/m^2) of PCDD/Fs were much higher for A and B than for G and H (3.15–16.04 ng I-TEQ/m^2 (dry deposition), 0.94–3.61 ng I-TEQ/m^2 (wet deposition)), which were located upstream of the stack flue gas. After different scenario simulation, the worst case was at farm A with 200 g feed and 5 g soil for duck intake, and the highest PCDD/F contributions from the feed, original soil and stack flue gas were 44.92, 47.81, and 6.58% respectively. Considering different uncertainty factors, such as flow rate variation of stack flue gas, errors from modelling and measurement, PCDD/Fs from the stack flue gas of EAFDT plant in a worst case scenario may increase up to twice the modelled amount (6.58%) and become 13.2%, which was still much lower than that from the total contribution fraction (86.8%) of both feed and original soil. Adding fly ashes into the duck feed by the farmers has the strong tendency to be major source for the duck daily intake. Therefore, the PCDD/F content in the feed and soil which was contaminated by illegal fly ash landfills should be paid more attention to.

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