

TRACING THE HAZARDS – THE SUBSTANCE FLOW ANALYSIS OF DIOXINS

Hwong-Wen Ma,* Ming-Lung Hung, Huie-Lun Hung and Chia-Wei Chao

Graduate Institute of Environmental Engineering
National Taiwan University
Taipei 106, Taiwan

Key Words: Dioxins, substance flow analysis, CalTOX, uncertainty analysis

ABSTRACT

Dioxins have caused numerous toxic and pollution incidents in Taiwan, where the management of dioxins has been focused mainly on the establishment of emission standards and inventory. The currently applied approach features source reduction and monitoring of annual emission trend of dioxins, but the comprehensiveness of management still needs to be enhanced, in addition to the insufficient understanding about the distribution of dioxins in the environment. This study employed the material flow analysis to simulate overall distribution of dioxins in Taiwan and incorporated the uncertainty analysis to take the uncertainty of each parameter into account, so as to improve the precision of estimation on dioxin distribution. In addition, CalTOX multi-media model was utilized to assess the distribution of dioxins emitted from the economical activities into the environment.

The research findings revealed that in 2003, dioxins emission from the anthroposphere in Taiwan was about 2480 g I-TEQ. By average, about 10.3% was released to the atmosphere, 88.6% to waste, 0.9% to soil, and others to water body. The industrial emission was the most significant, with municipal waste incinerator taking up the leading position of about 87%, followed by arc furnace. Moreover, emitted dioxins would undergo transport and distribution in the environment and mostly reside in the soil media. The management strategies should be expanded to consider the dioxins from various sources and in various environmental media in a more effective and efficient way.

INTRODUCTION

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans or dioxins, was first discovered in flue gas and fly ashes from general waste incinerator in 1977. The severe nature of this issue immediately gained international recognition. Dioxin is listed as one of Persistent Organic Pollutants under the Stockholm Convention.

With the rapid growth of industries, Taiwan has experienced many poisoning and polluting incidents caused by dioxins. In 1983, dioxin emission was generated by open burning of waste electrical wires and cables in Wan-li area of Tainan. The average amount of 2,3,7,8-TCDD in fly ash reached as high as 0.013 $\mu\text{g m}^{-3}$, causing permanent contamination in nearby soil and water resources. Moreover, in An-nan area of Tainan City, Anshun Plant of Taiwan Alkali Industrial Corp released toxic matters such as pentachlorophenol and dioxins. Not only fishes and shrimps were polluted but also blood dioxin levels of local residents

were found four times higher than habitants in adjacency to municipal incinerators. A significantly higher incident of cancer was also observed. At the end of 2004, ducks and eggs in Hsien-shi township of Chang-hua County were polluted by dioxins, causing severe damages to duck raisers. Taiwan's attitude toward dioxin management has been passive and incomprehensive with the focus only put on emission sources. Usually announced in response to the disclosure of dioxin-related incidents, control standards are unable to cope with the eventual distribution of dioxins in the environment, not to mention proactive prevention and management. Therefore, it is urgent to systematically analyze source conditions and dioxin distributions so as to develop management solutions.

Substance flow analysis (SFA) is used to characterize substance stocks and flows within and between the economy and the environment for a certain time period in a certain region [1-2]. SFA is usually used to evaluate the environmental burdens at different stages related to life cycle of specific substances. Many SFA

*Corresponding author
Email: hwma@ntu.edu.tw

have been carried out for various metals, including copper [3-7], cadmium [8-10], zinc [11-14], silver [15-16], and lead [11].

So far, several studies have been conducted on the material flow of dioxins. Two surveys on dioxin emissions from local industries of Denmark have been conducted in 1997 and later during the period between 2000 and 2002 [17]. The calculation was performed by assessing the emission factor and activity intensity of each industry. Finally, a comprehensive diagram was established to illustrate annual production of dioxins from individual industries, amount of recycling, and eventual residual. The amounts of dioxins emitted to atmosphere, soil, and water body by each industry were specified as well. The accomplished diagram facilitates the understanding about major emission sources and environmental media of dioxins in Denmark. Also, in the Tarragona province of Spain, a material flow analysis was conducted with risk evaluation [18]. A total of 11 subsystems were defined, including: environmental media (air, soil, sediments, etc.), materials, food chain (animals, fishes, and vegetables), and eventually, the human body. Based on 88 dioxins distribution patterns, the amount of dioxins accumulated in Tarragona province was estimated to fall between 62.6-159.5 g I-TEQ y^{-1} , mostly kept in sediments (27.9-74.6 g I-TEQ y^{-1}) and soil (35-80.8 g I-TEQ y^{-1}). Finally, health risk was estimated by identifying potential exposure to dioxins of human body.

In order to fulfill "integrated pollution prevention and control", it is necessary to conduct estimation on Taiwan's annual emission and distribution patterns to control the impact of dioxins on the ecosystem and human health. This requires the establishment of an overall dioxins material flow database. Besides the integration of emission sources in the economy via material flow analysis, multi-media model was also adopted to extend the scope to distribution and flow among environmental media. The purpose was to reveal the ultimate possible distribution in the environment and locate the media that contain high level of dioxins. Uncertainty analysis was also incorporated to enhance the credibility of results obtained from material flow analysis, so as to provide guidance on dioxins management in Taiwan.

DIOXINS IN THE ENVIRONMENT

Dioxins are not deliberately produced but are byproducts of many industrial activities and manufacturing processes aimed to enhance the convenience of our daily life. Dioxins can be derived from natural causes (such as volcanic eruption and forest fire), spin-offs of industrial material processing (such as chlorinated phenols), and burning as part of operations in specific industries. US Environmental Protection Agency [19] divided the already known and potential sources of dioxins into five categories: emission from

combustion (waste incineration, energy/power generation, other high-temperature sources, and minimally controlled or uncontrolled combustion), metals smelting, refining, and processing (ferrous metal smelting/refining, nonferrous metal smelting/refining, scrap electric wire recovery, drum and barrel reclamation), chemical manufacturing, natural sources and processes, and reservoirs.

Emitted from industrial processes and high temperature burning, most dioxins would enter the environment in the form of particles as pollutant transmitted among multiple media. In particular, gas and particles can be carried by the atmosphere to long distances. The distribution in each medium is determined by parameters of transport and transformation.

When emitted to the atmosphere, dioxins can not only be transported via air but also directly undergo deposition processes to reach and contaminate soil. Via wet deposition, they can also enter water body. Due to extremely low water-solubility, they can be easily absorbed by the sediment, rather than causing pollution to the water sources. After deposition to soil, dioxins can either be absorbed by plants or consumed by animals. Since dioxins are lipid soluble, they accumulate in body fat instead of being expelled. Moreover, dioxins in the water body can also be absorbed into bio-systems and eventually accumulate in the body of humans, which are located in the highest hierarchy of food chain.

In the atmosphere, dioxins can either be transported in the form of gas or absorbed in particles. However, due to low vapor pressure, dioxins are less likely to transform into gases than adhere to particles. Hence, they can stay longer in the atmosphere, spread via aerologic movements, and finally undergo dry and wet depositions to reach the ground and accumulate in soil, vegetation and water body. Combined with soil, dioxins can achieve long-term settlement with a half-life of about 25 to 100 years at a considerably slow pace of movement. Due to its lipid solubility, dioxins in soil are unlikely to enter ground water but can be dissolved in organic solvents (such as benzene and methylbenzene), or even carried by organic solvents to long distances. In water body, dioxins are mostly located in the sediments of lakes and rivers. In addition to the leakage or direction emission from factories (such as paper mill), dioxins would mostly enter water body via atmospheric deposition rather than evaporate into the atmosphere again.

International Agency for Research on Cancer has confirmed that dioxins may cause cancer; lesions in the liver, lungs, stomach, and the lymphatic system are all associated with dioxins. With its chemical structure similar to human hormones, dioxins may be mistaken by human body as hormone and thus cause hormone-like effects, interfering endocrine system and normal operation of the body, especially the reproductive function. Kimbrough [20] conducted an

animal test to verify the reproductive toxicity of dioxins, which may cause unwanted abortion, infertility, mutation, and decreased male hormone.

In addition to the impact on reproductive system, disturbed balance of hormone would further cause more toxic responses in cases of damaged physical function. It has been confirmed in a study by Puga et al. [21] that dioxins are powerful tumor promoter, especially for lungs and the skin. In addition to skin pigmentation and inflammation, chloroacne can also be incurred with hair follicles stimulated. Liver is the most sensitive organ of human body; dysfunction and hepatomegaly may occur from an exposure to dioxins. It was also observed in animal tests that, when exposed to dioxins, adult animals would stop gaining weight; exposure to a lethal or higher dose would cause drastic decrease in body weight [19]. According to World Health Organization, recommended daily uptake of dioxins is 1–4 pg kg⁻¹-body weight.

METHODOLOGY

By characterizing substance flow in the economy and between human and environmental systems, SFA can identify important sources of hazardous substances released to waste streams and eventually, the environment. SFA frameworks have been defined in the literature [2,22–23], and IN = OUT is a simple statement of the central paradigm of SFA [24]. The fundamental mass balance equation used in a system defined by temporal and spatial boundaries is as follows.

$$\text{Import} + \text{Production} + \text{Recycling} \\ = \text{Export} + \text{Accumulation} + \text{Consumption} \quad (1)$$

Dioxins are not natural resources but byproducts of human's industrial activities. Hence, life cycle of dioxins differ from those of other frequently studied matters, such as cadmium, lead, and copper. Material flow analysis can still be employed to examine dioxins; only that the source of emissions should be set as industrial activities. The focus would be put on the analysis of the major sources of emissions. Also, the environment can be incorporated into the analysis to understand the distribution of dioxins among environmental media. The research procedure can be stated as follows:

1. Scope

In this study, the spatial boundary concerning the emission and storage was set with local industry, trading activities, local livestock and poultry breeds, and environmental media. The temporal boundary was set in the year of 2003.

2. Material Flow Analysis of Dioxins

On the basis of dioxins life cycle, the pathways

linking the economy with the environment would be sketched, so as to facilitate data collection concerning industrial emissions. Analysis on the life cycle of dioxins would focus on the relation between environmental media and emissions from burning in the manufacturing (such as sintering and arc furnaces), operation of products (cigarette smoking and burning of fuel), and waste treatment (such as incinerator and burning of waste tires). Also, trading activities should be examined because distribution of dioxins is associated with transportation of certain commodities. Hence, the investigation into dioxins material flow would clearly connect industrial production, product utilization, and waste treatment with environmental media.

3. Database Establishment for Dioxins Material Flow

Dioxins material flow analysis of the economy consists of three parts: emissions from Taiwan local industries, amount of dioxins contained in commodities involved in trading activities, and dioxins carried in food locally produced in Taiwan. They are detailed as follows:

3.1 Dioxins produced in Taiwan's local industrial activities

With reference to the classification by US Environmental Protection Agency [19], Taiwan's local industries that are involved in the production of dioxins, excluding biological and photochemistry processing, are listed in Table 1.

Due to properties of dioxins, predominant industrial discharges are air emissions, and only a little amount would be attached to waste (e.g. bottom ash and fly ash) or be infused into water body. However, whether in cases of atmosphere, waste, or water body, the estimation on dioxins emission in this study was based on emission factor analysis, which can be stated as follows:

$$\text{Dioxins emission} = \text{emission factor} \times \\ \text{activity intensity} \quad (2)$$

The emission factor refers to the amount of dioxins related to per-unit production (or energy consumption and waste disposal). The data concerning emission factors were based on Taiwan's local records between 2000 and 2006; the missing figures were replaced with emission factors of similar industries in foreign countries. Further, the activity intensity refers to the production (or energy consumption and waste disposal) of each industry in 2003 by the unit of ton.

(1) Air emissions

In general, complete test reports associated with dioxin emissions to the atmosphere were available. The only exception was the open burning of agricultural waste. To amend the lack of data related to the amount of agricultural waste burned in 2003, reason-

Table 1. Data collected to evaluate the substance flow analysis of dioxins

Emission Source Category			Air	Water	Waste	Emission Source Category			Air	Water	Waste
COMBUSTION SOURCES						5. Minimally Controlled or Uncontrolled Combustion					
1. Waste Incineration						1. Landfill fires					
1. Municipal waste incineration			✓		✓				✓	✓	
2. Small-scale municipal waste incineration			✓		✓				✓		
3. Hazardous waste incineration			✓						✓		
4. Medical waste/pathological incineration			✓	?	✓				✓		
5. Industrial waste incineration			✓		✓				✓		
6. Crematoria			✓								
7. Tire combustion			✓								
2. Power/Energy Generation						METAL SMELTING/REFINING					
1. Coal combustion –residential/industrial/utility			✓			1. Ferrous metal smelting/refining					
2. Oil combustion – residential/industrial/utility			✓			1. Sintering plants			✓		
3. Vehicle fuel combustion						2. Coke production			✓		?
1. leaded/unleaded			✓			3. Electric arc furnaces			✓		✓
2. diesel			✓			4. Ferrous foundries			✓		
4. Other High Temperature Sources						2. Nonferrous metal smelting/refining					
1. Cement kilns			✓			1. Secondary copper			✓		
2. Asphalt mixing plants			✓			2. Secondary aluminum			✓		?
3. Petro. refining catalyst regeneration			✓		?	3. Secondary lead			✓		?
4. Kraft recovery boilers			✓			4. Secondary Zinc			✓		
						3. Metal recycling					
						1. Fly ash recycling plants					
						✓					
						CHEMICAL MANUFACTURING					
						1. Ethylene dichloride/vinyl chloride					
						✓					

✓ displays the data is quantifiable

? displays the data is not complete and not be calculated

able assumptions would be required. The estimation on the open burning of agricultural waste was determined as follows:

$$\text{Amount of open burning} = \text{farming area (ha)} \times \text{fuel loading factor (t ha}^{-1}\text{)} \times \text{ratio of agricultural waste to fuel} \quad (3)$$

(2) Waste

Emitted in the form of waste are residuals from industrial operations. Examples include bottom ash and fly ash left after incineration and remaining bottom ash after steel smelting. Such wastes may contain dioxins and eventually arrive in landfills to contact soil media.

As for the collection of data involved with waste treatment, the following were amenable to calculation: large waste incinerator, middle-to-small scaled waste incinerator, medical and infectious waste incinerator, industrial waste incinerator, bottom ash and fly ash, and ash generated from arc furnace. Among them, the amounts of bottom ash and fly ash from middle-to-small scaled waste incinerators, medical and infectious waste incinerators, and industrial waste incinerators were estimated because of the shortage in official registers. Since the proportion between waste treated and bottom ash produced in large waste incinerators of 2003 was known, the amounts of waste treated in the same year by middle-to-small scaled waste incinerators, medical and infectious waste incinerators, and industrial waste incinerators can be used to estimate the amount of bottom ash with the same proportion.

The amount of fly ash was assessed by the same method.

(3) Water body

Sampling reports related to dioxin emissions to water body are given less attention in Taiwan. The Industrial Waste Control Center is the only organization to report on bottom ash and acid leakage from sludge involved with the secondary refining of lead. However, the corresponding emission factor is unavailable. Therefore, this study only took dioxins contained in landfill leachate into account. Calculation on the amount of leachate was based on the study by Camobreco et al. [25]:

$$CF_{lea} = \frac{\text{rain} \times UF \times (20\% \times 1.5 + 6.6\% \times 4 + 6.5\% \times 4.5 + 0.04\% \times 90)}{(\text{Dep} \times UF \times BD) \times 100} \quad (4)$$

where CF: potential amount of leachate produced by waste per unit (L t⁻¹); Rain: annual rainfall; Dep: depth of the landfill; and BD: density of the landfill (t m⁻³)

3.2 Potential amount of dioxins contained in commodities in trading activities

Dioxins may derive from burning of many commodities, such as leathers and textiles, but such case is very rare during the normal life cycle of these products. In most cases, dioxin emissions from burning only occur when a commodity is used and sent into waste incinerator. Repeated calculation, therefore, should be avoided when the amount of dioxins gener-

ated from incinerator operation is concerned. In terms of trading activities, the consideration should be given only to dioxins contained in foods, including meat (port, beef, mutton, chicken, duck, goose), seafood (freshwater fish, marine fish, shrimp, and shellfish), dairy (fresh milk, mild power, and other diary products), oils (animal fats and vegetable fats), and eggs (chicken egg, duck egg, goose egg, preserved egg, and quail egg). Imported food, whether consumed (absorbed into human fat) or not (kept in diet fat), would increase the amount of dioxin compounds in the economy.

The dioxins level of traded food products can be calculated by the follows:

$$\text{Dioxins level of traded food products} = \text{amount of lipid of imported and exported food} \times \text{concentration of dioxins in foods} \quad (5)$$

The amount of imported and exported food was based on records by the Directorate General of Customs, Ministry of Finance. Despite the sophisticated classification of food products related to amount of traded goods, further calculations were conducted to assess the weight of fat contained in food. The amount of dioxins carried by traded food products was based on testing reports by The Department of Health.

3.3 Dioxin level of locally produced food in Taiwan

Exposed to dioxins distributed in the living environment, stocks may contain toxic matters. Therefore, local food supply by stock raising and fishing in Taiwan should also be taken into account. The calculation is the same as that of trading activities.

3.4 Distribution of dioxins in the environment

At the completion of dioxin material flow database, emissions from anthroposphere to environmental media (air and soil) can be outlined. Also, the CalTOX multi-media model [26] was adopted to investigate dioxin transfer from air and soil media to other environmental media, depicting dioxin distribution characteristics in the environment. CalTOX model combines multimedia transport and transformation modeling of pollutants in the environment and multiple pathway exposure modeling. This model is chosen because it is suitable for modeling regional environmental transfer between environmental media. The environment is separated into several compartments, including air, plants, surface water, ground water, sediment, ground-surface soil, root-zone soil, and vadose-zone soil.

CalTOX multi-media model incorporated three types of parameters into consideration, including chemical properties, landscape properties, and human exposure factors. In this study, local data related to landscape properties and human exposures in Taiwan were used.

3.5 Uncertainty analysis

Due to the uncertain nature of dioxins emission factors collected, uncertainty is an important parameter to be included in the results of material flow analysis. In light of this, statistical analysis of local database was conducted to derive probability distributions quantitatively, while uncertainty of foreign or missing data was assessed by Data Quality Indicators qualitatively as proposed by Frischknecht et al. [27]. With data assumed to feature lognormal distribution, six indicators to data quality can be applied: reliability, completeness, temporal correlation, geographic correlation, further technological correlation, and sample size. Finally, uncertainty factors obtained from the indicator system can be applied into the equation, so as to figure out the standard deviation in each group of data.

$$SD_{g95} = \sigma_g^2 = \exp \sqrt{[\ln(U_1)]^2 + [\ln(U_2)]^2 + [\ln(U_3)]^2 + [\ln(U_4)]^2 + [\ln(U_5)]^2 + [\ln(U_6)]^2 + [\ln(U_b)]^2} \quad (6)$$

where U represents uncertainty factor and subscript 1 to 6 represent reliability, completeness, temporal correlation, geographic correlation, further technological correlation and sample size, respectively. U_b represents the basic uncertainty factor.

Here, the U_b was specified by primary conditions of input or output for each environmental medium [27]. For example, dioxin emission into atmosphere caused by burning can be set as 3.0. Data quality indicators were employed to develop the standard deviation, so that the uncertainty associated with missing data, such as the emission factor of medical waste incinerators, can be integrated with the uncertainty that can be computed statistically, which included all other emission factors, dioxins content in traded food commodities, and the exposure and landscape parameters used in CalTOX. In the study, the Monte Carlo method along with the SFA described above was used to combine individual probability distributions of model parameters to produce a probability distribution of dioxin generation estimation. 1000 sets of simulations were conducted. Along with the Monte Carlo simulation, sensitivity analysis method is used to identify important information, whose uncertainty is a driving factor in the overall uncertainty of the outcomes of SFA.

RESULTS AND DISCUSSION

1. Estimated Dioxins Flows in Taiwan

Taiwan's dioxins material flow of 2003 can be constructed with emissions from industries in the economy to air and soil media, the amount transferred from air and soil media to other media, and the

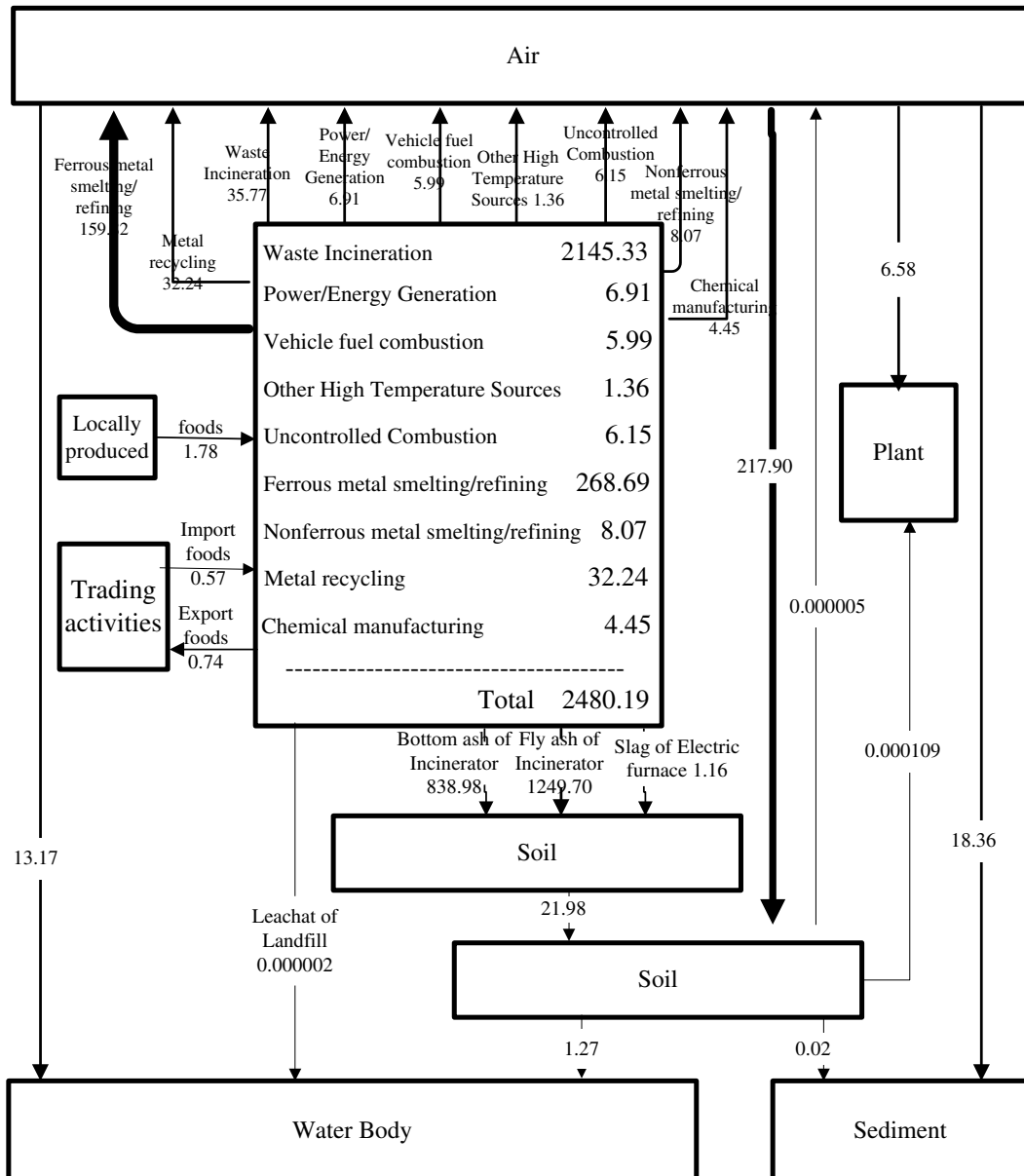


Fig. 1. Substance flow analysis of dioxins in Taiwan in 2003 (g I-TEQ y⁻¹).

amount contained in traded food products, local livestock, and poultry breeds. The average emissions of individual sources can be illustrated in Fig. 1.

It can be observed from Fig. 1 that, Taiwan's average dioxins emission from the economy in 2003 was 2480.2 g I-TEQ, in which 256.3 g I-TEQ was released to the atmosphere, 22.0 g I-TEQ to soil, 2197.5 g I-TEQ to waste, and the rest small amount to water body. In terms of category, the industrial emission was the most significant, with municipal waste incinerator taking up the leading position of 86.5%, followed by arc furnace (10.9%). Hence, it can be inferred that the major source of dioxins emission in Taiwan was municipal waste incinerator and arc furnace.

As for food, the average dioxin level of imported

food was 0.57 g I-TEQ, that of exported food was 0.74 g I-TEQ, and that of local livestock and poultry breeds was 1.78 g I-TEQ.

2. Dioxins Transfer in the Environment

When emitted to the environment, dioxins undergo multimedia transfer, in which each environmental medium exchange gains and losses till dynamic equilibrium is achieved.

Given the dioxin emission from the economy of 2003, the average amount transferred between environmental media can be as illustrated in Fig. 2. Hence, it can be inferred that, when steady state is achieved among environmental media, partial amount of dioxins would leave the system boundary Taiwan via at

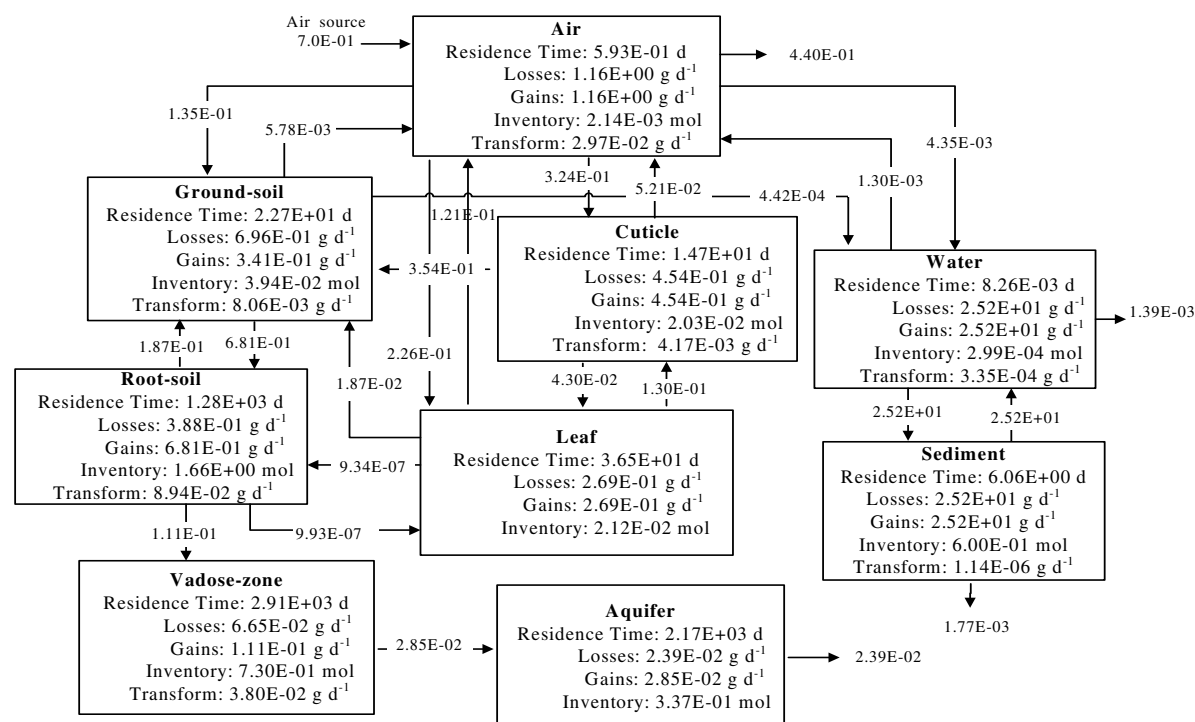
Fig. 2. Distribution of dioxins in environmental media (g I-TEQ d⁻¹).

Table 2. Contribution (in %) of dioxins emission into air from various pollution sources

Emission distribution pollution source	2.5%	5.0%	50.0%	95.0%	97.5%	Mean
Waste Incineration	3.1	3.3	5.0	17.1	23.3	13.7
Power/Energy Generation	1.6	1.8	3.0	2.6	2.3	2.7
Vehicle fuel combustion	0.7	0.9	2.0	2.9	2.9	2.3
Other High Temperature Sources	0.4	0.4	0.5	0.6	0.6	0.5
Uncontrolled Combustion	2.8	2.8	2.9	1.9	1.6	2.4
Ferrous metal smelting/refining	49.7	52.8	64.9	63.5	59.3	61.3
Nonferrous metal smelting/refining	2.2	2.6	3.7	2.7	2.6	3.1
Metal recycling	39.3	35.1	16.4	6.6	5.4	12.4
Chemical manufacturing	0.2	0.3	1.7	2.1	1.9	1.7

mosphere (0.44 g I-TEQ d⁻¹), water body (1.4×10⁻³ g I-TEQ d⁻¹), sediment (1.8×10⁻³ g I-TEQ d⁻¹), or groundwater (2.4×10⁻² g I-TEQ d⁻¹). In addition, dioxins can also decompose via transformation.

Dioxins emitted from the economy would eventually be allocated to the environmental media by the following approx proportions: 0.1% in the air, 1.0% in the leaf of vegetation, 1.0% in plant cuticle, 1.8% in surface soil, 52.7% in root-soil, 24.7% in deep soil, 0.01% in surface water, 5.5% in ground water, and 13.2% in the sediments. Hence it can be understood that dioxins would eventually locate in soil media, especially the root-soil.

3. Uncertainty Analysis

3.1 The economy

Emission sources in the economy include waste incineration, generation of electricity and energy, mobile sources, industries utilizing high temperature,

open burning, steel and metal smelting, smelting reduction of non-iron metal, precipitator ash recycling facilities, and manufacturing of vinyl chloride and other chemicals. It can be found from uncertainty analysis that, in 2003, the total amount of dioxins emitted from Taiwan's local industries to the atmosphere was about 98.5-628.9 g I-TEQ y⁻¹. The emission to soil was about 6.1-70.9 g I-TEQ y⁻¹, that of water body was 1.1×10⁻⁷-6.3×10⁻⁶ g I-TEQ y⁻¹, that contained in imported products was 0.29-0.88 g I-TEQ y⁻¹, that involved with exported products was 0.09-1.76 g I-TEQ y⁻¹, and that of local livestock and poultry breeds was 0.44-3.86 g I-TEQ y⁻¹.

Table 2 shows the ranking of dioxins emission into air by each emission source at various cumulative probability percentiles. The contribution patterns are generally quite similar for different percentiles. The contribution of steel and metal smelting surpassed the others by contributing over 50%, mainly because arc furnace, foundry plants, and sintering plants were

Table 3. Mass distribution (in %) of dioxins in environmental media under consideration of uncertainty analysis

Environmental media	Mass distribution (%)					
	Mean	2.5%	5.0%	50.0%	95.0%	97.5%
air	0.13	< 0.1	0.2	< 0.1	< 0.1	< 0.1
leaf	1.0	< 0.1	0.8	< 0.1	0.2	< 0.1
cuticle	1.0	0.2	0.2	0.2	0.9	< 0.1
ground-soil	1.8	0.3	0.8	0.8	0.8	0.6
root-soil	52.7	10.9	15.4	53.8	88.8	91.8
vadose-zone	24.7	36.3	35.6	7.5	7.1	5.2
surface water	< 0.1	< 0.1	< 0.1	< 0.1	0.00	0.00
sediment	13.2	36.7	30.3	37.4	2.0	0.3
aquifer	5.5	15.5	16.7	0.3	0.3	2.1

Table 4. Comparisons of people loading and land loading of different countries

Country	Year	Total amount of dioxins ($\mu\text{g I-TEQ yr}^{-1}$)	Population (person)	Area (km^2)	Population loading ($\mu\text{g I-TEQ population}^{-1}$)	Land loading ($\mu\text{g I-TEQ km}^{-2}$)	Data source
Taiwan	2003	min. 5.89E+08 max. 8.06E+09	22,689,100	35,570	26.0 378.8	16564 241594	this study
Denmark	2000- 2002	min. 7.20E+07 ave. 6.89E+08	5,430,000	43,094	13.3 126.9	1671 15988	[17]
China	2004	ave. 2.77E+09	1,299,880,000	9,600,000	2.1	289	[28]
U.S.	2000	ave. 1.35E+09	281,421,910	9,826,630	4.8	138	[19]
Japan	2000	min. 2.20E+09 max. 2.22E+09	126,974,630	377,835	17.3 17.5	5817 5870	[29]

taken into consideration. In particular, ferrous metal smelting/refining was the most contributive (61.3%), followed by waste incineration (13.7%) and metal recycling (12.4%). It can be observed that, at higher percentiles, contribution from waste incinerators becomes more significant, due to the influence of log-normal distribution of emission factors for large municipal waste and industrial waste incinerators.

3.2 The environment

Distribution of dioxins among environmental media is shown in Table 3. The average amount of dioxins in soil media was 77.4%, of which up to 52.7% was kept in the root-soil (10.9-91.8%), followed by vadose-zone of 24.7% (5.2-36.3%) and sediments of 13.2% (0.27-37.4%).

Major parameters related to the accumulation of dioxins in the environment can be assessed via sensitivity analysis. Such parameters can be categorized into three types: dioxin's physical and chemical characteristics, Taiwan's landscape parameter, and emission factors of local industries. Based on the sensitivity analysis on the accumulation of dioxins in each environmental medium, it was discovered that Taiwan's landscape parameter (such as depth of surface soil and soil disturbance rate) and dioxin's physical and chemical characteristics (such as dioxin's half-life dissipation in surface water), and emission factors of arc furnace have considerable influence on the estima-

tion of dioxins amount in the environment.

SUMMARY AND CONCLUSIONS

Since Taiwan's monitoring on dioxins focuses more on gas emission, data related to dioxins in soil and water body are insufficient. To assess soil pollution caused by dioxins, an examination was conducted on bottom ash and fly ash produced by large municipal waste incinerators, middle-to-small waste incinerators, medical and infectious waste incinerators, and industrial waste incinerators, as well as ash from arc furnaces. As for dioxin-polluted water body, landfill leachate was investigated. The other unavailable data were excluded from the calculation. Further information gathered in the future would enhance the credibility of evaluation results.

Because of variability of emissions, uncertainty of estimation parameters, and different degrees of data quality, uncertainty analysis was used to provide more completed information of dioxins flow. It was observed that in 2003 the leading contributor of dioxins emission was arc furnace, followed by industrial waste incinerator. Given strict monitoring and control over major emission sources such as arc furnace and sintering plants, the contribution of industrial waste incinerator and foundry plants would be highlighted. Hence, it is suggested that an adequate emission standard should be imposed on such sources, so as to facilitate the reduction and management of dioxins

emissions in Taiwan. The reduction of dioxin emissions should be achieved without causing transferring the problems between various environmental media.

A comparison was made with dioxins evaluation reports on other countries; the focus was placed on load per capita and soil load, as shown in Table 4. The obtained dioxin distribution was transformed into load per capita. Dioxin load per capita in Taiwan was about 26.0-378.8 μg I-TEQ; while that of Denmark was 13.3-126.9 μg I-TEQ, that of China was about 2.1 μg I-TEQ, that of the USA was about 5.1 μg I-TEQ, and that of Japan about 17.3 μg I-TEQ [29]. Dioxins distribution can also be transformed into soil load. Taiwan's soil load was about 16,560-241,590 μg I-TEQ km^{-2} ; that of Denmark ranged between 1,670 and 15,988 μg I-TEQ km^{-2} , that of the USA reached about 145 μg I-TEQ km^{-2} , that of the China reached about 289 μg I-TEQ km^{-2} , and that of Japan fell between 5,820-5,870 μg I-TEQ km^{-2} . The value of Taiwan's population and soil load is higher than that of China, the USA and Japan since the bottom ash and fly ash released to land was not considered in these countries. Whether released into the air or soil, almost 80% of total amounts of dioxins would eventually accumulate in soil. Due to the difficulty in soil pollution remediation, strategies should be made to facilitate monitoring and management of dioxins in soil, as well as regulation on and reduction from the sources.

REFERENCES

1. Guinee, J.B., J.C.J.M. van den Burgh, J. Boelens, P.J. Fraanje, G. Huppes, P.P.A.A.H. Kandelars, T.M. Lexmond, S.W. Moolenaar, A.A. Olsthoorn, H.A.U. de Haes, E. Verkuijlen and E. van der Voet, Evaluation of risks of metal flows and accumulation in economy and environment. *Ecol. Econ.*, 30(1), 47-65 (1999).
2. Brunner, P.H. and H. Rechberger, *Practical Handbook of Material Flow Analysis*. Lewis Publishers, Boca Raton, FL (2004).
3. Spatari, S., M. Bertram, K. Fuse, T.E. Graedel and H. Rechberger, The contemporary European copper cycle: 1 year stocks and flows. *Ecol. Econ.* 42(1-2), 27-42 (2002).
4. van Beers, D., M. Bertram, K. Fuse, S. Spatari, T.E. Graedel, The contemporary African copper cycle: One year stocks and flows. *J. S. Afr. I. Min. Metall.*, 103(3), 147-162 (2003).
5. Graedel, T.E., D. van Beers, M. Bertram, K. Fuse, R.B. Gordon, A. Gritsinin, A. Kapur, R.J. Klee, R.J. Lifset, L. Memon, H. Rechberger, S. Spatari and D. Vexler, Multilevel, Cycle of anthropogenic copper. *Environ. Sci. Technol.*, 38(4), 1242-1252 (2004).
6. Vexler, D., M. Bertram, A. Kapur, S. Spatar and T.E. Graedel, The contemporary Latin American and Caribbean copper cycle: 1 year stocks and flows. *Resour. Conserv. Recy.*, 41(1), 23-46 (2004).
7. Spatari, S., M. Bertram, R.B. Gordon, K. Henderson and T.E. Graedel, Twentieth century copper stocks and flows in north America: A dynamic analysis, *Ecol. Econ.*, 54(1), 37-51 (2005).
8. Van der voet, E., L. Vanegmond, R. Kleijn and G. Huppes, Cadmium in the European-community – A policy-oriented analysis. *Waste Manage. Res.*, 12(6), 507-526 (1994).
9. Lindqvist, A. and F. von Malmborg, What can we learn from local substance flow analyses? – The review of cadmium flows in Swedish municipalities. *J. Clean. Prod.*, 12(8-10), 909-918 (2004).
10. Hawkins, T.R., H.S. Matthews and C. Hendrickson, Closing the loop on cadmium – An assessment of the material cycle of cadmium in the US. *Int. J. Life Cycle Ass.*, 11(1), 38-48 (2006).
11. Palm, V. and C. Ostlund, Lead and zinc flows from technosphere to biosphere in a city region. *Sci. Total Environ.*, 192(1), 95-109 (1996).
12. Spatari, S., M. Bertram, K. Fuse, T.E. Graedel and E. Shelov, The contemporary European zinc cycle: 1-year stocks and flows. *Resour. Conserv. Recy.*, 39(2), 137-160 (2003).
13. Graedel, T.E., M. Bertram and B. Reck, Exploratory data analysis of the multilevel anthropogenic zinc cycle. *J. Ind. Ecol.*, 9(3), 91-108 (2005).
14. Graedel, T.E., D. van Beers, M. Bertram, K. Fuse, R.B. Gordon, A. Gritsinin, E.M. Harper, A. Kapur, R.J. Klee, R. Lifset, L. Memon and S. Spatari, The multilevel cycle of anthropogenic zinc. *J. Ind. Ecol.*, 9(3), 67-90 (2005).
15. Johnson, J., J. Jirikowic, M. Bertram, D. van Beers, R.B. Gordon, K. Henderson, R.J. Klee, T. Lanzano, R. Lifset, L. Oetjen and T.E. Graedel, Contemporary anthropogenic silver cycle: A multilevel analysis. *Environ. Sci. Technol.*, 39(12), 4655-4665 (2005).
16. Lanzano, T., M. Bertram, M. De Palo, C. Wagner, K. Zyla and T.E. Graedel, The contemporary European silver cycle. *Resour. Conserv. Recy.*, 46(1), 27-43 (2006).
17. Danish Environmental Protection Agency, *Substance Flow Analysis for Dioxin 2002*. Kobenhavn, Denmark (2003)
18. Fuster, G., M. Schuhmacher and J.L. Domingo, Human exposure to dioxins and furans – Application of the substance flow analysis to

- health risk assessment. *Environ. Sci. Pollut. Res.*, 9(4), 241-249 (2002).
19. U.S. Environmental Protection Agency (USEPA), An Inventory of Sources and Environmental Releases of Dioxin-Like Compounds in the United States for the Years 1987, 1995, and 2000 (2006).
 20. Kimbrough, R.D., Polychlorinated-biphenyls (PCBs) and human health – An update. *Crit. Rev. Toxicol.*, 25(2), 133-163 (1995).
 21. Puga, A., A. Maier and M. Medvedovic, The transcriptional signature of dioxin in human hepatoma HepG2 cells. *Biochem. Pharmacol.*, 60(8), 1129-1142 (2000).
 22. Baccini, P. and P.H. Brunner, *Metabolism of the Anthroposphere*. Springer Verlag, Berlin, Germany (1991).
 23. van der Voet, E., L. van Oers, J.B. Guinee and H.A.U. de Haes, Using SFA indicators to support environmental policy. *Environ. Sci. Pollut. Res.*, 6(1), 49-58 (1999).
 24. Kleijn, R., IN = OUT: The trivial central paradigm of MFA? *J. Ind. Ecol.*, 3(2-3), 8-10 (1999).
 25. Camobreco, V., R. Ham, M. Barlaz, E. Repa, M. Felker, C. Rousseau and J. Rathle, Life-cycle inventory of a modern municipal solid waste landfill. *Waste Manage. Res.*, 17(6), 394-408 (1999).
 26. McKone, T.E. and K.G. Enoch, CalTOX (registered trademark), A Multimedia Total Exposure Model Spreadsheet User's Guide Version 4.0 (Beta). Paper LBNL-47399, Lawrence Berkeley National Laboratory, Berkeley, CA (2002).
 27. Frischknecht, R., N. Jungbluth, H.J. Althaus, G. Doka, R. Dones, T. Heck, S. Hellweg, R. Hischier, T. Nemecek, G. Rebitzer and M. Spielmann, The ecoinvent database: Overview and methodological framework. *Int. J. Life Cycle Ass.*, 10(1), 3-9 (2005).
 28. Jun J., P. Hao, X. Tang, An inventory of potential PCDD and PCDF emission sources in the mainland of China. *Organohalogen Comp.*, 66, 852-858 (2004).
 29. Japan Ministry of the Environment, The Environmental Monitoring Report on the Persistent Organic Pollutants (POPs) in Japan, Tokyo, Japan (2002).
-
- Discussions of this paper may appear in the discussion section of a future issue. All discussions should be submitted to the Editor-in-Chief within six months of publication.
- Manuscript Received: July 7, 2007**
Revision Received: December 13, 2007
and Accepted: December 20, 2007