Comparison of PAH Emission from a Municipal Waste Incinerator and Mobile Sources

Hsiao-Hsuan Mi[1] *, Chow-Feng Chiang[2], Ching-Cheng Lai[3], Lin-Chi Wang[4], Hsi-Hsien Yang[2]

[1] *Department of Environmental Engineering and Health, Chia-Nan University of Pharmacy and Science, Tainan, Taiwan,*

[2] *Department of environmental Management, Chao-Yang University of Technology, Taichung, Taiwan,*

[3] *Department of Applied Chemistry, Providence University, Taichung, Taiwan,*

[4] *Department of Environmental Engineering, National Cheng Kung University, Tainan, Taiwan.*

PAH emissions from a municipal waste incinerator stack in central Taiwan were sampled and analyzed by gas phase and particulate phase samples. A modified Gaussian atmospheric dispersion model, industrial sources complex (ISCST3), was used to simulate the PAH concentration in ambient air, which was compared with the measured PAH concentration in the ambient air. Total-PAH emission factor for the municipal waste incinerator ranged between 748 and 992 mg/ton-waste and averaged 871 mg/ton-waste, while the total-PAH emission rate ranged between 21,000 and 27,900 mg/hr and averaged 24,500 mg/hr. The results of dispersion modeling indicated that the contribution of PAHs to the ambient air by the municipal waste incinerator only reached 0.98% of the background concentration. Consequently, other emission sources, especially mobile sources, impact air quality more significantly.

Keywords: PAHs, municipal waste incinerator, dispersion model, mobile sources

1. Introduction

Various investigations have recently been conducted to measure selected organic components in the emissions of municipal waste incinerator (MWI) in the past years [Colmsjo et al., 1986; Oehme et al., 1987; Yasuda et al., 1989; Li et al., 1995], and to assess the effect of these emission on human health [Zemba et al., 1996; Basham et al., 1999]. Polycyclic aromatic hydrocarbons (PAHs) and their derivatives are harmful compounds that are frequently generated by incomplete combustion, mostly from emissions cause by factitious activities. Airborne concentrations of PAH in Taiwan generally range from <200 ng/m^3 in rural areas, to 300-3,000 ng/m^3 in urban areas, and > 4,000 ng/m^3 in certain occupational settings [Mi, 1998]. However, PAHs emissions from the stack of MWI, exceed other industrial incinerators owing to their large treatment loading. Understanding the contribution of MWI to PAH emissions will help clarify the relationship between pollution sources and ambient air quality.

This study investigates the effect of using

*Corresponding author:
Tel.: +886-6-2660313
Fax: +886-6-2667323
E-mail address: mihh@mail.chna.edu.tw

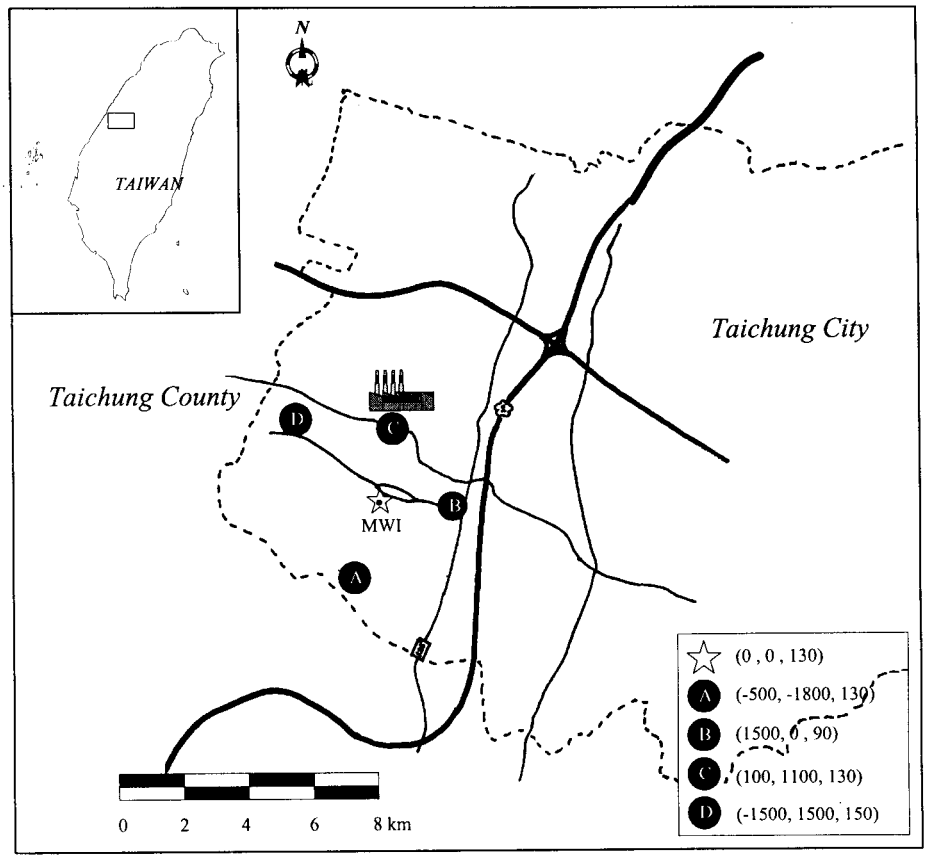

Figure 1. The locations of atmospheric sampling sites and the municipal waste incinerator in central Taiwan.

actual MWI stack emission, meteorological and receptors data on ambient air PAH-concentration, in predicting the contribution of emission from the MWI stack on ambient air PAH level.

2. Experimental Equipment and Method

2.1 Municipal Incinerator Stack

The target stack of municipal waste incinerator located in central Taiwan is with a maximum treatment loading of 900 ton/day. This MWI contains three equivalent stacks and the total flue gas flow rate after waste burning is 4.32 million m^3/day. The height and the diameter of the stacks are 120 m and 1.5 m, respectively. Meanwhile, the mean velocity of the flue gas in the stack is 15 m/s, and its average temperature is 185 °C. Twelve PAH samples were collected from these stacks and analyzed in statistical need of duplication experiments.

2.2 ISCST3 Model

This study aimed to identify and quantify PAHs emitted from a MWI and to examine the correlation with ambient air quality around MWI by a Gaussian plume model, Industrial Source Complex short-term, ISCST3, which was used to compute ambient air concentrations at specified receptor points. The stack PAH measurement was executed in 1996 and 1997, and was combined with hourly meteorological data obtained from the Central Weather Bureau in Taiwan for 1995-1997 inclusive. The dispersion parameters such as atmospheric stability and mixing height, were severally set to suitable stable class and

under 350 m high in ISCST3 model. The daily wind speed was found to vary from breezeless to 21.4 m/s and annual average wind speed was 1.49 m/s.

Land use within a 3.85 km radius of the MWI indicated that the ISCST3 "rural" modeling algorithm was suitable for this study. A 21 × 21 point grid with a resolution of 273 m between points was constructed, and gave receptor grid coverage of 5.5 km × 5.5 km and containing 441 receptor points. The four specific receptor points (X, Y, Z) relative to the location of MWI (0, 0, 130) is A point (-500, -1800, 130), B point (1500, 0, 90), C point (100, 1100, 130) and D point (-1500, 1500, 150) in meter scale (Fig. 1). The hilly terrain data is above the sea level and it was used to estimate PAH ground level concentration (Z coordinate: receptor height) in this study. However, for the selection of sampling locations, point B was chosen because it is close to a major highway and for point C, the effect of both traffic and industrial sources was considered. For the other two locations, points A and D were selected for their up- and downwind position in that area.

In this study, an important assumption made for the emitted PAHs in dispersion model developed is that PAHs will not decay during the dispersed and settling process from MWI stacks once to the ground. This reasonable condition is set to simulate the maximum ground concentration and to assess the PAHs contribution from the MWI.

2.3 PAH Sampling System for Stack Flue Gas

Graseby's modification of the USEPA's sampling method (MM5) (40CRF60) was adapted for the sampling. The flue gas was isokinetically sampled from the stack using the PAH sampling system, which was equipped with a sampling probe, a cooling device, a glass cartridge, a pump, a flow meter and a control computer. A PAH sampling system with a tube-type glass fiber filter (cleaned by heating to 450°C for 8 hours) was used to collect particulate and particle-phase PAHs. Meanwhile a glass cartridge packed with XAD-2 resin and supported by a polyurethane foam (PUF) plug was used to collect the gas phase PAHs. After each sampling cycle the sampling train was rinsed with *n*-hexane. Breakthrough tests were investigated by three stages of XAD-2/PUF cartridge [Yang et al., 1998], each of which was individually analyzed and compared in terms of the PAH mass collected. Breakthrough tests results revealed that no significant PAH mass was collected in the cartridge of the third stage. All the experiments were repeated at least three times to ensure that the results were reproducible.

2.4 PAH Sampling from Ambient Air

To compare the modeling results with ambient air in PAH contribution, both particle and gas phases of PAHs in the ambient air were collected from Nov. 1996 to June 1997 using a standard semi-volatile sampling train (General Metal Works PS-1). Four PS-1 sampler sets were simultaneously operated at four receptor points for 24 hours during the sampling periods to collect the PAHs. The sampler was placed more than 30 m away from the street to reduce the direct PAH contribution from traffic sources. Before each sampling, the PS-1 samplers with glass fiber filter and PUF/XAD-2 cartridge was pretreated following the same procedure as the sampling system for flue gas.

2.5 PAH Analysis

Each PAH-containing sample, including glass cartridge and the glass fiber filters, was separately Soxhlet extracted. The extract was then concentrated by purging with ultra-pure

nitrogen to 2 mL for the cleanup procedure, similar to previous studies (Mi et al., 1996; Sheu et al., 1997). The eluant collected from the cleanup procedure was then re-concentrated to 0.50 mL with ultra pure nitrogen (Mi et al., 1996; Sheu et al., 1997). A gas chromatograph (GC) (Hewlett-Packard 5890A) with a mass selective detector (MSD) (Hewlett-Packard 5972) and a computer workstation was used for the PAH analysis. This GC/MS was equipped with a Hewlett-Packard capillary column (HP Ultra 2 - 50 m x 0.32 mm x 0.17 μm), and PAHs was qualified by using the selected ion monitoring (SIM) mode. The method detection limitations of 21 individual PAHs were found between 0.023 and 0.524 ng.

The concentrations of the following 21 PAHs which were separated into three different molecular weight ranges: Low molecular weight (LMW) PAHs are 2 and 3 rings PAHs including naphthalene (Nap), acenaphthylene (AcPy), acenaphthene (Acp), fluorene (Flu), phenanthrene (PA), anthracene (Ant). Middle molecular weight (MMW) PAHs are 4 and 5 rings PAHs including fluoranthene (FL), pyrene (Pyr), cyclopenta (c,d)-pyrene(CYC), benz(a)anthracene(BaA), chrysene (CHR). Finally, high molecular weight (HMW) PAHs are 6 and 7 rings PAHs including benzo-(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(e)pyrene (BeP), benzo(a)pyrene (BaP), perylene (PER), indeno(1,2,3,-cd) pyrene (IND), dibenz(a,h)anthracene (DBA), benzo(b) chrycene (BbC), benzo(ghi)perylene (BghiP), and coronene (COR).

Table 1. PAH emission rate and emission factor of the municipal incinerator (n=12).

PAHs	PAH Emission Rate		PAH Emission Factor	
	Range (mg/hr)	Mean (mg/hr)	Range (mg/ton)	Mean (mg/ton)
Nap	18100 – 23700	20900	643 – 843	744
AcPy	56.1 – 134	90.2	2.00 – 4.77	3.21
Acp	32.9 – 187	116	1.17 – 6.65	4.13
Flu	160 – 415	266	5.70 – 14.8	9.45
Ant	1130 – 1840	1450	40.1 – 65.4	51.7
PA	39.5 – 57.9	49.2	1.41 – 2.06	1.75
FL	349 – 774	543	12.4 – 27.6	19.3
Pyr	152 – 330	231	5.42 – 11.8	8.22
CYC	6.24 – 13.9	9.99	0.22 – 0.49	0.36
BaA	13.1 – 24.6	20.0	0.47 – 0.87	0.71
CHR	28.8 – 44.4	37.0	1.02 – 1.58	1.32
BbF	53.5 – 299	181	1.90 – 10.7	6.45
BkF	20.6 – 42.2	30.8	0.73 – 1.50	1.09
BeP	15.6 – 32.4	23.3	0.52 – 1.15	0.83
BaP	53.3 – 84.0	69.9	1.90 – 2.99	2.49
PER	50.3 – 106	73.9	1.79 – 3.78	2.63
IND	44.7 – 82.0	60.5	1.59 – 2.92	2.15
DBA	70.9 – 130	102	2.52 – 4.62	3.64
BbC	48.6 – 144	89.0	1.73 – 5.13	3.17
BghiP	60.2 – 145	101	2.14 – 5.16	3.60
COR	0.89 – 29.4	12.8	0.03 – 1.05	0.46
Total PAHs	21000 – 27900	24500	748 – 992	871

Table 2. PAHs range and mean in the ambient air near the municipal incinerator

PAHs	A. (n=9)		B. (n=10)		C. (n=9)		D. (n=11)	
	Range (ng/m^3)	Mean (ng/m^3)	Range (ng/m^3)	Mean (ng/m^3)	Range (ng/m^3)	Mean (ng/m^3)	Range (ng/m^3)	Mean (ng/m^3)
Nap	208-750	451	98.7 - 580	353	162 - 1530	569	125 - 1140	345
AcPy	0.97 - 5.91	2.94	1.19 - 26.4	7.54	1.19 - 11.0	3.68	0.95 - 10.5	4.12
Acp	1.78 - 10.1	3.89	1.22 - 10.2	3.91	1.79 - 18.7	5.81	0.90 - 6.32	3.19
Flu	0.99 - 17.9	7.08	0.55 - 15.5	7.86	0.90 - 21.5	9.14	0.41 - 15.8	6.29
Ant	10.5 - 47.1	27.9	6.40 - 35.2	21.3	8.68 - 49.0	24.67	6.42 - 36.4	18.4
PA	1.24 - 9.16	3.38	0.90 - 6.70	3.20	1.04 - 6.98	3.59	0.67 - 2.99	1.85
FL	3.19 - 14.1	9.08	2.31 - 10.3	7.28	2.77 - 15.5	8.53	2.40 - 11.1	5.79
Pyr	4.30 - 16.2	8.16	2.24 - 9.98	7.12	2.50 - 17.0	8.69	2.12 - 10.7	5.07
CYC	0.27 - 3.23	1.41	0.35 - 2.89	1.04	0.46 - 4.27	1.76	0.26 - 1.74	0.81
CHR	0.70 - 14.1	4.36	0.78 - 8.81	3.50	ND - 9.98	3.87	0.63 - 3.57	1.87
BaA	ND - 5.77	2.75	ND - 13.1	4.02	ND - 18.5	5.25	ND - 6.34	3.25
BbF	0.69 - 7.89	4.78	0.72 - 15.7	5.80	1.76 - 15.9	6.03	1.31 - 5.31	3.14
BkF	ND - 8.47	1.83	ND - 6.63	1.23	ND - 4.46	1.12	ND - 2.97	0.73
BeP	0.54 - 5.09	3.11	0.64 - 17.3	7.22	0.72 - 20.4	6.38	0.78 - 5.69	2.88
BaP	0.64 - 8.80	3.71	0.64 - 42.1	10.3	0.72 - 34.0	9.27	0.31 - 9.19	3.95
PER	0.12 - 41.0	6.74	0.18 - 11.9	3.22	0.37 - 18.6	4.60	0.05 - 7.12	2.32
IND	1.19 - 27.5	10.3	0.21 - 74.5	24.8	0.87 - 57.7	18.3	1.79 - 64.8	14.8
DBA	0.28 - 40.1	9.50	0.92 - 86.2	19.3	1.46 - 62.2	17.5	ND - 28.6	10.3
BbC	ND - 1.04	0.42	ND - 9.25	1.83	ND - 4.73	0.85	ND - 1.00	0.41
BghiP	0.47 - 31.3	8.05	ND - 38.7	10.1	0.32 - 32.7	10.2	0.34 - 17.5	6.80
COR	ND - 1.56	0.52	ND - 19.4	4.19	ND - 16.9	3.33	ND - 3.01	0.83
Total PAHs	341 - 871	571	128 - 982	508	245 - 1940	722	145 - 1350	442

3. Results and Discussion

3.1 PAH Emission Factor

Table 1 lists the PAH emission rate and emission factor employed herein. By calculating both the volume concentration and the volume of exhaust per unit time (hr), the mean total PAH emission rate reached 24,500 mg/hr. The highest level among individual PAHs was 20,900 mg/hr of Nap, belonging to low molecular weight PAH. Others were under the level of 1.45 g/hr. Regarding bio-toxins, BaP (69.9 mg/hr), DBA (102 mg/hr) and BbF (181 mg/hr) have higher mean emission rates. Estimating PAH emission factor is made easier if the amount of treatment waste by the incinerator can be obtained. The treatment volume for this incinerator presently totals around 29 tons per hour. Consequently, the mean total PAH emission factor reaches 871 mg/ton-waste (Table 1). The highest level was 744 mg/ton-waste of Nap, followed by Ant (51.7 mg/ton-waste), FL (19.3 mg/ton-waste), Flu (9.45 mg/ton-waste) and Pyr (8.22 mg/ton-waste), respectively.

3.2 Atmospheric PAH Concentration

The four sampling locations include Assembly of God School of Theology (point A), Wen-Sun elementary school (point B), Chung-Shin Industrial Company (point C) and Taiwan Architecture Material Building (point D), respectively. To avoiding direct traffic source influence, PS-1 samplers were placed 30 meters away from the road. Table 2 lists the atmospheric PAH concentrations of four receptors for each of the 21 PAHs and the total-PAHs concentrations. Notably, these four receptors somewhat differ in the variation of atmospheric PAH concentration.

At point A, Nap made the greatest contribution

Table 3. Comparison of modeled and measured total-PAH concentration at the four monitoring sites.

Receptor Point	A (-500,-1800,130)		B (1500, 0, 90)		C (100,1100,130)		D (-1500, 1500,150)	
Date	Modeled Conc. (ng/m^3)	On-site Meas. Conc. (ng/m^3)	Modeled Conc. (ng/m^3)	On-site Meas. Conc. (ng/m^3)	Modeled Conc. (ng/m^3)	On-site Meas. Conc. (ng/m^3)	Modeled Conc. (ng/m^3)	On-site Meas. Conc. (ng/m^3)
Nov. 22, 1996	1.62	422	0	595	0	724	0	546
Dec. 12, 1996	0	871	0	727	0	699	0	419
Dec. 19, 1996	0	748	0	467	0	334	0	216
Feb. 21, 1997	0.34	534	0	579	0	723	0	485
Mar. 13, 1997	0	505	0	213	0	402	0	150
Mar. 14, 1997	0	341	0	128	0	450	0	129
Mar. 27, 1997	0.44	568	0	377	0	315	0	155
May 15, 1997	0	920	0.11	438	3.08	----[a]	0.31	461
May 30, 1997	1.40	398	0.96	607	0	----[a]	0	217

----[a]: not available.

to total-PAHs, with a concentration of 451 ng/m^3, and was followed by Ant, with a concentration of 27.9 ng/m^3. At point B, Nap was the greatest contributor to total-PAHs, with a concentration of 353 ng/m^3, while Ant was 21.3 ng/m^3. Meanwhile, Nap also had the highest concentration at point C, reaching 451 ng/m^3, and was followed by Ant (27.9 ng/m^3). Finally at point D, the concentration of Nap was 451 ng/m^3, while Ant was 27.9 ng/m^3. The total-PAH concentrations of these four receptors were 571 ng/m^3 (A), 508 ng/m^3 (B), 722 ng/m^3 (C) and 422 ng/m^3 (D), respectively. The comparable PAH concentrations were found between 314 to 735 ng/m^3 in urban areas [Mi, 1998]. Direct impact from traffic sources should increase the concentration of PAHs in ambient air up to between 1,520 and 3,920 ng/m^3 [Lee et al., 1995; Mi, 1998]. Considering the environmental conditions, the air quality of point B is significantly affected by traffic sources, while point C is affected by both industrial and traffic sources. Clearly, significant levels of traffic PAH indicators such as IND, DBA, BghiP and BbF [Mi et al., 1996] were found in ambient air monitoring samples (Table 2). However, the contributions of the traffic indicators in the atmosphere at sites, A and D, which are remote sites close to MWI, are similar to those for the areas with heavy traffic [Lee et al., 1995; Mi, 1998].

3.3 Comparison with ISCST3 Modeled Data

The simulated contribution concentration emitted from the stack of incinerators for four sampling locations are similar (Table 3). For point A, the total-PAH day-average contribution concentration exceeds zero in four simulated results out of nine. Meanwhile, while the results of the last two simulations exceed zero for point B, only one simulation exists in which the results are higher than zero for points C and D, respectively. Consequently, point A has the most significant positive effect on exhaust emission among the four sampling points in the modeling condition. This phenomenon is caused by the fact that point A is located in the downwind area, and the wind direction was primarily from the north-northwest, north, and north-northeast during the sampling period. For the results of the 9th simulation for point A, the highest total PAHs day-average contribution concentration was 1.40 ng/m^3, on May 30, 1997. Meanwhile for point B, the highest total-PAH day-average contribution

concentration was 0.96 ng/m^3, on May 30, 1997. This level was lower than that observed at point C, which was 3.08 ng/m^3, and is comparable to that at point D (0.31 ng/m^3, May, 15, 1997, Table 3).

Simulation results using the ISCST3 Model reveal that the highest total PAHs day-average falling contribution concentration totals 8.6 ng/m^3. Meanwhile, the position of falling is (3,-543). The modeling results display the position of maximum falling contribution concentration always occurs and within 1 km of the incinerator and to the south. A is located to the south-south-west, and wind direction is from the north and north-north-west during the sampling period. All of the above relationships are in accordance with the class B atmospheric stability.

While the sample analysis results of air dispersion testing are not intended to serve as a rigorous model evaluation, the influence of single stationary sources on ambient air quality appears insignificant. Traffic sources contribute far more pollutants to air than the stationary sources located in the nearby industrial park. Also, BbF, BaP, DBA, and BghiP were the PAH indicators mainly derived from mobile sources, and were found to have a high correlation with ambient air.

4. Conclusion

Based on the results in this study, we conclude the following:

1. The mean total-PAH (summation of 21 PAHs) emission rate was 24,500 mg/hr. Nap, the lowest molecular PAH, had the highest emission rate at 20,900 mg/hr. For other individual PAHs that are suspected of containing high level of bio-carcinogenic compounds, BaP, DBA, and BbF are 69.9 mg/hr, 102 mg/hr, and 181 mg/hr, respectively.
2. The mean total-PAH emission factor is 871 mg/ton-waste, and once again Nap has highest emission factor (774 mg/ton-waste), followed by Ant (51.7 mg/ton-waste), FL (19.3 mg/ton-waste) and Pyr (8.22 mg/ton-waste) for the MWI stack in central Taiwan.
3. The ambient air total-PAH concentrations collected from the sites near the incinerator reveal that the level of concentration varies significantly among 21 individual PAHs. Mean total PAHs concentration ranges between 422 and 722 ng/m^3. The air quality at those four sites was affected by environmental conditions, especially for the indicator pollutants (such as IND, DBA, BghiP and BbF) of traffic pollution sources.
4. The results of applying the ISCST3 Model reveal that stack flue gas from the incinerator has a minor effect on the four sampling sites. The single stationary pollution source (incinerator) thus has a relatively minor effect on its immediate environment less than that of traffic sources. The overall effect of the stack flue gas of the incinerator on ambient air quality is thus insignificant.

Acknowledgements

The authors would like to thank the National Science Council of the Republic of China for financially supporting this research under Contract No. NSC-89-2211-E041-021. We are also appreciative of both Dr. Lien-Teh Hsieh and Dr. Chien-Ming Chen for their great help. Thanks are also given to reviewers for their insightful comments in greatly adding to the quality of this manuscript.

References

Basham J. P., Whitwell I. (1999), Dispersion Modeling of Dioxin Releases from the Waste Incinerator at Avonmouth, Bristol, UK. Atmospheric Environment, 33: 3405-3416.

Colmsjo A. L., Zebuhr Y. U., Ostman C. E. (1986), Polynuclear Aromatic Compounds in Flue Gases and Ambient Air in Vicinity of A Municipal Incineration Plant. Atmospheric Environment, 20: 2279-2282.

Eschenroeder A., Lorber M. (1999), An Evaluation of EPA ISCST-Version 3 Model, Part 1. Air Dispersion of Dioxins. Organohalogen Compounds, 41: 547-552.

Lee W. J., Wang Y. F., Lin T. C., Chen Y. Y., Lin W. C., Ku C. C., Cheng J. T. (1995), PAH Characteristics in the Ambient Air of Traffic-Source. The Science of Total Environment, 159: 185-200.

Li C. T., Lee W. J., Mi H. H., Su C. C. (1995), PAH Emission from the Incineration of Waste Oily Sludge and PE Plastic Mixtures. The Science of the Total Environment, 170: 171-183.

Mi H. H. (1998), Emission Characteristics of Polycyclic Aromatic Hydrocarbons from Mobile Sources. Ph. D. Dissertation, National Cheng Kung University, Tainan, Taiwan, p.191-222.

Mi H. H., Lee, W. J., Lin T. C., Wu T. L., Chao H. R., Wang L. C. (1996), PAH Emission from a Gasoline-Powered Engine. Journal of Environmental Science and Health PART A-Environmental Science and Engineering, 31: 1981-2003.

Oehme M., Mano S., Mikalsen A. (1987), Formation and Presence of Polychlorinated and Polycyclic Aromatic Compounds in the Emission of Small and Large Scale Municipal Waste Incineration. Chemosphere, 16: 143-153.

Sheu H. L., Lee W. J., Lin S. J., Fang G. C., Chang H. C., You W. C. (1997), Particle-Bound PAH Content in Ambient Air. Environmental Pollution, 96: 369-382.

US-EPA, Method TO-13 (1988), The Determination of Benzo(a)pyrene [B(a)P] and other Polynuclear Aromatic Hydrocarbons (PAHs) in Ambient Air Using Gas Chromatographic (GC) and High Performance Liquid Chromatographic (HPLC) Analysis.

Yasuda K., Kaneko M., Sugiyama K., Yoshino H., and Otsuka Y. (1989), Basic Research on the Emission of Polycyclic Aromatic Hydrocarbons Caused by Waste Incineration. JAPAC, 39: 1557-1561.

Yang H. S., Lee W. J., Chen S. J., Lai S. O. (1998), PAH Emission from Various Industrial Stacks. Journal of Hazardous Materials, 60: 159-174.

Zemba S. G., Green L. C., Crouch E. A. C., Lester R. R. (1996), Quantitative Risk Assessment of Stack Emission from Municipal Waste Combustors. Journal of Hazardous Materials, 47: 229-275.

Received for review, June 30, 2000
Accepted, May 8, 2001
AAQR-2001-09