



Particulate matter characteristics during agricultural waste burning in Taichung City, Taiwan

Man-Ting Cheng^{a,*}, Chuen-Liang Horng^a, Yi-Ru Su^a, Li-Kai Lin^a, Yu-Chi Lin^b, Charles C.-K. Chou^b

^a Department of Environmental Engineering, National Chung Hsing University, 250, Kuo Kuang Road, Taichung 402, Taiwan, ROC

^b Research Center for Environmental Changes, Academia Sinica, Taipei 115, Taiwan, ROC

ARTICLE INFO

Article history:

Received 10 March 2008

Received in revised form

24 September 2008

Accepted 25 September 2008

Available online 2 October 2008

Keywords:

PM_{2.5}

Agricultural waste burning

Ionic species

Carbonaceous species

ABSTRACT

Agricultural waste burning is performed after harvest periods in June and November in Taiwan. Typically, farmers use open burning to dispose of excess rice straw. PM_{2.5} and PM_{2.5–10} measurements were conducted at National Chung Hsing University in Taichung City using a dichotomous sampler. The sampling times were during straw burning periods after rice harvest during 2002–2005. Ionic species including SO₄²⁻, NO₃⁻, NH₄⁺, K⁺, Ca²⁺, Cl⁻ and Na⁺ and carbonaceous species (EC and OC) in PM_{2.5} and PM_{2.5–10} were analyzed. The results showed that the average PM_{2.5} and PM_{2.5–10} concentrations were 123.6 and 31.5 μg m⁻³ during agricultural waste burning periods and 32.6 and 21.4 μg m⁻³ during non-waste burning periods, respectively. The fine aerosol ionic species including Cl⁻, K⁺ and NO₃⁻ increased 11.0, 6.7 and 5.5 times during agricultural burning periods compared with periods when agricultural waste burning is not performed. K⁺ was found mainly in the fine mode during agricultural burning. High nitrogen oxidation ratio was found during agricultural waste burning periods which might be caused by the conversion of Nitrogen dioxide (NO₂) to NO₃⁻. It is concluded that agricultural waste burning with low dispersion often causes high PM_{2.5} and gases pollutant events.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Biomass burning, the burning of living and dead vegetation, has become a global issue in the past decade [1,2] and is a significant source of atmospheric particles and gaseous pollutants [3–7]. Most of these particles come from residual waste burning including forests, grasslands and crops through natural or anthropogenic fires [8–11]. In general, numerous particulates (e.g., polycyclic aromatic hydrocarbons and other organics) and gaseous compounds (e.g., carbon monoxide (CO) and volatile organic compounds) that come from biomass burning are known to be hazardous to human health [12,13]. Metzger et al. [14] reported that mass concentrations of CO, Nitrogen dioxide (NO₂), PM_{2.5} (particulate matter with aerodynamic diameters less than 2.5 μm), organic carbon (OC) and elemental carbon (EC) in PM_{2.5} (fine particles) were significantly associated with emergency department visits at hospitals due to cardiovascular diseases.

Particulate matters are composed of secondary particles like organic carbon, nitrate and sulfate formed by homogenous or heterogeneous reactions with their precursor gases in the atmosphere. Biomass burning is one of the dominant sources of particulate

organic carbon [15,16]. Cao et al. [17] reported that emission rates of OC were 1.83–3.46 g kg⁻¹ from crop residues. Higher ionic contributions during combustion were observed in aerosols produced from combusted plant materials and soil suspended particles [3]. The aerosol composition of K⁺, NO₃⁻ and Cl⁻ ions increased during cereal waste burning periods and were found to be an independent source of NO₃⁻ not linked to SO₄²⁻ sources [18]. Potassium ion can be a trace species during biomass burning [19]. KCl and K₂SO₄ are formed by homogeneous nucleation from straw combustion [20]. The smoke from vegetation fires can act as condensation nuclei and indirectly change the radiation budget and albedo [21].

Taiwan is in a subtropical region with rice as one of the main farm products. There are two rice crops in June and November. The amount of rice straw produced every year is about 2.6 million tons [22]. Rice straw contains mainly total organic material (78.1–85.0%), K₂O (0.91–2.1%) and nitrogen (0.6–1.3%) [23]. Farmers use open agricultural burning to infuse the ground with nutrients for the next growing season. Agricultural waste burning emits lots of aerosols into the atmosphere and these emissions may have a significant impact on air quality. The objective of this research was to study the effect of agricultural waste burning on the particulate matter and water-soluble inorganic ions in the atmosphere by collecting samples of atmospheric particulate matter during the waste burning and non-burning periods.

* Corresponding author. Tel.: +886 422851984; fax: +886 422862587.
E-mail address: mtcheng@dragon.nchu.edu.tw (M.-T. Cheng).

2. Materials and methods

2.1. Sampling of $PM_{2.5}$ and $PM_{2.5-10}$

The ambient particulate matter during agricultural waste burning period was characterized at an urban site in Central Taiwan. Measurements were conducted on the roof of a seven-story building. The sampling site is located at National Chung Hsing University (NCHU) in Taichung City. There were two roads and no industrial emissions near the site. There were three sampling campaigns during agricultural waste burning periods (27 November 2002, 29 June 2004, and 25–27 November 2005) and non-waste burning periods (30–31 October 2002, 24 June 2004, 26–27 June 2004, and 24–27 October 2005), respectively. The collection time lasted 12 h, from 8:00 a.m. to 8:00 p.m. or from 8:00 p.m. to 8:00 a.m. The details for the sampling location are shown in Fig. 1. Twenty-six samples were collected from three agricultural waste burning and non-agricultural waste burning periods during 2002–2005. The season was autumn during periods I and III and summer during period II.

Two dichotomous samplers (Sierra & Andersen, model 241) using both Teflon membrane filters (R2PL037, PALL Life Sciences) and quartz fiber filters (2500QAT-UP, PALL Life Sciences) were used to collect fine ($PM_{2.5}$) and coarse aerosols ($PM_{2.5-10}$). The sampler equipped with Teflon membranes was used to analyze ions in the fine and coarse modes and the other one equipped with quartz fiber filters was used for the OC and EC. The total flow rate of the dichotomous sampler was 16.7 lpm. The unit was equipped with an inlet designed for 10 μm cut-point. The sampler contained a virtual impactor with a 2.5 μm cut-point used to separate entering particles into fine and coarse sizing ranges. NO_2 , nitrogen monoxide (NO), CO, sulfur dioxide (SO_2), ozone (O_3), temperature, wind speed and wind direction data were collected from the air quality monitoring station located near NCHU.

2.2. Water-soluble ion species and carbonaceous analysis

After each sampling, both $PM_{2.5}$ and $PM_{2.5-10}$ were analyzed for water-soluble ions and carbonaceous species content. The Teflon filter was placed in a vial with 10 ml of ultra-pure water and sonicated for 90 min. The extracted solution was then filtered through a 0.22 μm cellulose esters filter and stored at 4 °C until further analysis. All SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , Na^+ , Ca^{2+} and K^+ ionic extracts were determined using Dionex ion chromatography (Dionex, DX 100).

Before the carbon samples were collected, the quartz filters were pre-fired to 900 °C for 2 h before usage in order to remove the impurities. After the samples were collected, the filters were stored with refrigerator before analyzing the carbon contents in order to minimize the possible desorption of volatile organics from the particulates [24]. OC and EC were determined using IMPROVE

Thermo-Optical Reflection (TOR) as previously described by Chow et al. [25,26].

A Quality Assurance and Quality Control program for method detection limit (MDL), precision and accuracy were conducted during the experiments. The MDLs, calculated as three times the standard deviation of blanks, were 0.02, 0.02, 0.02, 0.02, 0.01, 0.01, 0.03, 0.01 and 0.02 $\mu\text{g m}^{-3}$ for SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , Na^+ , K^+ , Ca^{2+} , OC and EC. Precision of the chemical analysis was determined to be less than 10%. The average recoveries for all species appeared in the range of 100 \pm 5%.

3. Results and discussion

3.1. Concentrations of $PM_{2.5}$, $PM_{2.5-10}$ and gaseous pollutants

Fig. 2(a) shows daytime and nighttime concentration of $PM_{2.5}$, $PM_{2.5-10}$ as well as $PM_{2.5}/PM_{2.5-10}$ ratio during the three periods. The mean $PM_{2.5}$ concentrations during agricultural waste burning periods and periods when agricultural waste burning periods is not performed were 123.6 and 32.6 $\mu\text{g m}^{-3}$, respectively, whereas the corresponding concentrations for $PM_{2.5-10}$ were 31.5 and 21.4 $\mu\text{g m}^{-3}$ (Table 1). On average, $PM_{2.5}$ concentration was 3.8 times higher during agricultural waste burning compared with non-waste burning periods. Overall, $PM_{2.5}$ concentrations during

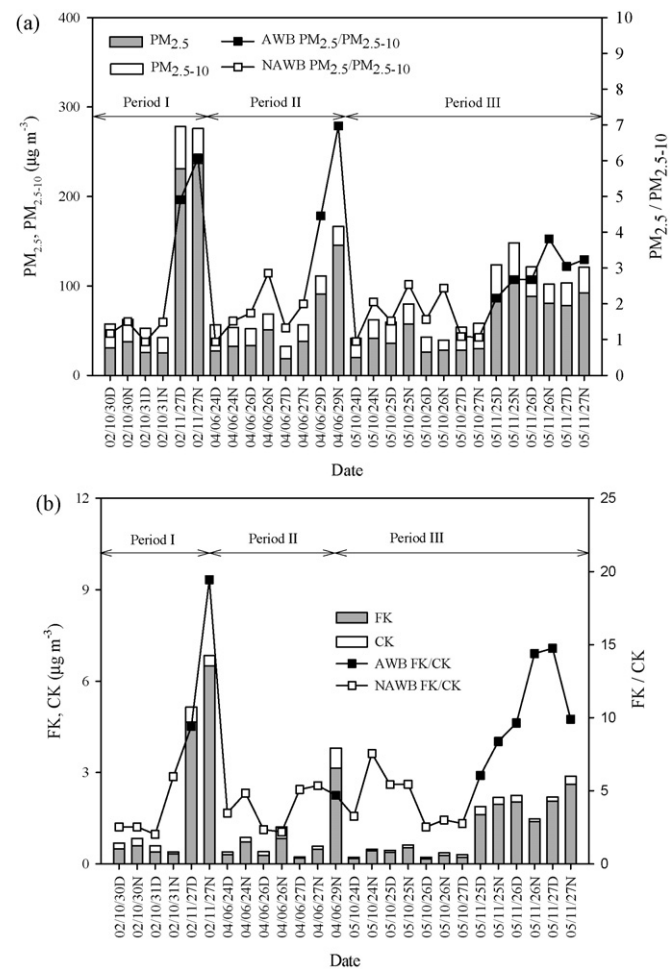


Fig. 2. (a) Daily concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ and ratio of $PM_{2.5}/PM_{10}$ and (b) daily concentrations of potassium in fine mode (FK) and coarse mode (CK) and ratio of FK/CK during the three periods of agricultural waste burning (AWB, Black square) and non-agricultural waste burning (NAWB, White square).

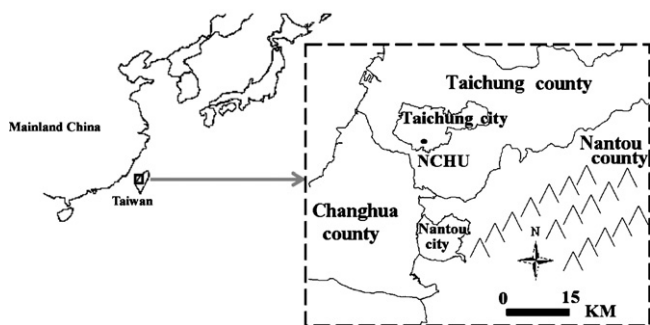


Fig. 1. Location of the sampling site at NCHU in central Taiwan.

Table 1
Concentrations of PM_{2.5}, PM_{2.5–10}, OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, K⁺, Ca²⁺, Na⁺, and Cl⁻ measured during the three agricultural and non-agricultural waste burning periods.

Sampling date	PM _{2.5}	OC	EC	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	K ⁺	Ca ²⁺	Na ⁺	Cl ⁻
AWB periods										
2002/11/27 (I) (N=2)	234.1	82.3	18.9	12.2	19.1	8.7	5.6	0.4	0.4	9.9
2004/6/29 (II) (N=2)	118.3	31.8	4.6	21.5	5.4	7.6	2.5	0.4	0.3	1.1
2005/11/25–11/27 (III) (N=6)	88.6	20.5	7.0	17.0	8.2	8.4	1.9	0.3	0.4	1.6
Average	123.6	35.1	8.9	16.9	9.8	8.3	2.8	0.3	0.4	3.1
S.D.	61.4	26.0	5.7	5.8	6.0	2.2	1.6	0.2	0.1	3.8
NAWB periods										
2002/10/30–10/31 (I) (N=4)	29.8	8.7	3.3	5.7	1.3	2.6	0.5	ND	ND	0.3
2004/6/24, 6/26–6/27 (II) (N=6)	33.3	13.4	2.5	4.3	1.0	1.3	0.5	0.3	0.5	0.2
2005/10/24–10/27 (III) (N=8)	33.4	7.4	3.6	6.9	2.6	2.7	0.4	0.1	0.2	0.4
Average	32.6	9.7	3.1	5.7	1.8	2.2	0.4	0.2	0.3	0.3
S.D.	9.9	4.0	1.1	2.0	1.8	1.3	0.2	0.1	0.2	0.3
Ratio of burning/non-burning	3.8	3.6	2.8	2.9	5.5	3.8	6.7	1.9	1.3	11.0
P value	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	0.31	0.01
Sampling date	PM _{2.5–10}	OC	EC	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	K ⁺	Ca ²⁺	Na ⁺	Cl ⁻
AWB periods										
2002/11/27 (I) (N=2)	43.0	8.4	1.5	1.1	2.4	1.6	0.4	1.1	0.3	0.9
2004/6/29 (II) (N=2)	20.6	0.7	ND	1.3	2.0	1.2	0.7	0.3	2.3	0.4
2005/11/25–11/27 (III) (N=6)	31.3	3.1	0.5	1.3	3.3	0.5	0.2	0.8	0.9	0.8
Average	31.5	4.0	0.7	1.3	2.9	0.8	0.3	0.8	1.1	0.7
S.D.	9.6	2.7	0.6	0.5	1.4	0.7	0.2	0.4	0.8	0.4
NAWB periods										
2002/10/30–10/31 (I) (N=4)	23.9	2.3	0.2	0.7	2.7	0.6	0.2	ND	1.6	1.6
2004/6/24, 6/26–6/27 (II) (N=6)	20.0	2.9	0.5	0.6	0.2	0.3	0.1	0.4	0.4	0.1
2005/10/24–10/27 (III) (N=8)	21.2	2.1	0.3	0.8	2.3	0.3	0.1	0.5	0.9	0.8
Average	21.4	2.2	0.3	0.7	1.7	0.4	0.1	0.4	0.8	0.7
S.D.	5.0	0.5	0.1	0.3	1.4	0.4	0.1	0.4	0.7	1.0
Ratio of burning/non-burning	1.5	2.3	2.5	1.7	1.7	2.3	2.5	1.8	1.4	1.1
P value	<0.01	<0.01	0.02	0.01	0.04	0.03	<0.01	0.04	0.46	0.86

N: Number of samples. Unit: $\mu\text{g m}^{-3}$. P value: ANOVA test.

agricultural waste burning periods exceeded U.S. Environmental Protection Agency (EPA) standards, and the highest PM_{2.5} concentration reached 234.1 $\mu\text{g m}^{-3}$ which was found in period I. The highest PM_{2.5} concentration on 27 November might be due to agricultural waste burning with calm or no wind (average wind speed = 0.5 m s^{-1} , Table 2) meteorological condition. The average of PM_{2.5}/PM_{2.5–10} ratios during waste burning and non-waste burning periods were 3.8 and 1.5, respectively. This value during the waste burning period was higher compared to Tsai and Cheng [27] and this high fine fraction ratio might be caused by agricultural waste burning.

The concentrations of atmospheric CO, SO₂, NO, NO₂, O_{3max} gases, temperature, relative humidity and prevailing wind direction during waste burning and non-waste burning days are shown in Table 2. The average concentrations of CO, SO₂, NO, NO₂ and O_{3max} were 1.7 ppm, 7.4, 22.3, 49.3 and 55.5 ppb during waste burning and 0.7 ppm, 2.8, 6.5, 24.2 and 39.3 ppb when no waste was burned. The prevailing wind direction was mainly from the northwest during periods I and III. The wind was from southwestward direction during period II. The ratios of CO, SO₂, NO_x, temperature and wind speed during waste burning days compared to non-waste burning days were 2.4, 2.6, 2.3, 0.9 and 0.6, respectively. For O_{3max} there was no significant difference between the agricultural waste burning and no waste burning periods. O₃ is formed by a chemical reaction between NO_x (NO + NO₂) and volatile organic compounds (VOC), in the presence of strong ultraviolet radiation [28]. Hence, the lower concentration of O_{3max} on 27 November might be due to a cloudy day and the lack of ultraviolet radiation. The highest concentrations of CO, SO₂, NO and NO₂ and the lowest values of temperature and wind speed were found during waste burning period I. Obviously, in this period a lot of gaseous pollutants were from agricultural waste burning. Crutzen and Andreae [8] reported that the CO, SO₂ and NO_x emission ratios were 10%, 0.3% and 12% from biomass burn-

ing. The wind speed during agricultural waste burning was slower compared to that during non-agricultural waste burning. Thus, the increase in CO, SO₂, NO and NO₂ might be due to agricultural waste burning combined with poor dispersion.

3.2. Species of PM_{2.5} and PM_{2.5–10}

The concentrations of fine and coarse OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, K⁺, Ca²⁺, Na⁺ and Cl⁻ aerosol during waste burning and non-burning periods are shown in Table 1. The average concentrations of fine OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, K⁺, Ca²⁺, Na⁺ and Cl⁻ were 35.1, 8.9, 16.9, 9.8, 8.3, 2.8, 0.3, 0.4 and 3.1 $\mu\text{g m}^{-3}$ during waste burning and 9.7, 3.1, 5.7, 1.8, 2.2, 0.4, 0.2, 0.3 and 0.3 $\mu\text{g m}^{-3}$ during non-waste burning periods, respectively. Organic carbon and sulfate were the major components of PM_{2.5}, representing on average 28% and 14% of PM_{2.5} during waste burning and 30% and 17% of PM_{2.5} during non-waste burning periods, respectively. The organic carbon contained only the mass of carbon in the organic aerosol material which also contained other species besides carbon. The mass of particulate organics matter was estimated by particulate organics matter = OC ÷ 0.6386 [29]. The percentage of the organics matter contributed more than 50% of mass in PM_{2.5}. The ratio of PM_{2.5} species during waste burning and non-waste burning periods showed that Cl⁻ (11.0), K⁺ (6.7) and NO₃⁻ (5.5) were the three dominant species during waste burning periods. Similarly, Ryu et al. [30] reported that during biomass burning Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺ and K⁺ were the dominant species.

The average concentrations of coarse OC, EC, SO₄²⁻, NO₃⁻, NH₄⁺, K⁺, Ca²⁺, Na⁺ and Cl⁻ aerosol were 4.0, 0.7, 1.3, 2.9, 0.8, 0.3, 0.8, 1.1 and 0.7 $\mu\text{g m}^{-3}$ during waste burning and 2.2, 0.3, 0.7, 1.7, 0.4, 0.1, 0.4, 0.8 and 0.7 $\mu\text{g m}^{-3}$ during non-waste burning, respectively. Organic carbon and nitrate were the most abundant components, representing 13% and 9% of PM_{2.5–10} during waste burning and

Table 2
Gaseous CO, SO₂, NO_x, NO, NO₂ and O_{3max} and meteorological temperature, RH, WS and PWD during the three agricultural and non-agricultural waste burning periods.

Sampling date	CO (ppm)	SO ₂ (ppb)	NO _x (ppb)	NO (ppb)	NO ₂ (ppb)	O _{3max}	Temperature (°C)	RH (%)	WS (m s ⁻¹)	PWD (°)
AWB periods										
2002/11/27 (I)	3.0	9.7	141.2	51.9	89.3	27.8	19.7	68.2	0.5	NW
2004/6/29 (II)	0.9	7.6	26.7	1.5	25.2	86.3	29.3	69.0	1.3	SW
2005/11/25–11/27 (III)	1.1	4.9	46.9	13.4	33.5	52.3	23.0	67.4	1.3	NW
Average	1.7	7.4	71.6	22.3	49.3	55.5	24.0	68.2	1.0	
S.D.	1.2	2.4	61.1	26.3	34.9	29.4	4.9	0.8	0.5	
NAWB periods										
2002/10/30–10/31 (I)	0.9	2.2	35.8	8.5	27.3	44.2	25.9	76.7	1.9	NW
2004/6/24.6/26–6/27 (II)	0.4	2.7	18.3	2.7	16.5	31.9	28.2	68.3	1.7	SW
2005/10/24–10/27 (III)	0.7	3.6	37.2	8.4	28.8	41.7	26.6	71.4	1.7	WNW
Average	0.7	2.8	30.7	6.5	24.2	39.3	26.9	72.1	1.8	
S.D.	0.3	0.7	10.0	3.3	6.7	6.5	1.2	4.2	0.1	
Ratio of burning/non-burning	2.4	2.6	2.3	3.4	2.0	1.4	0.9	0.9	0.6	
P value	<0.00	<0.00	<0.00	0.02	0.01	0.52	0.02	0.40	0.05	

N: Number of samples. P value: ANOVA test.

10% and 8% during non-waste burning periods, respectively. Thus, organic carbon is the most abundant component of PM_{2.5} and PM_{2.5–10} in Central Taiwan during both agricultural waste burning and non-agricultural waste burning periods. Organic carbon is generated by the condensation of low vapor pressure products during hydrocarbons photo-oxidation [31]. The ratio of PM_{2.5–10} species during waste burning and non-waste burning periods showed that EC (2.5), K⁺ (2.5), OC (2.3) and NH₄⁺ (2.3) were the four dominant species in PM_{2.5–10}. A previous study showed that K⁺ and Cl⁻ were the dominant species during biomass burning [21]. The K⁺ species was present mainly in the fine mode. Based on the amount of K⁺ and Na⁺ in PM_{2.5}, the portion of Cl⁻ were 80% of Cl⁻ in KCl and 20% in NaCl during waste burning period. However, the contents of Cl⁻ in PM_{2.5–10} was not enough to be distributed in NaCl and KCl. Since only large amount of Cl⁻ and K⁺ were found in PM_{2.5} during waste burning.

3.3. Agricultural waste burning aerosols

Correlation matrix statistical analysis was used to identify the relationship between mass and species in PM_{2.5}. A correlation matrix between PM_{2.5}, OC, EC and water-soluble ionic species is shown in Table 3. PM_{2.5} was highly correlated ($r=0.8–1.0$) and well

correlated ($r=0.6–0.8$) with OC, EC, NO₃⁻, and K⁺ during waste burning and non-waste burning periods, respectively. Duan et al. [32] reported that the correlation coefficient between increment OC and K⁺ was higher at a residential site compared to a background site. Thus, the higher correlation found in our study might be due to agricultural waste burning. K⁺ was highly correlated with PM_{2.5}, OC, EC, and Cl⁻ during waste burning periods. This result indicates that K⁺, Cl⁻, OC and EC were generated after agricultural waste burning.

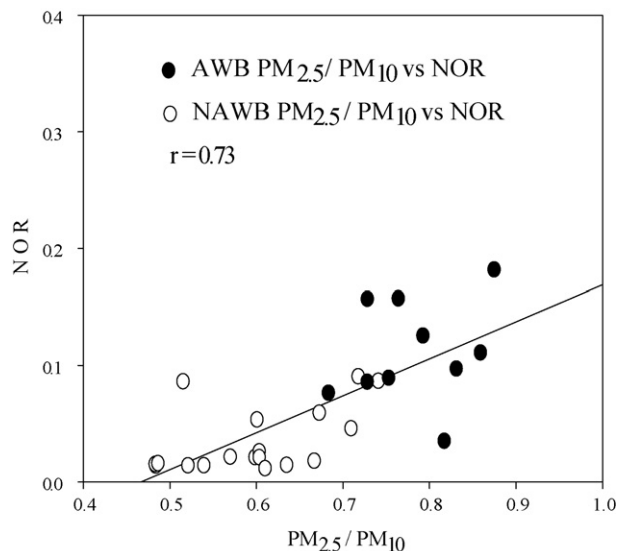
Table 4 shows comparisons of PM_{2.5} and OC, EC, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺ and Cl⁻ in PM_{2.5} in various areas. The EC, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺ and Cl⁻ data were in PM₁ measured in Helsinki [2]. The PM_{2.5}, EC, SO₄²⁻, NH₄⁺, K⁺ and Cl⁻ concentrations were higher in this area compared to other areas during the three agricultural waste burning periods [2,18,30,33]. This might be due to high emission of agricultural waste burning with poor ventilation during period I–III. Higher PM_{2.5}/PM_{2.5–10} ratio was observed during period I and II. A high correlation was found between nitrogen oxidation ratios (NOR) with PM_{2.5}/PM₁₀ (Fig. 3), where the NOR is defined as $N_{NO_3^-}/(N_{NO_3^-} + N_{NO_2})$ [27,34–36]. The unit of $N_{NO_3^-}$ and N_{NO_2} was $\mu\text{g N m}^{-3}$. The nitrogen oxidation ratio was used to evaluate the conversion from NO₂ to NO₃⁻. Therefore, the result indicated that the increase in PM_{2.5}/PM₁₀ ratios might be affected by nitrogen oxidation.

Table 3
Correlation matrix for PM_{2.5}, OC, EC and water-soluble ionic species during agricultural waste (AWB) and non-agricultural (NAWB) waste burning periods.

	PM _{2.5}	OC	EC	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	K ⁺	Ca ²⁺	Cl ⁻	Na ⁺
AWB										
PM _{2.5}	1.00									
OC	0.98	1.00								
EC	0.91	0.87	1.00							
SO ₄ ²⁻	-0.40	-0.50	-0.36	1.00						
NO ₃ ⁻	0.84	0.78	0.91	-0.46	1.00					
NH ₄ ⁺	0.09	-0.07	0.33	0.65	0.29	1.00				
K ⁺	0.95	0.97	0.86	-0.50	0.72	-0.07	1.00			
Ca ²⁺	0.53	0.43	0.31	-0.10	0.40	0.05	0.41	1.00		
Cl ⁻	0.89	0.90	0.92	-0.41	0.74	0.13	0.93	0.19	1.00	
Na ⁺	0.18	0.18	0.29	-0.46	0.34	-0.06	0.24	0.28	0.16	1.00
NAWB										
PM _{2.5}	1.00									
OC	0.63	1.00								
EC	0.75	0.15	1.00							
SO ₄ ²⁻	0.40	-0.18	0.59	1.00						
NO ₃ ⁻	0.80	0.18	0.86	0.49	1.00					
NH ₄ ⁺	0.60	-0.11	0.82	0.89	0.82	1.00				
K ⁺	0.74	0.80	0.38	-0.05	0.41	0.10	1.00			
Ca ²⁺	0.04	0.56	-0.55	-0.48	-0.27	-0.53	0.30	1.00		
Cl ⁻	0.53	0.25	0.57	0.31	0.67	0.57	0.40	-0.16	1.00	
Na ⁺	0.21	0.68	-0.21	-0.22	-0.13	-0.28	0.32	0.53	0.04	1.00

Table 4Comparison between mean concentrations ($\mu\text{g m}^{-3}$) of $\text{PM}_{2.5}$, OC, EC, NO_3^- , SO_4^{2-} , NH_4^+ , K^+ , Cl^- and $\text{PM}_{2.5}/\text{PM}_{2.5-10}$ in this study with some other published measurements.

Site	Period	$\text{PM}_{2.5}$	OC	EC	NO_3^-	SO_4^{2-}	NH_4^+	K^+	Cl^-	$\text{PM}_{2.5}/\text{PM}_{2.5-10}$	Reference
Helsinki, Finland ^a	24–29 April 2006	36.0	11.0	2.4	NA ^b	2.4	NA	0.3	NA	1.2	Saarikoski et al. [2]
Helsinki, Finland	15 May 2006	43.0	9.7	2.5	NA	4.9	NA	0.3	NA	1.1	Saarikoski et al. [2]
Gwangju, Korea	June 2003	67.9	20.9	3.4	6.6	10.8	4.6	1.7	1.0	3.6	Ryu et al. [28]
Southeastern Brazil	April 1999–February 2001	NA	NA	NA	0.5	1.9	0.3	0.4	0.2	NA	Allen et al. [31]
Victoria, northern Spain	10 September–15 October 1991	NA	NA	NA	1.0	3.2	0.8	1.3	0.8	NA	Ezcurra et al. [18]
Taichung, Taiwan	27 November 2002 (I)	234.1	82.3	18.9	19.1	12.2	8.7	5.6	9.9	5.5	This study
Taichung, Taiwan	29 June 2004 (II)	118.3	31.8	4.6	5.4	21.5	7.6	2.5	1.1	5.7	This study
Taichung, Taiwan	25–27 November 2005 (III)	88.6	20.5	7.0	8.2	17.0	8.4	1.9	1.6	2.9	This study

^a The species of OC, EC, NO_3^- , SO_4^{2-} , NH_4^+ , K^+ and Cl^- were from PM_{10} .^b NA represents not available.**Fig. 3.** Relation of NOR versus $\text{PM}_{2.5}/\text{PM}_{2.5-10}$ during the three periods.

The daily daytime and nighttime concentrations of potassium in the fine mode (FK) and in the coarse mode (CK) during the three periods of agricultural waste burning and non-burning periods are shown in Fig. 2(b). Agricultural waste burning aerosols can be related with enrichment of FK and can also be identified by the FK/CK ratio. The average FK/CK was 10.7 during agricultural waste burning and 3.9 during non-agricultural waste burning periods, respectively. Ryu et al. [30] reported that FK/CK above 9.2 can be a criterion for a biomass event. Hence, the FK/CK ratio used in this study might be a way to confirm agricultural waste burning.

During biomass burning periods the aerosol content of OC and K^+ displayed high portion both >10% and 1–10%, respectively [32]. We found that the mean portions of OC and K^+ accounted for 28% and 2% of $\text{PM}_{2.5}$ measured during agricultural waste burning, respectively. This result was similar to the value reported by Duan et al. [32]. Moreover, K^+/OC ranged from 0.08 to 0.10 which is close to the range between 0.04 and 0.13 used as an indicator to identify biomass burning reported by Echalar [37], Maenhaut et al. [38] and Andreae and Merlet [39]. Similarly, in this study the K^+/OC was 0.06–0.11 during agricultural waste burning and 0.02–0.05 for non-agricultural waste burning. These results indicate that species of K^+ and OC played an important role during agricultural waste burning periods.

4. Conclusions

This study was designed to investigate the $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ aerosol species in Taichung City during agricultural waste burning

periods. The results showed that gaseous CO , SO_2 and NO_x increased 2.4, 2.6 and 2.3 times during agricultural waste burning periods compared to non-waste burning periods. Moreover, the average $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ concentrations were 123.6 and 31.5 $\mu\text{g m}^{-3}$ during agricultural waste burning periods and two times higher than U.S. Environmental Standards (EPA). The highest concentration of $\text{PM}_{2.5}$ occurred in period I and the concentration reached 234.1 $\mu\text{g m}^{-3}$. The fine aerosol ionic species included Cl^- , K^+ and NO_3^- increased 11.0, 6.7 and 5.5 times during agricultural waste burning periods compared with non-waste burning periods. The K^+ was found mainly in the fine mode. High nitrogen oxidation ratio was found due to the faster conversion of NO_2 to NO_3^- . The data of this study showed that agricultural burning of rice straw can be a serious source of air pollutants which may influence ambient air quality. Thus, to manage the abundance of rice straw waste in Taiwan, other alternatives such as appropriate off-site use of straw by paper manufacturers, mushroom growers, and rope makers should be implemented in practice instead of burning in the open field. Evidently, more effort is needed to abate the air pollution caused by agricultural waste burning.

Acknowledgements

The authors would like to thank Academia Sinica and Environmental Protection Administration of Taiwan, Republic of China for financially supporting this research. Authors would also like to show our appreciation for the contributions from two anonymous reviewers for their constructive comments.

References

- L.C.C. Koe, A.F. Arellano Jr., J.L. McGregor, Investigating the haze transport from 1997 biomass burning in Southeast Asia: its impact upon Singapore, *Atmos. Environ.* 35 (2001) 2723–2734.
- S. Saarikoski, M. Sillanpää, M. Sofiev, H. Timonen, K. Saarnio, K. Teinilä, A. Karpinen, J. Kukkonen, R. Hillamo, Chemical composition of aerosols during a major biomass burning episode over northern Europe in spring 2006: experimental and modelling assessments, *Atmos. Environ.* 41 (2007) 3577–3589.
- A.G. Allen, A.H. Miguel, Biomass burning in the Amazon: characterization of the ionic component of aerosols generated from flaming and smouldering rainforest and savannah, *Environ. Sci. Technol.* 29 (1995) 486–493.
- Y. Yang, X.X. Zhang, T. Korenaga, Distribution of polynuclear aromatic hydrocarbons (PAHs) in the soil of Tokushima Japan, *Water Air Soil Pollut.* 138 (2002) 51–60.
- H.H. Yang, C.H. Tsai, M.R. Chao, Y.L. Su, S.M. Chien, Source identification and size distribution of atmospheric polycyclic aromatic hydrocarbons during rice straw burning period, *Atmos. Environ.* 40 (2006) 1266–1274.
- Y. Miura, T. Kanno, Emissions of trace gases (CO_2 , CO , CH_4 , and N_2O) resulting from rice straw burning, *Soil Sci. Plant Nutr.* 43 (1997) 849–854.
- H. Keshtkar, L.L. Ashbaugh, Size distribution of polycyclic aromatic hydrocarbon particulate emission factors from agricultural burning, *Atmos. Environ.* 41 (2007) 2729–2739.
- P.J. Crutzen, M.O. Andreae, Biomass burning in the tropics: impact on atmospheric chemistry and biogeochemical cycles, *Science* 250 (1990) 1669–1678.
- S. Yang, H. He, S. Lu, D. Chen, J. Zhu, Quantification of crop residue burning in the field and its influence on ambient air quality in Suqian China, *Atmos. Environ.* 42 (2008) 1961–1969.

- [10] T. Korenaga, X. Liu, Z. Huang, The influence of moisture content on polycyclic aromatic hydrocarbons emission during rice straw burning, *Chemosphere Global Change Sci.* 3 (2001) 117–122.
- [11] P.K. Gupta, S. Sahai, N. Singh, C.K. Dixit, D.P. Singh, C. Sharma, M.K. Tiwari, R.K. Gupta, S.C. Garg, Residue burning in rice-wheat cropping system: causes and implications, *Curr. Sci.* 87 (2004) 1713–1717.
- [12] J. Jacobs, R. Kreutzer, D. Smith, Rice burning and asthma hospitalizations, Butte County, California 1983–1992, *Environ. Health Perspect.* 105 (1997) 980–985.
- [13] K. Torigoe, S. Hasegawa, O. Numata, S. Yazaki, M. Matsunaga, N. Boku, M. Hiura, H. Ino, Influence of emission from rice straw burning on bronchial asthma in children, *Pediatr. Int.* 42 (2000) 143–150.
- [14] K.B. Metzger, P.E. Tolbert, M. Klein, J.L. Peel, W.D. Flanders, K. Todd, J.A. Mulholland, P.B. Ryan, H. Frumkin, Ambient air pollution and cardiovascular emergency department visits, *Epidemiology* 15 (2004) 46–56.
- [15] L.J. DeBell, R.W. Talbot, J.E. Dibb, J.W. Munger, E.V. Fischer, S.E. Frolking, A major regional air pollution event in the northeastern United States caused by extensive forest fires in Quebec Canada, *J. Geophys. Res.* 109 (2004) D19305.
- [16] S.Y. Ryu, J.E. Kim, H. Zhuanshi, Y.J. Kim, G.U. Kang, Chemical composition of post-harvest biomass burning aerosols in Gwangju Korea, *J. Air Waste Manage. Assoc.* 54 (2004) 1124–1137.
- [17] G. Cao, X. Zhang, S. Gong, F. Zheng, Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning, *J. Environ. Sci.* 20 (2008) 50–55.
- [18] A. Ezcurra, I. Ortiz de Zárate, P.V. Dhin, J.P. Lacaux, Cereal waste burning pollution observed in the town of Vitoria (northern Spain), *Atmos. Environ.* 35 (2001) 1377–1386.
- [19] J.G. Watson, J.C. Chow, Z. Lu, E.M. Fujita, D.H. Lowenthal, D.R. Lawson, Chemical mass balance source apportionment of PM_{10} during the Southern California air quality study, *Aerosol Sci. Technol.* 21 (1994) 1–36.
- [20] K.A. Christensen, H. Livberj, A field study of submicron particles from the combustion of straw, *Aerosol Sci. Technol.* 25 (1996) 185–199.
- [21] M.A. Yamasoe, P. Artaxo, A.H. Miguel, A.G. Allen, Chemical composition of aerosol particles from direct emissions of vegetation fires in Amazon Basin: water-soluble species and trace elements, *Atmos. Environ.* 34 (2000) 1641–1653.
- [22] J.H. Chen, K.H. Chang, The impact assessment of agricultural open burning on southern Taiwan's air quality using a multi-level nested photochemical model, in: A&WMA's 97th Annual Conference, Indianapolis, Indiana, USA, 2004, pp. 1855–1863.
- [23] M. Iranzo, J.V. Cañizares, L. Roca-Perez, I. Sainz-Pardo, S. Mormeneo, R. Boluda, Characteristics of rice straw and sewage sludge as composting materials in Valencia (Spain), *Bioresour. Technol.* 95 (2004) 107–112.
- [24] T. Novakov, S. Menon, T.W. Kirchstetter, Aerosol organic carbon to black carbon ratios: analysis of published data and implications for climate forcing, *J. Geophys. Res.* 110 (2005) D21205.
- [25] J.C. Chow, J.G. Watson, L.C. Pritchett, W.R. Pierson, C.A. Frazier, R.G. Purcell, The DRI thermal/optical reflectance carbon analysis system: description, evaluation and applications in US air quality studies, *Atmos. Environ.* 27A (1993) 1185–1201.
- [26] J.C. Chow, J.G. Watson, D. Crow, D.H. Lowenthal, T. Merrifield, Comparison of IMPROVE and NIOSH carbon measurements, *Aerosol Sci. Technol.* 34 (2001) 23–34.
- [27] Y.I. Tsai, M.T. Cheng, Characterization of chemical species in the atmospheric aerosols in a metropolitan basin, *Chemosphere* 54 (2004) 1171–1181.
- [28] W.L. Cheng, J.L. Pai, B.J. Tsuang, C.L. Chen, Synoptic patterns in relation to ozone concentrations in west-central Taiwan, *Meteorol. Atmos. Phys.* 78 (2001) 11–21.
- [29] Y.C. Chan, R.W. Simpson, G.H. Mctainsh, P.D. Vowles, D.D. Cohen, G.M. Bailey, Characterisation of chemical species in $PM_{2.5}$ and PM_{10} aerosols in Brisbane, Australia, *Atmos. Environ.* 31 (1997) 3773–3785.
- [30] S.Y. Ryu, B.G. Kwon, Y.J. Kim, H.H. Kim, K.J. Chun, Characteristics of biomass burning aerosol and its impact on regional air quality in the summer of 2003 at Gwangju, Korea, *Atmos. Res.* 84 (2007) 362–373.
- [31] J.C. Chow, J.G. Watson, Z. Lu, D.H. Lowenthal, C.A. Frazier, P.A. Solomom, R.H. Thuiller, K. Magliano, Descriptive analysis of PM_{25} and PM_{10} at regionally representative locations during SJVAQS/AUSPEX, *Atmos. Environ.* 30 (1996) 2079–2112.
- [32] F. Duan, X. Liu, T. Yu, H. Cachier, Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, *Atmos. Environ.* 38 (2004) 1275–1282.
- [33] A.G. Allen, A.A. Cardoso, G.O.da Rocha, Influence of sugar cane burning on aerosol soluble ion composition in Southeastern Brazil, *Atmos. Environ.* 38 (2004) 5025–5038.
- [34] S. Ohta, T. Okita, A chemical characterization of atmospheric aerosol in Sapporo, *Atmos. Environ.* 24A (1990) 815–822.
- [35] Y.I. Tsai, M.T. Cheng, Visibility and aerosol chemical compositions near the coastal area in Central Taiwan, *Sci. Total Environ.* 231 (1999) 37–51.
- [36] M.T. Cheng, Y.I. Tsai, Characterization of visibility and atmospheric aerosols in urban, suburban, and remote areas, *Sci. Total Environ.* 263 (2000) 101–114.
- [37] F. Echalar, A. Gaudichet, H. Cachier, P. Artaxo, Aerosol emissions by tropical forest and savanna biomass burning: characteristic trace elements and fluxes, *Geophys. Res. Lett.* 22 (1995) 3039–3042.
- [38] W. Maenhaut, I. Salma, J. Cafmeyer, H.J. Annegarn, M.O. Andreae, Regional atmospheric aerosol composition and sources in the eastern Transvaal, South Africa, and impact of biomass burning, *J. Geophys. Res.* 101 (1996) 23631–23650.
- [39] M.O. Andreae, P. Merlet, Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles* 15 (2001) 955–966.