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FTIR Spectra of Organic Functional Group Compositions in PM_{2.5} Collected at Chiang-Mai City, Thailand during the Haze Episode in March 2012

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Abstract: Organic functional groups of alcohol, alkane, alkene, alkyl halide, alkyne, amine, aromatic, ether, nitrile and nitro in PM_{2.5} were qualitatively identified by using FTIR techniques with the assistances of IRAffinity-1 Shimadzu. The PM_{2.5} were collected at the city center, sub-urban and rural area of Chiang-Mai city, Thailand during the haze episode in March 2012. The measured concentrations of PM_{2.5} at three air quality observatory sites were relatively high in comparison with those of other cities around the world during the agricultural waste-burning period. The predicted probabilistic Incremental Lifetime Cancer Risk (ILCR) of PM_{2.5} were consistently highest at the city center in both genders with the average values of 418±180 and 228±98 year⁻¹ for PM_{2.5} accumulated in male and female outdoor workers over exposure duration of 25 years, respectively. This indicates the influences of vehicular exhausts on the enhanced contents of PM_{2.5} irrespective of the “Haze Episode”. The results are in good agreement with those obtained from FTIR technique.

Key words: PM_{2.5}, Chiang-Mai, FTIR, incremental lifetime cancer risk

INTRODUCTION

The exceptional high levels of trans-boundary haze in ASEAN countries during the past few years encouraged several researchers, to critically assess the possible influences of biomass burning and haze on both ecosystem and human health (Huang *et al.*, 2013; Koe *et al.*, 2001; Nichol, 1997). Thailand, with its copious biomass resources, plays a leading role in the global biomass energy atlas. There is massive potential of biomass energy in northern part of Thailand due to abundant source of agricultural residues, woody biomass and other types of organic wastes (Thanarak, 2012). As a part of traditional/cultural practice, crop residue burning is repeated in the subsequent years in northern cities of Thailand for over the past few years. In Chiang-Mai, annual agricultural waste burning activities responsible for a widespread smoke-haze phenomenon that deteriorates public health quality, life style and incomes from tourism industries (Oanh and Leelasakultum, 2011). Although the effects of these regional-scale burning on physical environment are instant and evident, little is known about how chemical compositions such as organic functional groups are affected. By collecting this critical baseline information associated with the atmospheric contents of PM_{2.5}, trace gaseous species and other particulate organic functional groups will enable a more successful air quality management in Chiang-Mai and

afford a case study to better comprehend comparable incidences in other tropical cities around the world.

Several studies highlight airborne fine particulate matter PM_{2.5} as one of composite core air pollutants and Semi-Volatile Organic Compounds (SVOCs) as one of the major components of Persistent Organic Pollutants (POPs) has a crucial role on human health (Kalaierasan *et al.*, 2009; Pui *et al.*, 2014). As a consequence, there is an urgent need for comparatively rapid, easy operation and economically achievable analytical instrument for these organic chemicals. According to benefits of the Fourier Transform Infrared Spectroscopy (FTIR) technology, the infrared spectrum of organic functional groups in PM_{2.5} can be detected and thus used for both qualitatively and quantitatively analysis (Corry and Dillner, 2008, 2009; Maria *et al.*, 2002). Like other tropical cities, there is a lack of information associated with chemical compositions in particulate matter, particularly PM_{2.5}, in Chiang-Mai city. To our best knowledge, there are a limited number of publications connected with airborne chemical compositions in Thailand (Pongpiachan *et al.*, 2010, 2012a-c, 2013a, b; Pongpiachan, 2013a, b; Thumanu *et al.*, 2009). In this study, the author hypothesize that the application of FTIR coupled with various advanced statistical analysis assist in a better understanding of distribution pattern of organic functional compositions in PM_{2.5} which can be affected by not only biomass burnings but also traffic exhausts and other industrial

emissions. Overall the main principles of this study are: (1) To chemically characterize organic functional groups in $PM_{2.5}$ collected from three different observatory sites in Chiang-Mai city and (2) To evaluate the impacts of biomass burnings on the probabilistic Incremental Lifetime Cancer Risk (ILCR) of exposure on $PM_{2.5}$ during the haze episode in March 2012.

MATERIALS AND METHODS

Monitoring sites and sampling period: Collection of $PM_{2.5}$ was performed at three different air quality observatory sites in Chiang-Mai city. In all cases, the sampling period of individual sample was 24 h and carried out from 2nd to 31st March 2012 during the “Haze episode”. Intensive monitoring campaigns were conducted at three observatory sites namely Yupparat School Observatory Site (YOS; Yupparat School; 98.591917E, 18.472814N), Thanasarn Apartment Observatory Site (TOS; 98.591288E, 18.482055N) and Chiang-Mai City Hall Observatory Site (COS; 98.573558E, 18.501300N) (Fig. 1). The YOS is located in city center of Chiang-Mai. Air samples were collected at the air quality monitoring station of Pollution Control Department (PCD) representing the urban air mass of Chiang-Mai city (341 m above sea level). The TOS is

positioned at the rooftop of Thanasarn Apartment which is located in the middle of YOS and COS. The air samples collected at this site can be considered as the representative sub-urban air mass (341 m above sea level). The COS is situated at Chiang-Mai city hall which is approximately 5.33 km northwest of YOS. There were no obstructions in the vicinity of sampling equipment which was strategically positioned to be accessible to winds from all directions. In addition the sampling method is based on “Quality Assurance Guidance Document 2.12; Monitoring $PM_{2.5}$ in Ambient Air Using Designated Reference or Class I Equivalent Methods” (USEPA, 1998).

Sampling equipment: The MiniVol™ portable air samplers (Airmetrics) were used to collect $PM_{2.5}$ for 24 h at nine sampling sites. The MiniVol’s pump draws air at 5 L min^{-1} through a particle size separator (impactor) and then through a 47 mm filter. The $2.5\text{ }\mu\text{m}$ particle separation is achieved by impaction. Note that neither PM_{10} nor TSP samples were collected for this study.

FTIR and statistical analysis: An IRAffinity-1 Shimadzu equipped with the interferometer which is constantly optimized by a dynamic alignment mechanism and a built-in auto dryer assists ensure ease of maintenance,

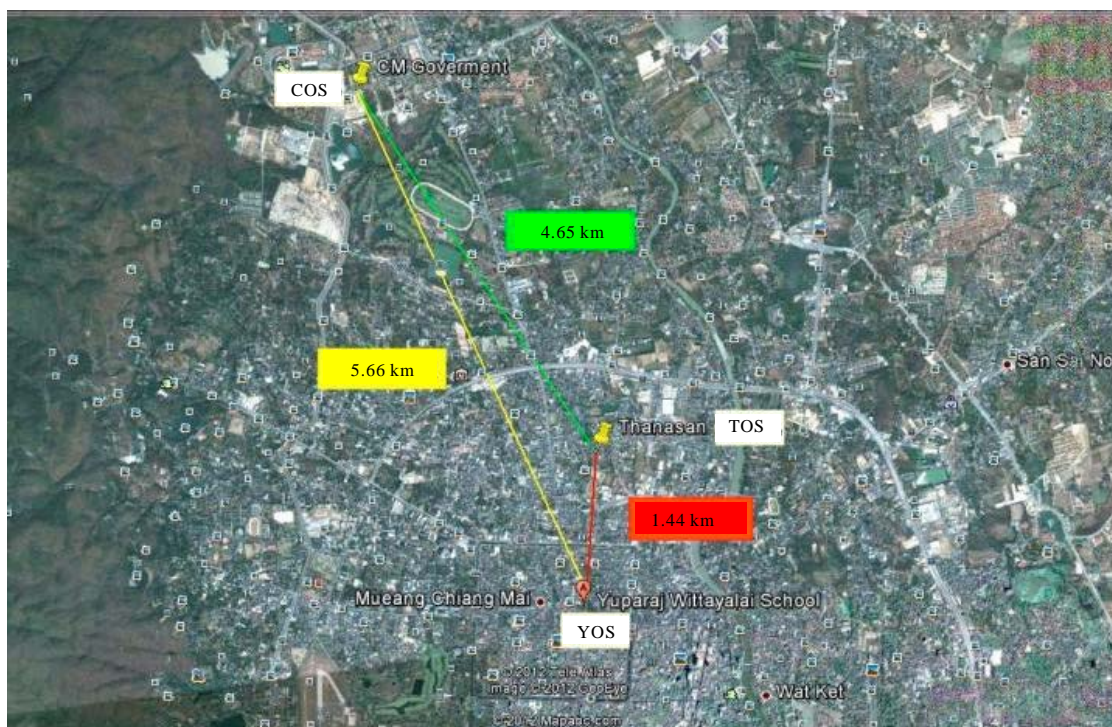


Fig. 1: Sampling positions of YOS, TOS and COS

offers the high S/N ratio (30,000:1, 1 min accumulation, neighborhood of 2,100 cm^{-1} , peak-to-peak), a maximum resolution of 0.5 cm^{-1} and compact dimensions. This instrument was operated by a high-performance LabSolutions IR software which provides a more easy performance for data processing and analysis. The major benefits of using LabSolutions IR software are its capability of detecting contaminants with a high degree of accuracy by combining Shimadzu's own algorithms (patent pending) with that of library spectra for common contaminants. Furthermore, this program makes pass/fail judgments associated with target samples in accordance with the tests specified in the library. The analytical procedure of FTIR transmission spectroscopy involves measuring the absorption of an infrared beam that is delivered straightforwardly through the sample and Quartz Fiber Filters (QFFs). While the subsequent absorption bands are distinctive to certain functional groups and are relative to the mass of sample exist, a functional group measurement can be conducted to provide for both quantitative and qualitative information

by applying FTIR. Subtraction of the blank (i.e., the spectrum of the empty QFFs) from the filter sample was conducted for each sample. Each spectrum was taken by averaging 16 scans at a resolution of 2 cm^{-1} . The spectra describe the absorbance of radiation as a function of wavenumber in the region of 4,000-400 cm^{-1} .

RESULTS AND DISCUSSION

Characteristic features of FTIR spectra: The characteristic IR absorption frequencies and its corresponding organic functional groups are illustrated in Table 1. The FTIR spectra for all samples at the absorption band in the 4,000-400 cm^{-1} intervals are compared in Fig. 2-6. Generally, the FTIR spectra of all samples are equitably similar to each other which can be described as follows:

- Faint broad declining absorption features in the 1050-1150 cm^{-1} intervals (C-O vibration of alcohol group (Fig. 2)) and 1000-1300 cm^{-1} intervals (C-O vibration of ether group Fig. 5))

Table 1: Characteristic IR absorption frequencies of organic functional groups (Silverstein *et al.*, 1981)

Functional group	Type of vibration	Characteristic absorptions (cm^{-1})	Intensity
Alcohol			
O-H	Stretch, H-bonded	3200-3600	Strong, broad
O-H	Stretch, free	3500-3700	Strong, sharp
C-O	Stretch	1050-1150	Strong
Alkane			
C-H	Stretch	2850-3000	Strong
-C-H	Bending	1350-1480	Variable
Alkene			
=C-H	Stretch	3010-3100	Medium
=C-H	Bending	675-1000	Strong
C=C	Stretch	1620-1680	Variable
Alkyl halide			
C-F	Stretch	1000-1400	Strong
C-Cl	Stretch	600-800	Strong
C-Br	Stretch	500-600	Strong
C-I	Stretch	500	Strong
Alkyne			
C-H	Stretch	3300	Strong, sharp
C=C	Stretch	2100-2260	Variable, not present in symmetrical alkynes
Amine			
N-H	Stretch	3300-3500	Medium (primary amines have two bands; secondary have one band, often very weak)
C-N	Stretch	1080-1360	Medium-weak
N-H	Bending	1600	Medium
Aromatic			
C-H	Stretch	3000-3100	Medium
C=C	Stretch	1400-1600	Medium-weak, multiple bands
Analysis of C-H out-of-plane bending can often distinguish substitution patterns			
Carbonyl			
C=O	Stretch	1670-1820	Strong (conjugation moves absorptions to lower wave numbers)
Ether			
C-O	Stretch	1000-1300 (1070-1150)	Strong
Nitrile			
C-N	Stretch	2210-2260	Medium
Nitro			
N-O	Stretch	1515-1560 and 1345-1385	Strong, two bands

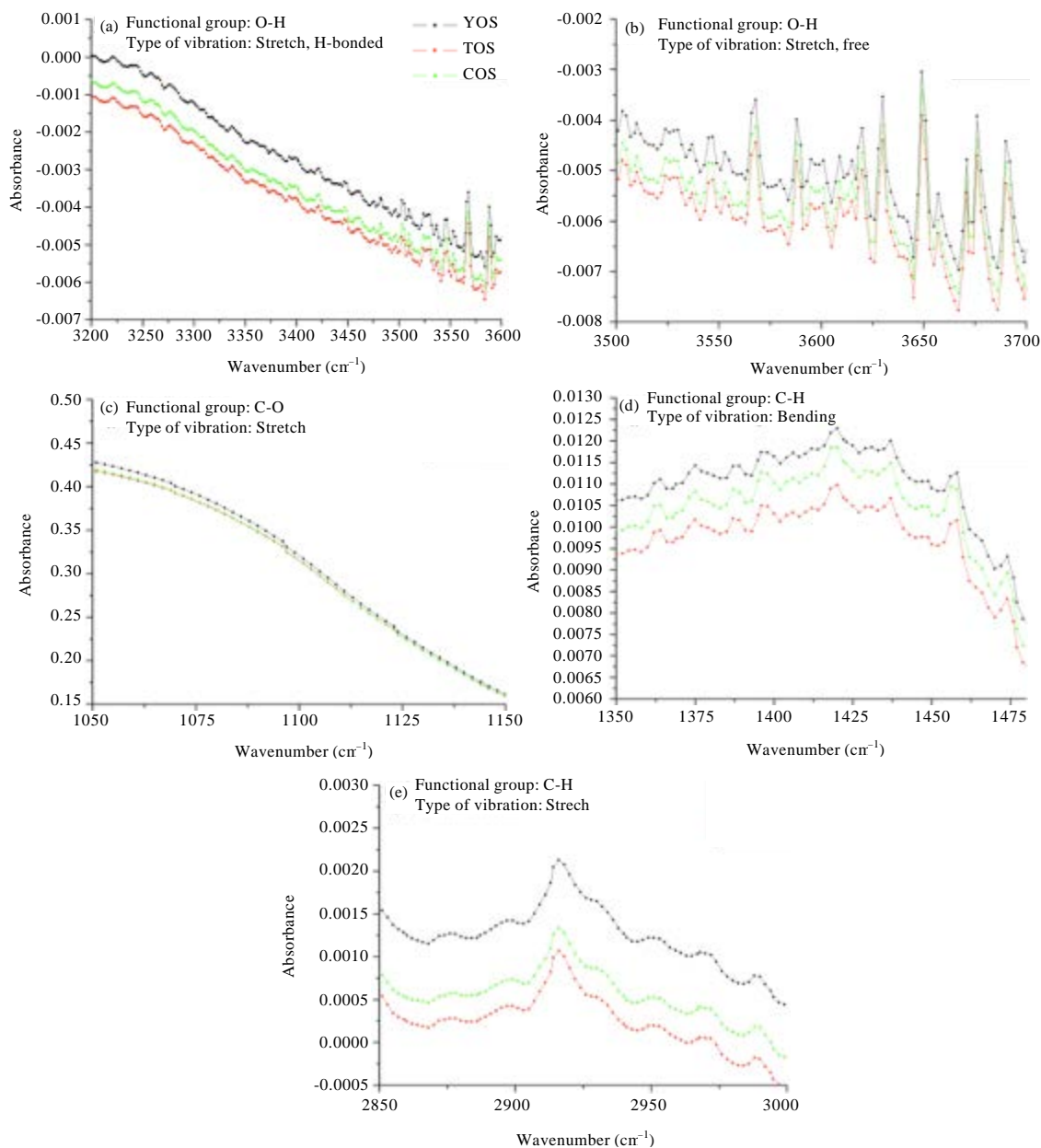


Fig. 2(a-e): FTIR spectra of alcohol for (a) O-H group at stretched H-bonded vibration, (b) O-H group at stretched free vibration, (c) C-O group at stretched vibration, (d) C-H group at bending vibration and (e) Spectra of alkane for C-H group at stretched vibration in $\text{PM}_{2.5}$ collected from YOS, TOS and COS

- Strong saw-tooth shaped absorption band in the 3500-3700 cm^{-1} intervals (O-H vibration of alcohol group (Fig. 2)), 1350-1480 cm^{-1} intervals (-C-H vibration of alkane group (Fig. 2)), 3010-3100 cm^{-1} intervals (=C-H vibration of alkene group (Fig. 3)), 1620-1680 cm^{-1} intervals (C=C vibration of alkene group Fig. 3)), 3000-3100 cm^{-1} intervals (C-H vibration of aromatic group (Fig. 5)), 1400-1600 cm^{-1} intervals (C=C vibration of aromatic group (Fig. 5)), 1670-1820 cm^{-1} intervals (C=O vibration of carbonyl group (Fig. 6)), 2210-2260 cm^{-1} intervals (C-N vibration of nitrile group (Fig. 6)), 1515-1560 cm^{-1}

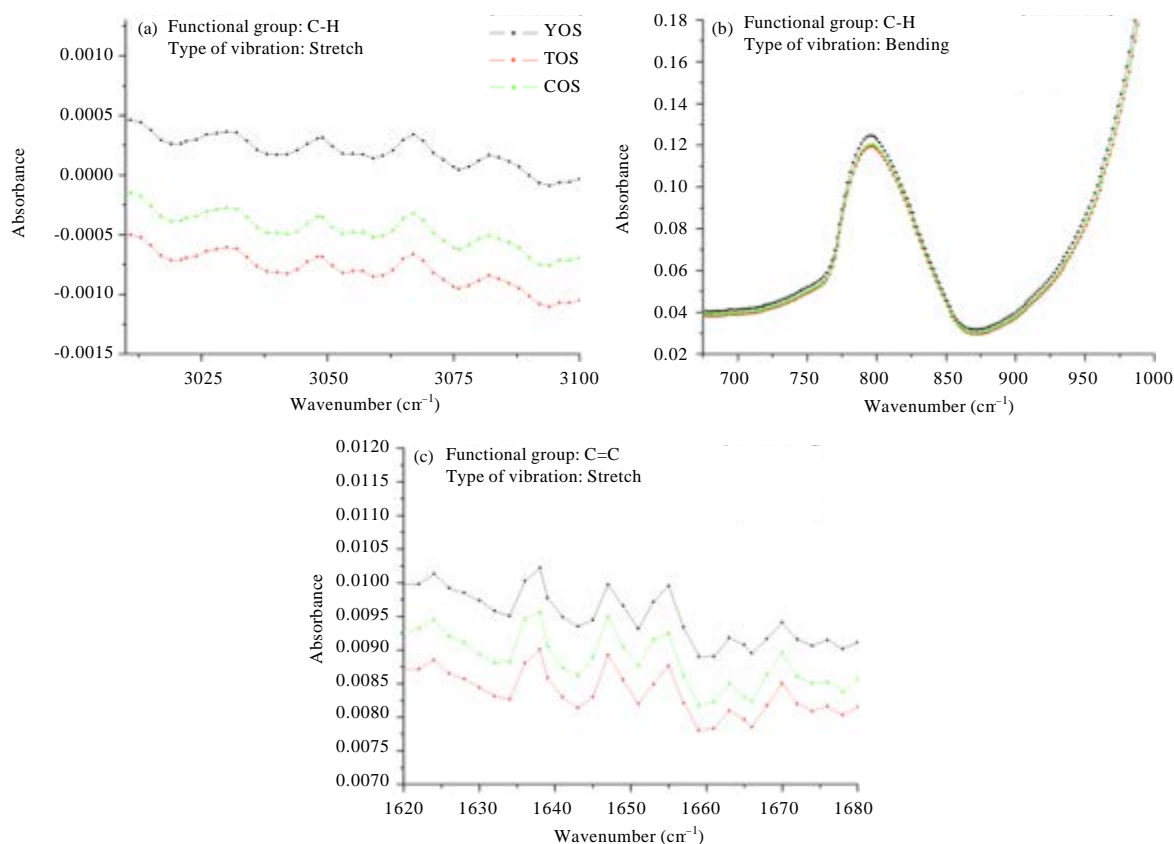


Fig. 3(a-c): FTIR spectra of alkene for (a) C-H group at stretched vibration, (b) C-H group at bending vibration and (c) C=C group at stretched vibration in PM_{2.5} collected from YOS, TOS and COS

intervals (N-O vibration of nitro group (Fig. 6)) and 1345-1385 cm⁻¹ intervals (N-O vibration of nitro group (Fig. 6))

- Asymmetric stretching of absorption peaks in the frequency range of 500-600 cm⁻¹ intervals (C-Br vibration of alkyl halide group (Fig. 4))
- Strong major absorption peaks in the frequency range of 750-850 cm⁻¹ intervals (C-H vibration of alkene group Fig. 3)) and 1000-1300 cm⁻¹ intervals (C-F vibration of alkyl halide group Fig. 4))

In spite of comparatively similar distribution pattern of FTIR spectra in the frequency range from 400-4000 cm⁻¹, some subtle differences were detected in the vibrational modes of organic components highlighting the dissimilarities of organic functional groups in PM_{2.5} collected from different emission sources. For instance, the prominent feature of the maximum intensity FTIR absorptions of YOS samples detected on the interval of 3200-3600, 1350-1475, 2850-3000, 3025-3100, 1620-1680, 2210-2260, 3300-3500, 3000-3100, 2210-2260 and

1350-1380 cm⁻¹ reflect the moderately high contribution of functional group OH (alcohol), C-H (alcohol), C-H (alkane), C-H (alkene), C=C (alkene), C-N (alkyne), N-H (amine), C-H (aromatic), C-N (nitrile) and N-O (nitro), respectively. Since, YOS is situated at the city center, this could be explained by the relatively high abundances of alcohol, alkane, alkene, alkyne, amine, aromatic, nitrile and nitro exist in traffic exhausts (Coronas *et al.*, 2009; Kotianova *et al.*, 2008; Slezakova *et al.*, 2013; Tsai *et al.*, 2013). This fact highlights the overwhelming influence of traffic emissions on the enhancement of these organic chemicals even during the agricultural waste-burning episode. On the contrary, the fairly homogeneous absorption spectra observed in the frequency range from 1000-1400 cm⁻¹, from 600-800 cm⁻¹ and from 500-600 cm⁻¹ reveal that functional group of alkyl halide C-F, C-Cl and C-Br appears to be completely unrelated to vehicle emissions.

PM_{2.5} concentrations and ILCR model: PM_{2.5} samples were collected effectively at three observatory sites

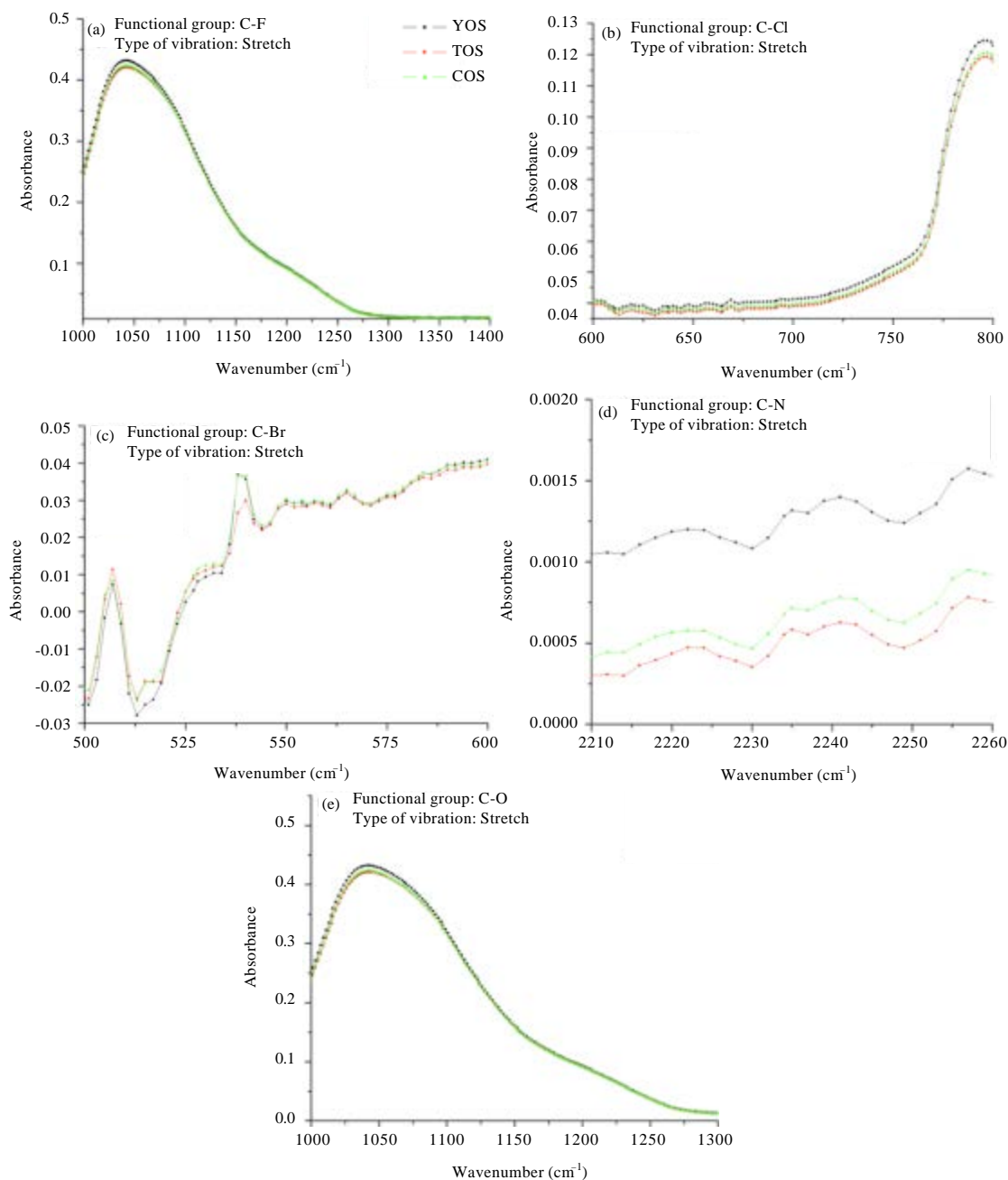


Fig. 4(a-e): FTIR spectra of alkyl halide for (a) C-F group at stretched vibration, (b) C-Cl group at stretched vibration, (c) C-Br group at stretched vibration, (d) Spectra of alkyne for C-N group at stretched vibration and spectra of ether for (e) C-O group at stretched vibration in $PM_{2.5}$ collected from YOS, TOS and COS

($n = 120$). Table 1 displays the statistical description of $PM_{2.5}$ detected during the “Haze episode” at YOS, TOS and COS. To evaluate the health risks connected with the occupational exposure to $PM_{2.5}$ of outdoor workers, the

Incremental Lifetime Particulate matter Exposure (ILPE) model was used and defined as:

$$ILPE = C \times IR \times t \times EF \times ED \quad (1)$$

Table 2: Atmospheric concentrations of PM_{2.5} (µg m⁻³) collected at different cities around the world

Country	City/ Province	Location	Year	Period	References	PM _{2.5}		ILCR-male (year ⁻¹)		ILCR-female (year ⁻¹)	
						Aver	Stdev	Aver	Stdev	Aver	Stdev
Thailand	Chiang-Mai	YOS	2012	March	This study	51	22	4.180E+02	1.803E+02	2.277E+02	9.822E+01
		COS	2012	March		49	19	4.016E+02	1.557E+02	2.188E+02	8.483E+01
	Bangkok	Dingdaeng	2010	Wet	Oanh <i>et al.</i> (2013)	25	6	2.049E+02	4.918E+01	1.116E+02	2.679E+01
			2010	Dry		57	13	4.672E+02	1.066E+02	2.545E+02	5.804E+01
China		Phaholyothin	2010	Dry		53	27	4.344E+02	2.213E+02	2.366E+02	1.205E+02
	Fujian	Yong'an	2007	Spring	Yin <i>et al.</i> (2012)	83	41	6.803E+02	3.361E+02	3.706E+02	1.830E+02
			2007	Autumn		90	30	7.377E+02	2.459E+02	4.018E+02	1.339E+02
			2008	Winter		79	22	6.476E+02	1.803E+02	3.527E+02	9.822E+01
Nigeria	Lagos	Mushin	2007	Week1 (July-August)	Ezeh <i>et al.</i> (2014)	10	3	8.197E+01	2.459E+01	4.465E+01	1.339E+01
			2007	Week 2 (July-August)		18	6	1.475E+02	4.918E+01	8.036E+01	2.679E+01
			2007	Week 3 (July-August)		9	3	7.377E+01	2.459E+01	4.018E+01	1.339E+01
			2007	Week 4 (July-August)		12	4	9.836E+01	3.279E+01	5.357E+01	1.786E+01
Argentina	Cordoba	Traffic area	2007	Week 5 (July-August)	Lopez <i>et al.</i> (2011)	5	1	4.098E+01	8.197E+00	2.232E+01	4.465E+00
		University campus	2009-2010	July-April		71	21	5.820E+02	1.721E+02	3.170E+02	9.376E+01
			2009-2010	July-April		67	18	5.492E+02	1.475E+02	2.991E+02	8.036E+01
			2003-2004			25	NA	2.049E+02	NA	1.116E+02	NA
Spain	Barcelona		2001		Pey <i>et al.</i> (2008)	22	NA	1.803E+02	NA	9.822E+01	NA
	Tarragona		2001		Moreno <i>et al.</i> (2006)	22	NA	2.213E+02	NA	1.205E+02	NA
	Barcelona		1995-2005		Querol <i>et al.</i> (2008)	27	NA	1.639E+02	NA	8.929E+01	NA
	Onda					20	NA	1.967E+02	NA	1.071E+02	NA
Greece	Alcora					24	NA	1.967E+02	NA	9.822E+01	NA
	Cartagena					22	NA	1.803E+02	NA	8.929E+01	NA
	Mallorca					20	NA	1.639E+02	NA	8.929E+01	NA
	Athens		1999-2000		Chaloulakou <i>et al.</i> (2003)	40	NA	3.279E+02	NA	1.786E+02	NA
Greece	Akrotiri (Crete)		2004-2006		Lazaridis <i>et al.</i> (2008)	25	NA	2.049E+02	NA	1.116E+02	NA
	Finokalia		2004-2006		Gerasopoulos <i>et al.</i> (2007)	18	NA	1.475E+02	NA	8.036E+01	NA
	Cairo		1999, 2002		Abu-Allaban <i>et al.</i> (2007)	86	NA	7.049E+02	NA	3.840E+02	NA
	Israel	Ashdod Urb	1999		Peled <i>et al.</i> (2005)	24	NA	1.967E+02	NA	1.071E+02	NA
Turkey	Ashkelon Urb		1999			24	NA	1.967E+02	NA	1.071E+02	NA
	Sderot Urb		1999			29	NA	2.377E+02	NA	1.295E+02	NA
	Erdemli		2001-2002			9.7	NA	7.951E+01	NA	4.331E+01	NA
	Izmir (Urban)		2004-2005		Kocak <i>et al.</i> (2007)	64	NA	5.246E+02	NA	2.857E+02	NA
Lebanon	Izmir (Suburban)				Yadkin and Bayram (2008)	24	NA	1.967E+02	NA	1.071E+02	NA
	HH		2006-2007			28	NA	2.295E+02	NA	1.250E+02	NA
	BH		2004-2005			39	NA	3.197E+02	NA	1.741E+02	NA
	Bliss		2003			41	NA	3.361E+02	NA	1.830E+02	NA

Department of health 2005 (average body weight), Male: 58.25±9.76, Female: 54.95±10.48, t: Daily Exposure time span (6 h day⁻¹, for two shifts), EF: Exposure frequency (250 day year⁻¹, upper bound value), ED: Exposure duration (25 years, upper-bound value), BW: Body weight, AT: Average time. Life expectancy (National Statistical Office Thailand, survey of population change 1995-1996); Male: 69.9 year, Female: 74.9 year. Inhalation rate, Male: 0.89 m³ h⁻¹, Female: 0.49 m³ h⁻¹

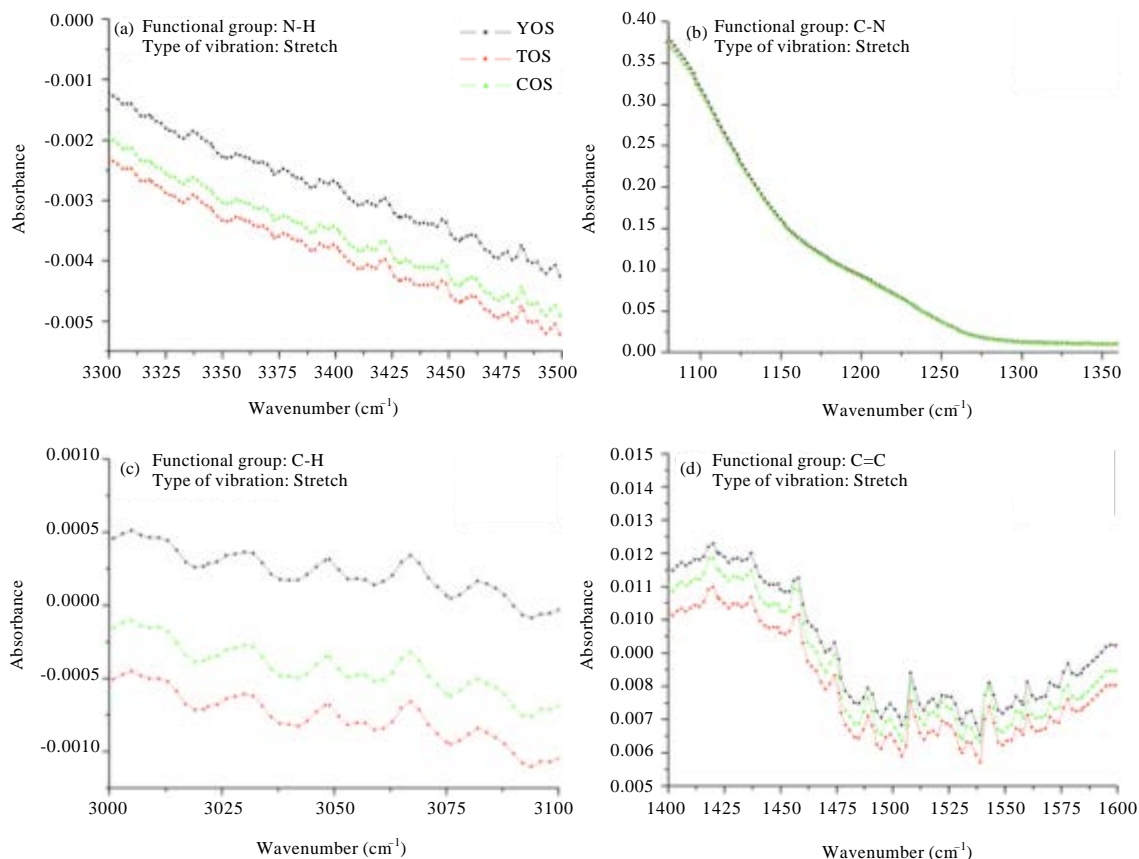


Fig. 5(a-d): FTIR spectra of amine for (a) N-H group at stretched vibration, (b) C-N group at stretched vibration. Spectra of aromatic hydrocarbon for (c) C-H group at stretched vibration and (d) C=C group at stretched vibration in $PM_{2.5}$ collected from YOS, TOS and COS

Where:

ILPE = Incremental lifetime particulate matter exposure (g)

C = $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$)

IR = Inhalation rate ($m^3\ h^{-1}$) (US EPA, 1994; Male: 0.89; Female: 0.49 $m^3\ h^{-1}$)

t = Daily exposure time span (6 h day^{-1} , for two shifts)

EF = Exposure frequency (250 days $year^{-1}$ ^a, upper-bound value)

ED = Exposure duration (25 years^a, upper-bound value)

Incremental Lifetime Cancer Risk (ILCR) can be described as follows:

$$ILCR = \frac{ILPE}{BW \times AT} \quad (2)$$

Where:

BW = Body Weight (kg) (Male: 58.25±9.76 kg; Female: 54.95±10.48 kg; Department of health 2005; Average Body Weight)

AT = Average Time (Year) (Life Expectancy for Male: 69.9 year; Female: 74.9 year; Source: National Statistical Office Thailand, Survey of Population Change 1995-1996)

The ILCR was adapted from the model used to assess traffic policemen exposure to PAHs during their work time in China (Hu *et al.*, 2007). The predicted ILCR levels in outdoor workers are illustrated in Table 2. The expected ILCR of $PM_{2.5}$ were constantly highest at YOS in both genders during the “Haze episode” with the average values of 418±180 and 228±98 $year^{-1}$ for $PM_{2.5}$ accumulated in male and female outdoor workers over

^aAdapted from Human Health Evaluation Manual

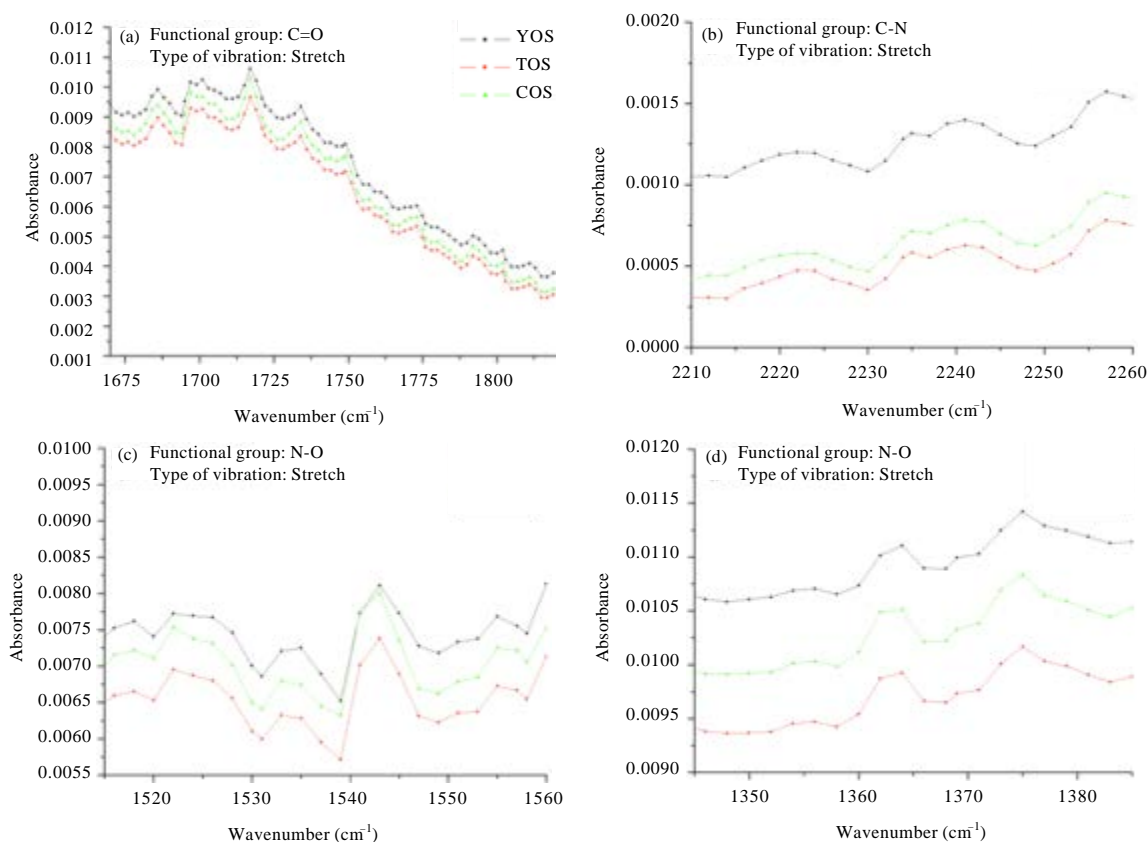


Fig. 6(a-d): FTIR spectra of (a) Carbonyl C=O group at stretched vibration, (b) Nitrile C-N group at stretched vibration, and (c and d) Nitro N-O group at stretched vibration for different intervals in PM_{2.5} collected from YOS, TOS and COS

exposure duration of 25 years correspondingly. This re-emphasizes the impacts of traffic emissions on the increased concentrations of PM_{2.5} irrespective of the “Haze episode”. By comparing with those of previous studies around the world, it is also interesting to note that the decreasing order of ILCR_{male} is 738±246 year⁻¹ (Fujian), 705 year⁻¹ (Cairo), 582±172 year⁻¹ (Cordoba), 467±107 year⁻¹ (Bangkok), 418±180 year⁻¹ (YOS), 402±156 year⁻¹ (COS), whilst the lowest values was observed in 41±8.2 year⁻¹ (Lagos). Similarly the declining order of ILCR_{female} is 402±134 year⁻¹ (Fujian), 384 year⁻¹ (Cairo), 317±94 year⁻¹ (Cordoba), 255±107 year⁻¹ (Bangkok), 228±98 year⁻¹ (YOS), 219±84 year⁻¹ (COS), whereas the smallest values was found in 22±4.5 year⁻¹ (Lagos). The relatively high ILCR values in YOS and COS can be ascribed as consequences of multiple reasons. Firstly, an inversion is more tending to occur during the cold period when the angle of the sun is extremely low and largely in mountainous provinces such as Chiang-Mai and could hypothetically be responsible for

the enhancement of PM_{2.5} concentrations in March 2012. Secondly, the increased hot spots coupled with vehicular exhausts could greatly affect the increased level of PM_{2.5} contents and thus the ILCR values during the observing period.

CONCLUSION

The investigation of FTIR spectra of PM_{2.5} collected during the “Haze episode” at three different air quality observatory sites in Chiang-Mai city was carried out from 2nd to 31st March 2012 by using IRAffinity-1 Shimadzu. Similar distribution pattern of FTIR spectra in the frequency range from 400-4000 cm⁻¹ were observed in PM_{2.5} samples at all observatory sites highlighting the effects of homogeneous mixing of air masses even during the agricultural waste-burning period. In spite of relative uniformity in its turbulence, the highest contribution of alcohol, alkane, alkene, alkyne, amine, aromatic, nitrile and nitro were detected at YOS indicating that traffic

emissions still play an important role in governing the organic function group contents in fine particles irrespective of agricultural waste-burning events.

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