Air Quality Measurements from the Southern Particulate Matter Supersite in Taiwan

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Abstract

This study introduces the Southern Particulate Matter Supersite in Taiwan, which began operating on April 1, 2005. The supersite has one core station and three satellite stations for monitoring the properties of particulate matter (PM) and emission sources in southern Taiwan. High-time resolution (1–30 minutes) data for physical and chemical properties of ambient PM are acquired continuously. Measurement data are as follows: (1) $PM_{2.5}$ (PM with aerodynamic diameters < 2.5 μm) and PM_{10} (PM with aerodynamic diameters < 10 μm) mass concentrations; (2) $PM_{2.5}$ compositions of sulfate, nitrate and carbon; (3) particle light scattering and absorption; (4) particle number concentrations in various size fractions between 10 nm and 20 μm; (5) related precursor gases such as NO_y, H_2O_2, and NH_3; and, (6) meteorological variables. Most measurements are unique to the study area and can be used to elucidate the causes of PM pollution and evaluate PM exposure and adverse health effects. In addition to describing the sampling location, measurements and data archiving, future challenges for the supersite are discussed as well.

Keywords: Supersite; Taiwan; $PM_{2.5}$; PM_{10}; Nitrate; Sulfate; Carbon.

INTRODUCTION

Epidemiological studies have identified the relationships between ambient PM concentrations and adverse health effects (e.g., Vedal, 1997; Guo *et al.,* 1999; Pope *et al.,* 2002; Brunekreef and Holgate, 2002; Englert, 2004; Harrison *et al.,* 2004a; Migilaretti and Cavallo, 2004; Anderson *et al.,* 2005; Peden, 2005; Chow *et al.,* 2006a;

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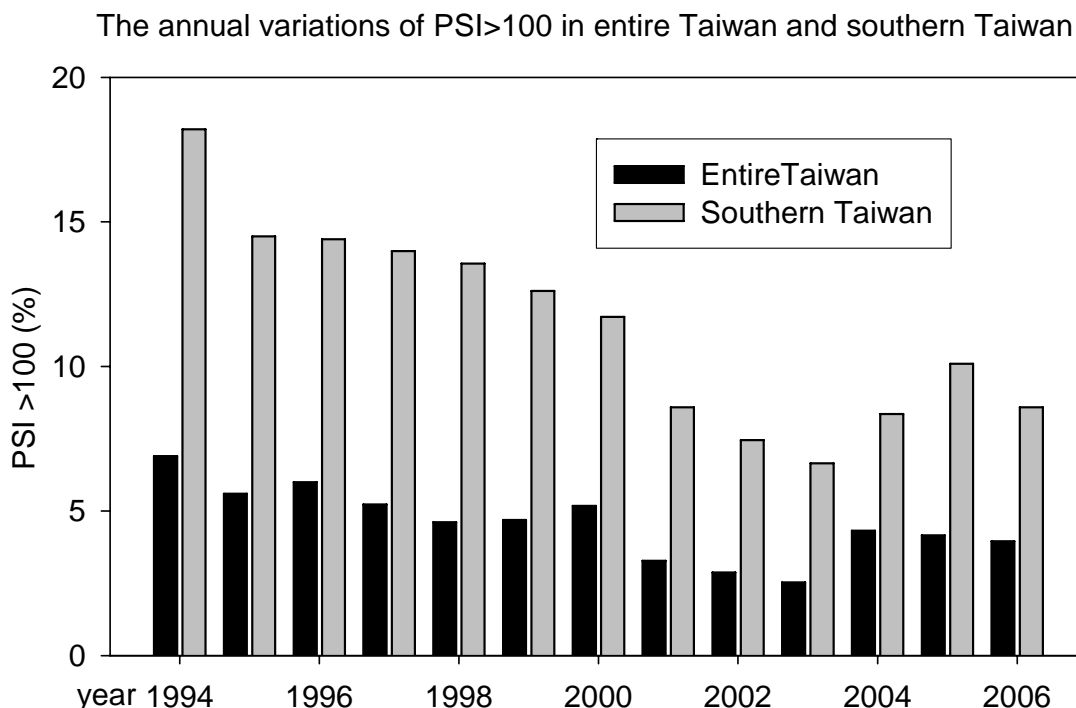E-mail address: chlin@mail.fy.edu.tw

Fig. 1. Annually occurring frequencies of PSI > 100 averaged over the whole of Taiwan and southern Taiwan, respectively, from the Taiwan Air Quality Monitoring Network (TAQMN) air stations during 1994-2006.

Pope and Dockery, 2006; Mauderly and Chow, 2008). To protect public health, the U.S. Environmental Protection Administration (EPA) implemented National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ in 1997 (Federal Register, 1997) and amended these standards in 2006 (Federal Register, 2006). The $PM_{2.5}$ mass, composition, and size-resolved particle number concentrations are important factors when developing effective emission control strategies and are needed when determining correlations between PM exposure and adverse health effects (National Research Council (RC), 1998; 1999; 2001; 2004). The U.S. EPA constructed Particulate Matter Supersites in 1999–2007 (U.S. EPA, 1999; 2000;

Solomon and Hopke, 2008) with objectives to: (1) evaluate advanced measurement methods, (2) further understand atmospheric processes, and (3) establish relationships between PM and adverse health effects (Watson *et al.,* 2000). Over 400 studies have focused on U.S. Supersites (e.g., Solomon and Hopke, 2008; Chow *et al.,* 2008; Solomon and Sioutas, 2008; Watson *et al.,* 2008). Several special journal issues (Brock *et al.,* 2004; Middlebrook *et al.,* 2004; Ondov *et al.,* 2004; Chow *et al.,* 2004; Stanier and Solomon, 2006; Wittig and Solomon, 2006; Geller and Solomon, 2006; Chow and Solomon, 2006; Solomon and Hopke, 2008) synthesize major findings.

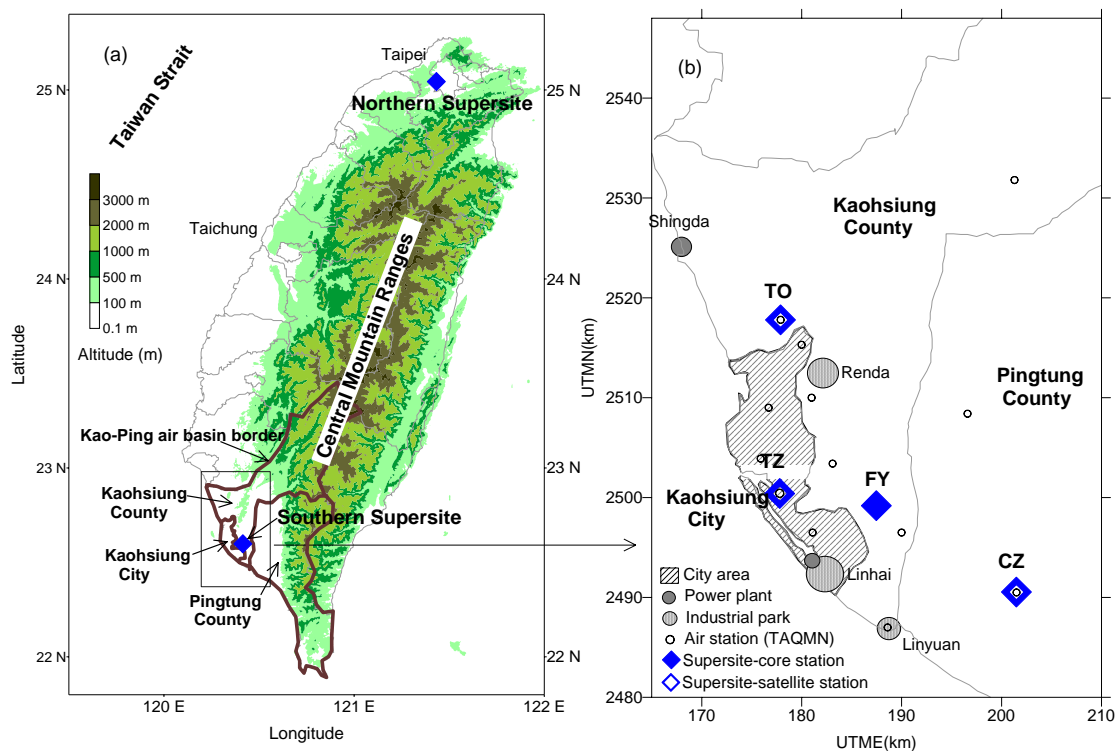Air pollution in Taiwan has been measured by the Taiwan Air Quality

Fig. 2. (a) Locations of the Northern and Southern Particulate Matter Supersites in Taiwan, and (b) The core (Fooyin, FY) and three satellite stations (Chiautou, TO; Chenjen, TZ; Choujau, CZ) of the Southern Supersite and its soundings. TAQMN stands for Taiwan Air Quality Monitoring Network.

Monitoring Network (TAQMN) since 1993 (Chang and Lee, 2006). The TAQMN consists of more than 70 air stations distributed throughout Taiwan (Taiwan EPA, 2007). Each air station measures hourly concentrations of criteria pollutants (i.e., SO_2, CO, NO_2, O_3, PM_{10}) and meteorological variables (i.e., wind speed, wind direction, temperature, relative humidity). The daily pollutant standards index (PSI) at each air station is calculated by the Taiwan EPA based on daily concentrations of criteria pollutants at each station. The calculated PSI value is proportional to pollutant concentrations with PSI = 100 equal to Taiwan's NAAQS. Therefore, the percentage of times the PSI >

100 is the frequency that Taiwan's NAAQS are violated. Fig. 1 shows annual percentage PSI > 100 during 1994–2006, averaged for all TAQMN air stations and in southern Taiwan. The annual percentages decreased from 1994–2003 (Fig. 1) as the Taiwan EPA has implemented many control strategies (e.g., air pollution fees, emission permits, and environmental impact assessments) over the last decade. However, these percentages have increased after 2003, suggesting the need for further control measures. This increase may be due to the increase in the number of vehicles, 6–10% per year (Lin *et al.,* 2002). An improved understanding of the causes of excessive air pollution and source-receptor relationships is needed. The

need for enhanced temporally, chemically, and size-resolved PM data prompted the establishment of Taiwan Particulate Matter Supersites (Chan, 2000). Currently, Taiwan has two supersites (Fig. 2), the Northern Particulate Matter Supersite (Northern Supersite) and Southern Particulate Matter Supersite (Southern Supersite). The Northern Supersite started monitoring in March, 2002 (Lee, 2002; Lee *et al.,* 2006; Chang *et al.,* 2007), and the Southern Supersite began in April, 2005 (Wu *et al.,* 2005; Lin *et al.,* 2006a). The objectives of the Southern Supersite are to characterize the physical and chemical properties of ambient PM, and provide high time-resolved measurements for the study of PM pollution, exposure and adverse health effects. This study reported on in this paper documents the locations, surroundings, and measurements of the Southern Supersite, as well as its future challenges.

SOURCES AND METEOROLOGICAL CHARACTERISTICS

The Southern Supersite (Fig. 2) is located in the Kao-Ping air basin in southern Taiwan (Wu *et al.,* 2005; Lin *et al.,* 2006a). The Kao-Ping air basin consists of three administrative districts—Kaohsiung City, Kaohsiung County, and Pingtung County—that encompass 5,700 km^2 and 3.7 million inhabitants. The western part of the basin is a coastal plain bordered by the Taiwan Strait. Most of the population and

industrial activities are located in the southwest, in Kaohsiung City and its suburbs. Most of the coastal plain is used for rice fields. The eastern part of the basin is bordered by the Central Mountain Range, which has altitudes of 2,500–4,000 m.

Pollution sources

Mobile and stationary sources are important in the Kao-Ping air basin. According to the Taiwan Emission Database 6.1 (Taiwan EPA, 2006), the 2003 NO_x emissions were 164,000 metric tons/year, of which 40% and 60% come from mobile and stationary sources, respectively. Mobile sources are concentrated in Kaohsiung City. Stationary sources are primarily located within three industrial parks, Linhai, Renda, and Linyuan (Fig. 2). Linhai, the largest industrial park, is located south of Kaohsiung City. Linhai has a coal-fired power plant (2,700 MW), steel mills, and petroleum refineries. Both Renda (next to the northeastern border of Kaohsiung City) and Linyuan (10 km from the southeastern border of Kaohsiung City) are petrochemical industrial parks. In addition, the Shingda coal-fired power plant (4,300 MW) (adjacent to the northwest border of the Kao-Ping air basin) is the single largest stationary source in southern Taiwan.

Meteorology

Southern Taiwan is located in a subtropical, coastal area. Fig. 3 shows the monthly variations in temperature, relative humidity and rainfall, based on a 30-year
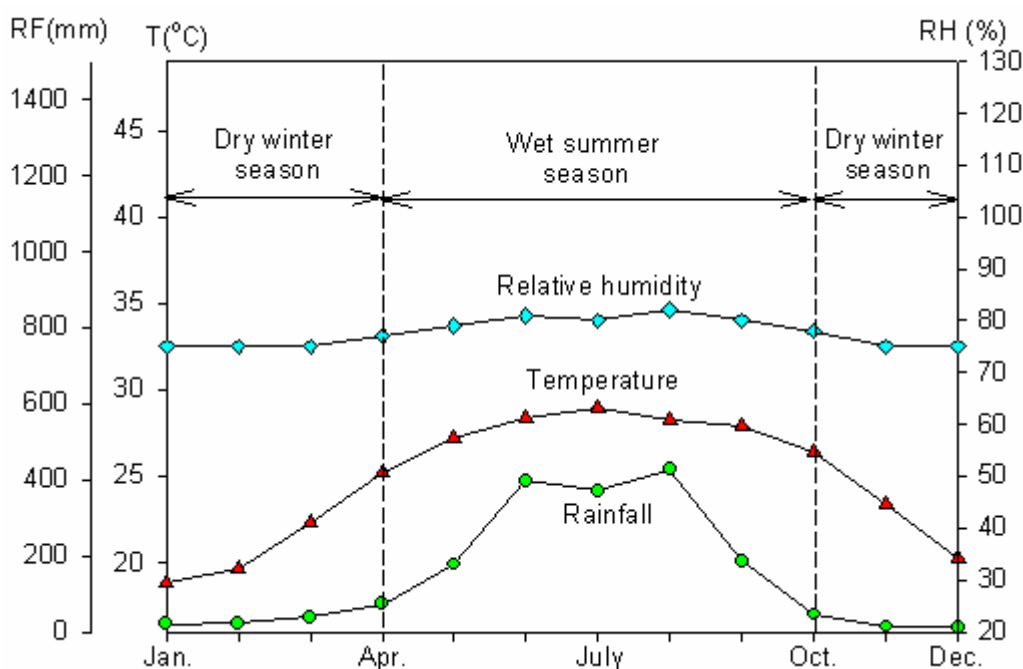
Fig. 3. Monthly averages of temperature, relative humidity, and rainfall based on a 30-year record for 1971–2000 at the Kaohsiung meteorological station in southern Taiwan.

record for 1971–2000 at the Kaohsiung meteorological station, which is maintained by Taiwan's Central Weather Bureau. Taiwan generally has only two seasons—wet (summer, April–September) and dry (winter, October–March) seasons. Rainfall is principally concentrated in summer. Temperature and humidity are relatively high in summer. However, variations in temperature (18.9–29.9°C), and humidity (75–82%) are relatively stable due to the coastal environment.

Air pollution levels are low in summer and high in winter (Chen *et al.,* 2004a). During summer, the synoptic weather is dominated by typhoons and Pacific anticyclones. The air is typically clean during typhoon periods due to the strong winds and heavy showers that impact

Taiwan. Pollution levels are also low during Pacific anticyclones due to clean southwesterly winds and deep daytime mixing depths. During winter, the synoptic weather is dominated by continental anticyclones traveling from China to the East China Sea. Western Taiwan experiences northerly winds due to channeling from the Central Mountain Range. Pollutants emitted in central and northern Taiwan can be transported southward. When the synoptic flow is easterly, western Taiwan is on the lee side of the Central Mountain Range. In this circumstance, upwind air pollution transport and accumulation of local emissions are conducive to pollution episodes in the Kao-Ping air basin.

Monitoring stations

The Southern Supersite has one core and three satellite stations. The core station, located at Fooyin University (22.603°N, 120.388°E) (FY), is approximately at the center of the industrial activities in the Kao-Ping air basin. The FY core station is 7 km from the Kaohsiung City center to the west, 8 km from the Linhai industrial park to the southwest, 15 km from the Renda industrial park to the north, and 13 km from the Linyuan industrial park to the south (Fig.2). This core station has two instrumented containers on the rooftop of a six-story building. Sampling inlets are 1.0–1.5 m above the containers. The three satellite stations are located at Chiautou (TO) (22.759°N, 120.298°E), Chenjen (TZ) (22.606°N, 120.301°E) and Choujau (CZ) (22.513°N, 120.529°E) (Fig. 2). These satellite stations are collocated with existing TAQMN air stations. The TO station, roughly 3 km from the northern border of Kaohsiung City and 19 km northwest of the core station, is an upwind site during the dry winter season. The TZ station, located in Kaohsiung City, is next to Kaohsiung Harbor and 8 km west of the core station. The TZ station characterizes PM in a vehicle-dominated environment. The CZ station, located in Pingtung County 18 km southeast of the core station, is a downwind site.

SUPERSITE MEASUREMENTS

Table 1 summarizes air quality and meteorological variables acquired at the Southern Supersite since April 1, 2005. These in-situ, continuous and high-time resolved measurements are as follows: (1) $PM_{2.5}$ and PM_{10} mass concentrations; (2) $PM_{2.5}$ chemical composition (i.e., sulfate [SO_4^{2-}], nitrate [NO_3^-] and organic and elemental carbon [OC and EC]); (3) $PM_{2.5}$ light scattering and absorption, and solar radiation; (4) particle number concentrations in various size fractions between 10 nm and 20 μm; (5) pollutants and PM precursor gases (i.e., O_3, CO, NO/NO_y, NH_3, H_2O_2, BTEX [benzene, toluene, ethylbenzene and xylene]); and, (6) meteorological variables (i.e., wind speed, wind direction, temperature and relative humidity). Except for PM mass concentrations and meteorological parameters, this is the first time that these variables have been continuously measured in southern Taiwan. As well, hourly criteria pollutants and meteorological measurements are available at the three satellite stations from the TAQMN. Table 2 lists the minimal detectable limits, precisions, and calibration methods for measurements.

Measurements

Both $PM_{2.5}$ and PM_{10} mass concentrations are measured by a tapered-element oscillating microbalance preceded by a sample equilibrium system (SES-TEOM) (Meyer *et al.,* 2000). The SES-TEOM, which is operated at 30°C to minimize volatilization, has a diffusion drier that

Table 1. Air quality and meteorological variables continuously monitored since April 1, 2005 at the core and three satellite stations of the Southern Supersite in Taiwan.

[a] Station code	[b] Observable	Instrument	Method	Particle Size or Measuring range	Averaging Time
	Particle mass concentrations				
FY,TZ, CZ,TO	PM_{10} Mass	RP 1400a (Thermo Scientific, USA)	SES-TEOM method	<10 μm	10 min
FY,TZ, CZ,TO	$PM_{2.5}$ Mass	RP 1400a (Thermo Scientific, USA)	SES-TEOM method	<2.5 μm	10 min
	Particle compositions				
FY,TZ, CZ,TO	[*] $PM_{2.5}$ SO_4^{2-}	RP 8400S (Thermo Scientific, USA)	Flash volatilization of sulfate and followed by SO_2 detection	<2.5 μm	30 min
FY,TZ, CZ,TO	[*] $PM_{2.5}$ NO_3^-	RP 8400N (Thermo Scientific, USA)	Flash volatilization of nitrate and followed by NO_x detection	<2.5 μm	30 min
FY,TZ, CZ,TO	[*] $PM_{2.5}$ OC/EC	Sunset OC/EC Field Instrument (Sunset Laboratory, USA)	Thermal/optical transmission method	<2.5 μm	30 min
	Particle light scattering and absorption				
FY	[*] Particle scattering coefficient	Ecotech M9003 (Ecotech, USA)	A single-wavelength (525 nm) nephelometer	<2.5 μm	1 min
FY	[*] Particle absorption coefficient	Magee AE-31 (Magee Scientific, USA)	A seven-wavelength (370, 470, 520, 590, 660, 880, 950 nm) aethalometer	<2.5 μm	5 min
FY	[*] Solar radiation	POM-01 (Kipp & Zonen, The Netherlands)	A seven-wavelength (315, 340, 380, 400, 443, 500, 675 nm) sky radiometer	[c]NA	10 min
	Particle number concentrations in different size fractions				
FY	[*] Fine particle number concentrations	Grimm 1.108 (GRIMM, Germany)	A 31 size bins of aerosol spectrometer	0.30-20 μm	10 min
FY	[*] Ultrafine particle number concentrations	Grimm SMPS+C (GRIMM, Germany)	A 256 size bins of aerosol sizer.	10-900 nm	7 min

Table 1. Air quality and meteorological variables continuously monitored since April 1, 2005 at the core and three satellite stations of the Southern Supersite in Taiwan. (Continued)

[a] Station code	[b] Observable	Instrument	Method	Particle Size or Measuring range	Averaging Time
	Precursor gases				
FY	Ozone (O_3)	Ecotech 9810 (Ecotech, Australia)	UV absorption method	0-20 ppm	1 min
FY	Carbon monoxide (CO)	Horiba APMA 360 (Horiba, Japan)	Infrared absorption method	0-10 ppm	3 min
FY	[*] Reactive nitrogen (NO/NO_y)	Thermo 42 CY (Thermo Env. Instruments, USA)	Chemiluminescent method associated with external converter	0-200 ppb	1 min
FY	[*] Ammonia (NH_3)	Thermo 17 C (Thermo Env. Instruments, USA)	Chemiluminescent method associated with external stainless steel converter	0-200 ppb	1 min
TZ,CZ, TO	[*] Ammonia (NH_3)	ESA AC32M (Environnement S.A, USA)	Chemiluminescence method associated with external stainless steel converter	0-200 ppb	1 min
FY	[*] Hydrogen Peroxide (H_2O_2)	Yankee AL-2021 (Yankee Env. System, USA)	Wet fluorescent detection method	0-200 ppb	0.5 min
FY,TZ	[d] BTEX	ChromaPID (Chromatotec, USA)	[d] GC/PID method	0-1 ppm	30 min
	Meteorological variables				
FY	Wind speed/direction	05305L (Met One, USA)	High-sensitivity wind vane and anemometer	[c] NA	1 min
FY	Temperature	CS500L (Met One, USA)	Platinum resistance sensor	[c] NA	1 min
FY	Relative humidity	CS500L (Met One, USA)	Capacitance sensor	[c] NA	1 min

[a] FY stands for the Fooyin core station and TO, TZ and CZ for the Chiautou, Chenjen and Choujau satellite stations, respectively.

[b] Hourly criteria pollutants (i.e., SO_2, CO, NO_2, O_3 and PM_{10}) and meteorological variables (i.e., temperature, relative humidity, rainfall, wind speed and direction) are available at the air stations of TAQMN for the three satellite stations.

[c] Not applicable.

[d] BTEX stands for benzene, toluene, ethylbenzene and xylene; GC/PID for gas chromatograph/ photo ionization detector.

[*] This variable are continuously measured for the first time in Southern Taiwan.

Table 2. Summary of minimal detectable limits, precisions, and calibration methods for the measurements at the Southern Supersite in Taiwan.

Measurement (Instrument)	Minimal detectable limit	Precision	Calibration and/or testing method
Particle mass concentration			
PM_{10} mass (RP 1400a)	1.5 μg/m^3	± 10%	Calibrated monthly by weighted filter substance.
$PM_{2.5}$ mass (RP 1400a)	1.5 μg/m^3	± 10%	Calibrated monthly by weighted filter substance.
$PM_{2.5}$ composition			
SO_4^{2-} (RP 8400S)	0.4 μg/m^3	± 15%	Daily zero and span testing with certified SO_2; monthly calibrated with certified SO_2 with dynamic dilution and SO_4^{2-} solution.
NO_3^- (RP 8400N)	0.4 μg/m^3	± 15%	Daily zero and span testing with certified NO; monthly calibrated with certified NO with dynamic dilution and NO_3^- calibration solution.
OC/EC (Sunset OC/EC)	0.2 μg/cm^2 filter	± 15%	Weekly methane gas injections checking; factory calibration when out of spec.
Particle light scattering and absorption			
Light scattering (Ecotech M9003)	0.5 M/m	± 10%	Monthly calibrated by certified CO_2.
Light absorption (Magee AE-31)	0.05 μg/m^3	± 15%	Weekly internal electronic checks; factory calibration when out of spec.
Solar radiation (Kipp & Zonen POM 01)	2 nm in wavelength	± 10%	Yearly factory calibration and when out of spec.
Particle number concentrations in different size fractions			
Fine particle (Grimm 1.108)	Particle size in 0.3-10 μm	± 15%	Daily internal electronic checks; yearly particle size comparisons with polystyrene latex suspension.

Table 2. Summary of minimal detectable limits, precisions, and calibration methods for the measurements at the Southern Supersite in Taiwan. (Continued)

Measurement (Instrument)	Minimal detectable limit	Precision	Calibration and/or testing method
Ultrafine particle (Grimm SPMS+C)	Particle size in 10-900 nm	± 15%	Daily internal electronic checks; yearly particle size comparisons with polystyrene latex; yearly factory calibrations.
Related gases			
O_3 (Ecotec 9810)	0.4 ppb	± 10%	Daily zero and span testing with internal ozone generator; quarterly calibrated with Dasibi 1003 PC UV photometer.
CO (Horiba APMA 360)	0.05 ppm	± 10%	Daily zero and span testing with certified CO; quarterly calibrated with certified CO mixture and dynamic dilution.
NO/NO_y (Thermo 42 CY)	50 ppt	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
NH_3 (Thermo 17 C)	1 ppb	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
NH_3 (ESA AC32M)	1 ppb	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
H_2O_2 (Yankee AL-2021)	50 ppt	± 20%	Monthly calibrated with internal H_2O_2 permeation device; six-monthly calibrated with liquid of H_2O_2 standards.
BTEX (Chromato ChromaPID)	3 ppb	± 20%	Monthly calibrated with certified CH_4 and reference gas
O_3 (Ecotec 9810)	0.4 ppb	± 10%	Daily zero and span testing with internal ozone generator; quarterly calibrated with Dasibi 1003 PC UV photometer.

Table 2. Summary of minimal detectable limits, precisions, and calibration methods for the measurements at the Southern Supersite in Taiwan. (Continued)

Measurement (Instrument)	Minimal detectable limit	Precision	Calibration and/or testing method
Related gases			
CO (Horiba APMA 360)	0.05 ppm	± 10%	Daily zero and span testing with certified CO; quarterly calibrated with certified CO mixture and dynamic dilution.
NO/NO$_y$ (Thermo 42 CY)	50 ppt	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
NH$_3$ (Thermo 17 C)	1 ppb	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
NH$_3$ (ESA AC32M)	1 ppb	± 20%	Daily zero and span testing with certified NO; quarterly calibrated with certified NO mixture and dynamic dilution.
H$_2$O$_2$ (Yankee AL-2021)	50 ppt	± 20%	Monthly calibrated with internal H$_2$O$_2$ permeation device; six-monthly calibrated with liquid of H$_2$O$_2$ standards.
BTEX (Chromato ChromaPID)	3 ppb	± 20%	Monthly calibrated with certified CH$_4$ and reference gas.
Meteorological variables			
Wind speed (Met One 05305L)	± 0.3 m/s	± 0.3 m/s	Six-monthly factory calibration; monthly comparison with synchronized motor.
Wind direction (Met One 05305L)	± 5°	± 5°	Six-monthly calibration with surveyor compass; monthly comparison with surveyor compass.
Temperature (Met One CS500L)	± 0.1^{o}C	± 0.1^{o}C	Six-monthly factory calibrations; monthly comparison with psychrometer.
Relative humidity (Met One CS500L)	± 2%	± 2%	Six-monthly factory calibrations; monthly comparison with psychrometer.

removes particle-bound water. Lee *et al.* (2005a; 2005b; 2005c) reported that the SES-TEOM and federal reference method (FRM) $PM_{2.5}$ are highly correlated ($R^2 =$ 0.89–0.95) and their regression slopes were near unity (0.86–1.07) at the Atlanta, Baltimore and Pittsburgh Supersites. However, the SES-TEOM inevitably loses some semi-volatile materials due to heating to 30°C (Lee *et al.,* 2005a; 2005b; 2005c). Notably, $PM_{2.5}$ SO_4^{2-}, NO_3^- and carbon (OC and EC) are the major $PM_{2.5}$ components in Taiwan (Lin, 2002; Chen *et al.,* 2003; Tsai and Cheng, 1999; 2004). Additionally, $PM_{2.5}$ SO_4^{2-} and NO_3^- are acquired by the RP 8400S and RP 8400N monitors (Thermo Scientific, USA), respectively. Particles in the air system are impacted onto platinum and Nichrome strips (Stolzenburg and Hering, 2000). The strip is then instantly heated to transform SO_4^{2-} into sulfur dioxide (SO_2) or NO_3^- into nitrogen oxides (NO_x) that are detected by high-sensitivity SO_2 or NO_x gas analyzers, respectively. Conversion efficiencies from particles to gases are determined monthly by testing pre-determined concentrations of SO_4^{2-} or NO_3^- solutions provided by the manufacturer. However, measurement results from the U.S. Supersites indicate that conversion efficiencies depend on the ambient PM composition, which varies for different times and locations (Wittig *et al.,* 2004; Harrison, *et al.,* 2004b; Chow *et al.,* 2008). Notably, conversion efficiencies from calibration solutions are not necessarily the same as those for ambient particles.

The continuous $PM_{2.5}$ OC and EC are measured by the Sunset OC/EC field instrument (Sunset Laboratory, USA). Bae *et al.,* (2004) reported that the Sunset monitor is comparable to filter-based laboratory analysis of $PM_{2.5}$ OC and EC concentrations with slopes of 0.93 and 0.95, respectively, at the St. Louis Supersite. Parallel comparisons between two Sunset monitors in Los Angeles demonstrated that they were very precise (R^2 of 0.98 and 0.97 for OC and EC, respectively) (Arhami *et al.,* 2006).

Particle light absorption (M/m) can be reported as black carbon (BC) concentration ($\mu g/m^3$) using an appropriate conversion factor (16.2 m^2/g as the default for an aethalometer) (Magee Scientific, USA). However, the conversion factor depends on absorption wavelength and the physical and chemical properties of particles. In Fresno, the conversion factor is 10 m^2/g, which was estimated by comparing light absorption with integrated filter analysis of EC (Park *et al.,* 2006a). This comparison can be used to acquire the local conversion factor in Taiwan. Measured solar spectral data can be post-processed to derive various atmospheric parameters, such as optical depth, scattering coefficients, aerosol distribution, and energy distribution (Kipp & Zonen, The Netherlands).

Collocated tests between the TSI scanning mobility particle sizer (SMPS) (TSI, USA) and Grimm SMPS (GRIMM, Germany) at the Fresno Supersite reveal that the two sizers are similar in the 30–50 nm particle

Table 3. Summary of data validity levels and flags.

Level	Description	Flag
0	Data transferred from the on-site instruments or downloaded from the data loggers without editing.	
1A	Missing values due to power supply failure.	1
1B	Values are removed due to maintenance.	2
1C	Values are removed due to calibration.	3
1D	Values are removed due to audits.	4
1E	Missing values due to unclear reasons.	5
1F	Negative values are removed.	6
1G	Values under the detection limits are removed.	7
1H	Values with error messages are removed.	8
1I	Hourly-averaged data, acquired when the ratio of effective samples, excluded Level 1A-1H data, in one hour is lager than 75%.	
2A	When hourly $PM_{2.5} > PM_{10}$.	9
2B	When hourly $SO_4^{2-} + NO_3^- + OC + EC > PM_{2.5}$.	10
2C	When hourly $NO_x > NO_y$.	11

range (Chow *et al.,* 2008). The Grimm 1.108 and Grimm SMPS can also report particle mass concentrations derived from the measured size distribution when particle shape and density are used. Therefore, measurement of local particle density is important to obtaining reliable PM concentrations.

Quality assurance and data validation

Monitors are maintained and calibrated by consulting firms according to standard operating procedures established by the Taiwan EPA (Taiwan EPA, 2005). Independent audits are performed by academic institutes (Wu *et al.,* 2005; Lin *et al.,* 2006a). Measurements are transferred to

data servers located at the main office of the Taiwan EPA in Taipei. Data are given validation codes (Table 3) (Wu *et al.,* 2005; Lin *et al.,* 2006a). Daily averages of each measurement are also calculated. The calculated daily data for the most recent year are available on the Taiwan EPA website (http://taqm.epa.gov.tw/emc/default.aspx?mod = PsiAreaHourly).

Seasonal variations

In this section, the seasonal trends of PM_{10} mass, as well as $PM_{2.5}$ mass, SO_4^{2-}, NO_3^-, OC, and EC measured in Southern Supersite are analyzed to demonstrate the possible benefit of the Supersite. The analysis of other measurements will be
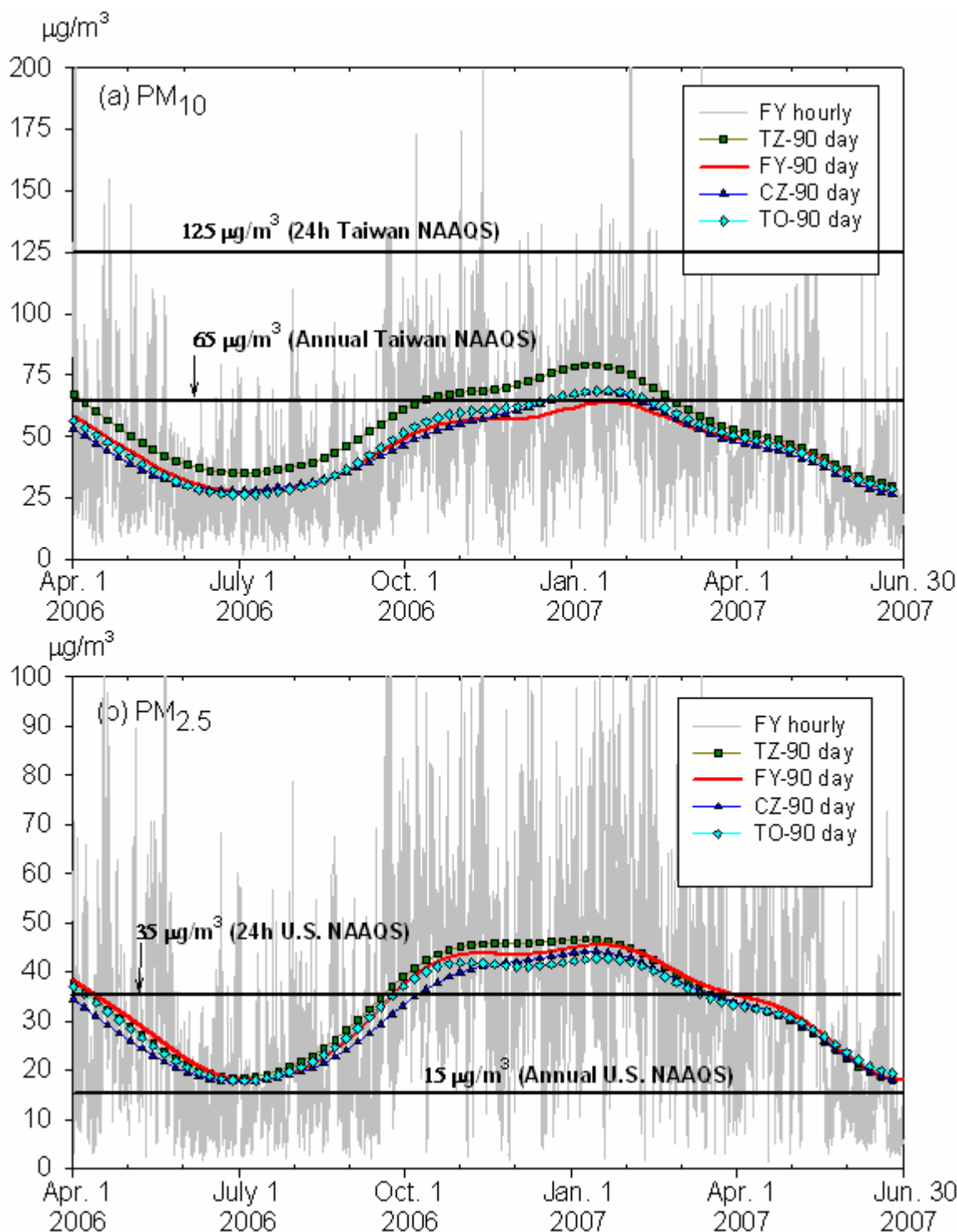
Fig. 4. Seasonal and hourly variations of: (a) PM_{10} mass, (b) $PM_{2.5}$ mass , (c) $PM_{2.5}$ sulfate, (d) $PM_{2.5}$ nitrate, (e) $PM_{2.5}$ organic carbon (OC), and (f) $PM_{2.5}$ elementary carbon (EC) between April 1, 2006, and June 30, 2007, at the Southern Supersite. Station codes are shown in Fig. 2. Notably, OC and EC at the FY station between February 1, 2007 and June 30, 2007 are removed due to failing to pass the data validation. As well, OC and EC at the CZ station between May 5, 2006, and October 23, 2006, are not measured.

given in future works. A low-pass Kolmogorov-Zurbenko (KZ) filter (Rao *et al.,* 1997; Eskridge *et al.,* 1997) was applied to ozone and PM concentrations to remove noise (e.g., Rao *et al.,* 1997; Vingarzan *et al.,* 2003; Wise and Comrie, 2005; Di Carlo *et al.,* 2007). The filter period, T, is determined by filter duration (t) and number of iterations (p); i.e., $T = t \times p^{1/2}$ (Eskridge *et al.,* 1997; Wise and Comrie, 2005). In this work, hourly data coupling with a filter period of T = 90 days with t = 40.25 days (966 hours) and p = 5 are used to produce seasonal tends for each considered measurement. Fig. 4 shows the calculated seasonal trends along with hourly concentrations for PM_{10} mass, as well as $PM_{2.5}$ mass, SO_4^{2-}, NO_3^-, OC, and EC between April 1, 2006, to June 30, 2007, at the core and the three satellite stations.

With the exception in Fig. 4(f), Figs. 4(a–e) show that PM_{10} mass, and $PM_{2.5}$ mass, SO_4^{2-}, NO_3^-, and OC concentrations have considerably seasonal variation with low values during summer and high values during winter. The strong ventilation and heavy showers via summer typhoons explain the low pollutant levels in summer, while relatively less ventilation and upwind pollution transport increase pollutions in winter (Chen *et al.,* 2004a; Lin *et al.,* 2005a). The PM_{10} mass levels before April 2007 at the TZ station were higher than those at the other stations (Fig. 4a), while $PM_{2.5}$ mass levels at the TZ station were close to those at the other stations (Fig. 4b). The increased coarse PM concentrations at the TZ station

are unclear and need further investigation in the future.

Notably, SO_2 is the primary precursor of sulfate and can be oxidized to form sulfuric acid (H_2SO_4) via gas- and liquid-phase reactions. Once H_2SO_4 forms, it can react quickly with NH_3 and thereby form nonvolatile ammonium sulfate ((NH_4)$_2SO_4$), which is mostly found in fine PM. The gas-phase oxidation rate is determined by levels of the ambient hydroxyl radical (OH), which is associated with photochemical activity (Seinfeld and Pandis, 1998). Liquid-phase oxidation is fast and occurs in clouds, rainwater, and within the water fraction of ambient aerosols. The liquid pathway is likely important in southern Taiwan as the monthly average humidity exceeds 75% (Fig. 3). Considerable sulfate differences among stations exist (Fig. 4c), indicating that sulfate is formed locally. Notably, SO_2 is primarily emitted from coal-fired power plants and large point sources in southern Taiwan. The relatively high levels of sulfate in winter at the northern TO station (Fig. 4c) is likely influenced by the SO_2 emissions originating from the upwind Shingda coal-fired power plant, 14 km from the TO station. However, further investigation is required to validate this hypothesis and evaluate other possible sources.

Fig. 5 compares the monthly variations of $PM_{2.5}$ mass, sulfate, nitrate, ozone and ammonia at the FY core station. The monthly averaged ozone concentration (Fig. 5) shows that photochemical activity peaks
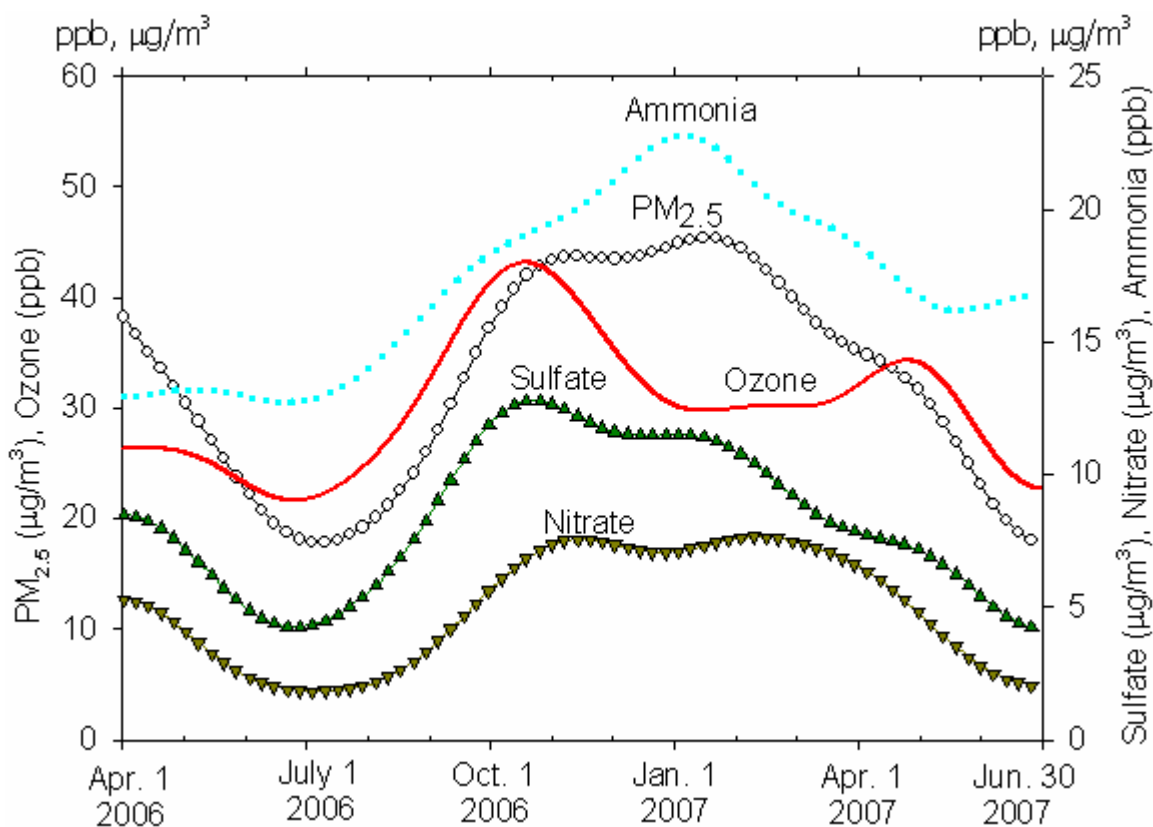
Fig. 5. Comparison of seasonal trends of $PM_{2.5}$ mass, $PM_{2.5}$ sulfate, $PM_{2.5}$ nitrate, ozone and ammonia between April 1, 2006 and June 30, 2007 at the core FY station. These seasonal trends are produced by the low-pass Kolmogorov-Zurbenko filter, as described in the section of seasonal variations in the text.

in October (major) and May (minor). The October ozone peak coincided with elevated $PM_{2.5}$ mass and sulfate (Fig. 5), implying that the increasing $PM_{2.5}$ mass resulted from excess production of sulfate through the gas-phase pathway due to strong photochemical activity.

The primary precursor of nitrate is NO_x. Both mobile and point sources are important NO_x sources in southern Taiwan. Similar to the gas-phase pathway for sulfate, NO_x can be oxidized by OH and thereby form nitric acid (HNO_3) (Seinfeld and Pandis, 1998). Additionally, NO_2 can be oxidized to nitrous

oxide (N_2O_5) by NO_3 radical during the nighttime and, subsequently, HNO_3 can be formed homogeneously or heterogeneously when N_2O_5 reacts with water (Seinfeld and Pandis, 1998). Notably, HNO_3 can react with NH_3 to form ammonium nitrate (NH_4NO_3); NH_3 can come from livestock and fertilizers. Considerable differences in nitrate concentrations exist among stations (Fig. 4d), indicating that NH_4NO_3 is produced locally. The relatively high nitrate concentrations in winter at the southern CZ station (Fig. 4d) likely resulted from abundant NH_3, with sulfate at the lowest

Table 4. Annual $PM_{2.5}$, PM_{10}, NO_3^-, SO_4^{2-}, OC and EC concentrations and their ratios relative to $PM_{2.5}$ mass at the Fooyin core station (FY) and the Chiautou (TO), Chenjen (TZ), and Choujau (CZ) satellite stations based on measurements between April 1, 2006 and March 31, 2007.

	FY	TO	TZ	CZ
Annual Average (mg/m^3)				
$PM_{2.5}$ mass	33.9	32.6	34.6	31.7
PM_{10} mass	47.2	48.8	57.1	47.1
$PM_{2.5}$ NO_3^-	5.2	4.7	3.4	5.0
$PM_{2.5}$ SO_4^{2-}	8.9	9.6	8.9	7.5
$PM_{2.5}$ OC	7.6	7.2	7.1	[a]Na
$PM_{2.5}$ EC	2.4	2.5	2.5	[a]Na
Ratios to $PM_{2.5}$ mass				
PM_{10} mass	1.39	1.50	1.65	1.49
$PM_{2.5}$ NO_3^-	0.15	0.15	0.10	0.16
$PM_{2.5}$ SO_4^{2-}	0.26	0.29	0.26	0.24
$PM_{2.5}$ OC	0.22	0.22	0.21	[a]Na
$PM_{2.5}$ EC	0.07	0.08	0.07	[a]Na

[a] Not applicable.

levels among the four stations (Fig. 4d). Therefore, small NH_3 was consumed by sulfate, explaining the elevated NH_4NO_3 concentration observed at the CZ station. Nitrate concentrations at the urbanized TZ station are much lower than those at the other three stations (Fig. 4d), possibly due to the lack of NH_3 in the city. Nitrate concentrations at the southern CZ and northern TO stations peak in January (Fig. 4d). These peaks are likely due to the high ambient NH_3 concentrations (Fig. 5) and low ambient temperature, which favor particle phase of NH_4NO_3.

Notably, EC is a primary pollutant and is generated by incomplete combustion of local sources (Fine *et al.,* 2008), while OC can be from primary and secondary sources. Elevated wintertime $PM_{2.5}$ OC and EC concentrations (Figs. 4e and 4f) can be attributed to the abundant vehicles in southern Taiwan. High amounts of carbonaceous aerosol at low temperatures suggest low combustion efficiency for internal combustion engines and enhanced condensation of organic vapors. The EC concentrations peak in January (Fig. 4f) can be explained by enhanced emissions and a relatively low mixing depth in cold months. Notably, the OC levels peak in December, one moth earlier than EC, likely due to the contribution of secondary OC (Fig. 5). The production rate of secondary OC peaks in

October and drops off after October, as inferred based on ozone variations (Fig. 5).

Table 4 shows the annual PM_{10} mass and $PM_{2.5}$ mass, NO_3^-, SO_4^{2-}, OC and EC concentrations and their ratios relative to $PM_{2.5}$ mass concentrations from April 1, 2006 to March 31, 2007. Generally, the concentrations of each measurement among the different stations are compatible, except for relatively high PM_{10} mass and low $PM_{2.5}$ NO_3^- concentrations at the TZ station, and the high $PM_{2.5}$ SO_4^{2-} concentrations at the TO station. The major components of $PM_{2.5}$ are SO_4^{2-} (24–29%), OC (21–22%) and NO_3^- (10–16%). This is similar to levels observed in the eastern U.S. (Fine *et al.,* 2008), but differ from those in the western U.S. where only NO_3^- and OC dominate (Chow *et al.,* 2008). The annual PM_{10} averages (Table 4), ranging from 47-57 $\mu g/m^3$, are lower than the Taiwan PM_{10} annual NAAQS (65 $\mu g/m^3$), but the annual $PM_{2.5}$ averages (Table 4), ranging from 32-25 $\mu g/m^3$, are much higher than the U.S. $PM_{2.5}$ annual NAAQS (15 $\mu g/m^3$). Figs. 4(a) and 4(b) present comparisons of these levels with seasonal trends of PM_{10} and $PM_{2.5}$ mass. The annual PM_{10} averages in 2006 were 88, 76 and 72 $\mu g/m^3$ at the TZ, TO, and CZ air stations (Taiwan EPA, 2007), which are much higher than the corresponding 57, 49 and 47 $\mu g/m^3$ measured at the satellite stations. The PM_{10} monitors used in the TAQMN are Model 650 PM_{10} beta attenuation monitors (BAMs) (Thermo Env. Instruments, USA), while the Southern Supersite uses the SES-TEOM. The high

PM_{10} concentrations measured by the BAMs are likely due to moisture interference. According to Tsai *et al.* (2006), when relatively humidity is > 85%, the BAM reading will increase as humidity increases. These situations frequently occur during nighttime in southern Taiwan due to the nature of high humidity (Fig. 3). The low PM_{10} concentrations measured by SES-TEOM are expected as the SES-TEOM loses some semi-volatile materials.

Figs. 6 and 7 compare hourly averages of PM mass and major components during the cold (October 2006 to March 2007) and warm (April–September 2006) seasons. The diurnal variations of PM mass and components are apparent during cold season and less important in warm months. The diurnal variations at the core and three satellite stations are similar (Figs. 6 and 7). For PM_{10} mass, $PM_{2.5}$ mass and $PM_{2.5}$ OC and EC concentrations, the major peaks were near morning rush hours, indicating that these are related to vehicle emissions. The TZ station, located in Kaohsiung City, has higher morning peaks of PM_{10} mass, and $PM_{2.5}$ mass, OC and EC concentrations than the other stations (Fig. 6). The major afternoon peaks of $PM_{2.5}$ SO_4^{2-} and NO_3^- concentrations indicate that they are related to photochemical activities. The relatively smaller diurnal variations in summer months can be explained by better atmospheric mixing.
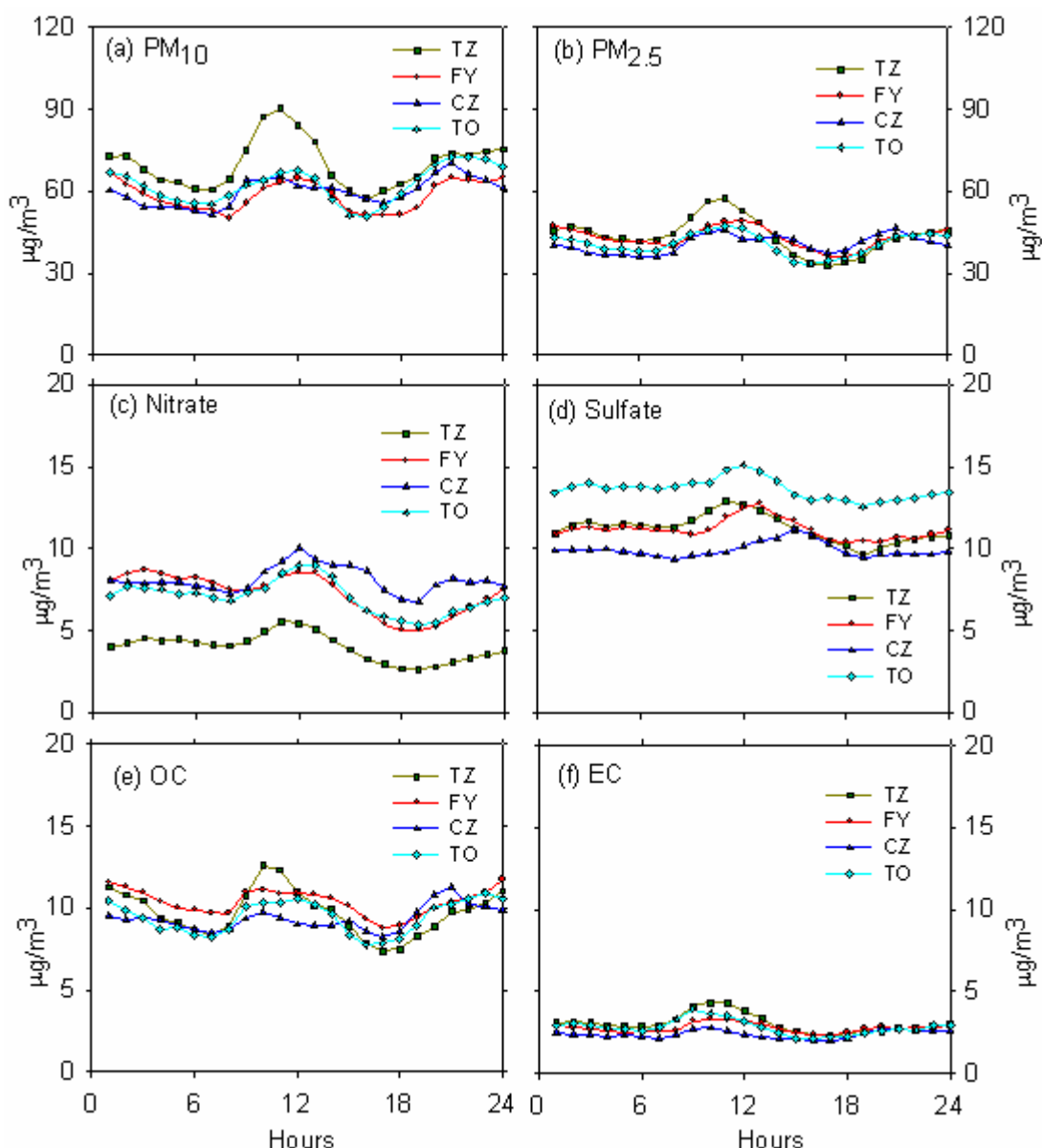
CHALLENGES FOR THE SOUTHERN SUPERSITE

Fig. 6. Wintertime diurnal variations of: (a) PM_{10} mass, (b) $PM_{2.5}$ mass, (c) $PM_{2.5}$ sulfate, (d) $PM_{2.5}$ nitrate, (e) $PM_{2.5}$ organic carbon (OC), and (f) $PM_{2.5}$ elementary carbon (EC), averaged from the hourly data between October 1, 2006 and March 31, 2007 at the Southern Supersite. Station codes are shown in Fig. 2.

Issues that need to be addressed in the future include: (1) loss of semi-volatile substances from the SES-TEOM, (2) efficiencies for the sulfate (RP 8400S) and nitrate (RP 8400N) monitors, (3) measurements of particle density and water,

(4) organic speciation, (5) size-resolved particle composition, and (6) new receptor modeling techniques and source profiles.

Loss of semi-volatile substances from the SES-TEOM

The SES-TEOM loses semi-volatile substances, especially nitrate (Lee *et al.,* 2005a; 2005b; 2005c), and thereby underestimates $PM_{2.5}$ and PM_{10} concentrations. The filter dynamic measurement system (FDMS) (Meyer *et al.,* 2002) accounts for volatilization (Grover *et al.,* 2005; Chow *et al.,* 2008). Concurrent measurements of HNO_3 and ammonium (NH_4^+) associated with current measurements of NH_3 and NO_3^- at the core station permit equilibrium calculations to determine limiting precursors for $PM_{2.5}$ NH_4NO_3 (Lin *et al.,* 2006b; Lin and Cheng, 2007).

Conversion efficiencies for the RP 8400S and RP 8400N

Wittig *et al.* (2004), Harrison *et al.* (2004b) and Chow *et al.* (2008) showed that conversion efficiencies acquired from calibration solutions are not representative of those for ambient sulfate and nitrate. Chow *et al.* (2006b) recommend site-specific comparisons between continuous monitor and filter measurements. On-site ion chromatographs (ICs) (Chow *et al.,* 2008) that measure anions and cations, such as Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+ and Ca_2^+, are an alternative. Good agreement existed between on-site ICs and filter measurements at several U.S. supersites (Hogrefe *et al.,* 2004; Grover *et al.,* 2006) and those in Taiwan (Chang *et al.,* 2006; 2007).

Measurements of particle density and water

Particle density facilitates conversion of number distributions to mass distributions and aerodynamic diameters related to Stokes diameter. Bulk particle density can be estimated from the PM chemical composition. Alternatively, effective particle density can be calculated when the following combinations are known: mobility size-aerodynamic size; mobility size-particle mass; or, aerodynamic size-particle mass (Ristimaki *et al.,* 2002; McMurry *et al.,* 2002). Particle water may account for some unresolved PM mass (Chow, 1995; Rees *et al.,* 2004). Rees *et al.* (2004) reported that water can contribute 8–16% of FRM $PM_{2.5}$ mass at the Pittsburgh Supersite. One of the most common methods for measuring particle water is to use hygroscopic tandem differential mobility analyzers (H-TDMAs) (Cocker *et al.,* 2001). Additionally, automatically measuring particle water is possible. One method has been successfully tested at the Pittsburgh Supersite (Khlystov *et al.,* 2005). Further investigation of seasonal variations of particle water is necessary to explain the unresolved PM mass in Taiwan, where numerous studies about PM compositions are based on filter analyses in the laboratory (Lin and Tsai, 2001; Lee and Chang, 2002; Lin, 2002; Lin and Lee 2004; Lin *et al.,* 2005b; 2007; Fang *et al.,* 2006; Chen *et al.,* 1999; 2001; 2003; 2004b; Tsai and Cheng, 1999; 2004; Tsai and Kuo, 2005; Chio *et al.,* 2004; Chang *et al.,* 2006; Chiang *et al.,* 2005).
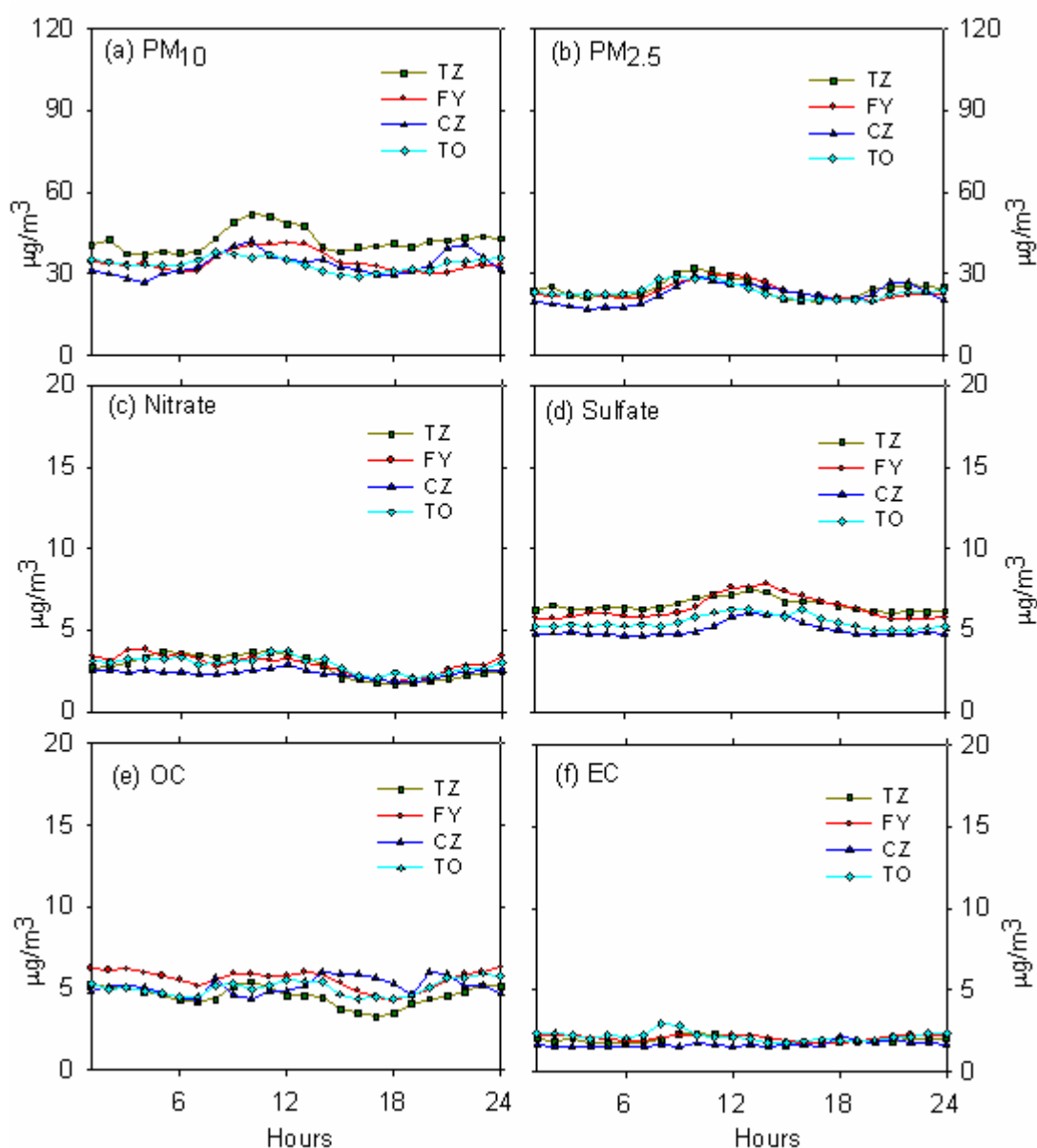
Fig. 7. Summertime diurnal variations of: (a) PM_{10} mass, (b) $PM_{2.5}$ mass , (c) $PM_{2.5}$ sulfate, (d) $PM_{2.5}$ nitrate, (e) $PM_{2.5}$ organic carbon (OC), and (f) $PM_{2.5}$ elementary carbon (EC), averaged from the hourly data between April 1, 2006, and September 30, 2006, at the Southern Supersite. Station codes are shown in Fig. 2.

Organic speciation

Organic compounds are emitted from combustion processes and natural sources, and formed in atmosphere as secondary organic aerosols (SOAs). Some organic compounds are good source markers, such as levoglucosan for biomass burning, cholesterol for meat cooking, 1,2-benzenedicarboxylic acid for SOA, and iso-nonacosane for cigarette smoke (Schauer and Cass, 2000; Chow *et al.,* 2007a; 2007b; Watson *et al.,* 2008). Therefore, organic

species analysis can enhance source apportionments and correlate combustion aerosols to adverse health effects (Mauderly and Chow, 2008). Analytical techniques for PM organic species in Taiwan are labor intensive and use water or solvent extraction followed by gas chromatography/mass spectrometry (GC/MS) (Lin and Lee, 2004). The recent advancement in thermal desorption following GC/MS analysis was examined by Chow *et al*. (2007b) and can be used as an alternative, cost-effective method. This method can acquire 130 non-polar organic species including n-alkanes, alkenes, cycloalkenes, hopanes, sternes, polycyclic aromatic hydrocarbons (PAHs) (Chow *et al.,* 2007b).

Size-resolved particle compositions

The evolution of PM size distribution provides insight into the nucleation process, secondary aerosol formation, and primary source attributions. Single particle mass spectrometers (SPMS) (Carson *et al.,* 1995; Johnston and Wexler, 1995; Nordmeyer and Prather, 1994; Jayne *et al.,* 2000; Thomson *et al.,* 2000; Middlebrook *et al.,* 2003) provide information on particle size (down to ~ 50 nm) and compositions (Wexler and Johnston, 2008). The micro-orifice uniform deposit impactor (MOUDI) (Marple *et al.,* 1991) and Nano-MOUDI (MSP, USA) are alternative devices that collect PM substrates for different size bins that can be analyzed in the laboratory (Lin *et al.,* 2005b; 2007; Tsai, *et al.,* 2005; Fang *et al.,* 2006; Chow and Watson, 2007).

New receptor modeling techniques and source profiles

Receptor models identify and quantify source contributions based on chemical fingerprints of various sources (Watson *et al.,* 2002; Chen *et al.,* 2007a; Watson *et al.,* 2008). Receptor modeling in Taiwan (Chen *et al.,* 1999; Chen *et al.,* 2001; Chio *et al.,* 2004; Chiang *et al.,* 2005; Wang *et al.,* 2007) has applied the effective variance solution to chemical mass balance (CMB) equations (Friedlander, 1973; Watson, 1984; Watson *et al.,* 1984). However, local source profiles are limited (Chen *et al.,* 1999; Chen *et al.,* 2001; Chio *et al.,* 2004; Chiang *et al.,* 2005; Wang *et al.,* 2007). Applying other CMB solutions, such as positive matrix factorization (PMF) (Paatero, 1997) and UNMIX (Henry, 1997), may provide source factors that can be compared with real source profiles (Chen *et al.,* 2007a; Watson *et al.,* 2008). The source-receptor hybrid pseudo-deterministic model (PDRM) should also be considered for use to detect and quantify elevated plume emissions and contributions (Park *et al.,* 2005; 2006b). All of these methods require that Taiwan increase its effort in obtaining source profiles that represent real-world emissions (Chen *et al.,* 2007b; England *et al.,* 2007a; 2007b).

SUMMARY

The Southern Supersite will increase an understanding of PM characteristics in southern Taiwan. Information provided is

related to: (1) PM composition, (2) PM optical properties (e.g., light scattering and absorption), (3) PM number concentrations in various size fractions from 10 nm to 20 μm, and (4) PM precursors gases (NO_y, NH_3 and H_2O_2). The daily averages of these measurements after validation are available on the Taiwan EPA website (http://taqm.epa.gov.tw/emc/default.aspx?mod = PsiAreaHourly). Receptor modeling analysis of measured data will provide source apportionment results for the development of PM pollution-control strategies. Additionally, these data sets will also benefit researchers investigating PM exposure and adverse health effects. The Southern Supersite should expand its measurements and related research to: (1) loss of semi-volatile substance from the SES-TEOM, (2) conversion efficiencies for the continuous sulfate (RP 8400S) and nitrate (RP 8400N) monitors, (3) measurement of particle density and water, (4) analysis of PM organic compounds, especially source markers, (5) analysis of size-resolved particle composition, (6) application of new receptor modeling techniques, and (7) measurement of source profiles for Taiwan pollution sources.

ACKNOWLEDGMENTS

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