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An overview of regional experiments on biomass burning aerosols and related pollutants in Southeast Asia: From BASE-ASIA and the Dongsha Experiment to 7-SEAS

Neng-Huei Lin [a,*], Si-Chee Tsay [b], Hal B. Maring [c], Ming-Cheng Yen [a], Guey-Rong Sheu [a], Sheng-Hsiang Wang [a,d], Kai Hsien Chi [e], Ming-Tung Chuang [f], Chang-Feng Ou-Yang [g], Joshua S. Fu [h,i], Jeffrey S. Reid [j], Chung-Te Lee [f], Lin-Chi Wang [k,l], Jia-Lin Wang [g], Christina N. Hsu [b], Andrew M. Sayer [b,m], Brent N. Holben [b], Yu-Chi Chu [n], Xuan Anh Nguyen [o], Khajornsak Sopajaree [p], Shui-Jen Chen [q], Man-Ting Cheng [r], Ben-Jei Tsuang [r], Chuen-Jinn Tsai [s], Chi-Ming Peng [t], Russell C. Schnell [u], Tom Conway [u], Chang-Tang Chang [v], Kuen-Song Lin [w], Ying I. Tsai [x,y], Wen-Jhy Lee [z], Shuenn-Chin Chang [n], Jyh-Jian Liu [n], Wei-Li Chiang [n], Shih-Jen Huang [aa], Tang-Huang Lin [bb], Gin-Rong Liu [bb]

[a] Department of Atmospheric Sciences, National Central University, Chung-Li, Taiwan
[b] NASA Goddard Space Flight Center, Greenbelt, MD, USA
[c] NASA Headquarters, Washington, DC, USA
[d] NASA Postdoctoral Program Fellow, Goddard Space Flight Center, NASA, Greenbelt, MD, USA
[e] Institute of Environmental and Occupational Health Sciences, National Yang Ming University, Taipei, Taiwan
[f] Graduate Institute of Environmental Engineering, National Central University, Chung-Li, Taiwan
[g] Department of Chemistry, National Central University, Chung-Li, Taiwan
[h] Department of Civil and Environmental Engineering, The University of Tennessee, Knoxville, TN, USA
[i] UTK-ORNL Center for Interdisciplinary Research and Graduate Education, Knoxville, TN, USA
[j] Naval Research Laboratory, Monterey, CA, USA
[k] Department of Chemical and Materials Engineering, Cheng Shiu University, Kaohsiung, Taiwan
[l] Super Micro Mass Research and Technology Center, Cheng Shiu University, Kaohsiung, Taiwan
[m] Goddard Earth Sciences Technology And Research (GESTAR), Universities Space Research Association (USRA), Columbia, MD, USA
[n] Taiwan Environmental Protection Administration, Taipei, Taiwan
[o] Institute of Geophysics, Vietnam Academy of Science and Technology, Hanoi, Viet Nam
[p] Department of Environmental Engineering, Chiang Mai University, Chiang Mai, Thailand
[q] Department of Environmental Engineering and Science, National Pingtung University of Science and Technology, Nei-Pu, Pingtung, Taiwan
[r] Department of Environmental Engineering, National Chung Hsing University, Taichung, Taiwan
[s] Institute of Environmental Engineering, Nation Chiao Tung University, Hsinchu, Taiwan
[t] WeatherRisk Explore Inc., Taipei, Taiwan
[u] NOAA ESRL Global Monitoring Division, Boulder, CO, USA
[v] Department of Environmental Engineering, National Ilan University, Yilan, Taiwan
[w] Department of Chemical Engineering and Materials Science, Yuan Ze University, Chung-Li, Taiwan
[x] Department of Environmental Resources Management, Chia Nan University of Pharmacy and Science, Tainan, Taiwan
[y] Department of Environmental Engineering and Science, Chia Nan University of Pharmacy and Science, Tainan, Taiwan
[z] Department of Environmental Engineering, National Cheng Kung University, Tainan, Taiwan
[aa] Department of Marine Environmental Informatics, National Taiwan Ocean University, Keelung, Taiwan
[bb] Center for Space and Remote Sensing Research, National Central University, Chung-Li, Taiwan

* Corresponding author.
E-mail address: nhlin@cc.ncu.edu.tw (N.-H. Lin).

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HIGHLIGHTS

- First comprehensive field study of biomass burning conducted in the northern SEA.
- First *in-situ* measurements of air chemistry and physics in South China Sea/East Sea.
- A conceptual model of circulation flows constructed for transport of biomass burning.
- Characterization of biomass-burning aerosols in source/sink region in northern SEA.
- Spatial distribution of Hg and POPs over northern SEA is illustrated.

GRAPHICAL ABSTRACT

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ABSTRACT

By modulating the Earth-atmosphere energy, hydrological and biogeochemical cycles, and affecting regional-to-global weather and climate, biomass burning is recognized as one of the major factors affecting the global carbon cycle. However, few comprehensive and wide-ranging experiments have been conducted to characterize biomass-burning pollutants in Southeast Asia (SEA) or assess their regional impact on meteorology, the hydrological cycle, the radiative budget, or climate change. Recently, BASE-ASIA (Biomass-burning Aerosols in South-East Asia: Smoke Impact Assessment) and the 7-SEAS (7-South-East Asian Studies)/Dongsha Experiment were conducted during the spring seasons of 2006 and 2010 in northern SEA, respectively, to characterize the chemical, physical, and radiative properties of biomass-burning emissions near the source regions, and assess their effects. This paper provides an overview of results from these two campaigns and related studies collected in this special issue, entitled "Observation, modeling and impact studies of biomass burning and pollution in the SE Asian Environment". This volume includes 28 papers, which provide a synopsis of the experiments, regional weather/climate, chemical characterization of biomass-burning aerosols and related pollutants in source and sink regions, the spatial distribution of air toxics (atmospheric mercury and dioxins) in source and remote areas, a characterization of aerosol physical, optical, and radiative properties, as well as modeling and impact studies. These studies, taken together, provide the first relatively complete dataset of aerosol chemistry and physical observations conducted in the source/sink region in the northern SEA, with particular emphasis on the marine boundary layer and lower free troposphere (LFT). The data, analysis and modeling included in these papers advance our present knowledge of source characterization of biomass-burning pollutants near the source regions as well as the physical and chemical processes along transport pathways. In addition, we raise key questions to be addressed by a coming deployment during springtime 2013 in northern SEA, named 7-SEAS/BASELInE (Biomass-burning Aerosols & Stratocumulus Environment: Lifecycles and Interactions Experiment). This campaign will include a synergistic approach for further exploring many key atmospheric processes (e.g., complex aerosol–cloud interactions) and impacts of biomass burning on the surface–atmosphere energy budgets during the lifecycles of biomass-burning emissions.

1. Introduction

Over the last several decades the region extending from the Maritime Continent and greater Southeast Asia (SEA) to Taiwan has seen massive economic growth, and with it, high particulate matter loads (Hopke et al., 2008; Kim Oanh et al., 2011). Anecdotal reports of increased air pollution levels over the last decade are prevalent in SEA, and a positive aerosol optical depth (AOD) trend has been seen offshore (Zhang and Reid, 2010; Hsu et al., 2012). At the same time, biomass burning is a recurring problem, further reducing air quality, with a deleterious effect on human health (e.g., Dawud, 1998; Aditama, 2000; Kunii et al., 2002; Tipayarom and Kim Oanh, 2007; Wiwanitkit, 2008; Pengchai et al., 2009; Hyer and Chew, 2010; Silva et al., 2010; Kim Oanh et al., 2011; Pavagadhi et al., 2013). Even so, at times, SEA can exhibit some of the lowest lower troposphere aerosol particle concentrations in the world (Reid et al., 2013). Such a sharp contrast presents an excellent natural laboratory for studying aerosol–meteorology interactions in tropical to sub-tropical environments. While extremely interesting and complicated atmospheric phenomena occur throughout SEA, the region's high cloud cover, heterogeneous land/ocean surface, and complex pattern of emitters pose massive observability challenges to the scientific community (Reid et al., 2013). With SEA's importance to global climate change and atmospheric sciences as a whole, an extensive effort is required to consolidate and evaluate previous measurement datasets and guide the next generation of targeted observations.

While isolated field measurements of SE Asian smoke are presented in the literature (e.g., Gras et al., 1999; Ikegami et al., 2001; Okada et al., 2001; Balasubramanian et al., 2003; See et al., 2007; Betha et al., 2012), overall there have been limited comprehensive or wide coverage *in-situ* experiments conducted to characterize the physical, radiative, and chemical properties of biomass-burning pollutants in SEA. There have been even fewer field efforts to assess biomass burning's regional impact on meteorology, the hydrological cycle, the radiative budget, and climate change. To some

extent, the NASA TRACE-P (Jacob et al., 2003) and UNEP ABC/EAREX2005 (Nakajima et al., 2007) campaigns conducted in spring 2001 and 2005, respectively, cover a few topics and part of the SE Asian region. Until recently, NASA's BASE-ASIA (Biomass-burning Aerosols in South-East Asia: Smoke Impact Assessment; *cf.* http://smartlabs.gsfc.nasa.gov/; Tsay et al., 2013) field campaign, operated at a rural site in Phimai, Thailand, was the only comprehensive biomass-burning measurement suite in the region. Several trace gases, aerosol optical and microphysical properties, surface radiation, and meteorological parameters were measured with the NASA/SMART (Surface-sensing Measurements for Atmospheric Radiative Transfer) and COMMIT (Chemical, Optical, and Microphysical Measurements of *In-situ* Troposphere) mobile laboratories. To cope with the lack of adequate field data, the Seven South-East Asian Studies (7-SEAS; Reid et al., 2013) project was established to perform interdisciplinary research in aerosol–meteorology and climate interaction in the SE Asian region, with particular emphasis on the impact of biomass burning on cloud, atmospheric radiation, hydrological cycle, and regional climate. As part of 7-SEAS, numerous sun photometer sites have been established and the first strategically placed intensive studies have been conducted. These include the establishment of a supersite at the National University of Singapore in 2009, to assess boreal summertime smoke in the Maritime Continent, research cruises in 2011 and 2012, and a comprehensive international campaign centered at Dongsha Island (aka Dongsha Experiment, http://aerosol.atm.ncu.edu.tw/DongSha; Wang et al., 2011) in the northern South China Sea (or East Sea; hereafter referenced as SCS/ES) during March–June 2010. The Dongsha Experiment provided a relatively complete dataset of aerosol chemistry and physical observations conducted in the source/sink region for the marine boundary layer and lower free troposphere of biomass burning/air pollutants in the northern SEA. Relevant papers on data collected in these campaigns can be found in this special issue as well as a previously published special issue in *Atmospheric Research* (Volume 122, 2013).

In this overview paper, we concentrate primarily on results from northern SEA, summarizing results from the BASE-ASIA and 7-SEAS/Dongsha Experiment campaigns presented in this special issue. Additional papers from field work under 7-SEAS and related studies in the Maritime Continent are also included. The twenty-eight papers collected in this volume can be categorized into (1) 3 overview papers, to summarize the experiments and regional weather/climate; (2) 9 papers on aerosol chemistry and trace gas analyses, to characterize biomass-burning pollutants in source and sink regions; (3) 4 papers on air toxics, to explore the measurements of atmospheric mercury (Hg) and Dioxins in source and remote areas; (4) 8 papers on aerosol physical characterization, to study microphysical, optical, and radiative properties of biomass burning pollutants; and (5) 4 papers on modeling and impact studies, to assess the effects of biomass-burning pollutants in this region. Taken together, these papers advance our present knowledge regarding the characterization of biomass burning near the source regions, and of the physical and chemical processes along the transport pathways (i.e., high- and low-altitude transport). In addition to the conventional approach to aerosol chemical characterization, observations of Hg, dioxins, and organic acids associated with biomass burning are also presented. These chemical species have rarely been studied in SEA, particularly using network measurements. The NASA mobile laboratories were operated during the BASE-ASIA and Dongsha Experiment providing 3 continuous months of observations over northern SEA and SCS/ES, respectively, for the first time.

This paper provides a brief overview of most of the studies in this special issue. The results from mobile laboratories equipped with a suite of remote sensing and *in-situ* measurements along with satellite data analysis are reviewed in a separate paper (Tsay et al., 2013). In this paper we first introduce the BASE-ASIA and 7-SEAS/Dongsha Experiment and their experimental designs, and briefly describe regional weather and climate. Secondly, we present an overall description of the chemistry of biomass-burning aerosols and related pollutants, and air toxics on a regional scale. Thirdly, we summarize the physical, optical, and radiative properties of aerosols observed by remote sensing techniques deployed at surface platform and network sites. Finally, we discuss model performance and provide a preliminary impact assessment of biomass burning.

2. Field experiments of biomass burning in northern SEA

In recent years, several international and national field experiments such as INDOEX (Ramanathan et al., 2001a, 2001b), ACE-Asia (Huebert et al., 2003), TRACE-P (Jacob et al., 2003), APEX (Nakajima et al., 2003), PEACE (Kondo et al., 2004), ABC (Ramanathan et al., 2007), and EAREX (Nakajima et al., 2007) have been conducted in Asia, to characterize aerosols and precursor trace gases, as well as study their environmental and climatic effects. For example, Asian aerosols were generally found to be a mixture of anthropogenic air pollutants and mineral dust particles in spring season, causing a strong negative surface radiative forcing (e.g., Nakajima et al., 2007). Only very limited regional campaigns (e.g., TRACE-P, Jacob et al., 2003) were focused on biomass-burning aerosols in SEA. More recently, BASE-ASIA (Tsay et al., 2013) and 7-SEAS/Dongsha Experiment (Wang et al., 2011, 2013b) characterized the chemical, physical, optical, and radiative properties of biomass-burning aerosols and related air pollutants over the northern SEA. Fig. 1 shows the site locations of BASE-ASIA and 7-SEAS/Dongsha Experiment and collaborating networks. Table 1 further lists more detailed information of site location and type of measurements at individual sites. The instrumentation, deployment, and objectives of these two experiments are related to the broader 7-SEAS project, which are described briefly below.

2.1. BASE-ASIA

Located at Phimai on a vast agricultural plain in central Thailand, the BASE-ASIA campaign was conducted late February to early May in 2006 which corresponds to the monsoonal dry season and a maximum in agricultural and forest fire activity (e.g., Giglio et al., 2006; Reid et al., 2005a, 2013; Tsay et al., 2013). In addition to the major objective of extending the current understanding of how biomass-burning aerosols affect atmospheric remote sensing and radiative energetics, BASE-ASIA provided an essential foundation in regional capacity building (Tsay et al., 2013).

NASA's COMMIT mobile laboratory as well as instrumentation for aerosol composition from University of Hawaii were deployed at the Phimai observatory and radar station (15.18°N, 102.56°E, 206 m MSL), which is operated by Thailand's Bureau of Royal Rainmaking and Agricultural Aviation. Thus, comprehensive surface measurements of various trace-gas concentrations, aerosol chemical/microphysical/optical properties, and meteorological parameters were acquired (*cf.* Table S1 of Tsay et al., 2013 for a detailed listing of the remote sensors and *in-situ* instruments). Because Phimai is relatively distant from the major transport routes of biomass-burning emissions from the northern region of SEA, BASE-ASIA measurements at Phimai often represent typical local background conditions. Phimai is under the influence of prevailing northeast flow from November to mid-March and southwest monsoonal flow from mid-May to September, seasonal aerosol loadings at Phimai consist of local biomass-burning smoke and anthropogenic aerosols transported from distant sources.

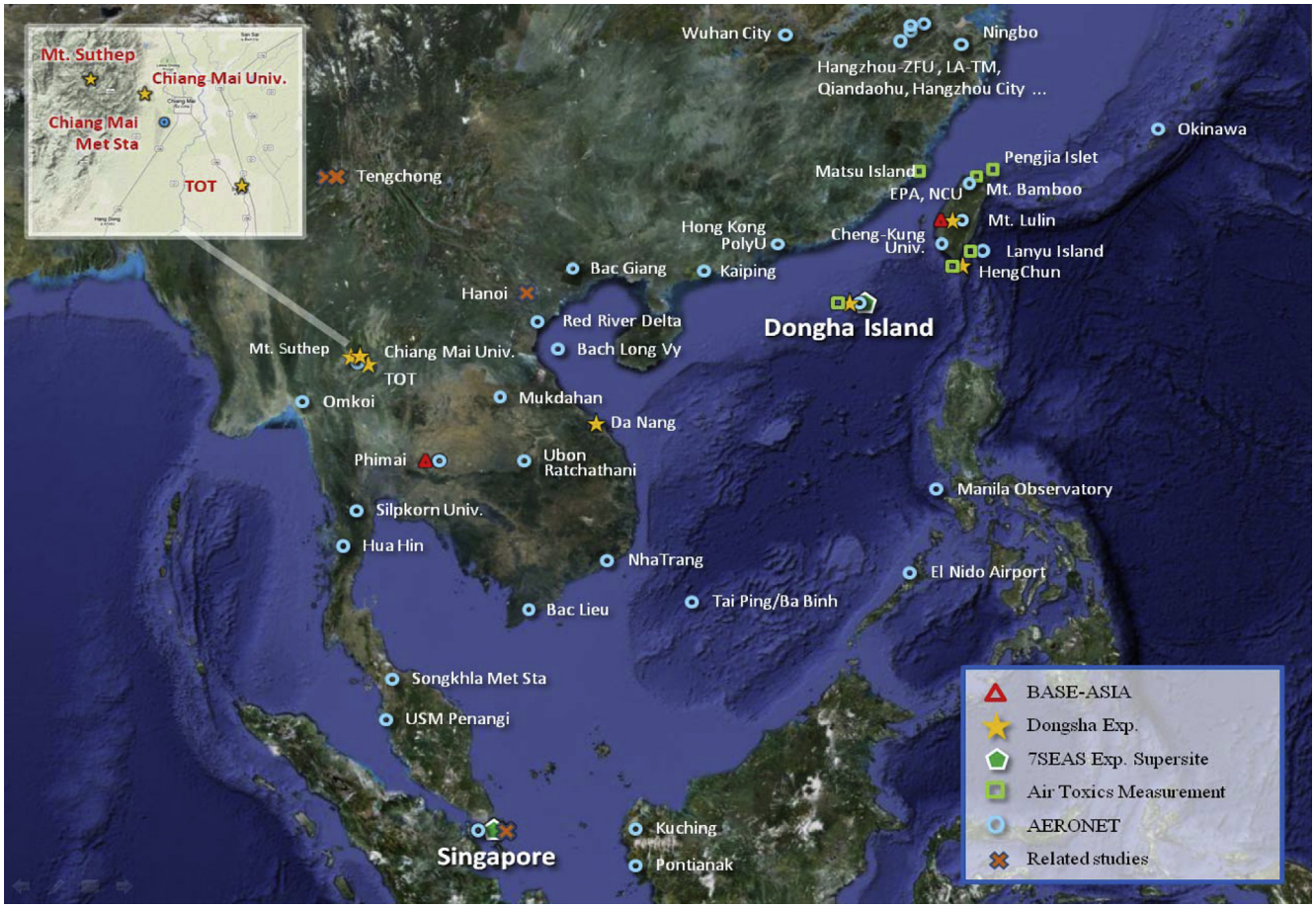

Fig. 1. Location of sites for BASE-ASIA, 7-SEAS/Dongsha Experiment and collaborative networks.

2.2. 7-SEAS and the Dongsha Experiment

2.2.1. Summary

Initiated in 2007, the 7-SEAS project (*cf.* whitepaper in http://7-seas.gsfc.nasa.gov/; http://www.nrlmry.navy.mil/aerosol_web/7seas/7seas.html; Reid et al., 2013) seeks to perform interdisciplinary research in the field of aerosol–meteorology and climate interaction from Java through the Malay Peninsula, and Southeast Asia to Taiwan. The primary scientific goal is to investigate the impact of biomass burning on clouds, atmospheric radiation, the hydrological cycle, and ultimately regional weather and climate. Countries with participating scientists and programmatic elements include Indonesia, Malaysia, Philippines, Singapore, Thailand, Taiwan, Vietnam, and the USA (e.g., NASA; Naval Research Laboratory, NRL; Office of Naval Research, ONR; Office of Naval Research-Global, ONRG; and many universities). The 7-SEAS is a grass-roots style program which promotes collaboration, builds capacity, and shares information throughout the region on science, field data, and observing systems. The study region, which ranges from the tropics to subtropics, has significant gradients in air pollution varying from near pristine to heavily-polluted. Thus, SEA's geography provides a unique natural laboratory for atmospheric measurements.

The Virtual Biomass Burning Experiment (VBBE) in August 2007 (*cf.* http://www.nrlmry.navy.mil/aerosol/7-SEAS/) was executed to collect and archive a dataset over SEA enabling 7-SEAS participating members to become familiar with experiment protocols and products. Measurements in the southern domain commenced in boreal summer-fall 2009 with the development of the National University of Singapore supersite. During the spring seasons of 2010–2012, a series of pilot IOPs in the northern regions of the 7-SEAS domain were successfully conducted, which included the Dongsha Experiment in 2010 (*cf.* http://aerosol.atm.ncu.edu.tw/DongSha/ and Wang et al., 2011; Tsay et al., 2013). Since the analysis of measurements made during the Son La Campaign (*cf.* http://aerosol.atm.ncu.edu.tw/SonLa/) during 2011/2012 boreal spring seasons is incomplete, results from these two deployments, which characterize long-range transport of biomass-burning aerosols over northwestern Vietnam, will be presented in a subsequent publication.

A monitoring network for ground-based measurements in remote/suburban area has been established, which includes the Lulin Atmospheric Background Station (LABS, 2862 m MSL) in central Taiwan, Hengchun (HC, coastal background station) in the very southern tip of Taiwan, Dongsha Island (DSI) in northern SCS/ES, Da Nang (DN, near coastal region) in central Vietnam, and Chiang Mai (CM, at Doi Suthep, 1396 m MSL) in northern Thailand. Two additional sites, located in urban and industrial zone in Chiang Mai, were operated for comparison. Measurements at Doi Suthep and Mt. Lulin are used to study the source/receptor relationship of active biomass burning along the high-altitude long-range transport pathway associated with the prevailing westerlies. By contrast, a sampling line from Hengchun, Dongsha Island to Da Nang enabled observation of the intrusion of air pollutants associated with Asian continental outflow (and weakening northeast monsoon flow) into SCS/ES and SEA via low-altitude transport.

The Dongsha Experiment was conducted in northern SEA during March–June 2010 as the first field campaign of 7-SEAS. The main

Table 1
Site location and instrumentation of the BASE-ASIA, 7-SEAS/Dongsha Experiment and related studies.

Experiment/sites	Location	Instrumentation			
		Radiation/remote sensing[a]	Aerosols[b]	Trace gases[c]	Toxics[d]
BASE-ASIA					
Taiwan					
Mt. Lulin[e]	23.47°N, 120.87°E; 2862 m	AOD, SR	Mass, chemical, optical	CO, O_3	Hg
Thailand					
Phimai	15.18°N, 102.05°E; 220 m	AOD, SRL	Mass, chemical, size, optical	CO, O_3	
Dongsha Experiment					
Taiwan					
Mt. Lulin[e]	23.47°N, 120.87°E; 2862 m	AOD, SR	Mass, chemical, size, optical	CO, O_3, SO_2, NH_3, NO_x/NO_y, GHGs	Speciated Hg, dioxins
Dongsha Island[f]	20.70°N, 116.73°E; 5 m	AOD, SRL	Mass, chemical, size	CO, O_3, SO_2, NO_x, HC, GHGs	Hg, dioxins
Hengchun	22.06°N, 120.70°E; 6 m		Mass, chemical	CO, O_3	Hg, dioxins
Ocean Research No. 1	Cruise mission between Hengchun and Dongsha Island	AOD	Mass, chemical	CO, O_3, SO_2, HC	Hg
Thailand					
Doi Suthep	18.81°N, 98.89°E; 1396 m		Mass, chemical		Hg, dioxins
Chiang Mai University	18.80°N, 98.95°E; 340 m		Mass, chemical	CO, O_3	
TOT	18.69°N, 99.05°E; 299 m		Mass, chemical	CO, O_3	
Vietnam					
Da Nang	16.04°N, 108.21°E; 7 m		Mass, chemical	CO, O_3, SO_2	Hg, dioxins
Air Toxics Measurement					
Taiwan					
Mt. Bamboo	25.21°N, 121.56°E; 1080 m				Hg, dioxins
Hengchun	22.06°N, 120.70°E; 3 m				Hg, dioxins
Pengjiayu Islet	25.63°N, 122.07°E; 3 m				Hg, dioxins
Lanyu Island	22.04°N, 121.57°E; 300 m				Hg, dioxins
Matsu Island	26.16°N, 119.95°E; 50 m				Hg, dioxins
Dongsha Island[f]	20.70°N, 116.73°E; 5 m	AOD, SRL	Optical	GHGs	Hg, dioxins
Mt. Lulin[e]	23.47°N, 120.87°E; 2862 m	AOD, SR	Mass, chemical, size, optical	CO, O_3, SO_2, NH_3, NO_x/NO_y, GHGs	Speciated Hg, dioxins
AERONET					
Hong Kong					
Hong Kong Polytechnic University	22.30°N, 114.18°E; 80 m	AOD			
Taiwan					
Nation Central University	24.97°N, 121.19°E; 144 m	AOD, SRL			
Chen-Kung University	23.00°N, 120.22°E; 50 m	AOD			
Dongsha Island[f]	20.70°N, 116.73°E; 5 m	AOD, SRL	Optical	GHGs	Hg, dioxins
Mt. Lulin[e]	23.47°N, 120.87°E; 2862 m	AOD, SRL	Mass, chemical, size, optical	CO, O_3, SO_2, NH_3, NO_x/NO_y, GHGs	Speciated Hg, dioxins
Thailand					
Chiang Mai Met. Station	18.77°N, 98.97°E, 312 MSL	AOD			
Mukdahan	16.61°N, 104.68°E; 166 m	AOD			
Phimai	15.18°N, 102.56°E; 220 m	AOD			
Silpakorn University	13.80°N, 100.49°E; 72 m	AOD	SRL		
Vietnam					
Bac Giang	21.29°N, 106.23°E; 15 m	AOD			
Bac Lieu	09.28°N, 105.73°E; 10 m	AOD			
Other sites marked in Fig. 1.					

[a] SR = Solar radiation, SRL = Solar radiation and lidar.
[b] Mass = $PM_{2.5}/PM_{10}$ mass concentrations; chemical = ions, EC/OC and elemental metals; size = size spectrum; optical = absorption and scattering.
[c] GHGs = Greenhouse gases measured with flask samples for NOAA/ESRL/GMD/GGCC network (*cf.* http://www.esrl.noaa.gov/gmd/ccgg/flask.html).
[d] Speciated Hg = Gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particulate mercury (PHg).
[e] Mt Lulin = Lulin Atmospheric Background Station (LABS), established since 13 April, 2006. Instruments were added in various years.
[f] Dongsha Island = Dongsha Background Station, established since 6 January, 2009, and is collaborative with AERONET. It served as a supersite during 7-SEAS/Dongsha Experiment and was fully equipped with aerosol and air quality instruments in March–June, 2010.

goals of the Dongsha Experiment were to develop Dongsha Island (about 2 km^2, 20°42′52″N, 116°43′51″E) in the SCS/ES as an atmospheric observation platform for atmospheric chemistry, radiation and meteorology, and to characterize the chemical and physical properties of biomass burning aerosols from the northern SE Asian region. In addition to the five primary sites listed above and two complimentary sites, the Mobile Air Quality Station of Taiwan EPA and NASA/COMMIT were shipped to Dongsha Island for continuous measurements of CO, SO_2, NO_x, HC, O_3, aerosol particles, and aerosol optical properties and vertical profiles. Two IOPs were conducted, one during 14–30 March and the other 10–20 April 2010. Aerosol chemistry and air toxics instrumentation deployed at five sites are listed in Table S1. Detailed descriptions of the instruments deployed at DSI and LABS are listed in Tables S2 and S3, respectively. Concurrent with IOP-1, the Taiwanese R/V Ocean Research No. 1 (ORO) cruised from southern Taiwan to SCS/ES during 14–19 March. Sounding balloon launches were increased to 4 times per day at Dongsha Island.

Flask samples of whole air were collected weekly using two 2 L glass flasks along with a portable sampling unit (PSU) at DSI and LABS starting in March 2010 and August 2006, respectively. These samples were analyzed by the NOAA/ESRL/GMD/Carbon Cycle Greenhouse Gases Group (CCGG) in a manner identical to samples from their other network sites. Detailed descriptions of the sampling and analytical methods can be found on the NOAA/ESRL/GMD webpage (*cf.* http://www.esrl.noaa.gov/gmd/ccgg/flask.html).

2.2.2. Air Toxics Measurement

The Air Toxics Measurement (ATM) project funded by Taiwan EPA was established in 2008. Atmospheric mercury and dioxins were measured in remote areas and islands in and around Taiwan. Fig. 1 shows the location of six remote sampling sites (Chi et al., 2013). Samples were intentionally collected during southwest and northeast monsoon flow. At the same time, samples were collected at LABS to enable comparison between the boundary layer and the LFT (Chi et al., 2010; Sheu et al., 2010). The site at ATM was operational during 7-SEAS/Dongsha Experiment.

2.2.3. Space-borne remote sensing and regional surface networks

The NASA/EOS program (*cf.* http://eospso.gsfc.nasa.gov/, *EOS Mission Profiles* for details of satellite and instrumentation), polar-orbiting satellites/sensors (e.g., SeaWiFS, Terra, EO-1, Aqua, ICESat, OMI, POLDER, CALIPSO, and Suomi NPP) provided comprehensive measurements of regional aerosol properties during BASE-ASIA and 7-SEAS operations. In addition, international and regional ground-based networks also provide data, which enrich the studies of biomass-burning aerosols in SEA. These networks include: AERONET (*cf.* http://aeronet.gsfc.nasa.gov/ and Holben et al., 1998), MPLNET (*cf.* http://mplnet.gsfc.nasa.gov/ and Welton et al., 2001), SKYNET (*cf.* http://atmos.cr.chiba-u.ac.jp/ and Sano et al., 2003), Silpakorn University's Solar Radiation Monitoring Network (Janjai et al., 2009, 2012), and the ABC Project (Ramanathan et al., 2007) sites in the region supported by the United Nations Environment Programme.

3. General description on regional climate and weather conditions during field campaigns

In this section, we present the climatology and mean atmospheric flow patterns for March and April emphasizing the northern SE Asian region, which controls the transport and dispersion of biomass-burning plumes. A similar discussion for the Maritime Continent can be found in Reid et al. (2012). Fig. 2 depicts a 32-year (1979–2010) monthly mean atmospheric circulation with National Centers for Environmental Prediction–II (NCEP-II) reanalysis 2.5° longitude × 2.5° latitude winds (Kanamitsu et al., 2002) for the lower troposphere at 700, 850 and 925 hPa levels, depicted with streamlines, indicating the East Asian high (or rather the cold surge anticyclone) over East China retreats northward and weakens at 925 hPa from March to April. Due to the presence of mountain ranges located at the east and west coast of the SEA peninsula, the wind at 925 and 850 hPa is channeled toward the north over the central peninsula's vast plain. At 700 hPa, the winds are westerly as a result of the dominating winter tropical SE Asian high (Chen et al., 2002) located south of 20°N over the region of northern SEA-SCS/ES in March. This westerly wind weakens in April. The streamlines in Fig. 2 indicate the coupling of southerly winds at 925, and 850 hPa over SEA peninsula during March and April. This large-scale circulation environment appears to transport biomass-burning pollutants from SEA toward East Asia (EA) and even eastward across the Pacific.

According to the annual rainfall climatologies, the SEA Peninsula has a relatively cooler dry season from mid-December to mid-April before the commencement of the Asian summer SW monsoon (Yen et al., 2013). An upward branch of the local East–West cell/circulation lifts biomass-burning pollutants to approximately 700 hPa, and is enhanced by a well-organized convergent center that forms over SEA at 925-hPa in March. A modeling study by Cheng et al. (2013) also suggested a similar condition favorable for forcing the uplift of biomass-burning emissions. Thus, these pollutants can be

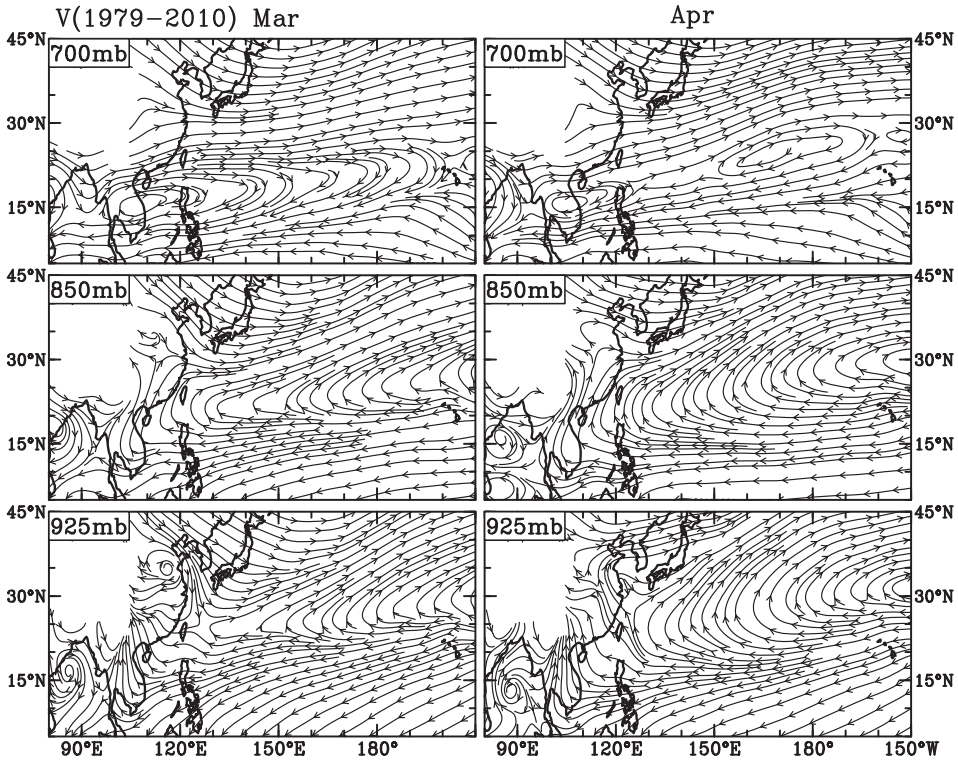

Fig. 2. Monthly mean streamline charts of large-scale circulation in SEA for 700-, 850-, and 925-hPa levels, respectively, in March and April for the period 1979–2010.

further transported downwind to the east at high-altitudes in the vicinity of Taiwan (Wai et al., 2008; Sheu et al., 2010; Lee et al., 2011; Ou-Yang et al., 2012; Chuang et al., 2013a; Chang et al., 2013).

The preceding climatological analysis is summarized in Fig. 3a. Surprisingly, Yen et al. (2013) discovered that during the period of the 7-SEAS/Dongsha Experiment, biomass-burning pollutants at high-altitudes were brought down to the surface in southern Taiwan by the downward branch of the transient local East–West circulation, driven by the subsidence of a cold surge anticyclone and mountain effect (See Fig. 3b). This analysis was based on a composite analysis of six events (*cf.* Fig. 8 in Yen et al., 2013) with ground-based air quality measurements and qualitatively confirmed using the Hybrid Single-Particle Lagrangian-Integrated Trajectory (HYSPLIT) particle dispersion forward model. However, based on continuous measurements at Dongsha, Atwood et al. (2013a) found no indication that the copious amounts of smoke aloft was being transported in the SCS's marine boundary layer.

4. Overall chemistry of biomass burning and related pollutants

4.1. Aerosol chemistry

Some studies investigated biomass-burning chemistry near source regions in SEA (see Reid et al., 2013 for a list). Others characterized the deterioration of local air quality, for instance, See et al. (2006), Yang et al. (2013) in Singapore, Wangkiat et al. (2003) in Thailand, Salam et al. (2003) in Dhaka – Bangladesh, Robinson et al. (2009) and Mahmud (2013) in Malaysia, and Hai and Kim Oanh (2013) in Vietnam. The BASE-ASIA and 7-SEAS/Dongsha Experiment provide pioneering and comprehensive datasets of biomass-burning aerosol chemistry near the source region. Fig. 4 presents the mass, major ions, organic and elemental carbon, and levoglucosan concentrations of $PM_{2.5}$ (except for CMU, TOT, Phimai and Da Nang for PM_{10}) measured during BASE-ASIA and Dongsha Experiment, and from southeastern Tibetan Plateau (Sang et al., 2013) and Hanoi (Hai and Kim Oanh, 2013).

Overall, $PM_{2.5}$ mass concentrations were relatively high (above 45 μg m^{-3}) near biomass-burning source regions (Doi Suthep, Tengchong-1) and an urban area (Hanoi). The $PM_{2.5}$ mass concentrations were below 15 μg m^{-3} at Dongsha Island and background sites (Hengchun and LABS). The PM_{10} in city areas (Chiang Mai, Hanoi and Da Nang) ranged from 70 to 110 μg m^{-3}, at least twice more than those in remote and background sites. The ratios of $PM_{2.5}/PM_{10}$ were greater than 0.70 for Doi Sethup and Hanoi, while below 0.45 for Dongsha Island and Hengchun, indicating that fine particles for the former were elevated due to the proximity of burning source(s) and/or secondary aerosol formation. This ratio was close to unity for LABS. Evidently, fine particles accounted for most of aerosol mass at LABS during the biomass burning season.

To our knowledge, Chuang et al. (2013a) is the first study to characterize the chemistry of biomass-burning aerosols near the source region in the northern SEA, i.e., border of Thailand and Myanmar (at Doi Suthep, near Chiang Mai city) where very intense fires burn nearby. Their results suggest the biomass-burning aerosols are mainly produced by smoldering softwood, which is also common in southern Tibet, China (Sang et al., 2013). Tsai et al. (2013) attributed episodic pollution in Chiang Mai to frequent biomass burning events at approximately the same time as Dongsha Experiment.

The spatial distribution of biomass burning activities from northern SEA to southern Tibet can be characterized by tracer

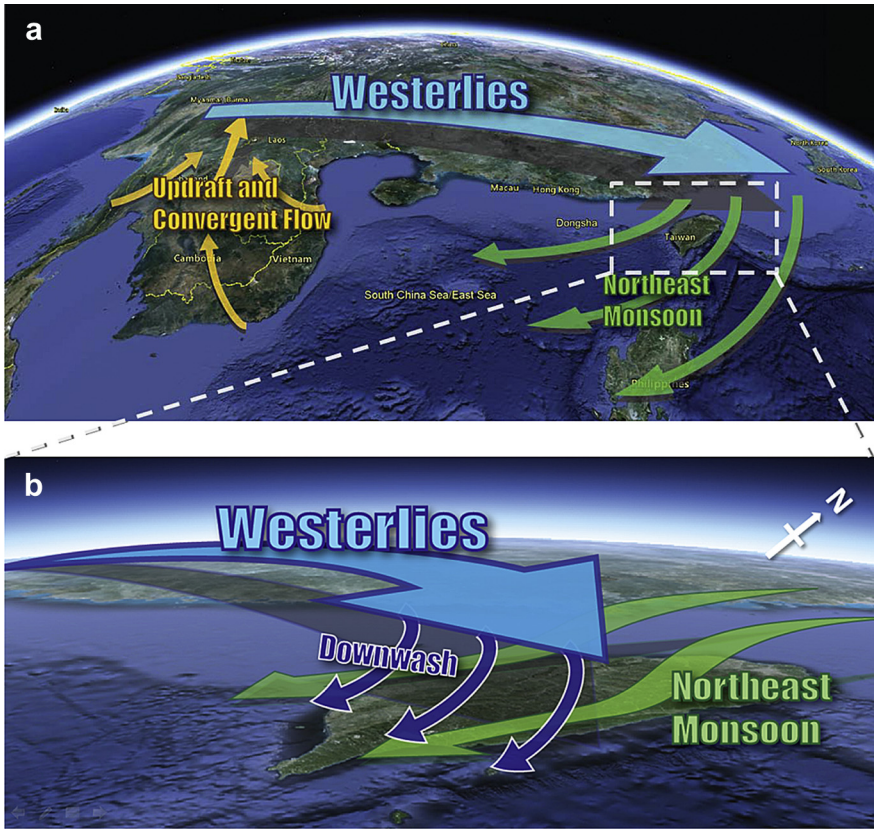

Fig. 3. Conceptual model for (a) climatological circulation pattern in March for high-altitude and low-level transport pathways by westerly and northeast monsoon flows, respectively, and (b) the integrated downwash branch over the downstream regions, e.g., Taiwan area.

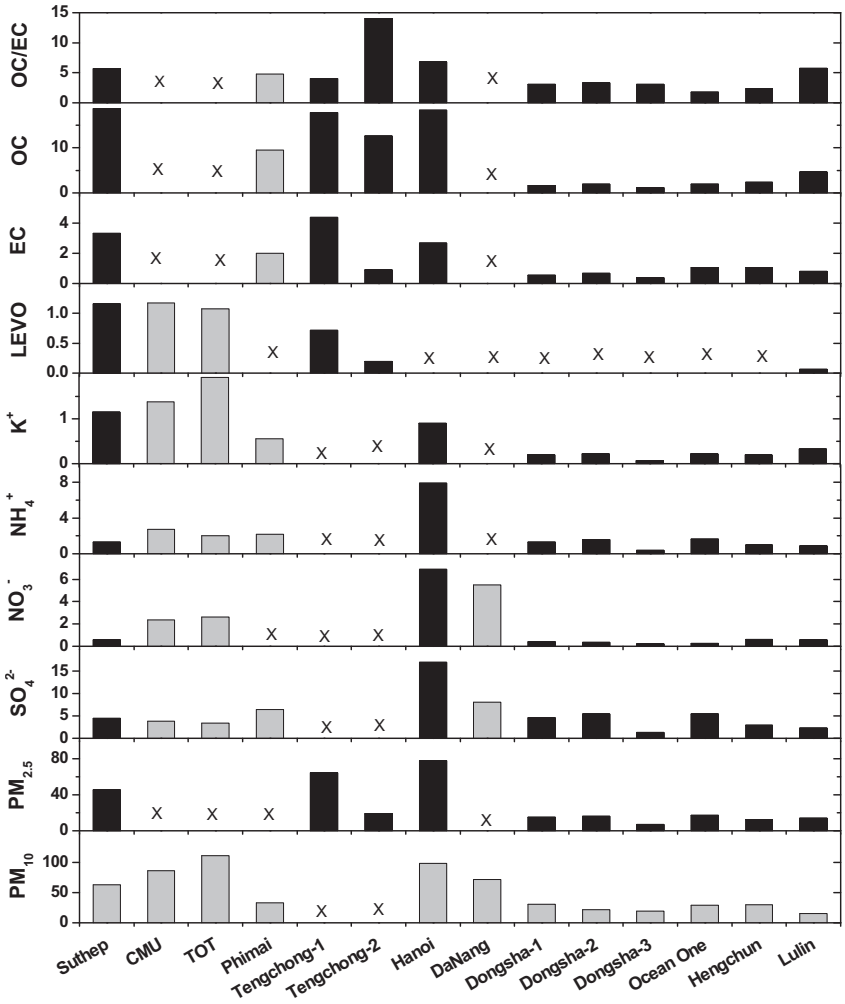

Fig. 4. Comparison of aerosol chemistry measured at sites of BASE-ASIA, 7-SEAS/Dongsha Experiment, and related studies. Black and gray histograms represent $PM_{2.5}$ and PM_{10}, respectively. Unit: μg m^{-3}. 'X' stands for no data available for that species. Suthep (near BB source): Chuang et al. (2013a); CMU (urban site) and TOT (industrial site): Tsai et al. (2013); Dongsha-1 (air masses from Asian continent), Dongsha-2 (air masses from Philippine) and Dongsha-3 (air masses from Pacific Ocean): Chuang et al. (2013b); Hanoi: Hai and Kim Oanh (2013); Lulin: Lee et al. (2011) ; Hengchun: Tsai et al. (2012); Tengchong-1 (suburban site) and Tengchong-2 (mountain site): Sang et al. (2013).

species, for instance, K^+ and levoglucosan (Cachier et al., 1991; Simoneit and Elias, 2001). The K^+ was substantially elevated at Doi Suthep and Chiang Mai city, as shown in Fig. 4. The K^+ in $PM_{2.5}$ at the near-source site reached 2.5 ± 0.3%, compared with about 1.5% in Chiang Mai city, and about 1.1% in Hanoi where biomass burning sources are the combustion of hardwood, softwood, and crop residue (Hai and Kim Oanh, 2013). In contrast, the ratios of K^+ to $PM_{2.5}$ are generally below 1.0% for most field and laboratory studies. The biomass-burning aerosols in the northern SEA obviously distinct from those measured in previous studies.

Among anhydrosugar species, levoglucosan has been confirmed as the most abundant species in biomass-burning aerosols in many studies (Simoneit and Elias, 2001; Engling et al., 2006; Pio et al., 2008). As shown in Fig. 4, levoglucosan in $PM_{2.5}$ is 1.2 ± 0.5 μg m^{-3} for Doi Suthep. In Chiang Mai city, levoglucosan in PM_{10} reached 1.2 ± 0.5 μg m^{-3} during biomass burning episodes. In Tengchong, over the southeastern Tibetan Plateau, levoglucosan in $PM_{2.5}$ were 0.7 ± 0.3 and 0.2 ± 0.1 μg m^{-3} at a mountain site and a suburban site, respectively. The levoglucosan concentrations appear to decrease from biomass burning sources to downwind areas. Although the degradation of levoglucosan has been widely hypothesized to be caused by oxidation with hydroxyl radical (OH) (Holmes and Petrucci, 2007; Hoffmann et al., 2010), direct observations of this reaction in the atmosphere are still lacking. Nevertheless, this mechanism may explain certain part of production of organic carbon, for example the humic-like substances (Schmidl et al., 2008). Other organic compounds such as dicarboxylates (e.g., oxalate, malonate, succinate, glutarate) and water-soluble organic carbon (WSOC) are also produced in biomass-burning aerosols (Yang et al., 2013). WSOC in aerosols may enhance cloud droplet production and growth, enhancing the aerosol indirect effect. Recent studies suggest the production of WSOC is related to aerosol aging (Aggarwal and Kawamura, 2009; Miyazaki et al., 2009). The WSOC in biomass-burning aerosols at both near-source and downwind areas was determined during the Dongsha Experiment to investigate the production of WSOC during the aging process (Lee, personal communication, in preparation for publication).

Particulate organic matter accounts for up to 85% of biomass burning particle mass (see Reid et al., 2005b; Akagi et al., 2011 for a list of studies). Organic carbon (OC) contains species like anhydrosugars and water-soluble species which can influence the hygroscopicity. Furthermore, elemental carbon (EC) usually has a high correspondence with black carbon (BC) (Lavanchy et al., 1999). Thus EC can also be considered as an indicator of absorption of solar radiation. Therefore, OC and EC measurements are of importance for understanding the impact of biomass-burning aerosols. Fig. 4

shows OC and EC are abundant in SEA and the southern China, but much less abundant outside SEA. In Fig. 4, the ratios of OC/EC were higher in SEA during Dongsha Experiment than other places, except at LABS, suggesting that LABS is affected by biomass-burning emissions from SEA. Besides, biomass-burning aerosols are highly related to the formation of clouds (Hsu et al., 2003). As described in Reid et al. (2005b, 2013), there are significant amounts of sulfate in biomass-burning aerosols, which is very hygroscopic. Indeed, the small inorganic fraction of smoke particle mass is seen as the best indicator of particle hygroscopicity (Carrico et al., 2010). Sulfate together with other water-soluble ions like ammonium and nitrate observed in Hanoi were higher than other locations (Fig. 4) implying the urban and industrial sources of hygroscopic aerosols is greater than that from biomass burning during the wintertime in Hanoi.

The biomass-burning plume from northern SEA was sampled at several locations along the transport pathway. Plume signatures were observed at LABS in central Taiwan (Lee et al., 2011), above boundary layer over Hong Kong (Chan et al., 2003; Deng et al., 2008), and possibly even near the surface in the southern Taiwan (Tsai et al., 2012; Yen et al., 2013). Modeling studies by Tang et al. (2003), Satake et al., 2004, Verma et al., 2009, and Fu et al. (2012) suggested SEA to EA biomass burning emissions can spread over southeastern China, SCS/ES, and migrate to Central Pacific, or shift to northeastern China, even to Japan. Wang et al. (2007) modeled the radiative forcing of these biomass-burning aerosols in March of 2001 during the TRACE-P experiment. The monthly mean clear-sky direct shortwave radiative forcing ranged from −1.9 to 0.4 W m^{-2} at the top of the atmosphere and from −0.5 to −12.0 W m^{-2} at the surface, resulting in an increase in the atmospheric heating rate from 0.01 to 0.3 °C day^{-1}. Atwood et al. (2013a) inferred the biomass-burning plume was transported aloft over the boundary layer from northern Vietnam and Dongsha Island, to Taiwan. Thus the near-surface aerosol concentrations observed at Dongsha Island were relatively unaffected by biomass burning during the experiment. This observation is consistent with Chuang et al. (2013b) who noted biomass tracer K^+ was hardly ever observed at Dongsha Island during the 7-SEAS experiment. Nevertheless, Tsai et al. (2012) observed the biomass-burning plume from SEA can impact air quality in the southern Taiwan, which was confirmed by Yen et al. (2013) for the cold surge events.

Aside from the transport of biomass-burning plume by high-altitude westerly winds, air masses traveling into SCS/ES via low-level transport (See Fig. 3) have significantly different characteristics. Air quality and atmospheric chemistry are rarely investigated in this oceanic region. Zhang et al. (2007) reported water-soluble ions and trace elements in atmospheric aerosols collected during two cruise missions in northern SCS/ES in 2003. Hsu et al. (2007) investigated marine aerosol during the northeastern monsoon on two wintertime cruises between Taiwan and Dongsha Island, suggesting that massive amounts of air pollutants from China and South Asia/SEA affected air quality in SCS/ES. They found nearly 90% chlorine loss from marine aerosol. Similar chlorine loss was also found for ORO cruise mission during the Dongsha Experiment. The high chlorine loss is due to heterogeneous reactions between sea salt aerosols and nitric and sulfuric acids produced by anthropogenic sources. For comparison, Chuang et al. (2013b) found chlorine loss at Dongsha Island to be about 70% for $PM_{2.5}$. This difference may result from the measurement locations for Hsu et al. (2007) being nearer anthropogenic sources.

Fig. 4 shows the $PM_{2.5}$ chemistry for Dongsha-1 (Asian source) and −2 (Philippine source) (Chuang et al., 2013b), ORO (NNW and SW airflows were prevailing for the first and second half period during 7-day cruise mission, respectively), and HC (NE and SW airflows were prevailing during two IOPs) are similar, particularly for aerosol mass and most of water-soluble ions, while were significantly lower than those measured in SEA. In contrast, $PM_{2.5}$ mass (~7 μg m^{-3}, on average) and all chemical compositions for Dongsha-3 (which is associated with clean air mass from West Pacific) were about 40–60% of the aforementioned measurements in SCS/ES and the southern Taiwan. For the above studies in the northern SCS/ES, the observed acidic pollutants represent the major anthropogenic contaminants to the region, especially sulfate (SO_4^{2-}). On average, SO_4^{2-} ions were 4.4 and 12.3 μg m^{-3} during two cruises near the coast of the southern China (Zhang et al., 2007); 1.4–6.4 μg m^{-3} during wintertime (Hsu et al., 2007) and 4.2–6.7 μg m^{-3} during springtime (this study) ORO cruise missions; and 1.3–5.5 μg m^{-3} at Dongsha Island (Chuang et al., 2013b). The NO_3^- ions were nearly half of SO_4^{2-} for the two cruises, reported by Zhang et al. (2007), and only 10–20% of SO_4^{2-} in the other studies. Obviously, the emissions of S and N compounds from the Chinese continent to SCS/ES are significant. By contrast, SO_4^{2-} ions ranged in 0.21–4.3 μg m^{-3} (average: 1.9 μg m^{-3}) and were more than 3 times NO_3^- during a cruise mission in fall in Central India Ocean, representing a relatively polluted condition in region (Balasubramanian et al., 2013).

The biggest dust storm on record to reach Taiwan occurred during the Dongsha Experiment. This dust event had its genesis in China on 19–20 March 2010 (Bian et al., 2011; Li et al., 2012), and reached the southern tip of Taiwan Island (Tsai et al., 2012) and Dongsha Island (Wang et al., 2011; Chuang et al., 2013b) on 21 March 2010. Both Bian et al. (2011) and Han et al. (2012) applied air quality models to simulate the dust event on 19–22 March 2010, and confirmed airborne dust can dramatically impact SCS/ES and SEA. The hourly and daily PM_{10} average concentrations surprisingly reached 570 and 94.1 μg m^{-3}, respectively (Wang et al., 2011) at Dongsha Island. A number of studies suggest the events, which transport dust to SCS/ES, are usually accompanied by frontal passages in springtime (Cohen et al., 2010; Atwood et al., 2013a). In addition, Asian dust was occasionally observed at Mt. Lulin in central Taiwan during the peak of biomass burning (Lee et al., 2011) suggesting the atmospheric dust layer reaches as high as 3 km. Lin et al. (2009) and Wang et al. (2012) characterized the impact of Asian dust aerosol on biogeochemistry in SCS/ES using satellite data. Wang et al. (2011) suggested the dust aerosols in heavy dust events contain bioavailable iron and enhance the phytoplankton growth in the oligotrophic northern SCS/ES.

4.2. Trace gases

During the 7-SEAS/Dongsha Experiment, overall mean mixing ratios of ozone were 44.2 ppb at LABS, 40.9 ppb at Dongsha Island, and 40.8 ppb at Hengchun (Table 2). At Chiang Mai, the source region for episodic pollution ($PM_{10} \geqq 120$ μg m^{-3}), the mean level of ozone was approximately 39.6 ppb and NO_x 17.9 ppb (Tsai et al., 2013), typical of urban conditions. Satellite data indicates biomass burning in SEA usually peaks in March, coincident with the CO seasonal maximum based on surface measurement at Srinakarin (14.36°N, 99.12°E, 296 m MSL) in rural Thailand (Pochanart et al., 2004). Ozonesonde measurements over Hong Kong as well as aircraft observations of ozone detected increases in ozone in air impacted by biomass burning over SEA in spring during the PEM-West B (Pacific Exploratory Mission-West B) and TRACE-P (Chan et al., 2003; Kondo et al., 2004). At the LABS mountain top site, which is downwind of biomass burning emissions from SEA, the seasonal maximum of CO and ozone were regularly observed in March (Ou-Yang et al., 2009, 2012). However, at sea level, e.g., Hengchun and Dongsha Island, elevated CO and ozone (i.e., 60 ppb) concentrations indicate strong northeasterly winds during the winter Asian monsoon, which transport polluted air masses from

Table 2
Averaged levels of trace gases measured at Dongsha Island, Hengchun, and LABS during the 7-SEAS/Dongsha Experiment.

Compound	Overall (10th March–26th June)			Spring (10th–31st March)			Summer (1st–26th June)		
	Dongsha	Hengchun	Lulin	Dongsha	Hengchun	Lulin	Dongsha	Hengchun	Lulin
In-situ measurements									
O_3 (ppb)	40.9[b]	40.8[a]	44.2	44.6[b]	43.3[a]	53.0	30.9[b]	30.9[a]	30.1
NO_x (ppb)	3.29[b]	4.29[a]	–	3.59[b]	3.96[a]	–	3.15[b]	4.42[a]	–
CO (ppb)	226.4[c]	178.4[a]	189.1	259.9[c]	222.9[a]	234.5	140.2[c]	97.4[a]	120.2
Flask air samples[d]									
CO (ppb)	213.7	–	187.0	243.9	–	227.9	109.5	–	134.0
CO_2 (ppm)	395.0	–	390.1	395.4	–	390.4	390.6	–	387.9
CH_4 (ppm)	1.86	–	1.84	1.88	–	1.85	1.81	–	1.83
N_2O (ppb)[e]	324.2	–	324.0	324.0	–	323.7	324.1	–	324.0
SF_6 (ppt)[e]	7.51	–	7.14	7.54	–	7.16	7.38	–	7.10

– Not applicable.
[a] Taiwan EPA air quality station.
[b] Taiwan EPA monitoring mobile.
[c] NASA/COMMIT.
[d] NOAA/ESRL/GMD CCGG cooperative sampling network.
[e] Defined by NOAA/ESRL/GMD CCGG as preliminary data.

the northern Asian continent. In contrast, ozone concentrations were lower (i.e., 30 ppb) when the monsoon subsided. This ozone level is typical for marine air. Shipborne measurements (11–16 July 2011) in northern SCS/ES also showed low ozone and CO concentrations (e.g., 22.5 ppb and 164.7 ppb, respectively). Close agreement ($R^2 = 0.76$) was found between the 24-h moving averages of ozone and CO for Hengchun and Dongsha Island, suggesting these sites experienced similar atmospheric influences (Ou-Yang et al., 2013). Both CO and ozone at Dongsha Island were significantly enhanced during the period of outflow from the Asian continent (Chuang et al., 2013b). This phenomenon was successfully simulated using an air-quality model (i.e., Taiwan Air Quality Model, TAQM) (Ou-Yang et al., 2013).

No diurnal variations of NO_x were seen at Dongsha Island or Hengchun during this campaign. The mean NO_x concentration at Dongsha Island was 3.3 ppb and at Hengchun was 4.3 ppb. These concentrations represent regional background values for the northern SCS/ES. At Chiang Mai, an important source region, the mean concentration for NO_x was 19.8 ppb during non-episodic pollution ($PM_{10} < 120$ μg m^{-3}, Tsai et al., 2013), which is about 4.6–6.0 times the NO_x observed at downwind surface sites in the Taiwan area. TAQM model calculations showed the ozone difference between runs with and without Taiwan emissions at Dongsha Island to be only 0.6 ppb on average. Thus Dongsha Island appears to be only minimally affected by Taiwan's emissions and is an ideal baseline site in northern SCS/ES (Ou-Yang et al., 2013).

Flask samples for greenhouse gases were taken weekly at LABS starting in August 2006 and Dongsha Island starting in March 2010. During the period of the 7-SEAS 2010 campaign, negligible differences (<3.0%) in the GHG concentrations were found between Dongsha Island and LABS, excepting CO (Table 2). Based on measurements from 2010 to 2012, distinct and similar seasonal patterns of CO_2, CH_4, and CO were observed at both Dongsha Island and LABS with a springtime maximum and a summertime minimum. Steady trends in growth of N_2O and SF_6 were found at both sites. From 2006 to 2011, SF_6 at LABS increased 0.28 ppt yr^{-1}, which is similar to Mauna Loa (0.29 ppt yr^{-1}). However, N_2O at LABS grew at a rate of only 0.77 ppb yr^{-1}, while at Mauna Loa N_2O increased at 0.86 ppb yr^{-1} during the same period.

Flask samples collected at Dongsha Island and Kumukahi (surface site in Hawaii) showed SF_6 levels in the northern SCS to be lower and more stable during the southwest monsoon than near Hawaii. Similar to the SF_6 data, the lowest concentrations of the CH_4 at Dongsha Island are lower and more stable than those measured at Kumukahi in mid-summer. Methane is higher at Dongsha Island than Hawaii during the winter when the air is coming off the Asian continent. Flask sample data showed a wintertime excess of CO_2 and CO from fossil fuel combustion and biomass burning had permeated the SCS/ES Basin. In spring and summer, vegetation growth controls the CO_2 concentration at Dongsha Island which is as low and stable as that measured at Kumukahi. The N_2O data at Dongsha Island showed a very similar trend to that of Kumukahi, but concentrations elevated when air masses come from the Asian continent.

Air quality at different altitudes was controlled by different sources and transport patterns. Based on real-time measurements and flask samples from Dongsha Island, trace gases were primarily controlled by the Asian continental outflow from the north driven by the winter monsoon. But at LABS, ~3 km asl, the seasonal pattern of the trace gases showed signs of biomass burning emissions from SEA. In contrast, oceanic influences from the Pacific Ocean dominated at the 7-SEAS sites both at the surface and in the LFT during summer.

4.3. Air toxics

4.3.1. Atmospheric mercury

Pacyna et al. (2006) estimated 2190 Mg of anthropogenic Hg was emitted during 2000, with Asian countries contributing about 54%. Moreover, Friedli et al. (2009) estimated biomass burning released 675 ± 240 Mg yr^{-1} of Hg to the atmosphere, of which 57 and 192 Mg yr^{-1} were emitted from SEA and equatorial Asia, respectively. In addition to anthropogenic emissions, biomass burning could be another important atmospheric Hg source in Asia. Exports of Asian anthropogenic and biomass-burning Hg emissions have been detected downwind between Japan and Taiwan in the marine boundary layer (Jaffe et al., 2005; Nguyen et al., 2010) and the free troposphere (Friedli et al., 2004; Sheu et al., 2010). However, the distribution and transport of atmospheric Hg in SEA and SCS/ES is poorly understood (Fu et al., 2010).

During the 7-SEAS/Dongsha Experiment, atmospheric Hg was measured at multiple sites in spring to characterize the distribution and transport of atmospheric Hg in SEA and northern SCS/ES. The 7-SEAS/Dongsha Experiment sampling sites included Hengchun, Mt. Lulin and Dongsha Island (Taiwan), Da Nang (Vietnam), Chiang Mai (Thailand) and aboard a research vessel (ORO) navigating over the northern SCS/ES between southern Taiwan and Dongsha Island. Concentrations of atmospheric Hg were also measured at ATM sites, i.e., Mt. Bamboo and Matsu Island (Taiwan) in the spring of 2010. Moreover, Hg measurements at Lanyu Island and Pengjiayu

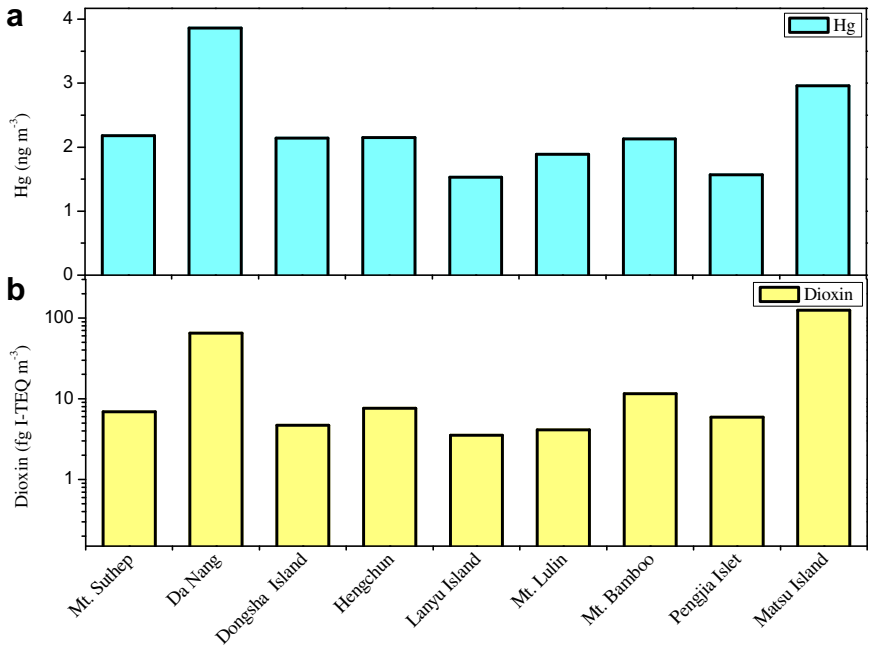

Fig. 5. Comparison of total Hg and dioxin concentrations measured at sites of 7-SEAS/Dongsha Experiment and ATM. Blue and yellow histograms represent Hg and dioxin, respectively. Unit: ng m^{-3} and fg I-TEQ for Hg and dioxin, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Islet (Taiwan) during 2008–2009 under ATM are included to better characterize the regional atmospheric Hg distribution and transport. The results are illustrated in Fig. 5a. The overall mean atmospheric Hg concentrations were 2.15, 2.14, and 2.01 ng m^{-3} at Hengchun, Dongsha Island, and ORO in the northern SCS/ES, respectively. The ranges and mean values of atmospheric Hg measured at Dongsha Island and ORO were quite similar, suggesting relatively homogeneous distribution of atmospheric Hg over the northern SCS/ES. Since no major anthropogenic Hg emission sources exist in Dongsha Island or over the northern SCS/ES, the measured atmospheric Hg concentrations likely primarily reflect the export of the East Asian Hg emissions by the northeast monsoon (Sheu et al., 2013). Export of atmospheric Hg from EA to East China Sea (ECS) in spring was observed at Matsu Island with a mean concentration of 2.96 ng m^{-3} and at Cape Hedo, Japan with a mean concentration of 2.04 ng m^{-3} (Jaffe et al., 2005). The Cape Hedo mean value was close to that at Dongsha Island and ORO, while the Matsu Island mean value was significantly higher likely due to its proximity (about 10 km) to the coast of southeastern China. Thus the springtime export of atmospheric Hg from the East Asian continent to downwind oceanic regions, including ECS and the northern SCS/ES, by regional monsoon activity is substantial.

Summer measurements of atmospheric Hg were conducted in 2008 at Dongsha Island and averaged 1.37 ng m^{-3}; further, at Lanyu Island the mean was 1.53 ng m^{-3} (Sheu et al., 2009). Both these values are close to Northern Hemisphere background concentrations of 1.5–1.7 ng m^{-3} (Slemr et al., 2003). At Dongsha Island, the 2008 summer mean value was 0.77 ng m^{-3}, lower than the 2010 spring mean value. Back-trajectories indicate summer air masses were mainly from the deep SCS, representing relatively clean marine air masses. Fu et al. (2010) also found lower Hg concentrations were usually associated with air masses from the open ocean, including the deep SCS/ES and the west Pacific Ocean. Sheu et al. (2013) indicate the atmospheric Hg concentration over the northern SCS and the west Pacific Ocean in summer is close to the background value. However, Hg concentrations can be enhanced in spring due to the export of the East Asian Hg emissions by regional monsoon activity.

At Da Nang, Hg concentrations ranged between 2.08 and 6.83 ng m^{-3} with an overall mean of 3.86 ng m^{-3}, which was mainly attributed to the influence of local emissions (Sheu et al., 2013). At Doi Suthep, Hg concentrations ranged between 1.66 and 2.95 ng m^{-3}, with an overall mean of 2.18 ng m^{-3}. Detailed chemical analyses of concurrently sampled aerosols indicated aerosols were enriched with biomass-burning tracers, while the influence of anthropogenic sources was negligible (Chuang et al., 2013a). The sampling site is located in a region of intense biomass burning. Trajectory analysis further indicated air masses frequently passed over the biomass-burning source region in southern Myanmar before reaching the sampling site. Therefore, atmospheric Hg measurements at Doi Sethup likely reflect the influence of local and regional biomass-burning Hg emissions.

Sheu et al. (2010) demonstrated the influence of springtime biomass-burning emissions from SEA on atmospheric Hg concentrations measured at LABS, a high-elevation receptor site. During the 7-SEAS/Dongsha Experiment, measurements of atmospheric Hg were conducted concurrently at Doi Sethup and LABS. The mean concentrations were 2.18 and 1.89 ng m^{-3}, respectively. Concentrations of Hg were always higher at Doi Sethup than LABS, which suggests that Mt. Lulin is under the influence of Southeast Asian biomass-burning Hg emissions in spring due to the transport of high altitude westerlies (Yen et al., 2013).

4.3.2. Dioxins

Persistent organic pollutants (POPs) such as PCDD/Fs, PCBs, PBDD/Fs, PBBs and PBDEs are persistent, bio-accumulative, toxic, and susceptible to long-range transport. The Stockholm Convention on POPs aims to protect human health and the environment by restricting and ultimately eliminating the production, use, release and unsafe disposal of POPs. Moreover, Stellman et al., 2003 showed PCDD/Fs are impurities found in commercial defoliants, especially in Agent Orange. These compounds, found in soils and sediments, have been shown (Masunaga et al., 2001) to be another source of PCDD/F contamination, in addition to those from combustion and industrial processes. Some natural processes, which

generate PCDD/Fs in small amounts, have been identified to be associated with biomass burning (Kolenda et al., 1994). It is well known that the main areas of wild fires lie in the tropical and subtropical areas of Africa, South America, SEA, and Australia. Previous studies also indicated (Gullett and Touati, 2003) the PCDD/F emission factor during biomass burning to range from 15 to 25 ng I-TEQ kg^{-1} burned. The mass-specific emissions are about 20 times higher than the concentration in the extracted biomass, suggesting that PCDD/F emissions are not simply a result of vaporization of cuticle-bound PCDD/Fs but are formed predominantly during biomass combustion.

Fig. 5b combines the atmospheric POPs measured (Chang et al., 2013; Chi et al., 2013; Thuan et al., 2013) at ATM and during the Dongsha Experiment, illustrating the background and elevated levels at remote sites and source region, respectively. Thuan et al. (2013) present the first detailed measurements of atmospheric PCDD/F in central Vietnam, they report relatively high atmospheric PCDD/F concentrations (23.4–146 fg I-TEQ m^{-3}) in Da Nang city. Additionally, significantly higher fractions of PCDDs (82 ± 12%) were also observed in Da Nang city. In Vietnam, dioxin contamination by Agent Orange can be characterized by the predominance of PCDDs, the contaminant of the herbicide 2,4,5-T. Hence, the high fraction of PCDDs observed in Vietnam in this study probably originated as anthropogenic emission from specific sources in Vietnam.

Chi et al. (2013) indicated that the highest PCDD/F concentrations (10.2–65.2 fg I-TEQ m^{-3}) and PCDD/F contents (132–620 pg I-TEQ/g-TSP) in ambient air were measured in northern Taiwan and at two islands in the Taiwan Strait close to mainland China during the northeast monsoon periods. Recent studies (Li et al., 2007a, 2007b) further indicated 972–51,200 fg-I-TEQ m^{-3} and 223–3454 fg-I-TEQ m^{-3} PCDD/Fs were found in the vicinity of electronic plant wastes in Guangdong and Zhejiang provinces of China, respectively. These values are dramatically higher than others measured around the world. Since Taiwan is an island located in the subtropics, PCDD/F concentrations should be relatively low. However, in the winter and spring, Taiwan and its surrounding areas are often influenced by northeast monsoon flow from October to March that originates from central Asia. Chi et al. (2013) conclude the significant increase in atmospheric PCDD/Fs measured during the northeast monsoon period was attributable to long-range transport of emissions from the coastal regions of mainland China.

Chang et al. (2013) is the first study to comprehensively explore the long-range atmospheric transport behavior of the five groups (PCDD/Fs, PCBs, PBDD/Fs, PBBs and PBDEs) of POPs and investigate the atmospheric characteristics of the POPs near the biomass-burning source regions of Chiang Mai in northern Thailand and Da Nang in central Vietnam along with LABS as a receptor of SE Asian biomass burning. The affected atmospheric POP concentrations at LABS were generally at least one order of magnitude lower than those in other two sites, except for elevated brominated POP (PBDD/Fs, PBBs and PBDEs) concentrations, especially PBDEs, which were attributed to biomass burning and were considered unlikely to be related to the local use of brominated flame retardants.

During the ATM and Dongsha Experiment (Chi et al., 2013; Chang et al., 2013; Thuan et al., 2013), mean PCDD/F concentrations in ambient air were 13.7 and 52.2–65.2 fg I-TEQ m^{-3} in Chiang Mai and Da Nang, respectively. These two sites were significantly affected by the biomass-burning and anthropogenic activities. In contrast, the atmospheric PCDD/F concentrations ranged from 1.43 to 35.2 fg I-TEQ m^{-3} at the background sites in Taiwan and several remote islands in Taiwan Straight, ECS, western Pacific Ocean and SCS/ES. These concentrations were considerably lower than those measured in other Asian countries (Lee et al., 2007; Makiya, 1999). However, the mean PCDD/F concentration of 35.2 fg I-TEQ m^{-3} measured in Matsu Island in Taiwan Straight (only 10 km from Mainland China, but 210 km from Taiwan Island) was significantly higher than that measured at any other site, especially during the northeast monsoon periods. This elevated mean PCDD/F concentration observed during the northeast monsoon periods can be attributed to the transport of anthropogenic emissions from mainland China. Meanwhile, atmospheric PCDD/F concentrations increased by a factor of 2–3 during the Asian dust storm event (March, 2010; see Wang et al., 2011) at the background site (from 3.06 increased to 6.69 fg I-TEQ m^{-3}) at Hengchun in southern Taiwan and Dongsha Island (from 1.43 increased to 4.20 fg I-TEQ m^{-3}) in SCS/ES. The PCDD/F concentrations at these two sites were mostly unaffected by local sources and possibly considered as background levels. Evidently, the elevated PCDD/F concentrations should be through the long-range transport of Asian dust that originated from northern China and traveled along coastal China (Wang et al., 2011). The dust particles mixed with anthropogenic aerosols, which greatly facilitated the transfer of PCDD/Fs emitted from local facilities in the coastal area of China to the solid phase.

5. Aerosol physical and optical properties

Aerosol optical and microphysical properties are important parameters, which are commonly used for interpreting aerosol loading, types, and hygroscopicity, as well as to estimate aerosol radiative effects. It is valuable to link those parameters, which can be measured *in-situ*, with satellite observations (Reid et al., 2013). However, the relationship between *in-situ* and satellite measurements over northern SEA is not straightforward due to the complex nature of aerosol dynamics and the dearth of *in-situ* measurements in this region (e.g., Tsay et al., 2013; Given an example as in Fig. 6). Further, consistency between *in-situ* measurement of bulk chemistry, microphysical, and optical properties with a theoretical aerosol model in the dynamic aerosol-rich environment at Dongsha Island is elusive (Bell et al., 2013). Understanding the discrepancy between measured and modeled parameters is important for advancing remote sensing and climate studies.

From satellite observations, the regional picture of biomass burning in northern SEA has been described. Based on analysis of inter-annual variations of AOD and fire counts from MODIS, Hyer et al. (2013) and Gautam et al. (2013) report significant biomass-burning activities encompassing Myanmar, Laos, and Thailand usually reaching maximum intensity in March (also shown in Fig. 7). A strong gradient in aerosol loading and vertical distribution was observed over SEA from the relatively clean equatorial environment to heavy smoke-laden northern SEA (Gautam et al., 2013). Smoke near source regions was typically confined to the lower 2 km boundary layer, but was sometimes found as high as 4 km (e.g., Huang et al., 2011; Campbell et al., 2013; Gautam et al., 2013).

To help bring some consistency to comparisons across northern SEA, AERONET data are provided and compared to other observations in the literature. Fig. 7 shows the AERONET sites (also see Fig. 1) in the 7-SEAS experimental region and gridded climatology (2003–2011) fire counts in March from the MODIS. A more detailed discussion of site-by-site differences can be found in Reid et al. (2013). Fig. S1 shows the monthly variations of AOD during 2008 and 2010 for five selected AERONET sites. The Chiang Mai, Silpakorn University, and Mukdahan sites are considered as source regions, while LABS and Dongsha Island are considered more remote. We found the five sites showed consistent seasonal variations, suggesting a consistent source–receptor relationship between these sites. During March at Chiang Mai and LABS, AODs decreased (Fig. S1) while single scatter albedo and asymmetry parameter

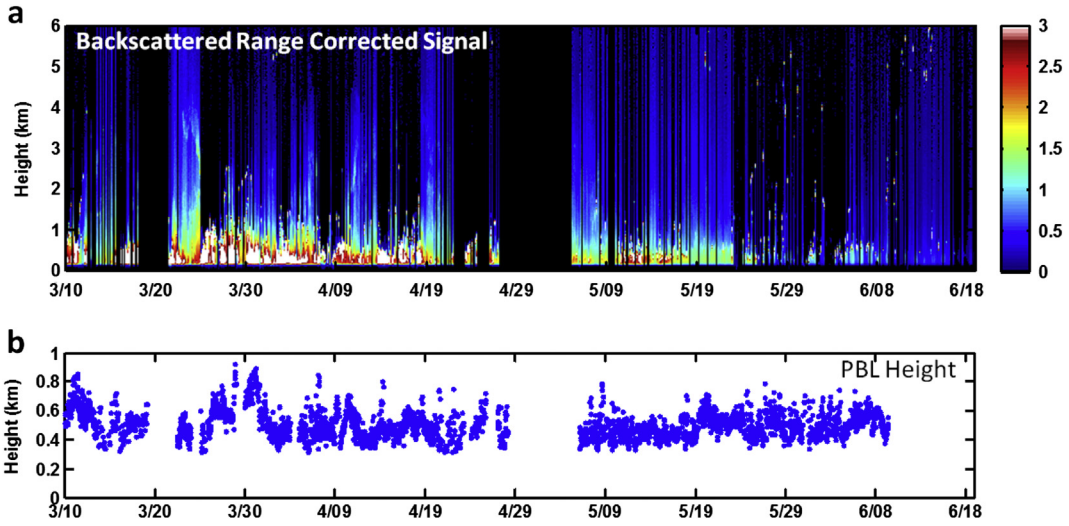

Fig. 6. Time series of (a) vertical distribution of the backscattered range corrected signal and (b) PBL height at Dongsha Island during 7-SEAS/Dongsha Experiment.

increased (Fig. S2) during aerosol transport. By contrast, Salinas et al. (2013) observed high AOD during dry months of August and September in 2011 at Kuching, East Malaysia, which was attributed to regional episodes of biomass burning. Atwood et al. (2013b) also showed that biomass-burning emissions were the likely source of the elevated AODs and surface aerosol measurements in the 7-SEAS supersite located in Singapore between August and October 2009 which coincided with a moderate El Nino event and enhanced tropical burning, particularly in peatlands.

Very few direct *in-situ* measurements of aerosol microphysical properties have been made in SEA up into the present 7SEAS/Dongsah Experiment (Reid et al., 2013). In this special issue, three papers present results from comprehensive *in-situ* aerosol measurements carried out in Phimai/central Thailand (Li et al., 2013), Dongsha Island/northern SCS/ES (Wang et al., 2013b), and LABS/central Taiwan (Chen et al., 2013). The aerosol at these three sites was dominated by submicron particles, consistent with high Ångström exponent from AERONET observation (Table S4). Aerosol observations in Phimai showed complex characteristics in terms of composition and properties, attributed to diverse aerosol sources (agricultural fires, industrial and vehicular emissions) and hygroscopic growth effects (Li et al., 2013). In addition, aerosol optical properties over the SEA were found to exhibit a significant diurnal cycle, which may relate to the evolution of the atmospheric boundary layer (Li et al., 2013; Gautam et al., 2013; See Fig. 6). Results from the Dongsha Experiment imply high aerosol concentrations observed at Dongsha Island are typical of continental origin, mainly from northern China to the northern SCS/ES, passing the coastal area and being confined in the marine boundary layer (Wang et al., 2013a). Observed moderately absorbing ($\omega \sim 0.94$ at 500 nm) and hygroscopic aerosol (f(RH) $\sim$ 2.1 at 85%) implies a mixture of marine and anthropogenic particles. Aerosol size distribution and size-resolved chemical composition (including biomass-burning derived fine potassium) were measured at LABS during summer and winter seasons (Chen et al., 2013).

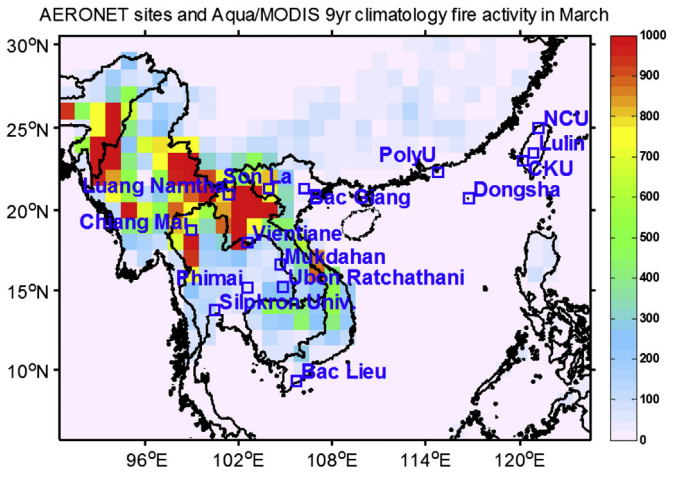

Fig. 7. The locations of AERONET sties collaborating with 7-SEAS project in northern SEA. Also shown are the gridded climatology (2003–2011) fire counts in March from the MODIS instrument aboard the Aqua satellite (http://disc.sci.gsfc.nasa.gov/neespi/data-holdings/myd14cm1.shtml).

6. Modeling and impact studies

6.1. Model simulation of biomass-burning pollutants transport

Under the framework of BASE-ASIA, the Models-3/Community Multiscale Air Quality (CMAQ) model was utilized to study the impact of biomass burning on SEA and EA. FLAMBE (Reid et al., 2009) was used as the biomass-burning emission inventory input for model during the BASE-ASIA study period of March to May, 2006 (Fu et al., 2012; Huang et al., 2013). Biomass-burning emissions in the spring of 2006 peaked in March–April when most intense biomass burning occurred in Myanmar, northern Thailand, Laos, and parts of Vietnam and Cambodia.

Model performance was evaluated using both ground-based measurements and satellite observations. CMAQ simulated the spatial pattern and temporal variations of CO reasonably well in comparison to both satellite and ground measurements. Overestimation or underestimation occurred in different regions due to large uncertainties in the biomass-burning emission inventory. The largest discrepancy between modeled and observed CO occurred in northern Thailand, where peak episodes were overestimated by a factor of 2–3. Evaluation of several datasets, i.e., EANET (Acid Deposition Monitoring Network in East Asia), AERONET, and Taiwan supersites' data, illustrated distinct regional differences in aerosol chemical and optical properties. Local biomass burning, anthropogenic sources, and long-range/regional transport were the

main factors controlling the aerosol chemistry. The highest concentrations of particulate K^+, SO_4^{2-}, NO_3^-, and NH_4^+ were observed at Hanoi in Vietnam. Correspondingly, the model estimated the highest AOD and AAOD (absorption AOD) at Bac Giang in Vietnam. The contribution of biomass burning to AOD was estimated to be over 56%. In Thailand, the magnitudes of major aerosol chemical components, AOD, and AAOD were much lower than in Vietnam. The contribution of biomass burning to AOD was about 18–50%. The contribution of biomass burning to AOD at Hong Kong and Taiwan was also significant (within the range of 26–62%). The observed monthly mean single scattering albedo was around 0.90 during intense biomass-burning periods, suggesting high production of strongly absorbing substances (i.e., EC) due to biomass burning. Modeled concentrations of aerosol chemical components were biased low in most circumstances, and the modeled AOD values were biased low by about a factor of 2, probably due to underestimation of biomass burning emissions.

The Asian spring monsoon facilitated the impact of biomass burning extending from SEA to the SCS/ES, the Taiwan Strait, and some provinces in southern China. Different spatial distribution patterns of biomass-burning AOD between March and April were caused by variable wind fields. In March, the average wind fields at the latitudes of 17°N–25°N blew primarily from west to east, which carried pollutants eastward. Below 17°N, winds blew from the east over the western Pacific and then circulated to higher latitudes which formed an anticyclone, carrying large quantities of biomass-burning emissions over the SCS/ES. Biomass burning contributed about 30–60%, 10–20%, and 20–70% to the total CO, O_3 and $PM_{2.5}$ concentrations, respectively. In April, the wind fields changed and mostly blew from low to high latitudes, which pushed pollutants north and eastward, while the SCS/ES was less impacted. Biomass burning CO reached the Yangtze River Delta with concentrations of 60 ppb (10–20%). High concentrations of O_3 extended farther in April. The O_3 concentration reduction ranged from 9 to 11 ppb in the Pearl River Delta region, the Guangxi province in China, and large areas of the SCS/ES. $PM_{2.5}$ was transported more widely and influenced major areas of southern China, with particulate contributions from biomass burning ranging from 10 to 30%.

Two intensive episodes during BASE-ASIA were further analyzed. For the first episode on March 27, the influence of biomass burning in the source region contributed to CO, O_3, and $PM_{2.5}$ concentrations as high as 400 ppb, 20 ppb, and 80 μg m^{-3}, respectively. They were reduced by 10–60%, 10–20% and 30–70%, respectively, when no biomass burning occurred. Long-range transport spread these pollutants over the southeastern parts of mainland China, including the Pearl River Delta region and the Fujian province in China. Biomass burning in this region resulted in 160–360 ppb CO, 8–18 ppb O_3 and 8–64 μg m^{-3} $PM_{2.5}$, respectively; and the percentage could reach 20–50% of CO, 10–30% of O_3, and as high as 70% of $PM_{2.5}$. During the second episode period on April 13, transport was weaker to the southern areas of China than the first episode, probably reflecting differences in both biomass-burning intensity and wind patterns. The amount of biomass burning derived CO and O_3 in SEA and southern China reached approximately 100 ppb and 6 ppb, respectively, while amount of $PM_{2.5}$ was not very significant.

Two cross-sections at 15°N and 20°N were selected to compare the vertical flux of biomass burning. In the source region (SEA), CO, O_3, and $PM_{2.5}$ concentrations showed transport from the surface to high altitudes (Also referred to Yen et al., 2013). The transport became strong from 2 to 8 km in the free troposphere, and the pollutants were quickly transported eastward due to a strong westerly. The subsidence during the long-range transport contributed 60–70% CO, 20–50% O_3, and 80% $PM_{2.5}$, respectively, to the surface in the downwind area.

6.2. Impact studies of biomass burning

In SEA, biomass-burning smoke often degrades air quality and alters aerosol composition. For example, Mahmud (2013) evaluated PM_{10} levels in August 2004, the peak month of biomass-burning activity, at seven air quality stations in Sarawak state that lies along the northern coast of Borneo, Malaysia, and are located downwind from the centers of intensive fires. The daily average PM_{10} concentration limit of 50 μg m^{-3} by WHOAQG (World Health Organization Air Quality Guidelines) was exceeded by 74–90% for six stations, and by 29% for one station distant from the fire source. Meanwhile, relatively high AOD, greater than 0.9 was detected from the last week of August when the intense biomass-burning activities occurred in the western, central and southern Borneo.

In Singapore, particulate matter is significantly elevated by transboundary biomass-burning smoke during peak fall season (Hyer and Chew, 2010; Wang et al., 2013a; Atwood et al., 2013b). For example, it increased by about 40% for both average $PM_{2.5}$ and PM_{10} mass concentrations for the one-month experiment during 8 September to 5 October, 2008 (Yang et al., 2013). During the smoky period (19–30 September), levoglucosan exhibited an averaged 154.9 ng m^{-3}, which was around 5.6 times of its concentration (27.6 ng m^{-3}) when smoke was absent. Similar values were found for 2009 (Atwood et al., 2013b). The C2–C5 total dicarboxylates (TDCAS, the sum of dicarboxylic acids (DCAs) and dicarboxylate salts (DCS)) increased more than twofold on average. Transboundary smoke increased malic acid concentrations more than 3.5 times, which is the largest relative increase among the measured TDCAS. Considering that malic acid is mainly generated through ambient photooxidation, such a significant increase in malic acid demonstrates extensive photooxidation of smoke.

Permadi and Kim Oanh (2013) estimated an emission inventory for biomass open burning (OB) sources including forest, agro-residue and municipal solid waste (MSW) in Indonesia for year 2007, covering toxic air pollutants and greenhouse gases (GHGs). It was estimated that crop residue OB contributed more than 80% of the total biomass OB emissions, followed by forest fires at 2–12% (not including peat soil fire emission) and MSW (1–8%). The total net GWP (Global warming potential) of warming species from biomass OB in the country constituted about 0.9–1.1% of that from the global biomass OB. BC released from OB was the third most important warming agent, contributing 21% (20 year horizon) and 12% (100 year horizon) of the total global warming forcing as compared to 30 and 60%, respectively, for CO_2 emissions.

Atmospheric BC emitted from SEA fires could increase Himalayan glacier melting through BC deposition, the so-called snow darkening effect. Yasunari et al. (2013) estimated BC dry deposition rates ranging from 270 to 4700 mg m^{-2} over the southern Himalayas in spring. Results from sensitivity tests suggest the surface roughness parameter and wind speed are the most critical factors in determining BC deposition rates on Himalayan glaciers.

7. Concluding remarks and perspectives

This paper has provided an overview of results from the BASE-ASIA and 7-SEAS/Dongsha Experiment and related studies from twenty-eight papers collected in the present special issue entitled "Observation, modeling and impact studies of biomass burning and pollution in the SE Asian Environment." It summarizes the experiments and regional weather/climate, chemical characterization of biomass-burning aerosols and related pollutants in source and sink regions, measurements of air toxics (atmospheric mercury and dioxins) in source and remote areas, aerosol physical characterization, and modeling and impact studies. The major scientific findings are highlighted below:

1. A conceptual model of atmospheric circulation over the large-scale environment in SEA, favorable for transport of biomass-burning pollution has been constructed. The coupling southerlies at 925 and 850 hPa levels over northern SEA during March and April convey the air pollutants due to active biomass burning over SEA. Later, an upward branch of the local East–West longitude-height circulation is enhanced by a well-organized convergent center that forms over northern SEA at 925-hPa, particularly in March, allowing the biomass-burning pollutants to be uplifted to the free troposphere and immediately transported downwind to the east. Meanwhile, atmospheric pollutants originating from the Asian continent can be conveyed into SCS/ES and even northeastern SEA via low-level transport by northeast monsoon flows. Surprisingly, it is first discovered that during the period of 7-SEAS/Dongsha Experiment, the biomass-burning pollutants at high-altitudes were brought down to the surface of southern Taiwan by downward branch of the transient local East–West circulation, which was induced by the subsidence of a cold surge anticyclone. Both surface observations and model studies have confirmed the enhancement of local air quality in southern Taiwan.
2. The 7-SEAS/Dongsha Experiment is the first study to characterize the chemistry of biomass-burning aerosols near the source region in northern SEA, and compare it with those acquired from concurrent measurements in background and remote sites. Overall, $PM_{2.5}$ mass concentrations were relatively high near burning source regions, while much lower in Dongsha Island and background sites. The K^+ ion and levoglucosan, as the markers of biomass-burning smoke, were abundant near source regions, while decreasing from biomass-burning sources to downwind areas. For the Dongsha Experiment, OC and EC were also abundant in northern SEA and southern China, but much less elsewhere. The ratios of OC/EC were also higher in northern SEA than other places except those at LABS. The biomass-burning aerosols in northern SEA present different characteristics from previous studies in other regions.
3. During the Dongsha Experiment, the $PM_{2.5}$ chemistry for background Dongsha Island and Hengchun sites, and one cruise mission, were similar, particularly for mass and most of water-soluble ions, while were significantly lower than those measured in northern SEA. The observed acidic pollutants represent the major anthropogenic contaminants to the region, especially sulfate (SO_4^{2-}). A super dust storm was serendipitously observed to invade into SCS/ES on 21 March 2010. It was suggested the dust aerosols in heavy dust events contain readily bioavailable iron and enhance the phytoplankton growth in the oligotrophic northern SCS/ES.
4. Air quality at different altitudes was dominated by different sources during the 7-SEAS/Dongsha Experiment. Trace gases at Dongsha Island were mainly affected by the Asian continental outflows driven by the NE monsoon flows. However, trace gases at higher altitudes, such as the elevation of LABS ($\sim$3 km), revealed signals of biomass burning arising from northern SEA. At the surface sites, e.g., Hengchun and Dongsha Island, strong northeasterly winds arising from the winter Asian monsoon may have transported polluted air masses to the northern SCS/ES, as indicated by elevated ozone levels of approximately 60 ppb. This level of ozone was significantly higher than the mean level of 39.6 ppb seen at the Chiang Mai urban site. Relatively low levels of NO_x with mean concentrations of 3–4 ppb and the lack of distinct diurnal features were seen at Dongsha Island and Hengchun during the campaign, representing the baseline conditions for the region of northern SCS/ES during the monsoon season. In contrast, the average NO_x level was 19.8 ppb at Chiang Mai urban site during the non-episodic period. As a result, the NO-titration was suspected to be the cause of low ozone seen at the Chiang Mai urban site.
5. At LABS, situating at the downwind path of biomass burning emissions from northern SEA, the seasonal maxima of CO_2, CH_4, CO and ozone were repeatedly observed during March as the result of the transport of air masses from the source region of SEA. During the period 2006–2011, continued increase in the growth rates of N_2O and SF_6 was observed at both Dongsha Island and LABS.
6. During the Dongsha Experiment a relatively homogeneous distribution of mean atmospheric Hg from southern tip of Taiwan to the northern SCS/ES, including cruise mission, were measured in a range of 2.0–2.15 ng m^{-3}. This is comparable to 2.04 ng m^{-3} observed at Cape Hedo (Japan) in East China Sea (ECS). On the other hand, atmospheric Hg observed at Matsu Island, with a mean of 2.96 ng m^{-3}, was significantly higher than Dongsha likely due to its proximity (about 10 km) to the coast of southeastern China. This indicates that the export of atmospheric Hg from the East Asian continent to the downwind oceanic regions, including ECS and the northern SCS/ES, by regional monsoon activity is an extensive phenomenon in spring. In contrast, regional summertime atmospheric Hg concentrations over open oceans are close to the Northern Hemisphere background concentration of 1.5–1.7 ng m^{-3}. For northern SEA, mean Hg concentrations measured at Da Nang and Chiang Mai were 3.9 and 2.2 ng m^{-3}, respectively, which were mainly attributed to the influence of regional and local emissions, particularly from biomass-burning activities for the latter.
7. From satellite observations, significant biomass-burning activities encompassed Myanmar, Laos, and Thailand, usually reaching maximum intensity in March. A strong gradient in aerosol loading and vertical distribution was observed from the relatively clean equatorial environment to heavy smoke-laden northern SEA. Smoke near source region was found to be typically confined to the lower 2 km boundary layer, and elevated as high as 4 km upon strongest over regional mountain ranges. Ground-based lidar observations at Dongsha Island indicated that most aerosols distributed within the marine boundary layer (MBL) of around 0.5 $\pm$ 0.09 km, representing a typical MBL height for a sub-tropical maritime environment during springtime. Meanwhile, biomass-burning aerosols from northern SEA were transported in the lower free troposphere (3–4 km) over the northern SCS/ES. Further lidar parameter of asphericity suggested these biomass-burning aerosols to be moderately non-spherical. It was also found the stratiform clouds are prevalent in the shallow boundary layer with cloud tops at $\sim$2 km and depths of $\sim$1 km, overlaid the low-level anthropogenic aerosol layer over the northern SCS/ES. The complex vertical distributions of aerosols and clouds have important implications for remote-sensing observations and atmospheric radiative transfer.
8. The CMAQ model simulation for SEA during BASE-ASIA period indicated that the contribution of biomass burning to AOD over northern Vietnam was estimated to be over 56%, a significant influence; about 18–50% over Thailand, a moderate impact; and downwind at Hong Kong and Taiwan within the range of 26–62%, a significant contribution. The modeled AOD was biased low about a factor of 2, probably due to the underestimation of biomass-burning emissions. The model results captured the transport pathway of biomass-burning pollutants well, showing that in source region the pollutants had a strong upward transport from surface to high altitudes, and were quickly transported eastward due to a strong westerly. The subsidence during the long-range transport contributed 60–

70% CO, 20–50% O_3, and 80% $PM_{2.5}$, respectively, to surface in the downwind area. Model results are evidently consistent with the conceptual model of springtime circulation flows over the large-scale environment in SEA we constructed, based on climatology and observations.

We have presented our recent 7-SEAS deployments and major scientific findings of regional biomass-burning studies that advance the current fundamental understanding with regards to dynamical, chemical, optical, microphysical, and radiative characteristics of aerosol and clouds over northern SEA. In conjunction with satellite overpasses, as elucidated in Tsay et al. (2013), the strategic deployments of ground-based supersites (e.g., SMARTLabs mobile laboratories) combined with distributed networks (e.g., AERONET/MPLNET) and regional contributing measurements near/downwind of aerosol source regions and along transport pathways, offer a synergistic approach for further exploring many key atmospheric processes (e.g., complex aerosol–cloud interactions) and impacts of biomass burning on the surface–atmosphere energy budgets during lifecycles from source to receptor areas. These methodologies, developed through our years of experience, serve as a call to action, baselining potential pathways to an improved understanding of the regional aerosol effects, which continue to be one of the largest uncertainties in climate forcing. The first such action is a joint international effort, named as the 7-SEAS/BASELInE (Biomass-burning Aerosols & Stratocumulus Environment: Lifecycles and Interactions Experiment), which is scheduled to commence in springtime 2013 in northern SEA. Thus, 7-SEAS/BASELInE will focus on addressing the following key questions:

- ***What processes, interactions, and feedbacks control the lifecycle of biomass-burning aerosols from source to receptor regions in boreal spring Southeast Asia?***
- ***How do droplet nucleation and the resulting products (e.g., cloud albedo, water content, precipitation efficiency, etc.) of warm rain processes in the cloud-resolving model simulations respond with respect to key aerosol physicochemical observables (e.g., number/mass concentration, size/shape distribution, and chemical composition) over a regional scale?***
- ***How do changes to aerosol–cloud interactions in boreal spring Southeast Asia provide feedbacks to regional climate through exchanges of energy, water, gases, and particulate matter between the surface and troposphere?***

Although it is unlikely to be able to tackle all these questions in one deployment, through integrated and synthesized space-borne and ground-based measurements and comprehensive modeling efforts, 7-SEAS/BASELInE would provide a basis for a deeper improvement of the understanding of the lifecycles of regional biomass-burning aerosols, and their interactions with stratocumulus clouds during the Asian pre-monsoon season. The results from these planned studies, in turn, would shed new light on the range of potential effects on water cycle due to regional anthropogenic activities.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.04.066.

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