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Atmospheric Chemistry and Physics
Discussions

Aerosol meteorology and Philippine receptor observations of Maritime Continent aerosol emissions for the 2012 7SEAS southwest monsoon intensive study

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ABSTRACT:
The largest 7 Southeast Asian Studies (7-SEAS) operations period within the Maritime Continent occurred in the 2012 August-September biomass burning season. Included where an enhanced deployment of Aerosol Robotic Network (AERONET) sun photometers, of multiple lidars, and of a Singapore supersite. Simultaneously, a ship was dispatched to the Palawan Archipelago and Sulu Sea of the Philippines for September 2012 to observe transported smoke and pollution as it entered the southwest monsoon monsoonal trough. Here we describe the nature of the overall 2012 southwest monsoon biomass burning season, but focus on the findings of the research cruise and the aerosol meteorology of this convectively active region. This 2012 cruise followed a 2 week cruise in 2011, and was in part consistent with the findings of that cruise for how smoke emission and transport relate to monsoonal flows, the propagation of the Madden Julian Oscillation (MJO), tropical cyclones, and covariance between smoke transport events and the atmosphere's thermodynamic structure. Aerosol observations in the 2011 cruise also highlighted the importance of squall lines and cold pools as they propagate across the South China Sea, scavenging aerosol particles in their path. For 2012, the cruise experienced differing environments. The monsoonal flow direction was perturbed by easterly waves, leading at times to total flow reversal in the South China Sea. Two category 5 typhoons just east of the Philippines also modulated flow patterns and convection. Whereas in 2011 large synoptic scale aerosol events transported high concentrations of smoke into the Philippines over days, in 2012, measured aerosol events exhibited a much more short term variation, sometimes only over 3-12 hours. Navy Aerosol Analysis and Prediction System (NAAPS) simulations captured longer wavelength aerosol events quite well, but largely failed to capture the timing in high frequency phenomena. Also observed were nucleation events in cleaner and polluted conditions, as well as in urban plumes. Combined, observations indicate pockets of high particle counts are not uncommon in the region. Perhaps most interestingly, several cases of squall lines heralding major aerosol events were observed, as opposed to the previous observations in 2011 of these lines largely scavenging aerosol particles from the marine boundary layer. We hypothesize that these phenomena may originate from weakly forced convection ahead of polluted land breeze fronts caught in strong monsoonal flows. Ultimately, the research findings of the 2012 cruise nicely complement the narrative started by the 2011 research cruise, and point to the importance of small scale phenomena such as sea breezes and squall lines embedded in the large scale monsoonal flow patterns in dominating aerosol lifecycle and potentially effects. "Pure" biomass burning plumes are relatively rare and are usually mixed with significant amounts of anthropogenic pollution.

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1.0 INTRODUCTION AND BACKROUND

The 7 Southeast Asian Studies (7SEAS) program has motivated observation and analysis of aerosol/meteorology interactions in the Maritime Continent (MC) that have led to significant advances in understanding the life cycle of aerosol particles in this region. While linkages between biomass burning to the El Nino Southern Oscillation (ENSO) have long been identified (e.g., Nichol 1998; Field and Shen 2008), the relative importance of the Madden Julian Oscillation (MJO), equatorial waves, tropical cyclones and even features as fine as boundary layer dynamics and the sea breeze have been recently connected (e.g., Reid et al., 2012; 2015; Atwood et al., 2013; Campbell et al., 2013; Wang et al., 2013; Xian et al., 2013; Ge et al., 2014). Yet the complexity of MC meteorology continues to pose great challenges to quantitative characterization and prediction of MC atmospheric composition and potential climate impacts. Based on the early findings of the Coordinate Regional Climate Downscaling Experiments (CORDEX; http://www.cordex.org/; Giorgi et al., 2012), climate model simulations for the Southeast Asian domain diverge widely and have strong biases in regard to temperature and precipitation At the same time, large uncertainties in diagnosed precipitation estimates have been identified based on the challenges of extrapolating across a complicated meteorological regime (Jamandre and Narisma, 2013). Atmospheric models have difficulty representing complex tropical waves in the MC, on scales ranging from Kelvin waves through the Madden Julian Oscillation and the regional monsoon (e.g, Misra and Li, 2014; Zhang, 2014). Near ubiquitous cloud cover hampers many aspects of remote sensing observation (Reid et al., 2013; Campbell et al., 2016). Field measurements in the MC are difficult to obtain and quality assure. Ultimately, the complexity and high degree of variability of the MC aerosol environment poses great challenges in determining how aerosol particles, weather, and climate interact.

The 7SEAS program was formed in the late 2000’s to enhance communication between scientists interested in Southeast Asian aerosol impacts on weather and climate (*Reid et al.*, 2013; *Lin et al*. 2013). 7SEAS quickly developed into a grass-roots organization, with a strong focus on collecting information on aerosol properties and lifecycle in a region with limited data. Central to this effort were studies that employed surface observations to verify and interpret satellite remote sensing data, which in turn was assimilated into, or used to evaluate, models. In this manner, 7SEAS allowed for some of the first end-to-end aerosol lifecycle studies in Southeast

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Asia, and in the MC in particular. Persistent observations across the region suggest a natural boundary between the regions' boreal spring northeast monsoon and summer/fall southwest monsoon. 7SEAS data collection and analysis also tended to follow these monsoonal lines, with one component emphasizing the springtime Peninsular Southeast Asia biomass burning season of Myanmar, Thailand, Laos and Cambodia with Vietnam and Taiwan as receptors (e.g., see review Lin et al., 2013), and another component emphasizing the MC.

Although all seasons are of interest, within the MC, 7SEAS field measurements and lifecycle studies have focused on the August through October biomass burning season in Indonesia and Malaysia, with Singapore and the Philippines as key receptors (e.g., Reid et al., 2012; 2015; Atwood et al., 2013; Chew et al., 2011; 2013; Salinas et al., 2013; Xian et al., 2013; Wang et al., 2013; Yang et al., 2013). Enhanced deployments of Aerosol Robotic Network (AERONET; Holben et al., 1998) sun photometers to the MC began in 2007 (Salinas et al., 2009), followed by the development of a Singapore based supersite (Atwood et al., 2013) and long term installations of MicroPulse Lidar Network (MPLNET) lidars in 2009. Each year, progressively more instrumentation or intensive measurement initiatives were added to the region. This measurement effort reached a crescendo in the summer of 2012, where a series of additional sun photometers, lidars and other aerosol instruments were deployed to the MC in an above-average biomass burning year.

Included in the 2012 deployment was a research cruise by the M/Y *Vasco*, a 35 m Philippine flagged vessel to study aerosol properties on the edge of the southwest monsoonal trough. Studied were "natural" particles, as well as biomass burning and industrial emissions, being transported from Indonesia, Malaysia and Singapore into the eastern South China and Sulu Seas. The *Vasco* has previously been used in support of 7SEAS in late September 2011, performing a research cruise in the northern Palawan Archipelago (~11^{o} N; 119^{o} E) and sampling two major aerosol events originating from Borneo and Sumatra (*Reid et al.*, 2015). This 2011 cruise was a trial for the more substantial 2012 effort described here, and provided the means to conduct a detailed seasonal examination of how biomass burning emissions and lifecycle related to tropical waves. Together, these two cruises provide the first ever, to our knowledge, measurements of aerosol properties in the remote southwest monsoon Southeast Asia region, increasing our

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understanding of their relation to regional meteorology and transport patterns.

Findings of the 2011 cruise were consistent with the conceptual analysis of MC aerosol lifecycle in the monsoonal flow, as put forth by Reid et al. (2012) with supported mesoscale simulations by Wang et al., (2013). In particular, the 2011 cruise highlighted the role of the Madden Julian Oscillation (MJO) in regulating aerosol emissions and transport. While relationships between the MJO and aerosol loadings have long been hypothesized (e.g., Tian et al., 2008; Beegum et al., 2009; Reid et al., 2012), these research cruises were direct verification of what was occurring in observed environments under the clouds. The 2011 cruise showed that incorporated into the MJO signal was the associated tropical cyclone (TC) cyclogenesis relationship put forth by Maloney and Hartman (2001). Reid et al. (2012) and Wang et al. (2013) suggested that this relationship strongly influences the development of aerosol events advecting into the South China and Sulu Seas. Indeed, these TCs were in large part responsible for modulating monsoonal flows and large scale aerosol transport during the 2011 cruise. Further, the 2011 *Vasco* cruise highlighted the importance of finer-scale features such as squall lines in regulating over-ocean wet deposition. Ultimately, a key finding of the 2011 cruise was that while monsoonal scale flow patterns and convection are important, short-lived phenomena can strongly modulate cloud condensation nuclei (CCN) concentrations, resulting in pockets of clean and polluted air that are difficult to account for in both observational and modeling studies of aerosol interaction. The demonstrated covariance of thermodynamic structure and aerosol properties in convective environments highlighted the need for any study of aerosol, cloud, and precipitation interaction to control for meteorological factors.

The 2012 research cruise continues the narrative set forth from the previous year. In this paper, we provide an overview of the 2012 biomass burning season and the observations of the September 2012 research cruise. A description of the cruise track, instrumentation, and overall sampled aerosol environment is given. As in the previous paper covering the 2011 cruise, an overview of the overall aerosol and meteorological environment for 2012 is provided for context. Results will focus on the relationships between observed aerosol and meteorological patterns, along with a discussion of the implications for aerosol observability and predictability. Details on aerosol microphysics will be provided in related papers-notably Atwood et al. (2016, to be

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submitted) delves deeply into variability in variability in particle size distributions. While the two-week 2011 cruise occurred at a location and during a period that fit nicely with conceptual models and numerical model simulations, the aerosol and meteorology of the 2012 cruise was more far reaching, with many high-frequency events observed that were not well simulated in the models used in previous studies. The 2011 and 2012 data are compared in detail in the discussion section.

2.0 CRUISE INSTRUMENTATION, TRACK, AND SUPPORTING DATA

The 4-29 September 2012 research cruise was conducted on the same vessel with largely the same instrumentation and measurement configuration as the 2011 cruise (Reid et al., 2015). The vessel used was the Cosmix Underwater Research Ltd M/Y *Vasco*, a 186 ton/35 m ship used for regional diving applications, including salvage, tourism and research. However, the cruise was 12 days longer in duration and ventured further south, as far as Balabac Island on the southern tip of the Palawan Archipelago (7.6 N; 117.0 E) that was less than 100 km from the northern tip of Borneo (Figure 1 (a)). Details on the *Vasco* and its instrumentation can be found in Reid et al., (2015), although a brief overview and notable changes are described below. This is followed by further descriptions of the cruise track and sampling, and finally of the ancillary data used in this analysis.

2.1 Vasco Instrumentation

Instrumentation for 2012 largely mimicked 2011. A bow mast provided high-rate sampling at 50 Hz for turbulent fluxes using a Campbell CSAT3 3-D sonic anemometer and LI-COR H_2O/CO_2 gas analyzer. An Inertial Measurement Units (IMU) consisting of a GPS, a gyro stabilized electronic compass, and accelerometers are used to characterize the ship position and orientations for ship motion removal from the turbulence measurements. Mean meteorological measurements were made by an RM Young propeller anemometer, a Campbell ventilated temperature and humidity probe and a barometer for static pressure. Duplicate measurements for mean meteorology and precipitation were made by a Vaisala Weather Transmitter WXT520 for quality assurance purposes. Differences between all sensors for temperature was within 0.3^{o}C, RH within 5%, pressure within 0.5 hPa, and within 0.5 m s^{-1} for winds. Downwelling short and longwave radiation was measured by Kipp and Zonen CMP 22 and CGR4 instruments,

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respectively. Cloud cover was monitored via a Vaisala C31 ceilometer. InterMet 1-AB radiosondes were released two to three times a day when the ship was at a moorage. New to the 2012 cruise was an OTT Parsivel disdrometer for the measurement of rain rate and droplet size distribution.

Atmospheric composition measurements made on the bow also largely mimicked the 2011 cruise. $PM_{2.5}$ filters were collected by 5 lpm Minivol Tactical Air Samplers (TAS), and analyzed by gravimetric, XRF and ion chromatography at the Desert Research Institute. A second set of filters was analyzed for organic and black carbon, by the thermal-optical method of Chow et al. (1993). The period of filter sampling ranged from one to two-and-a-half days depending on estimated aerosol concentration. Size-resolved elemental data from Na through Pb was provided by XRF analysis at the Lawrence Berkeley Lab Advanced Light Source (ALS) on samples collected by an 8 stage Davis Rotating-drum Uniform size-cut Monitor (DRUM) sampler (unheated PM10 inlet and cut points at 5 µm, 2.5 µm, 1.15 µm, 0.75 µm, 0.56 µm, 0.34 µm, 0.26 µm, and 0.07; Cahill et al., 1985). Due to an electronics failure during ship installation, this instrument was manually rotated to provide data during specific periods. While total mass concentrations are still in analysis, we have high confidence in PM1 elemental ratios presented in this paper. As in Reid et al. (2015), sulfur, potassium, and vanadium ratios are used as markers to help separate industrial anthropogenic from biomass burning particles. For trace-gas analysis, 95 whole-air gas samples were collected for gas chromatography analysis by the University of California Irvine (See Colman et al. 2001 for a list of 60+ compounds provided, details on analysis methods and relative uncertainties). Of these, 85 passed internal quality assurance tests.

Aerosol microphysics instrumentation was located in a forward locker fed by a 3 cm diameter/4 m long inlet from the top of the ship. Wind directional data were used to ensure that only periods with air moving over the bow were used to remove periods of contamination and self-sampling from the dataset. Periods of residual self-sampling were also abundantly clear from CN and total particle counts. Like the previous cruise, a base set of aerosol scattering, absorption number and size were made and processed by two TSI three wavelength (450, 550, 700 nm) nephelometers (Anderson et al., 1996) one ambient and one hearted dry (50% RH), a Radiance Research three wavelength (440 nm, 523 nm, 660 nm) Particle Soot Aerosol

Photometer (PSAP; Bond et al. 1999), TSI Condensation Nuclei Counter (CPC), a combined DMT bench top Passive Cavity Aerosol Sizing Spectrometer (PCASP) and a TSI Aerodynamic Particle Sizer (APS; Reid et al., 2006) for fine and coarse model particle sizing, respectively. A Maritime Aerosol Network Microtops hand-held sun photometer (MAN; Smirnov et al., 2011) was brought on board for measuring Aerosol Optical Thickness (AOT) on those rare cloud free occasions that permitted solar observation.

More significant additions were made to 2012 cruise relative to 2011 in regard to aerosol microphysics. First, data from previous campaigns showed that the lab-bench PCASP instrument was prone to calibration drift, and so a second PCASP configured in an aviation pod and heated inlet was placed on the *Vasco* top mast in a manner as described in Reid et al. (2006). This instrument proved to be much more reliable and steadfast in calibration of the two, and hence is used in this paper's analysis. Also supplementing particle size in 2012 was a combined electrostatic classifier cloud condensation nucleation counter (CCNc) package to measure the size-resolved CCNc characteristics of aerosol particles. This system and its analysis are described in detail in Atwood et al. (2016; to be submitted) but are summarized here. This system provided aerosol particle size distributions and hygroscopicities across a size range of 17-500 nm using a size-resolved CCN system similar to Petters et al. (2009). Coarse-mode particles were first removed using a URG cyclone with an approximate 1 um and 50% size cut before being dried using a Permapure poly-tube Nafion column. Approximately 1.1 lpm was drawn through a TSI 3080 Electrostatic Classifier with a 0.071 cm orifice impactor (approximate 0.69 um 50% cutpoint diameter) and Model 3081 long DMA column, with a sheath air flow rate of 5 lpm for 30 bin midpoint sizes between 17 and 500 nm in diameter. Sampled air was then split between a TSI 3782 Water Condensation Particle Counter (CPC) with a flow rate of 0.6 lpm, and a DMT CCN Counter (CCNc) with a flow rate of 0.5 lpm. The CCN counter was operated at five supersaturation set points between 0.14% and 0.85% and was calibrated using Ammonium Sulfate. Calibration and inversion of the raw measured data followed the procedures described by Petters et al. (2009). Number concentrations for both the CPC and CCNc counter were corrected using actual flows measured by a Gilibrator. Hygroscopicity was assessed with the kappa parameter (Petters and Kreidenweis, 2007) using three-parameter activated fraction fits similar to Rose et al. (2010).

Finally, in parallel to the PSAP, multi-spectral absorption was measured with the newly-developed NOAA Continuous Light Absorption Aerosol Photometer (CLAP). The CLAP is a filter-based system of similar configuration to the PSAP, although with seven sequential filters loaded that eliminates the need for frequent filter changes. Its design requirements were driven by the high sensitivity necessary to monitor aerosol absorption in more pristine conditions at Global Atmospheric Watch stations. Both laboratory and field comparisons between the CLAP and PSAP show that they agree to within 10% (John Ogren, *NOAA manuscript in preparation*). For the 2012 cruise, the CLAP was integrated with the dry nephelometer.

2.2 Vasco Cruise Track and Sampling Schedule

The *Vasco* cruise track in mid-cruise is superimposed on a MODIS Terra image in Figure 1 (a). Like the previous cruise, the *Vasco* home ported from Navotas, Manila Bay, Philippines. Aerosol sampling was also performed in a manner similar to the 2011 cruise, conducted in a series of anchorages that were protected from the swell yet provided unobstructed sampling of the ocean. Typically these anchorages were behind reef zones or small islands that had little breaker activity or swell. Aerosol sampling was also conducted when the air was flowing within +/-50^o of the bow on both transit and at a moorage. Given the consistent nature of winds while at moorage, the *Vasco* naturally weather-vaned with the bow pointed into the wind.

While on deployment, the ship took on provisions once at Liminangcong (11.0 N,119.3 E) near the El Nido anchorage on Sept 10, and twice at Puerto Princesa, Palawan Island (9.7 N, 118.8 E) overnight on Sept 13 and 19. The Puerto Princesa port calls divided the cruise into thirds, which were distinct in geographic region and the sampled environment. In the first phase of the cruise (Sept 4-13, 2012), the *Vasco* sampled the same locations as the 2011 cruise. From Manila Bay, the *Vasco* transited to Apo Reef for a day of sampling (12.7 N,120.5 E; Sept 6, 2012), followed by a day at Coron Island (11.9 N, 120.3 E, Sept 7, 2012) and finally four days on a station at El Nido behind the Guntao Islands reef (11.1 N, 199.2 E; Sep 8-12, 2012)). On the 12th, the *Vasco* transited the east side of Palawan Island to provision in Puerto Princesa overnight on the 13th. Given the prevailing winds at the time, the second phase of the cruise brought the *Vasco* to the very southern tip of the Palawan Archipelago to the western side of Balabac Island in the

Balabac Great Reefs (7.9N, 116.9E; Sept 15-19), 100 km north of the northern tip of Borneo. This site provided excellent shelter from waves and breakers, while experiencing unobstructed sampling of air from the southern South China Sea. Returning to Puerto Princesa on Sept 19 and departing on Sept 20, the Vasco then entered its third phse of sampling; the middle of the Sulu Sea at Tubbataha Reef (8.8 N, 199.9 E) at two very close moorages Sept 21-26. The *Vasco* then had a final return to Navotas Manila Bay Sept 26-29, 2012.

2.3 Regional AERONET Measurements

The maximum extent of 7SEAS aerosol observations throughout the MC occurred during the 2012 Southwest Monsoon season, collinear with cruise activities. Specifically relevant to the analysis of 2012 *Vasco* cruise, over fifteen AEROSOL Robotic Network sun photometers were in operation, with start dates typically in mid to late July through October. Of these, eight were of particular use for the *Vasco* analysis. These are, as displayed in Figure 1(b): (a) Jambi, Sumatra and (b) Palangkaraya, southern Kalimantan in core biomass burning source regions of Indonesia; (c) Singapore, (d) Tahir, Malay Peninsula, Malaysia, (e) Pontianak, western Kalimantan and (f) Kuching, Sarawak as key coastal exit sites; and g) Marbel University, Mindanao and (h) Nha Trang, Vietnam as outer boundaries and receptors. While there are many other sites in the region, they are not used here because of instrumentation failure or excessively high cloud cover. To track overall smoke or pollution transport, we utilize the AERONET operational 500 nm daily averaged fine-mode AOT derived from Level 2.0 Spectral Deconvolution Algorithm (SDA) Version 4.1 (*O'Neill et al.*, 2003). Use of the SDA allows us to track fine-mode particles such as from biomass burning or anthropogenic sources (Kaku et al., 2015) while at the same time removing the influence of thin cirrus contamination which can be large in this region (*Chew et al.*, 2011).

2.4 Ancillary Model and Satellite Data

To capture the nature of the 2012 cruise period a number of model and satellite products were used. The Navy Global Atmospheric Prediction System (NOGAPS; *Hogan and Rosmond*, 1991) is used to provide baseline meteorological data for analysis as well as drive offline Navy Aerosol Analysis and Prediction System (NAAPS) aerosol simulations. While NOGAPS horizontal resolution is ~0.5^o in this region, spectral files were truncated to 1° x 1° for modeling aerosol

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transport of this season in a configuration consistent with the NAAPS reanalysis (*Lynch et al.*, 2015; submitted). In NAAPS four species are simulated: dust, biomass burning smoke, anthropogenic/biogenic fine, and sea salt. In the reanalysis configuration, smoke fluxes are driven from the MODIS smoke source function drawn from a MODIS-only version of the Fire Locating and Monitoring of Burning Emissions (FLAMBE; Reid et al., 2009). NAAPS capability for smoke characterization in the Maritime Continent region has been demonstrated (e.g., Hyer and Chew, 2010; Reid et al., 2012; 2015; Xian et al, 2013), and has been even further improved upon in this reanalysis version. To account for the multitude of anthropogenic and biogenic sources of fine particles, a combined anthropogenic and biogenic fine (ABF) species is generated, based on a combination of Monitoring Atmospheric Composition and Climate-City (MACCity) and Model of Emissions of Gases and Aerosol by Nature (MEGAN) emissions inventories. Quality-assured AOT retrievals from MODIS and MISR observations are assimilated into the model using a two-dimension variational (2D var) technique (*Zhang et al.*, 2008; Shi et al., 2011; Hyer et al., 2013). To better constrain aerosol particle wet deposition, the Climate Prediction Center (CPC) MORPHing product (CMORPH, *Joyce et al.*, 2004) is used instead of the native NOGAPS precipitation field for aerosol scavenging (Xian et al., 2009).

For tropical cyclone information on track and intensity, we utilize the best track statistics from the Automated Tropical Cyclone Forecasting (ATCF) system (Sampson and Schrader, 2000). Air mass histories are also evaluated using the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Version 4.9 Model (*Draxler & Hess*, 1997, 1998; *Draxler*, 2004) based on the GDAS1 1° × 1° global meteorological dataset. Back trajectories were typically run for 72 hours. These trajectories do not directly enter in the narrative here, but are presented in the analysis by Atwood et al. (2016; to be submitted). It is well understood that such trajectory analysis can be problematic in regions such as the Maritime Content, with strong high frequency features such as land sea breeze and orographic flows (Atwood et al., 2013; Wang et al., 2013). Thus, here we only use such analyses at the qualitative level.

The *Vasco* data analysis was enhanced by using geostationary MTSAT satellite products (visible, IR, cloud heights, scatterometer, etc.) as found on the NEXSAT website (*Miller et al.*, 2006; http://www.nrlmry.navy.mil/nexsat-bin/nexsat.cgi). Regional precipitation was also monitored by the aforementioned CMORPH product. MODIS active fire hotspot analysis were utilized based on the similar analysis structure of Reid et al. (2012; 2015), with context as laid out in Hyer et al. (2013). NASA MODIS Col 6 level 3 data were also used (Levy et al., 2013). Due to the heavy cloud cover there is limited space-based CALIOP data available for smoke events over the South China Sea corresponding to cruise periods.

3.0 RESULTS I: REGIONAL METEOROLOGICAL AND AEROSOL CHARACTERISTICS OF THE 2012 BURNING SEASON

The 2012 *Vasco* cruise covered most of September, which corresponds to the second half of the August-September high season for burning in Borneo and southern Sumatra (Reid et al., 2012). This period is generally associated with strong southwesterly winds up the South China Sea region and eastward progression of monsoon onset across the MC over time (Chang et al. 2005; Moron et al. 2009). Smoke and pollution emissions on the islands are then transported across the South China, Sulu and Celebes Seas by the prevailing monsoonal winds (Reid et al., 2012; Wang et al., 2013; Xian et al., 2013). It is not necessary to go into details of smoke transport patterns and their relationships to key meteorological indicators, as these are covered in the above papers as well as in the 2011 *Vasco* cruise case, which fit many conceptual models quite well. However, 2012 did show some anomalous behavior which is worth a description here.

The overall NOGAPS-derived monsoonal flow, over-plotted with the CMORPH satellite precipitation dataset of the core burning months, August and September 2012, is provided in Figure 2, along with the MODIS Terra MOD08 average cloud cover. Corresponding information on MODIS fire detections (MOD and MYD 14) and their relationship to the MJO and regional TCs is provided in Figure 3. At first glance, overall flow patterns are as expected with southeasterly surface winds over the South China and Sulu Seas, veering with height to near westerlies at 700 hPa. South of Borneo, winds are more easterly, wrapping around the islands of Borneo and Sumatra. Precipitation gradually increases from the south to the north, with a maximum east of the Philippines in association with the overall monsoonal trough MODIS cloud cover is spatially correlated with precipitation, ranging from over 50% in southern Borneo to near 100% around Luzon. Other notable cloud and precipitation enhancements include an area west of Sumatra in association with a local vorticity maximum as winds transition from easterly

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to westerly (Wu et al., 2009; Reid et al., 2012), the Bay of Bengal in association with monsoonal flow convergence onshore of Peninsular Southeast Asia, and the region west of Luzon, Philippines, a result of where the monsoonal flows intersect the land (e.g., Cruz et al., 2013). Regions of enhanced fire prevalence (Figure 3(a)) were consistent with the fire baseline of Reid et al. (2012), and are understandably opposite of precipitation. The most typical fire hotspots of Central Sumatra/Riau, Southern Sumatra/Sumatera Selatan and Southern Kalimantan provinces of Borneo are clear.

Key meteorological features of concern for biomass burning in the MC include: a) the interannual El Nino Southern Oscillation (ENSO), which has broad correlation to precipitation anomalies and hence fire (e.g., Nichol, 1998; Field and Shen 2008; Reid et al. 2012); b) the seasonal migration of the monsoon; and c) the ~45 day Madden Julian Oscillation (MJO) that brings longer periods of regional convection and breaks (Zhang, 2005), which in turn has a strong influence on fire emissions and aerosol lifecycle (Reid et al. 2012). Correlated with these are numerous other phenomenon, including the onset and length of the monsoon season (Staub et al., 2006; Cook and Buckley, 2009; Tong et al., 2009), tropical cyclone activity (e.g., Maloney and Hartman, 2001), diurnal cycle of precipitation (e.g., Peatman et al., 2014) and off-island airflow, which that are related to aerosol activity and transport (Reid et al., 2012; Wang et al., 2013). For the 2012 southwest monsoon season, the mean Oceanic Nino Index (ONI) was slightly warm, at +0.3 to +0.4^{o}C for July through October coming out of a ~0^{o}C neutral index in the boreal spring. While in a warmer phase, the burning season is still considered neutral ENSO conditions based on the commonly-used 0.5^{o}C threshold. In comparison, the 2011 southwest monsoon showed decreasing ENSO ONI index temperatures of ~-0.3^{o}C to -0.7 ^{o}C throughout the season. Thus, we expect overall enhancement in 2012 burning activity. Reid et al. (2012) noted that there appeared to be a correlation between positive ENSO phase and earlier monsoonal transition from the Southwest to the northeast phases. In 2012, the monsoonal transition appeared to be consistent with this warmer ENSO phase, with northeasterly winds returning by the end of first week of October. Based on the fire count baseline of Reid et al. (2012), overall burning in Central Sumatra was within ~10% of the Terra + Aqua MODIS time-series average, while Southern Sumatra biomass burning was at a level about 0.5 standard deviations (~20%) above the time-series average.

Overall fires in greater Indonesian and Malaysian Borneo were lower by a half to a third compared to the large El Nino events of 2004, 2006, and 2009, but still ~50% higher than all other negative or neutral ENSO years since combined MODIS measurements could be made (2003). This is expected for a borderline high ENSO anomaly season. In comparison to the 2011 cruise, which had much higher burning than one would expect for a negative ENSO phase, fire prevalence for the season 2012 was ~10 -20% higher than 2011.

If ENSO and the seasonal monsoon migration set the bounds of the burning season, the MJO often regulates the temporal variability and transport within that period. This is demonstrated in a plot of the phase and amplitude of the Wheeler and Henderson (2004) index, Figure 3(b), along with seasonal fire activity in Figure 3 (c), which shows a five-day box car average time series of combined Terra and Aqua MODIS fire hotspot data for June through October that are broken down into the five key regimes defined in Reid et al., (2012; 2015): Central Sumatra, Southern Sumatra, Indonesian Kalimantan-Borneo (an aggregate of western, southern and eastern Kalimantan), Malay Borneo (Sabah and Sarawak), a combined Sulawesi, Java, and Timor aggregate, and finally far-eastern Maluku and the entire island of New Guinea.

In 2012, MJO activity during the burning season was generally weak, although nevertheless present. The burning season was initiated by a moderately strong MJO from May into early June, with amplitudes well above 1, especially for the drier phases in the MC (phases 5-8). This MJO was related to the first major burning event that emerged from Central Sumatra in June. This short but dramatic pulse is a common feature in the MODIS record, and is likely the result of agricultural burning, or fuels which had been stacked and ready for the first sufficiently dry southwest monsoon period (Reid et al., 2012), which in this case, a strong late phase MJO event provided. While much attention has been paid to the “anomalous” June 2013 burning and smoke event in Sumatra which severely reduced air quality on the Malay Peninsula, this is actually a recurring feature (Reid et al., 2012), although the impacts to Singapore for that year are still unprecedented for June.

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The next MJO cycle, while starting strong in the Indian Ocean in mid-June (Phase 1 & 2) failed on approach to the MC. A weak MJO event replaced this, although it could be argued that it is really the same event. As this MJO progressed to later phases, a second greater peak in August burning in Central Sumatra and Western Kalimantan (apparent in a peak in the greater Indonesian Kalimantan domain) appeared. From this point, the burning pattern was eastward-propagating through the season, which is consistent with the eastward migration of the monsoon as described by Moron et al. (2009).

The third relevant MJO cycle, which also coincided with the *Vasco* cruise, formed in the Indian Ocean in mid-August, with MJO amplitudes as high as 2 through early Phase 3 (the MJO entering the MC). Upon reaching the MC in the last week of August, however, the amplitude weakened significantly, to less than 1(below the significance level), and propagation was slow from Phase 3 to Phase 6 (the period when the MJO is transiting the MC) and subsequently died off by the end of September. Nevertheless, fire activity tracked this event, reaching a peak at the end of September. This event was then followed by another weak, although complete, event in October and November (not shown); not an uncommon occurrence for MJO propagation across the MC (Zhang, 2005; Peatman et al., 2014). The implication for 2012 is that more significant smoke generation and transport events advecting across the South China Sea would have been observed had this MJO progressed in a more typical fashion. Instead, the stalling of the MJO over the Maritime Continent would bring additional rainfall over the ocean (e.g., Chen and Houze 1997; Peatman et al., 2014), further shortening aerosol lifecycles.

Embedded in the MJO signal is the influence of TCs. When TCs are located just west of the Philippines or transiting westward into the South China Sea, two significant meteorological impacts on aerosol lifecycle often occur (Reid et al., 2012). First, large scale upper-level subsidence around the TC can inhibit convection and rainfall in the region, thus promoting biomass burning (and enhancing its observation from space). But, simultaneously a long area of monsoonal enhancement akin to an inflow "arm" often forms to the south and west of the TC, leading to enhanced flow from biomass burning source regions up the South China Sea but also within the monsoonal enhancement, increased convection leading to scavenging. The balance of these two factors in production, advection and regional changes to precipitation allows TCs to

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simultaneously modulate multiple aspects of aerosol lifecycle. From ATCF, the 2012 Western Pacific had 25 named storms compared to 26 in the long term average. Noteworthy is that when TCs were east of the Philippines (as marked in Figure 3(b)) this coincided with peaks in observed biomass burning activity.

Moving from burning activity to resultant AOT, from a biomass burning perspective, the 2012 season showed higher burning-region AOTs in comparison to the 2011 cruise and the ENSO neutral baseline provided in Reid et al. (2012). Overall core August and September burning season monthly AOTs from Terra MODIS and NAAPS are provided in Figure 4(a)-(d). Because of the widespread cloud cover (e.g., Figure 2(b)), both satellites and models have extreme difficulty characterizing aerosol in the region (Reid et al., 2013; Sessions et al., 2015). Ultimately, it is difficult to perform a pairwise analysis. Nevertheless, these products provide qualitative insight into regional aerosol behavior.

In agreement with the temporal series of biomass burning in Figure 3, we see a shift from high AOTs in Central Sumatra in August to more southern Sumatra and southern Kalimantan in September. The highest AOTs tend to be over or near the islands. Moderately high AOTs (~>0.2) do cover the greater part of the South China Sea over the course of the entire season. Differences between MODIS and NAAPS are a result of a number of causes, including sampling differences (there are very few MODIS retrievals available, whereas NAAPS data is continuous), and underestimates in modeled emissions due to cloud cover that limits FLAMBE efficacies. Notable in the NAAPS data in particular, is that, in August (Figure 4(c)), smoke from Central Sumatra propagates up across the entirety of the South China Sea, whereas in September (Figure 4(d), AOTs are higher in the southern South China and Celebes Sea. As discussed in Section 4, this is a result of easterly waves and the influence of TC induced monsoonal enhancements and inflow arms.

The best indicator for regional AOTs however is from AERONET, which can, through the SDA method, minimalize thin cirrus contamination (Chew et al., 2011). Included in Figure 4 are the time series from AERONET sites deployed for the 2012 season, including the first AOT measurements made in the heart of the burning areas on Sumatra and Kalimantan, Borneo. The

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500 nm fine-mode AOT from key AERONET sites is also shown in Figure 4 for August through October when the full network was deployed. Sites provided include: (e) Jambi, Sumatra and (f) Palangkaraya, Central Kalimantan, respectively in core Sumatran and Borneo burning areas. Also shown are: (g) Singapore and (h) Tahir on the Malay Peninsula and (i) Pontianak and (j) Kuching on the western and northwestern coasts of Borneo, respectively. These sites provide AOTs as smoke plumes leave the islands. Finally, long-range receptor sites at (k) Notre Dame of Marbel University, Mindanao and l) Nha Trang, Vietnam are given. These sites are also marked on Figure 4(a).

Regional fine-mode AOTs, to a certain degree, were correlated with the fire signal, but not exceedingly so; perhaps due to the high coverage of cirrus clouds that can affect satellite fire observations more than it affects AERONET with its SDA extraction of fine-mode AOT (*e.g.*, see observability review by Reid et al., (2013)). Also, the weaker MJO periods or TCs may have perturbed the transport of smoke off-island. Singapore was the only site operating for the mid-June burning event; it showed slightly-elevated fine 500 nm AOTs on the order of 0.5 above the background of 0.3 (not shown). For the greater August through September burning season, in the source regions of Sumatra and Kalimantan, the Jambi and Palangkaraya AERONET sites showed multiple events of 500 nm AOT greater than one; reaching over three at the end of the season just before the monsoonal transition. As demonstrated in the comparison of the Singapore and Tahir site AOTs, smoke transport off Sumatra favored the southern end of the Malay Peninsula, with the exception of a large event in early August. The Pontianak and Kuching sites on the western side of Borneo showed moderately high AOTs for the entire season, with 8+ events having AOTs over 0.5, peaking just before the end of the monsoon. Finally the typically clean receptor location of Marbel University, Mindanao showed several significant enhancements in fine-mode AOT values above 0.2 in mid-September and early October. Nha Trang, Vietnam typically showed background levels, with the only enhancement being by continental Asian emissions in north/northwesterly flow at the end of the monsoon.

4.0 RESULTS II: THE METEOROLOGICAL AND AEROSOL ENVIRONMENT MEASURED DURING THE VASCO CRUISE

Based on the Section 3 analysis, at first glance one would expect a nominally "typical" year for a slightly-warm ENSO phase. Elevated fire activity and emissions superimposed on a near climatologically average wind field should systematically yield large smoke events propagating across the South China and Sulu seas in a manner very similar to the 2011 cruise as reported in Reid et al. (2015). However, while the averages are close to normal, the daily meteorology during the 2012 was quite variable, with two extremely weak monsoonal flow periods separated by the influence of two category 5 typhoons that slowly propagated northward east of the Philippines. As is presented in Section 3.3, this modulation in monsoonal strength coupled with a longer cruise extent resulted in observation of much more variable aerosol behavior in 2012 than 2011.

4.1 The regional meteorological environment during the Vasco cruise

The overall meteorological and aerosol behavior during the 2012 cruise period is best described by simultaneous examination of Figures 5 and 6. Figure 5 provides combined daily average NOGAPS winds at the surface (black) and 700 hPa (magenta) overlaid with the CMORPH daily average precipitation for four representative days during the cruise, marked with the *Vasco*'s location. For each day, the 2:32 UTC (~9:32-10:32 LST) MTSAT false-color visible image is also provided, annotated with the *Vasco* location and TCs in the area. Figure 6 then provides the corresponding *Vasco* data time-series of key meteorology, including ventilated temperature, RM Young wind, and disdrometer-derived precipitation. Also shown are key aerosol and gas data, during appropriate sampling conditions. Included is the particle counter concentrations, PCASP derived aerosol volume, and whole-air gas can-sampled carbon monoxide (CO). Finally, the derived NAAPS fine-mode surface aerosol concentrations of biomass burning (gold) and anthropogenic fine (red) are also provided.

The most notable aspect of the 2012 cruise relative to the 2011 counterpart, or indeed all other 7SEAS regional studies, was the extreme variability in the monsoonal flows. While at the seasonal mean level, wind patterns seemed near normal, during the entire month of September 2012, daily flow patterns over the South China Sea changed measurably. These periods were largely defined by the migration of two tropical cyclones (TC 17 W Sanba and TC 18 W Jelawat), separated by easterly waves with significant flow reversals (e.g., Figure 5). During the

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very first week at sea, monsoonal flow across the SCS was weak, associated with a westward propagating wave consistent with the features of a tropical depression/easterly wave. The day of departure (Sept 4, 2012) the Sulu Sea was already under the influence of an easterly wave, with southeasterly winds at the surface and nearly calm winds through the mid troposphere. Over the next two days, winds shifted with the propagation of the wave, from southeasterly, to westerly and northwesterly into the eastern part of the SCS, followed by full westerly and even northerly winds by Sept 7 (Figure 5 (a)). Winds at 700 hPa even became strong northeasterly. Thus for the first five days of the cruise at Apo Reef, Coron, and the first several days at El Nido, sampled winds were largely not from the South China Sea, but rather off of the Philippine islands of Luzon, Mindoro, and Iloilo. This is evident in the *Vasco* time series where winds are generally light with often a northerly or even easterly component.

Monsoonal flow and precipitation began developing on Sept 10th in association with the formation of what would become super-typhoon Sanba (TC 17 W) at Palau, while the *Vasco* was stationed in El Nido. By Sept 12, TC Sanba grew to typhoon strength, followed by rapid intensification to category 5 the very next day. The slow northward migration of TC Sanba resulted in enhnaced westerly components of the marine boundary layer and mid-tropospheric winds, as well as enhanced precipitation in a well-defined zonally-aligned monsoonal enhancement/inflow arm. The *Vasco* transited to Puerto Princesa on Sept 12th and 13th, still under significant influence of the inflow arm of TC Sanba. Upon departure from port on the 14th, heading for Balabac Island at the southern tip of the Palawan Archipelago, the slow moving super-typhoon was still west of Luzon, with a well-defined inflow arm across the South China Sea. Even through Sept 16th (Figure 5(c)), the influence of TC Sanba lingered as a inflow arm extended from the Malay peninsula through Luzon.

For Sept 17th through 21st, monsoonal winds slackened yet again, with westerly winds in the northern region, and even some northerly components in the lower free troposphere. This is consistent with the propagation of a second easterly wave across the region. During this period the *Vasco* transited back to Puerto Princesa (Sept 19th) and finally out to its last station at Tubbataha Reef in the middle of the Sulu Sea. While on station there, monsoonal flow returned, with the formation of another super-typhoon, Jelawat (TC 18 W). Jelawat had a similar lifecycle

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to Sanba, forming over Palau and slowly migrating up the eastern side of the Philippines with a well-defined inflow enhancement across the South China Sea (e.g., Sept 24th Figure 5(d)). Also like TC Sanba, TC Jelawat intensified rapidly, becoming a category 5 super-typhoon; the strongest of the season. While surface winds had a typical southwesterly direction, winds in the free troposphere had significant northerly components across the South China Sea; unusual relative to the more typical westerly to southwesterly winds. The *Vasco* then returned back to home port in Navotas, Manila (September 27-29th) on the southern edge of the northward propagating inflow of TC Jelawat. At this same time period a tropical low was forming in the northern South China Sea which would eventually become tropical storm Gaemi, further enhancing monsoonal flow along the Palawan Archipelago.

The meteorology described above for the 2012 cruise resulted in some marked differences from the measured meteorological environment on the *Vasco* on the 2011 cruise. Perhaps the most important difference was that in 2011, monsoonal flows were consistent southwesterly across the South China Sea, with enhancements in low-level wind and precipitation found in inflow arms, and clearer air and high aerosol loadings in regions with suppressed convection. In contrast, the 2012 cruise witnessed monsoonal enhancement due to tropical cyclone inflow arms, which induced winds that were largely zonal in nature. The observed easterly waves also resulted in much less coherent wind and precipitation patterns. Thus wind direction and speeds at the ship were more variable, and precipitation occurred most days (e.g., Figure 6(b)-(d)).

Similarities between the 2011 and 2012 cruise periods were also noted. Perhaps the most important was the clear presence and importance of convection and squall lines. 2012 saw an increase in the frequency of convection of the South China Sea region over 2011, there was also an increase in the frequency of telltale signs of cold pools, including frequent temperature drops by as much as 5^{o}C within several minutes, in combination with spikes in one-minute average wind speeds to 15 m s^{-1}, and one second values as high as 25 m s^{-1}. As discussed in Section 6, these cold pools also have "aerosol washout" air, dropping particle concentrations significantly. Similarities and differences between these events and the cold pool events described in Reid et al. (2015) will be discussed in Section 6.

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4.2 The measured aerosol environment during the Vasco cruise

The far-reaching extent of the 2012 cruise, coupled with the more variable monsoon meteorology, resulted in much more dynamic aerosol behavior sampled during the 2012 cruise relative to 2011. In the 2011 cruise, when the *Vasco* was anchored outside of El Nido, two 2-3 day large-scale aerosol events with ~15-30 μg m^{-3} fine-mode mass concentrations and CN= 1000-4000 cm^{-3} were sampled, separated by periods of very clean conditions (<1 μg m^{-3}, CN=<150-300 cm^{-3}). The nature of aerosol events in 2012 followed similar pathologies to 2011, with TCs enhancing winds and into the inflow arms. The balance between precipitation and convective inhibition due to large scale subsidence from the TCs, or dry layers advected from the Indian Ocean, induced these aerosol events, which were qualitatively captured by the NAAPS aerosol simulations. Additional high-frequency drops in aerosol loading were also found in association with cold pools; some associated with long lasting convective squall lines.

While some low frequency phenomena occurred during 2012, the 2012 cruise also showed variability in aerosol events over short time scales. Inspection of the 2012 cruise time series of aerosol-relevant parameters in Figure 6 shows a great deal of variability in aerosol and CO loadings. For example, while there was one period of overall aerosol enhancement from September 13th through the 18th, the most significant pulses (Sept 14 & 25) in inferred aerosol mass concentration were generally less than 6 hours in duration; as opposed to two to three day event lengths for 2011. In addition, multiple short pulses of less than 3 hours were also observed.

Significant enhancements in CO as measured by the whole-air samples, as high as 200-240 ppbv, were found for the most significant Sept 14 & 25 cases. Indeed, the air from the open ocean exhibited a very distinct “smoky” aroma for these periods. Further, while there were more data outages in 2011, owing to more variable wind direction across the bow while in transit, the *Vasco* did observe several high-frequency pulses of condensation nuclei, which we are confident were not due to self-sampling contamination. Of particular note were spikes in CN measured when the *Vasco* was moored at Apo Reef and Coron, in air masses moving offshore of the islands of Mindoro and Luzon. Also, spikes were observed entering port or in the vicinity of island cities, such as September 13th and 20th while the *Vasco* was downwind of Puerto Princesa.

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Also notable in 2012 was the higher baseline for "background" particle concentrations in the marine boundary layer. Whereas in 2011 the background ranged from 150-500 cm^{-3}, in 2012 the CN concentration rarely dropped below 500 cm^{-3}. Part of this difference is 2012's closer proximity to Borneo source regions along the cruise track. Further, for periods when the *Vasco* was in the northern region, winds were anomalous and precipitation reduced due to the presence of easterly waves. Thus, there was more sampling of Philippines islands, and regionally reduced wet scavenging.

In comparison to observations, the NAAPS model simulations of aerosol loadings near the *Vasco* exhibited mixed performance relative to the outstanding comparisons for 2011. For example, NAAPS did simulate some aspects of aerosol transport, such as the broader aspects of the Sept 13-18th period. However, the model had difficulty capturing the most significant pulses, such as observed spikes on September 14th and 25th. NAAPS also included other moderate events that did not materialize, such as Sept 10-11, and Sept 24th and 29th. NAAPS did simulate the presence of some high-frequency events, although these had notable time displacement. The use of satellite precipitation to constrain scavenging processes in NAAPS improves representation of variability in aerosol loadings in high emission and high convection environments, although finer scale features unresolvable in a 1x1 degree transport model are clearly important.

Aerosol and gas chemistry measurements provided additional insights into aerosol properties and their sources. Detailed aerosol and gas chemistry is a subject of a subsequent paper, but important aspects that assist in the aerosol meteorology analysis are discussed here. As the filter datasets required sampling times of one-to-two days to ensure sufficient loading, much of the temporal characteristics were washed out with the background. Filter-based XRF metals also had a higher noise floor and thus were inconclusive. However, the DRUM data does provide elemental ratios which, in combination with the PCASP data, can provide higher resolution information on elemental chemistry. The 2012 cruise included a larger number of whole-air samples as compared to those sampled during the 2011 cruise.

A summary of observed aerosol chemistry is provided in Figure 7. Shown in Figure 7 (a) is derived non sea-salt $PM_{2.5}$ gravimetry from filters, with corresponding quartz filter analyses of organic and black carbon. For comparison, an inferred 30 minute PCASP-derived dry mass concentration using an assumed density of 1.4 g cm^{-3} is presented. This value of density provided good closure (unity slope; r^2=0.8) between the temporally-integrated PCASP and gravimetric values, and is close to the density for dry biomass burning of 1.35 g cm^{-3} as measured by Reid et al. (1998). Zero values of filter mass are associated with no-sampling periods due to the relative wind direction over the bow. Shown in Figure 6(b) are the Teflon filter-derived K and SO_4 values, followed by 6(d) elemental ratios of vanadium and potassium to sulfur from the DRUM sampler used as an indicator to separate aerosol industrial and biomass burning origin. Also provided in Figure 7 are key whole-air gas sample species. While there are no unique chemical identifiers to isolate natural, biomass burning and other anthropogenic sources, several species warrant attention. Included are: 6(d) CO and CH_4; 6(e) benzene and methyl-iodide as commonly used key indicators for biomass burning (Akagi et al., 2011); 6(f) i- and n-pentane as well as their ratios , with enhanced ratios suggesting more industrial rather than biomass burning sources (McGaughey et al., 2004; Simpson et al., 2014); and finally, 6(g) isoprene and 2-BuONO, a photo-oxidation product of butane and indicator of photochemistry.

Figure 7 illustrates several notable aspects of atmospheric chemistry in this region. For example, organic mass fractions vary significantly between the aerosol events. For the largest smoke event on Sept 14th, the vanadium-to-sulfur ratio is extremely low, while the potassium-to-sulfur ratio was higher, suggesting this air mass was dominated by biomass burning. The organic and black carbon mass fractions were ~50% and 5%, respectively; these values are very reasonable for biomass burning (*Reid et al.,* 2005; *Akagi et al.,* 2011). This also suggests also that the bulk of the sulfate here is indeed a result of biomass burning emissions; perhaps secondary in origin from primary SO_2 emissions of peat burning (Reid et al., 2013). However, there are significant reductions in OC and BC for the later burning events, on the order of 25% and 2.5% for organic and black carbon, respectively, and simultaneously vanadium was enhanced. The difference was made up through sulfur/sulfate enhancements, ranging from 20% for the 14th event, to 30-40% for the others. The sulfate-to-organic carbon ratio then increases from 0.45 for the 14th, to 1.4-1.8 for the subsequent filters. Clearly, these major events had significantly different inorganic

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mass fractions, and perhaps enhancement for MC industrial emissions. In all cases, potassium was at 1.5-2% from the filters, although the potassium to sulfur ratio from the DRUM sampler did show potassium enhancements consistent with increased biomass burning. All of these findings as well as those from the 2011 cruise point out that "pure" biomass burning plumes are relatively rare. The bulk of the polluted air mass is more often a combination of biomass burning and other anthropogenic emissions.

Periods of pure pollution were also observed in the early days of the cruise. When *Vasco* was under the influence of flow from the Mindoro and Luzon Islands, organic carbon mass fractions were higher, at 60-to-80%, and there were very high vanadium to sulfur ratio values(~0.1). In one case, Sept 11, derived OC was higher than gravimetric mass. In this case, the gravimetry matched the PCASP; a sure sign that this is organic artifact. But this nevertheless suggests perhaps a change in aerosol organic chemistry associated with regional anthropogenic emission. Indeed in the first period, sulfate-to-organic carbon ratios were low, on the order of 0.4-0.6. Potassium was particularly enhanced with vanadium against sulfate from Sept 6th. However, this was a very dry period in the mission, and perhaps un-oxidized SO_2 was prevalent in the atmosphere.

Gas chemistry can further help distinguish biomass burning from anthropogenic industrial emissions. From a gas chemistry point-of-view, indeed the largest aerosol mass events are associated with CO and consistent with biomass burning (Figure 7(d)). However, the correlations between other gas species were highly variable. For example, correlations between benzene and CO as commonly used biomass burning indicators are quite good (slope 0.0008 and r^2=0.72) with the exception of an isolated spike at ~0Z Sep 20 while entering Puerto Princesa. Methyl iodine, sometimes used as a burning tracer, showed numerous spikes over an order of magnitude above the nominal baseline for CO and benzene (Figure 6(e)). Indeed, at times very high spikes of CH_3I are present with little increase of CO. This may reflect other potential sources of CH_3I such as coastal macrophyytic algae (Moore and Zafiriou, 1994); not unreasonable in the highly-productive waters of SE Asia.

For separation of burning emissions from non-burning anthropogenic emissions, we can look to information in the concentration of i- and n-pentane and their ratio (Figure 6(f)). When the *Vasco* entered or exited Puerto Princesa's urban plume, values of i-pentane increased markedly, along with the ratio of i- to n- isomers, thus providing a good indicator of local city or village sources. Outside of these areas, or in background conditions, n-pentane was enhanced. The significant variability in i- to n-pentane demonstrates the significant variability in sources in the region.

From a photochemistry point of view, spikes in isoprene were frequently found in the vicinity of islands (Figure 6(g)), but also occasionally a day's distance from shore. At concentrations near 100 pptv, these levels are rather low compare to terrestrial source regions, where values on the order of 1-5 ppbv are expected and measured (e.g., Wiedenmyer et al., 2005; Hu et al., 2015). However, spot cans on the interior of islands taken as part of the 2011 Vasco cruise did reach 1 ppbv (Reid et al., 2015). Similarly, 2-$BuONO_2$, an indicator of photochemistry, also showed sporadic behavior, in this case associated with both smoke events and urban plumes alike. Finally we observed sporadic cases of strongly enhanced methane, which in general did not correlate with CO or any other species. This very easily could be indicative of gas hydrate derived methane production in under-ocean cold seeps in the South China Sea (*Suess*, 2014).

5.0 RESULTS: AEROSOL METEOROLOGY OF SIGNIFICANT AEROSOL EVENTS

From Section 4, the measured aerosol and meteorological environment during the 2012 cruise was found to be much more complex than the 2011 counterpart was. The meteorology was more variable, and additional aerosol phenomenon such as from urban plumes and nucleation events, were sampled. Ultimately, the aerosol events were relatively short lived compared to 2011. Indeed, more prevalent high frequency phenomenon, such as particle concentration drops due to cold pool or the occasional spike in CN are also observable (Figure 7). In this section, we delve into more detail on the aerosol meteorology of key aerosol events. To help inter-compare aerosol events, particle concentrations and key whole-air can samples with associated aerosol particle concentrations are provided in Table 1. Thermodynamic data for soundings collected in three key events are given in Figure 8. Atwood et al., (2016; to be submitted) go into much greater detail on the implications of these events to aerosol microphysics.

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5.1 Biomass Burning Events

One can interpret the PCASP-inferred mass, aerosol elemental ratios, and CO versus the ratio of *i*-to-*n*-pentane as indicative of two very clear biomass burning-dominated event periods sampled on the *Vasco*: Sept 14-17th and 26th. There are also multiple small aerosol and CO enhancements visible, especially late in the cruise. While we say these are biomass burning events, we must emphasize that it is likely that other species were transported with the open burning emissions, including urban and shipping fossil fuel emissions and biofuel. Regardless, the two major event periods have every indication of being dominated by open burning (including the smoke we could smell near the ship) and warrant special attention, as they give key insight into aerosol lifecycle in the South China and Sulu Seas.

5.1.1 Puerto Princesa to Balabac Sampling: The Sept 14th-17th event

Details of the Sept 14th - 17th event are provided in Figure 9 for the time period from the *Vasco* leaving Puerto Princesa through its Balabac anchorage and start of its return home. To describe the lead up of the event, included are MTSAT visible satellite images for Sept (a) 13th and (b) 14th at 4:32 Z with the combined Terra and Aqua 550 nm AOT for that day. Also recall that wind, precipitation, and satellite imagery for the middle of the event are presented in Figure 5(c). Included in Figure 9 is the *Vasco* time series of several key parameters, including (c) PCASP volume distributions, (d) temperature, (e) wind speed, and (f) precipitation. In terms of duration, and of fine particle and (based on Figure 7) CO concentrations, the Sept 14-16th event was the most significant burning event sampled during the 2012 cruise. Peak values for particle number and CO concentration reached as high as 2000 cm^{-3} and 250 ppbv respectively, just as the Vasco moved south from Puerto Princesa. Whole-air sample data taken at this point give all of the key VOC markers of biomass burning dominated aerosol loading, including very high ethene and benzene. The ratio of i-to-n-pentane was ~1.3, also suggesting biomass burning over other anthropogenic emissions.

Based on the spike in AERONET AOT at Marbel University, Mindanao (Figure 4 (k)), this smoke event extended through the Philippines into the Pacific Ocean with a fine-mode AOT of 0.34. Particle size distributions were fairly constant, with a dry volume median diameter of ~0.3

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µm. While the transported smoke into the region was immediately noticed upon departure from Puerto Princesa ~0 Z on the 14th, based on NAAPS data, it is quite possible that the event began to arrive just after the *Vasco* entered the harbor for resupply 18 hours earlier. No doubt, we also measured Puerto Princesa influence in the hour entering and hour leaving the harbor. However, the particle and CO concentrations continued to increase as the *Vasco* transited to the south toward Balabac Island, well away from any local sources. At 12 Z on the 14th, a very rapid drop in particle concentrations occurred while the *Vasco* was approaching the southern tip of Palawan Island (down to 500 cm^{-3}), with partial particle and CO recovery when the *Vasco* made anchor at Balabac Island. There, higher particle concentrations remained for another two days with a slow decay to cleaner conditions of 500 cm^{-3}, bringing the event to a close. A final peak was observed and modeled in NAAPS as the *Vasco* departed for a return to Puerto Princesa near 23Z on the 19th.

For the transit south to Balabac, weather was relatively stormy, with moderately high winds and periods of rain. On anchorage, isolated cells of precipitation were frequently observed in the vicinity. The first radiosonde released occurred on the arrival at Balabac Anchorage on Sept. 15, and showed generally moist conditions in the lower free troposphere. However, air was dry in the upper troposphere, perhaps due to large scale subsidence in association with TC Sanba. This dry air aloft may have inhibited some of the deep convection, thus allowing the transport event to persist.

The Sept 14-17th event has several interesting characteristics. First, while the NAAPS model generally predicted this event, the initial peak concentration was significantly underestimated. Second, the dramatic drop in particle concentration ~ 12 hours into the event would normally imply a cold pool. However, the particle decline occurred over a period of 45 minutes as opposed to the minute or two which one would expect from a cold pool event. While there was a temperature drop associated with the particle reduction, it was not as dramatic or rapid as some other events. Indeed, there are actually several significant temperature drops in the hours during the high concentration period with only moderate perturbations to the PCASP and CN data. The NAAPS model is somehow, however, picking up on the particle concentrations in the recovery. Clearly, this is not a typical cold pool event as was demonstrated in the 2011 cruise. Third and

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finally, on the last day on station in Balabac Island, the cleanest periods of the cruise were sampled. While part of this observation may be attributable to the rainfall observed at the *Vasco*, the lowest concentrations were between rain events. Given the location of this ship right off of the northwest tip of Borneo, higher particle concentrations were expected.

We hypothesize that the dynamics of this particular event are based on two meteorological components coupled with an orography effect from Palawan Island. The first is related to coastal and orographic flows in western Borneo. The September 14-16th event was initiated with a large biomass burning outflow event on September 13 (e.g., AERONET data at Kuching in Figure 4(j) and Figure 9(a)). Fine-mode AOTs at Kuching peaked at 1 on this date, while AOTs at Pontianak, further south were constant at ~1. In the absence of significant burning in the vicinity of Kuching, smoke must be transported from western and central Kalimantan regions. Throughout the mission however, as seen in the model data in Figure 5, the NOGAPS model had very low surface wind speeds right offshore of Kuching. In the lower free troposphere where winds are higher, they tended to be westerly, thus preventing smoke above the boundary layer from being advected offshore into the South China Sea. Thus in the model, the smoke does not get advected offshore nearly as far as it should and smoke clings to the coast. However based on MODIS AOT in Figure 9(a), we see that in fact the smoke was transported hundreds of kilometers offshore. This plume feature may also have contributions from Sumatra, also unresolved in the global model. As hypothesized in Reid (2012), and then demonstrated in mesoscale simulations by Wang et al., (2013), the sea/land breeze and orography play a significant role in modulating smoke transport on and off the islands of the Maritime Continent. We hypothesize that orography and land breezes coupled with additional enhancement in monsoonal flows due to TC Sanba resulted in this significant ejection event. This phenomenology resulted in the significant smoke loadings at the *Vasco* as it left Puerto Princesa and simultaneously missed in the model. As the *Vasco* moved south, the model was able to account for the smoke that was transported closer to the Borneo coast.

The second significant feature of the Sept 14-17th event, the precipitous drop in smoke particle concentrations on September 14 at 1230Z, was due to the remnant of a massive squall line, clearly visible in the Figure 9(b) satellite image. Based on inspection of the MTSAT data, this

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was formed from a series of isolated cells aligned from south of the southern tip of Vietnam to the Malay Peninsula the night before. Cold pools from these cells were advected to the west as part of accelerated winds over the South China Sea in association with TC Sanba, eventually resulting in a clear squall line that was nearly 700 km long before daybreak on the 14th. MTSAT imagery suggests the arrival of this squall line at Palawan Island at ~12Z on the 14th, coincident with the drop in particle concentration. Since the wind speed of the steering flow aloft is greater than that of the smoke-laden marine boundary layer air, the squall line created a large patch of clean air at the surface. We also hypothesize that Palawan Island broke up this particular squall line and its associated cold pool, thus slowing the more typical rapid temperature and particle drop.

The third interesting aspect of the September 14-17th event is the relatively clean conditions experienced at its end on September 17th. This was partially seen in the NAAPS model, but the model nevertheless over predicted mass concentrations in what was the cleanest air sampled during the mission, even though the *Vasco* was very close to Borneo. We believe this situation was a result of the northern propagation of TC Sanba during the event. By September 15, the TC's inflow arm was in the middle of the South China Sea. Surface winds along the northwest Borneo coast slackened considerably, even taking on a northerly component by late on the 16th. At this same time, coastal convection began to increase. By the 17th, surface and lower free tropospheric winds at the *Vasco* were coming from the Central China Sea, allowing for more clean conditions to be sampled. NAAPS resolved a very strong gradient in particle concentration right at the *Vasco* anchorage. At 1x1 degree resolution, even a single grid-box difference made a significant difference in predicted values.

5.1.2 Puerto Princesa to Tubbataha Sampling and the Sept 25th-26th event

The second largest biomass burning event was identified in association with the final stage of the cruise while moored at Tubbataha Reef. The *Vasco* departed from Puerto Princesa to Tubbataha Reef in the Sulu Sea on September 21st, and ended its sampling with the return voyage back to Manila on Sept 27th. While there were sporadic peaks in particle and CO concentration on Sept 23rd and 24th, the event was sampled for a 12 hour period over Sept 25th-26th. This event had peak particle concentrations and CO values nearly as high as the September 14-17 event, but was

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considerably shorter in duration-nominally only eight hours long. There were also a number of minor events flanking either side of the primary event. NAAPS also suggested a peak in smoke concentrations, but the model predicted peak concentration 12-18 hours earlier than observed, and over predicted smoke and pollution thereafter.

More detailed data from the Sept 25-27 period is presented in Figure 10 in a manner similar to Figure 9. Some aspects of the Sept 25th-26th event mimic the earlier Sept 14-16th event. Particle size distributions, with a volume median diameter of ~0.3 μm, were similar. Key VOC markers, as listed in Table 1, looked similar to the Sept 14th event. A TC (here Jelawat) was just east of the Philippines, with an extensive inflow arm reaching to Southern Vietnam (e.g., Sept 24th meteorology and imagery in Figure 5(d)). A day later, as TC Jelawat migrated northward, a large aerosol ejection event occurred along northwestern Borneo into the South China Sea, in this case visible in both the Kuching and Pontianak AERONET time series. At the same time, NAAPS and MODIS AOT data suggest that for this case, a large event also departed Sumatra, which we speculate may have also been part of the sampled airmass of the principle event or perhaps the secondary event that appeared 12Z on the 26th. Regardless, neither the modeling nor the remote sensing data provide enough information to confidently make this attribution. Also like the previous Sept 14-16th event, the TC's continued northward migration also ended the event. Soundings were similar between the two events, being relatively moist in the lower troposphere, with some drying aloft. Further, CMORPH data suggested development of coastal convection early on the 26th.which also likely resulted in additional particle scavenging.

The comparison of the weather and PCASP time series for this event does show some interesting features. Most notably, the most significant increase in particle concentration at 18Z on the 25th was heralded by a cold pool with near instantaneous temperature drop and increased wind speeds. Generally, we think of cold pools being associated with convectively washed-out air. But in this case, particle concentrations increased, and the magnitude of the temperature, wind and precipitation perturbations were quite small. Thus, the event may have been associated with some minor convection along the leading edge of the airmass. But the observation does nevertheless pose a question regarding a generalized view of clean air behind cold pools and squall lines. Indeed, while a focus of the Sept 14th case was in the clean air behind the squall

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line, at 6 hours before that event there were multiple temperature drops with corresponding peaks in winds and precipitation, but little effect on particle concentration. These too may be indicative of cold pool remnants. It is possible that the convection causing the cold pool had dissipated leaving the propagating density current. As this feature moved, it may have entrained air with higher particle counts. Such events may be consistent with land breezed induced coastal convection.

5.2 Local aerosol events

While the primary focus of the 2012 Vasco cruise was to observe the nature of long-range biomass burning and anthropogenic aerosol transport from Borneo and Sumatra to the Philippines, we were mindful of the potential impact of local aerosol sources. During the cruise, two significant types of local sources were observed; a nucleation event at Apo Reef on Sept. 5th -6th, and a series of urban plumes as the *Vasco* neared the vicinity of port towns such as Coron and Puerto Princesa. These events are discussed in more detail below.

5.2.1 Apo Reef Nucleation Event

The first anchorage reached after departing Manila was at Apo Reef in the middle of the Mindoro Strait, on September 5th and 6th. As noted in Section 4, during this period the South China Sea was experiencing strong break in the southwest monsoon. The atmosphere was relatively dry above 700 hPa (Figure 8), with only scattered cumulus and congestus in the region. Boundary layer and lower free-tropospheric winds were generally northwesterly to easterly in the northern Philippines on these days, instead of the much more typical southwesterly monsoonal flow. Consequently, sampled air masses on the *Vasco* were downwind from Luzon and/or Mindoro. On September 6th at ~1Z (~9:00 AM local) the *Vasco* sampled a significant spike in condensation nuclei, in excess of 1500 cm^{-3}. At the same time, filter and PCASP-inferred particle mass concentrations were low, only perhaps 1-3 µg m^{-3}, and CO was only slightly above background at ~100-110 ppbv. During this early period in the cruise, the SMPS was still operational and resolved the aerosol particle size dynamics from 0.02-0.5 µm (Figure 11). Three whole-air samples were also collected during the event, including 22:20Z Sept. 5th, as a pre event can, 2:45Z Sept 6th in the middle of the event, and 7:30Z Sept 6, for post event sample (Table 1).

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The aerosol dynamics for Apo Reef show the characteristics of a classic nucleation event (e.g., see review by Kulmala et al., 2004). Leading up to the event, particle concentrations were at ~500 cm^{-3}, with an estimated mass concentration <1 µg m^{-3}. The fine-mode particle number distribution was fixed to a count median diameter of 0.1 µm, but with significant enhancements throughout the event. Clearly, an airmass change occurred at ~ 23:00Z Sept 5, with a dramatic increase in particle concentration to 1000 cm^{-3}, and a slight fine-mode particle volume increase to ~1.5 µg m^{-3}. Nucleation was indicated at Sept 6 1Z, or nominally 9:00 local time, as solar radiation was increasing throughout the morning. Total concentration peaked at 1800 cm^{-3}. The count median diameter of the ultrafine mode initialized at 0.02 µm, growing to 0.05 µm in five hours. By 6:00Z (14:00 LST), the bimodal nature of the fine-mode aerosol population ended, with a strong 0.1 µm number mode in place. The fine mass concentration was estimated to be ~ 3-5 µg m^{-3} throughout the core of the event. While the ultrafine mode may have grown into this fine mode, there were additional modal shifts to 0.08 µm over the next two hours, which may actually be more representative of the airmass. Also noteworthy is that at ~3:00Z a simultaneous enhancement in both the fine and ultrafine mode occurred, suggesting a covariance in both the fine mode particles and the nucleation event precursor gases. Indeed, this is consistent as the nucleation event occurred along with a strong increase in fine particle concentration.

While a separate paper will be devoted to the whole-air samples from the 2011 and 2012 cruises, it is noteworthy here that the VOC profile during the event is consistent with the nucleation event, coinciding with reactive anthropogenic gas emissions (Table 1). During the event there are significant enhancements in reactive alkanes, notably roughly a factor of two enhancements in ethene and propene. Also notable were a factor of 2.5 increase in dimethyl sulfide over background, and a slight increase in CH_4, *i*-pentane and *i*-$PrONO_2$ and a decrease in CH_3I. All other species were relatively constant. Interesting, isoprene was near detectable limits, as were pinenes, as were pinenes, suggesting that terrestrial biogenic influences had been photochemically removed upwind. Also missing are enhancements in biomass burning markers. CO was fairly constant at 95-100 ppbv, as was benzene. Further the ratio of i- to n-pentane increased from 1.3 to 1.7. All of this data then points to the likelihood that the nucleation precursors were anthropogenic in origin from Luzon and/or Mindanao.

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5.2.2 Puerto Princesa Plumes

A second class of observed local aerosol phenomena was in association with urban plumes. While the *Vasco* cruise track was purposely away from population centers, there were times during transit that the influences of population centers were considerable. These ranged from small boat activity and emissions around coastal villages, to observation of the urban plume of Puerto Princesa as the *Vasco* entered and exited to take on supplies. Focusing on Puerto Princesa, the *Vasco* visited on ~Sept 13 and 20th, leading to four sets of observations. On three of these occasions, significant enhancement in particles due to the Puerto Princesa plume were clearly observed (for the exit on Sept 14, the local aerosol environment was dominated by the Borneo smoke event).

As the *Vasco* was nearing Puerto Princesa on September 13th, 2:00Z UTC (~10:00 LST) at approximately 30 km out, CN concentrations rapidly increased to instrument saturation at 10,000 cm^{-3}. At this point, the crew immediately suspected self-sampling, and quickly shut instrumentation down. However, it was then realized that the wind was in fact traveling directly over the bow. Some of the instrumentation was then restarted, and a whole-air sample was collected. While the boundary layer was clearly biomass burning-dominated upon departure the following day, the crew was prepared for sampling the plume the next visit Sept 19-20. In this case, concentrations peaked at only ~2200 cm^{-3}.

The pair of visits, while relatively isolated samples, nevertheless provide some insight into the nature of particle populations within the Philippines Islands. Key particle and gas measurements are included in Table 1, and can be compared to the Apo Reef and biomass burning events. Most importantly, the very high particle concentrations for arrival on Sept 13 have every indication of being a nucleation event. Unfortunately, as the SMPS was inoperative for this portion of the cruise, we cannot directly compare size distributions with the Apo reef case. But comparison of PCASP and CN count showed a substantial aerosol population with diameters less than 0.1 μm. Winds were clearly in an outflow region for the city, and solar radiation was fairly intense in the late morning under only moderately cloud free skies. On the subsequent visit on September 19, a sample was taken just before arrival, and a subsequent sample was collected as the *Vasco* entered

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the harbor. Clear enhancements in particle concentrations were observed. Although, as reported by Atwood et al. (2016; to be submitted), there was no nucleation mode in this case, suggesting these particles were primary; this is not unexpected given the earlier time of arrival (~8:00 AM LST) and full cloud cover. Similarly, upon departure in the 21st, at 6:00 AM LST, particle concentrations were low (<400 cm^{-3}), even before the morning commute.

Whole-air sample data for these cases provide us with other useful information. First, and most notably, the use of the ratio of *i*-to-*n*-Pentane in previous studies seemed to be justified, with values above 2 being clearly associated with the urban plume, and also slightly enhanced in the Apo Reef plume. Also hexane, a gasoline derivative, also appears to be a strong signature for Puerto Princesa. But in general, for most species, the differentiation between "urban" and "biomass burning" in older plumes is not so straightforward.

6.0 DISCUSSION: COMPARISON OF THE 2011 AND 2012 STUDIES

This paper had three primary objectives: 1) provide a broad overview of the 2012 *Vasco* cruise, including instruments carried, cruise track, and the general characteristics of the regional environment sampled; 2) apply the 2012 *Vasco* as a vehicle for continuing the narrative put forth in the 2011 effort on the nature of aerosol populations associated with the southwest monsoon, thus bridging climatological indicators commonly used to assess aerosol lifecycle to real world meteorology; and 3) relate how aerosol properties co-varied with regional meteorological phenomenon in order to establish the extent to which biomass burning or industrial pollution from the southern Maritime Continent can be transported towards or into the boreal summer southwest monsoonal trough. To our knowledge, these cruises provide the first published aerosol field measurements in the boreal summertime South China and Sulu Sea regions.

The similarities and contrasts between the 2011 and 2012 burning seasons and cruise observations portray key aspects of the southwest monsoon aerosol system, pointing to a number of pressing observational and prediction challenges. Certainly from an inter-seasonal, seasonal, and even monthly time period, the conceptual models of aerosol lifecycle in the southwest monsoon of Reid et al. (2012) largely holds. At an inter-seasonal scale, ENSO phase is related to precipitation deficits and hence more burning in the MC. This ENSO cycle also effects monsoon

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onset and transition. Within the season, the MJO, in part, regulates large-scale precipitation patterns, which then affects event timing. Tropical cyclones develop well-defined areas of monsoonal enhancement/inflow arms with accelerated surface winds that help draw smoke further into the monsoonal flow, and but may also lead to enhanced scavenging. Major land/sea breeze events can lead to significant aerosol ejection off island. Ultimately, multi-day events are possible, such as the two events for 2011, and the Sept 14-17 event for 2012. Finally, while near "pure" biomass burning events are possible, there is more typically a mixture of biomass burning and other anthropogenic emissions. Both the comparison of the seasonal behavior and the measurements on the cruises bear these similarities out.

Several key differences between long-range transport characteristics in 2011 and 2012 are highly noteworthy. First, while the monsoon frequently has weak and strong phases, the 2012 case clearly showed how strong the effect of tropical waves moving through the region can be on low-level flow patterns. Indeed, the first week of the 2012 cruise coincided with uncommonly clear skies and even northerly winds. Such clear areas provide some of the rare opportunities for satellite observations. Yet from a climatological point of view provide, this clear sky bias fundamentally represents a skewed portrait of the aerosol system (Reid et al., 2013). Additional studies are already underway to ascertain the specific representativeness of aerosol particles in the region.

A second significant difference between 2011 and 2012 is that while a multi-day event occurred, many shorter events were observed in the latter year. This is likely in part due to the closer proximity of the *Vasco* to Borneo, and we speculate that a more significant role of convection along the coast of Borneo led to more pockets of smoke. Further in 2012, the *Vasco* did not experience a regional severe clear day as there was at the end of the 2011 cruise that was caused by a TC propagating across Luzon and into the South China Sea. This then leads to suspicion that many pockets of polluted air can be migrating through the region, in between convective cells but still obscured from satellite detection, on a regular basis.

High-frequency events in 2012 also included observation of two nucleation events and urban plumes. While it is often thought that these types of nucleation events only occur in the presence of gas precursors but few aerosol particles (e.g., Mäkelä et al., 1997; Kulmala et al., 2004; Boy et al., 2008), for the tropics and subtropics, nucleation events have also been noted in polluted

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urban environments (e.g., Cheung et al., 2011; Betha et al., 2013; Kanawade et al., 2014, Brines et al., 2015) and even dense tropical smoke plumes (Reid et al., 2005). The *Vasco* observed both kinds. The Apo reef nucleation event seemed to follow the more traditional relationship, with precursor gases in the presence of low aerosol particle surface area. Indeed, while in clean mid-latitude marine conditions, Covert et al., (1992) observed explosive nucleation events and discounted local or transported sources. Instead, they suggested such an event being a natural outcome for a marine boundary layer having low particle surface area. This later argued that nucleation in some of these remote sub-tropical to mid latitude areas are assisted by ion-mediated nucleation events (IMEs) formed by the ionization of molecules by cosmic rays (Yu et al., 2008). While Yu et al., (2008) considers such nucleation generally unfavorable in tropical regimes and the interior of the MC, they did predict significant nucleation on the periphery- notably west of the northern Philippines, south of Java and east of New Guinea. Indeed, Yu et al., (2018) placed a nucleation hotspot right at our point of observation. Aided by anomalously clear skies, and thus likely high photolysis rates we see this nucleation mechanism as being reasonable contributor to the event. Indeed, it was the only such observed event in the two Vasco cruises.

For the *Vasco* case, we indeed had low particle concentrations, but we must consider enhancements due to the presence of reactive VOCs. Indeed, parts of Luzon are heavily populated, such as Manila, and there are many other urban centers, such as Batangas, ~200 km northwest of where we were anchored. In between are countless small villages, large shipping, and small fishing boat activity, all of which are capable of producing photochemical precursors and hence promoting nucleation events such as those observed. Thus, while particle mass concentrations (and hence AOT) may be low, there are likely many pockets of high CCN concentration generated from remote islands.

The second type of nucleation event, in the outflow of a polluted urban plume, was also observed by the Vasco outside of Puerto Princesa. Nucleation events with concentrations this high have been reported in urban tropical air in late morning (e.g., Cheung et al., 2011; Betha et al., 2013; Kanawade et al., 2014, Brines et al., 2015). For Singapore, enhancements of higher than 10,000+ cm^{-3} were frequently observed, dramatically shifting the size distribution to smaller sizes (Betha et al., 2013). Ultimately, whether in clean or more polluted conditions, aerosol nucleation events are probably not uncommon in the Maritime Continent.

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In addition to nucleation events, the *Vasco* in 2012 intersected many small plumes, as well as the strong urban plume of Puerto Princesa (population =~250,000). These observations remind us that while many of the islands of the MC are thought of as "remote", outside of the megacities, they can nevertheless harbor reasonably sized populations. Given the significant use of biofuel or highly polluted engines, these islands can clearly emit significant amounts of CCN. The relationship between these nucleation events and urban plumes to CCN dynamics is discussed at length in Atwood et al., (2016; to be submitted).

Finally, and perhaps most interestingly, the 2012 cruise demonstrated a new relationship between aerosol events and convective cells and more organized squall lines. In 2011, drops in particle concentration were coincident with temperature; consistent with the notion that cold pool air was advecting into the region with aerosol particles already deposited out. As the profiles show wind shear and variable wind speeds, the steering winds of the squall lines roll over polluted airmasses underneath. Thus, these squall lines could be likened to "lawn mowers" ingesting or scavenging aerosol particles as they propagate.

Based on the work of Seigel and van den Heever (2012), which showed that dust generated ahead of cold pools on the leading edge of thunderstorms is lifted to mid-levels where the potential impact of aerosol particles as CCN was minimal, the 2011 cruise suggested that the nature of convection in the region often insulated itself from potential aerosol impacts. Certainly, the *Vasco* observed some of this behavior in 2012, but also observed the opposite; cases where the telltale cold pool signs of rapid temperature drop and spikes in wind heralded the coming of a polluted airmass. Indeed, during the 14th-18th September period in Figure 9, both clear air and polluted air followed cold pools. While the wiring diagram for larger scale features is largely well known, and to some extent can be qualitatively captured by a coarse-grid model such as NAAPS with additional constraint from satellite precipitation products, there remains much to understand about aerosol lifecycle in the vicinity of convective cells and squall lines. We suspect that a clue to the behavior where air pollution is following cold pool event lies in the rather shallow temperature drops (1-2^{o}C versus 5-6^{o}C). This may be an indicator that the convection is not so strong, or that may in part be a remnant. Such events may also be related to the nature of the initial formation of convection or a squall line relative to a polluted airmass. The origin of the convection, whether from a coastal ejection event or a large convective system,

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may play a role. Or, steering winds and wind shear are such that some moisture convergence occurs on the leading edge of an ejection event leading to weak convection along the boundary. However, this situation thus far has not been observable from satellite.

To speculate, these events of thick aerosol plumes behind convection seem to be consistent with a land breeze origin that was propagated much further than normal by the monsoonal flows. Certainly the temperature change and high aerosol loading behind a cloud top front matches aircraft observations of large land breeze ejection events in the Arabian Gulf (e.g., Reid et al., 2008). In the MC case, cloud development along land breeze fronts is much larger, leading to significant convection offshore of islands (e.g., Liberti et al., 2001; Qian 2008; Virts et al., 2013).

While the propagation of such land breeze fronts in a strong monsoonal flow cannot be observed by satellite due to the collinear presence of cirrus, dramatic events when the monsoonal flow is weak are frequently observed in imagery. Some examples of the Sarawak Borneo coast are presented in Figure 12, which were observed by Terra MODIS Sept 22nd and October 1st and 3rd 2012. During these periods monsoonal winds slackened somewhat before and after the passing of TC Jelawat to the east. The land breeze fronts are visible as clear areas just offshore of the island, with a long cloud border parallel to the island, which in geostationary data is propagating outward. Within the imagery, heavy aerosol loadings are visible within the land breeze zone, although currently none of operational satellite aerosol products can conduct a retrieval three due to off colored ocean surface and the presence of thin cirrus. Based on MOD06 cloud products, clouds along the leading boundary are generally low, with top at ~2-3 km, with periodic cells to the 0^{o}C melting level. It is likely that such deep warm deep clouds are precipitating-but it is unknown for sure. Occasionally, cumulonimbus with tops to 14 km are visible, in good agreement with the climatology of Virts et al., (2013). Such storms are visible based on the telltale cirrus blowing off to the southwest. Certainly these are precipitating clouds, and large enough to generate their own cold pools and may also explain the temperature drops associated with aerosol event arrival as well as help sustain the line of convection. These three events all have the same overall phenomenon observed on the Vasco; namely, an aerosol front behind a thin line of convection, some of which precipitating. The question is, once monsoon flow are enhanced, are these the phenomenon the ones ultimately responsible for the aerosol fronts

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observed on the Vasco. A next step for this science tem is to then attempt to model such phenomenon.

6.0 CONCLUSIONS

This paper provides an overview of the meteorological and aerosol environment measured by the *M/Y Vasco*, which sampled Maritime Continent air in September 2012 along the entire length of the Palawan Archipelago, Philippines. This cruise was a longer follow-on to a similar research cruise the previous year (Reid et al., 2015) and was a significant component of the 2012 7 Southeast Asian Studies (7SEAS) southwest monsoon intensive period-a high water mark for observations throughout the Maritime Continent. The Palawan region for this research cruise was selected for being a receptor of smoke and anthropogenic emissions from Borneo, Sumatra and the Malay Peninsula as emissions were advected by southwesterly monsoonal flow into the seasonal monsoonal trough east of the Philippines. Also presented in this manuscript for context is an overview of the 2012 Maritime Continent burning season, and its relationship to key meteorological features, including ENSO, monsoonal flows, the MJO and tropical cyclones. The key conclusions of this study are as follows:

1) 2012 exhibited slightly above-average burning activity in the Maritime Continent, consistent with a warm-neutral ENSO phase. Several pulses of burning and emissions occurred monthly in June through October, consistent with the migration of three moderate to weak MJO events. Particularly strong pulses were associated with large-scale upper tropospheric subsidence and regional flow enhancement provided by the inflow arms of tropical cyclones. This and previous work point to the close coupling between tropical waves and the regional aerosol system. Aerosol Optical Thickness (AOT) from AERONET sites frequently registered above 1 at 500 nm in source regions, with the strongest long-range events reaching 0.34 in Mindanao.

2) The 2012 cruise home ported at Manila, Philippines, and sampled three major regions: (a) the upper Palawan chain and El Nido for September 4-13, 2012; (b) the southern Palawan chain and Balabac Island on the southern tip of the Palawan chain, ~100 km north of the northern tip of Borneo, Sept 14-20; and (c) the Sulu Sea and Tubbataha Reef Sept 21-29, 2012. In the northern

locations, the atmosphere was under the influence of an easterly wave, bringing unseasonable north-to-northeasterly winds and air from the northern Philippine islands of Luzon and Mindoro. Observations included a pronounced particle nucleation event in relatively clean conditions in a region where ion mediated nucleation was predicted by Yu et al., (2008). In the southern and Sulu Sea locations, biomass burning and anthropogenically-polluted air masses were sampled, largely modulated by enhancement in monsoonal flows associated with two category 5 tropical cyclones. Fine particle concentrations reached ~35 μg m^{-3}, and CO was as high as 250 ppbv. Finally, while transiting through Puerto Princesa for supplies, the city plume was also sampled, including a second nucleation event in more polluted conditions with CN concentrations of 10,000 cm^{-3}. In comparison, "background" values of aerosol particle concentrations were on the order of 500 cm^{-3}, roughly 50-100% higher than the 2011 cruise period.

3) The large-scale relationships between aerosol emissions, aerosol transport and regional meteorology during the cruise broadly matched the conceptual models of Reid et al. (2012) regarding relationships to the MJO and tropical cyclones. However, easterly waves resulted in significant weakening of the monsoonal flow, and two slow moving category 5 tropical cyclones located southeast of the Philippines resulted in monsoonal winds that had anomalous northerly and enhanced westerly components at times.

4) While a multi-day biomass burning event was observed mid-cruise, in comparison to 2011, aerosol events showed much higher frequency behavior. Even in the middle of the Sulu Sea, pulses of aerosol particles on the order of 3-6 hours were observed. This behavior is likely in part due to influence of scattered convection, leaving pockets of polluted and clean air masses. In addition, the aforementioned nucleation events and urban plumes added additional high-frequency signals. This high frequency behavior further complicates an already complex aerosol and cloud system.

5) Finally, the 2011 cruise pointed to the important role of organized squall lines and cold pools in scavenging aerosol particles from the marine boundary layer. While very clean air was observed behind the squall lines, there were many cases where the opposite relationship was observed. That is, a rapid temperature drop and spike in wind heralded not clean air behind a squall line, but highly polluted air following behind. This difference may be a result of squall line origin, meteorology, and/or lifecycle. Some of the effects may be a result of remnant cold pools. Or, steering winds and wind shear are such that some moisture convergence occurs on

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the leading edge of an ejection event leading to weaker convection along the boundary. However, our prevailing hypothesis is that these events are a result of convection forming from a coastal land breeze ejection event that is caught in enhanced monsoonal flows. Clearly, understanding the dynamics of aerosol particles around such organized convective features is a high priority for future work.

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Table 1. Aerosol and whole-air sample concentrations and associated particle data for key aerosol events during the study. Units are pptv unless otherwise noted. BDL=Below detectible limits. Local standard time is 8:00 ahead of UTC

Specie	Back-ground	Biomass Burning		Apo Reef			Puerto Princesa			
		Sep 14 6:38Z	Sep 26 0:52Z	Pre-Event Sept 5 22:20Z	Early Event Sept 6 2:45Z	Late Event Sept 6 7:45Z	Inside Sep 13 2:47Z	Outside Sept 19 22:55Z	In plume Sep 19 23:52Z	Depart Sep 20 22:00 Z
CN (cm^{-3})	~400	2500	2100	490	1100	1450	>10000	400	2230	317
~PM_1($\mu g\ m^{-3}$)	~1.5	30	21	1.2	1.8	1.9	10	1.5	8	2.5
CO (ppbv)	77+/-3	244	209	100	98	92	144	111	207	136
CH_4 (ppmv)	1.77+/-0.01	1.77	1.76	1.768	1.81	1.77	1.77	1.94	1.82	1.82
DMS	24+/-12	27	31	7.3	19	9.1	8.6	13.2	9.8	11.1
CH_3I	0.7+/-0.7	1.38	1.48	0.95	0.55	2.9	0.42	1.25	0.43	0.48
Ethane	348+/-55	877	824	399	504	369	525	514	619	434
Ethene	47+/-32	376	490	101	212	69	274	381	880	235
Ethyne	68+/-18	561	501	107	131	102	315	116	953	123
Propane	67+/-33	137	166	86	90	68	140	111	374	78
Propene	24+/-12	76	148	49	80	76	62	249	236	62
i-Pentane	57+/-33	35	51	23	40	30	241	49	537	64
n-Pentane	40+/-23	26	43	18	24	19	106	50	258	56
Hexane	31+/-14	21	38	15	12	9	49	48	123	52
Benzene	32+/-27	140	162	36	40	23	96	42	198	40
Isoprene	22+/-36	4	BDL	6	3	BDL	3	18	59	14
i-$PrONO_2$	1.1+/-0.3	2.3	2.7	1.7	2.0	1.4	2.2	3.1	1.9	2.6
n-$PrONO_2$	0.3+/-0.2	0.4	0.4	0.3	0.2	0.5	0.5	0.2	0.3	0.6
$BuONO_2$	1+/-0.7	1.6		1.3	1.6	1.2	2.7	2.1	1.9	2.0
Ratio of i to n	~1.4	1.3	1.2	1.3	1.7	1.6	2.3	1	2.08	1.2

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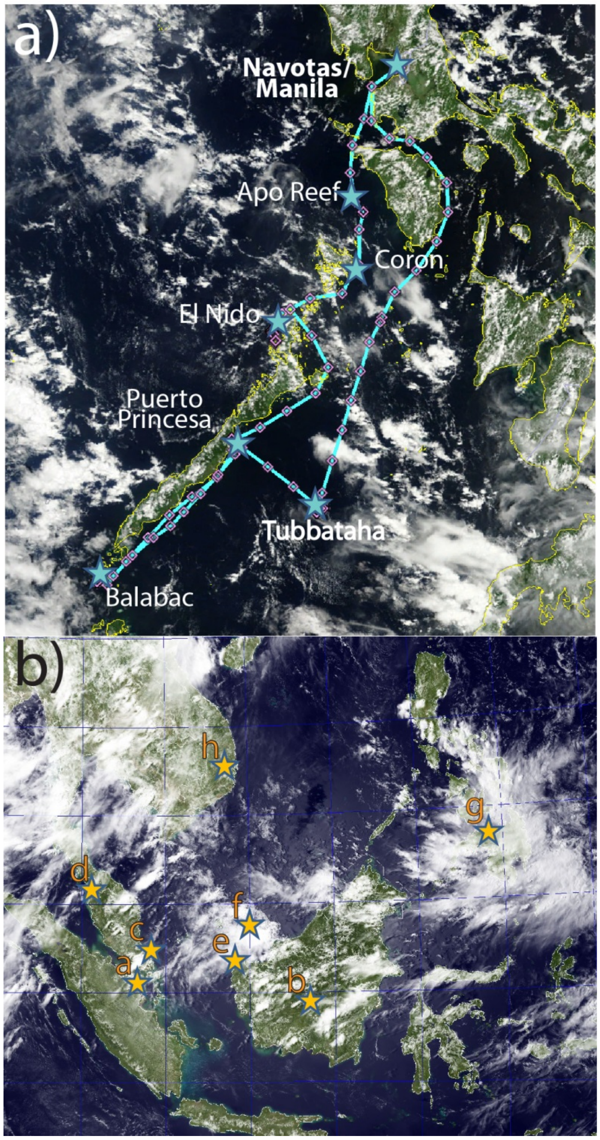

Figure 1. a) Cruise track with major sampling locations stared overlaid on Terra MODIS 7 Sep 2012. b) AERONET sun photometer sites used in this analysis overlaid on MTSAT including false color visible imagery for 7 Sep 2012 6:32 UTC, a-Jambi; b-Palangkaraya; c-Singapore; d-Tahir; e-Pontianak; f- Kuching; g-Marbel University; h- Nha Trang.

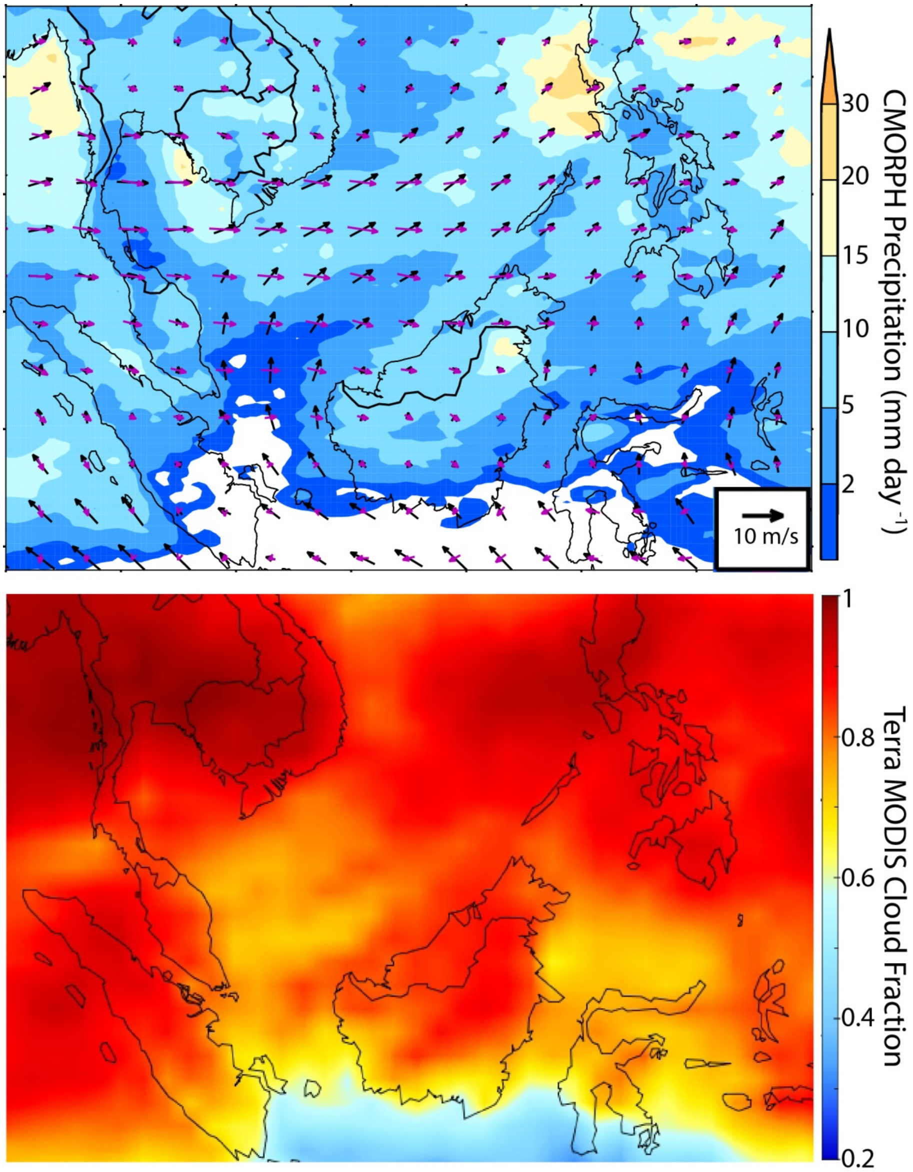

Figure 2. Average August & September 2012 NAVGEM Surface (Black) and 700 hPa (Magenta) winds overlaid on average CMORPH derived rain rate.

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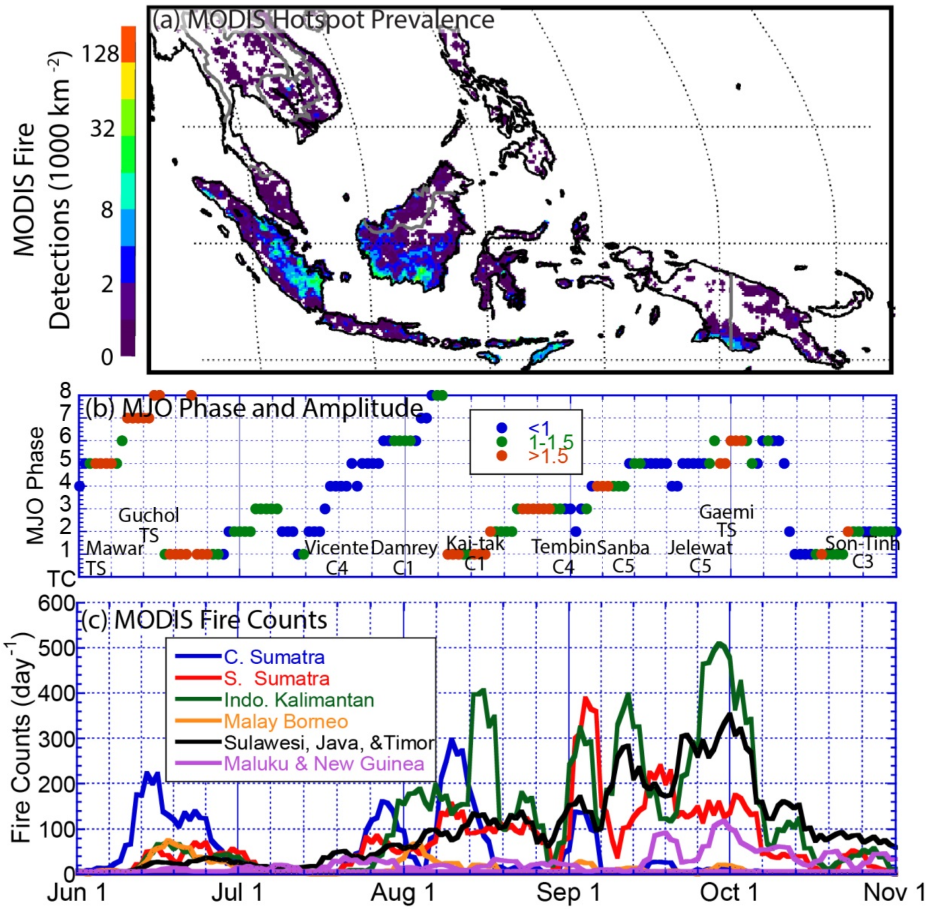

Figure 3. (a) Overall Terra and Aqua MODIS detected fire prevalence for June-October 2012; (b) Wheeler index of MJO phase and color coded amplitude. Amplitudes above one are considered statistically significant. (c) A 5 day box car average of observed Terra and Aqua combined active fire hotspot detections for the 2012 Maritime Continent burning season by region as defined in Reid et al., (2012; 2015).

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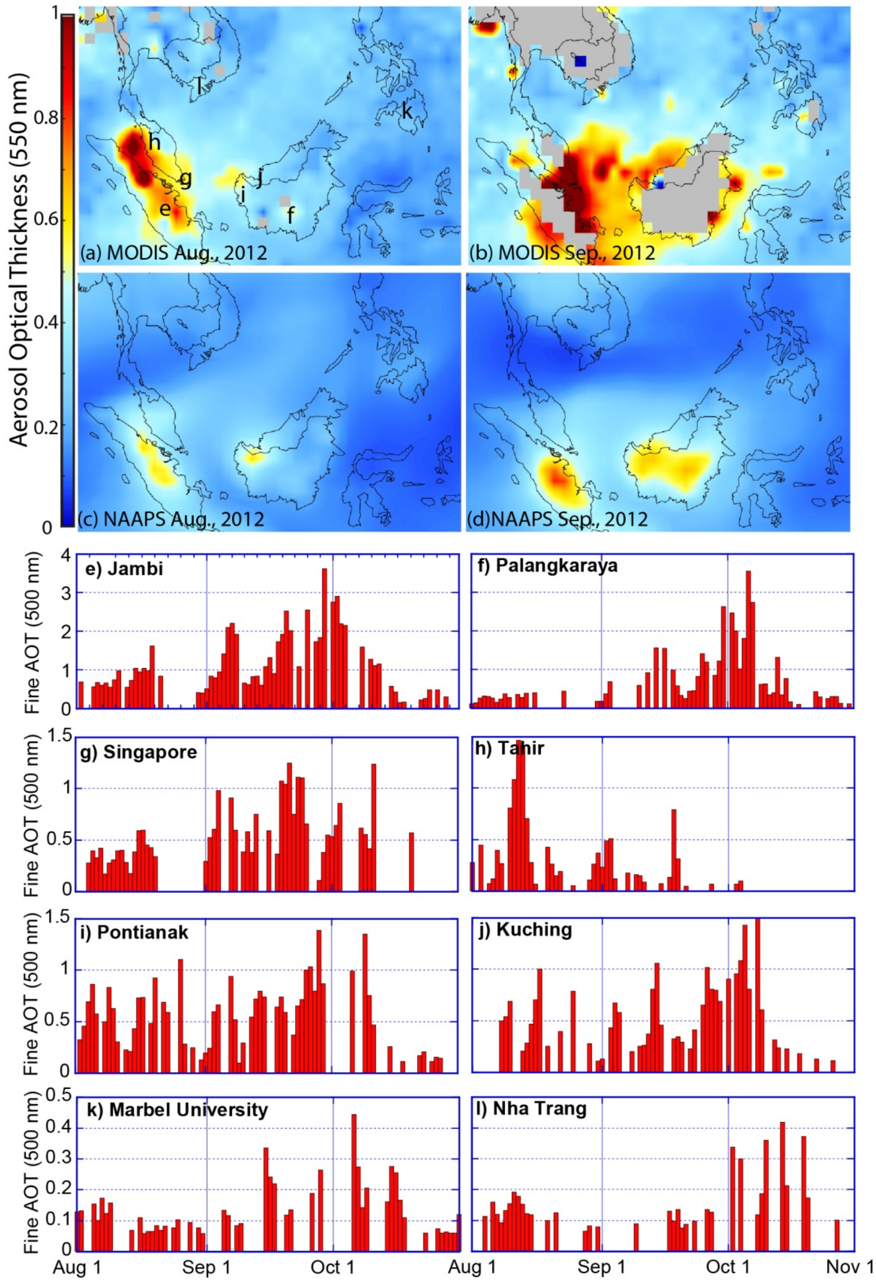

Figure 4. August and September 2012 monthly average 550 nm total Aerosol Optical Thickness (AOT) from (a) and (b) MODIS and (c) and (d) NAAPS reanalysis. Also shown are (e)-(l) daily average AERONET 500 nm fine mode AOTs. Site locations are also marked in (a).

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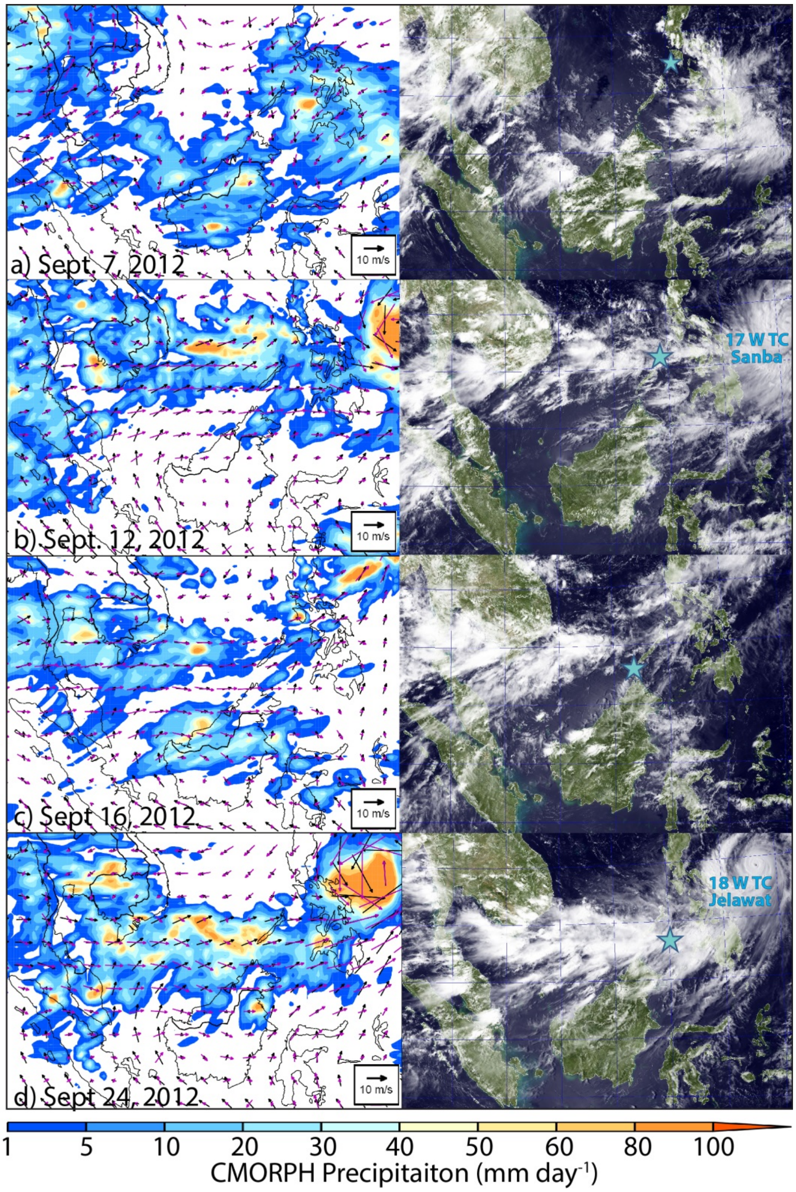

Figure 5. Model, satellite precipitation and MTSAT visible imagery for four representative days at major anchorages throughout the *Vasco* cruise. Left column provides daily averaged NAVGEM wind (black surface, magenta 700 hPa) overlaid on CMPRPH dial averaged precipitation. Right column is the 02:32 UTC (nominal noon local time) MTSAT false color visible image, annotated with Vasco position (blue star) and active TCs influencing regional weather. Selected dates are at major sampling moorings/anchorages (a) Sept 7, at Apo Reef, (b) Sept 12 at El Nido, (c) Sept 16 at Balabac Island, and (d) Sep 24th at Tubbataha Reef.

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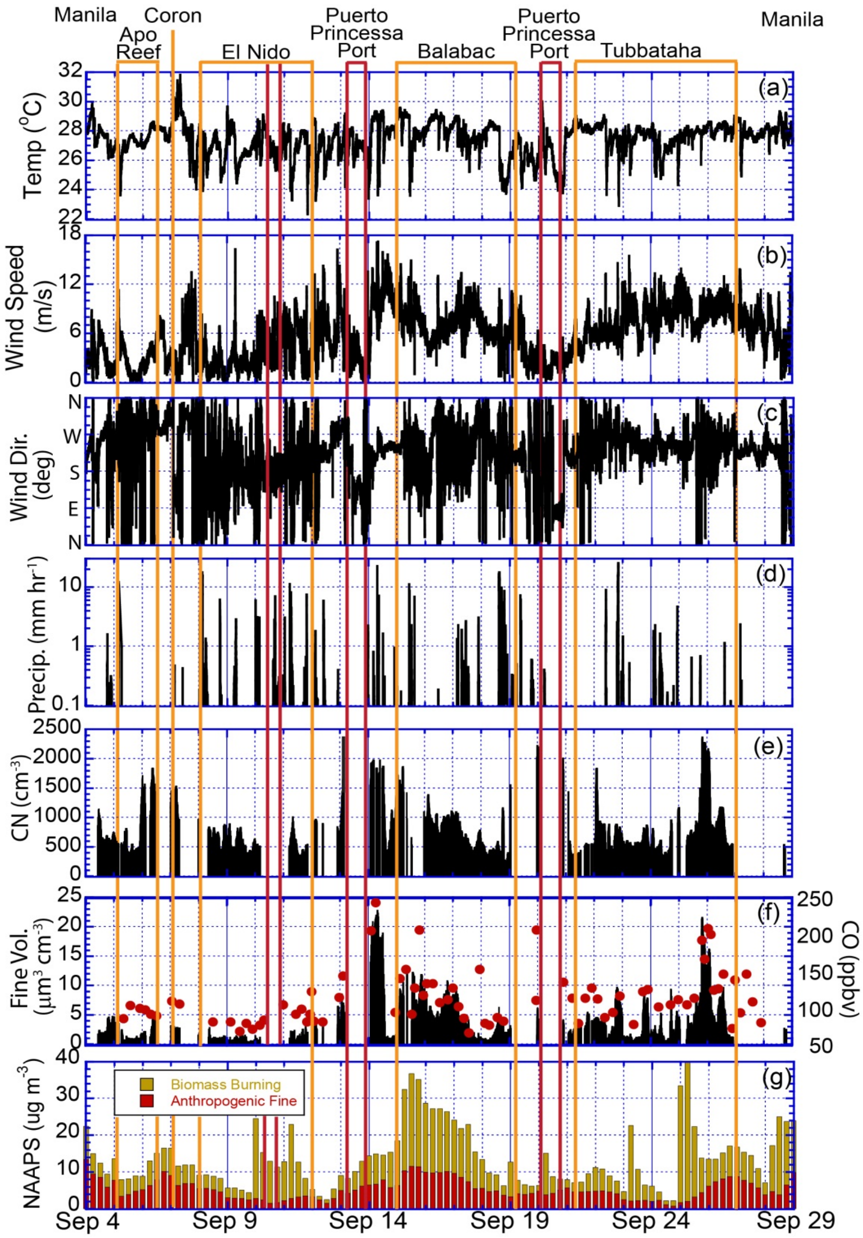

Figure 6. Time series of key meteorological and compositional data along the Vasco track (UTC Time). Locations of stationary sampling and in port periods are bound in orange and red brackets, respectively. Included are a) temperature [^{o}C] ; b) wind speed [m s^{-1}]; c) wind direction; d) precipitation rate [mm hr^{-1}]; e) condensation nuclei count [cm^{-1}]; f) Fine mode volume as derived by the PCASP [black µm^{3} cm^{-3}] and the gas can CO [red dots ppbv]. Also shown is the combined NAAPS model derived fine mode mass sampled along the *Vasco* track.

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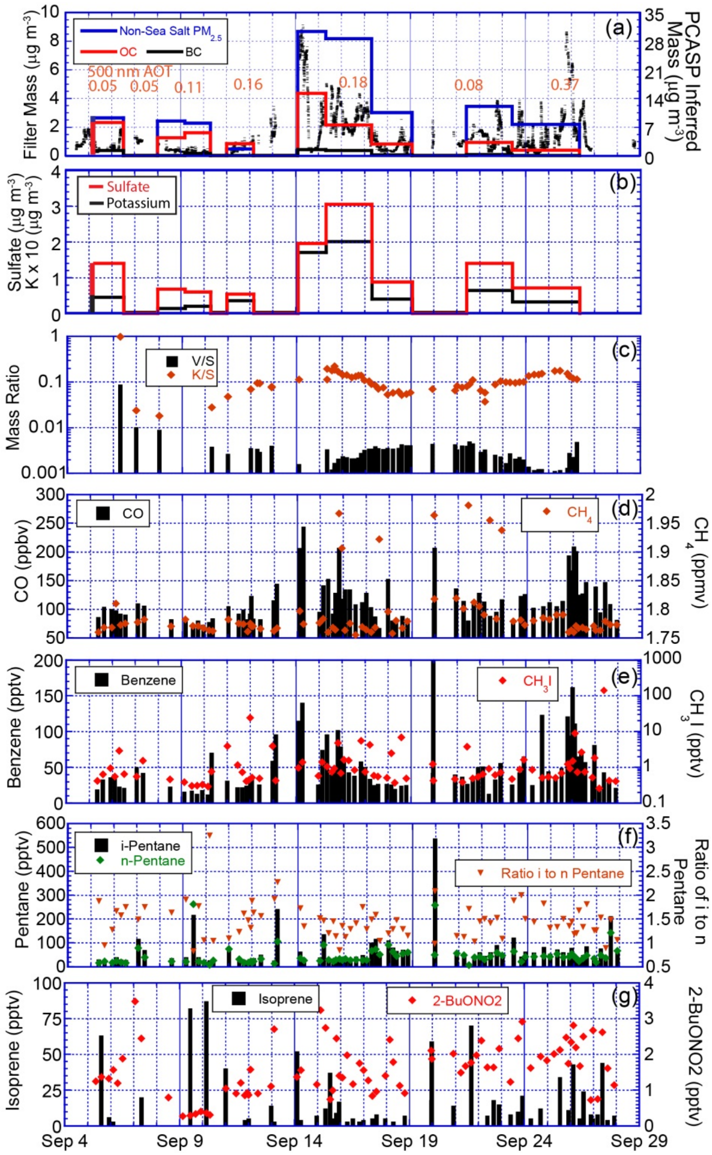

Figure 7. Time series of chemistry measurements including (a) Filter based gravimetry and Organic-OC and Black-BC carbon. Also shown is 30 minute averaged mass inferred from the PCASP; (b) filter sulfate and potassium; (c) DRUM sampler elemental ratios of vanadium and potassium to sulfur; (d) whole-air sampled CO and methane; (e) whole-air sampled benzene and methy iodide; (f) whole-air sampled i- and n-pentane with their ratio; (g) whole-air sampled isoprene and 2-butanalkyl nitrate.

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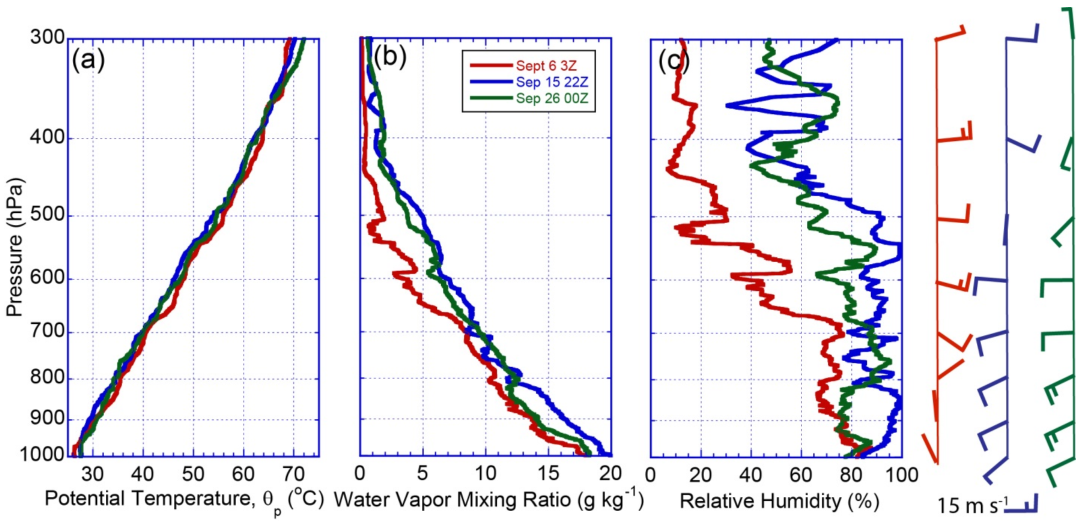

Figure 8. Sounding profiles of (a) Potential temperature; (b) Water vapor mixing ratio; and (c) Relative humidity for profiles corresponding the nucleation event on Sept 6 at Apo reef, and the biomass burning cases of September 15 and 26th at Balabac island and Tubbataha Reef, respectively. Wind flags for these cases are marked on the far right.

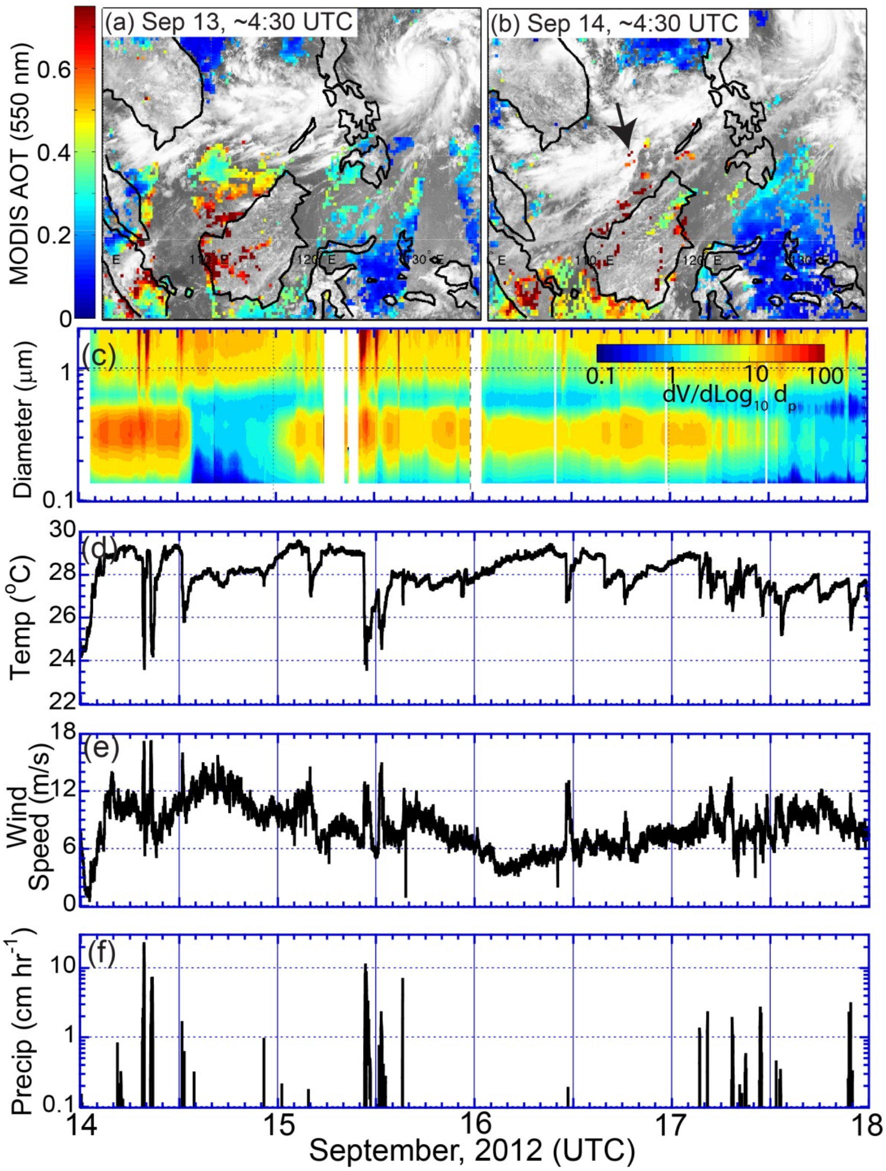

Figure 9. Satellite data and Vasco one-minute data time series describing the Sept 14-17 smoke event. (a) and (b), combined Terra and Aqua MODIS C6 550 nm AOT overlaid on MTSAT visible channel for September 13 and 14, respectively; (c) PCASP volume distribution; (d) Temperature; (e) Wind speed; (f) Precipitation rate. The presence of a large squall line originating from a massive thunderstorm over the Malaya Peninsula that resulted in the Sept 14 12Z clean period as evident in (c) is marked with an arrow in (b).

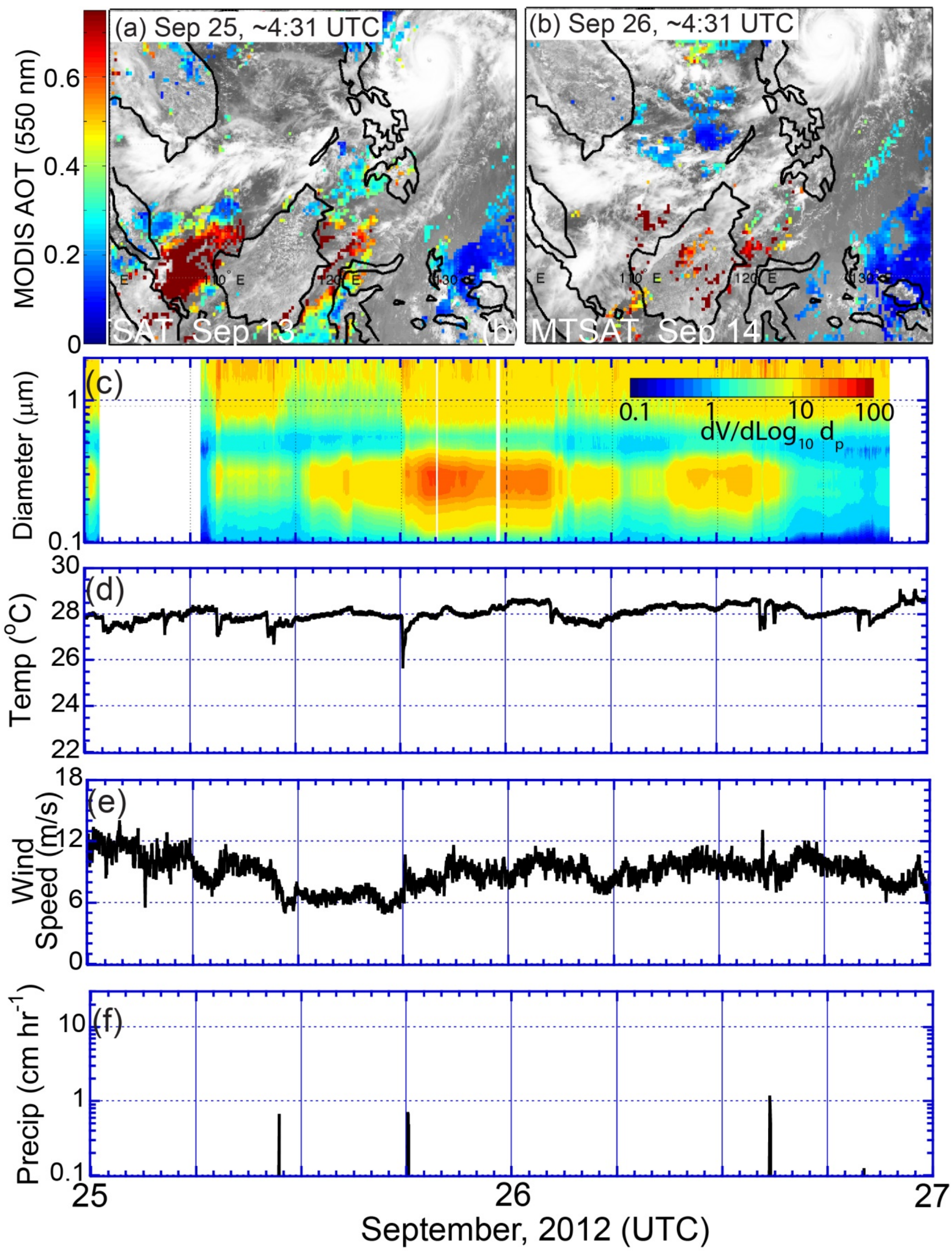

Figure 10. Same as Figure 9 but for the Sept 25-26, 2012 smoke event.

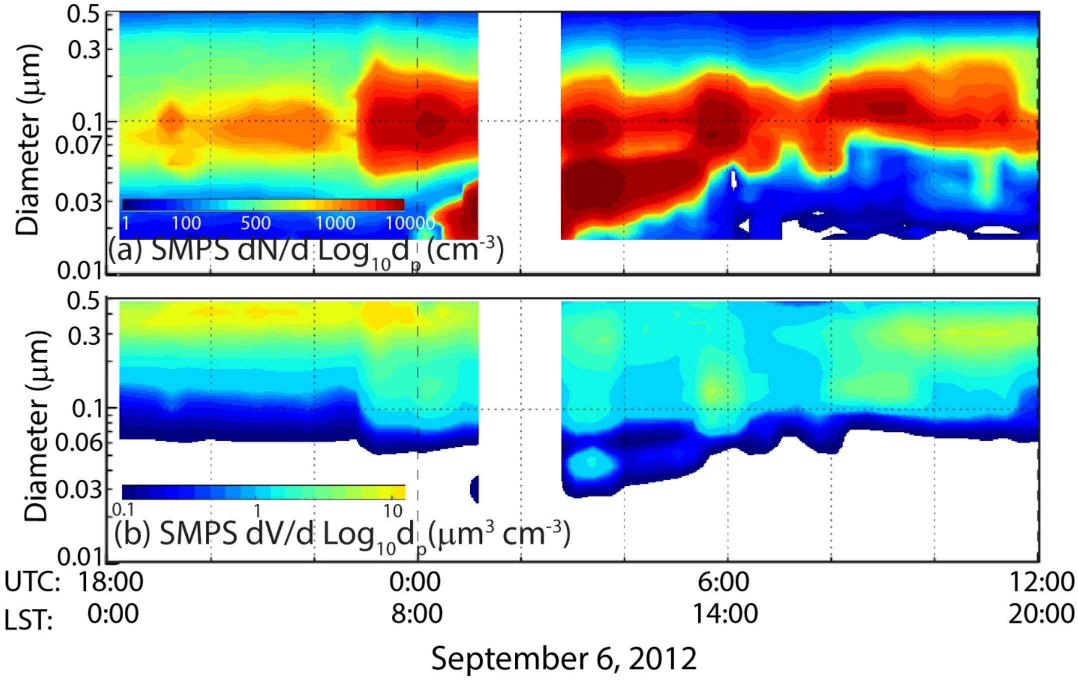

Figure 11. SMPS particle number and volume distribution for the Sept 6th nucleation event at Apo reef. Corresponding whole-air samples are listed in Table 1 including 22:20Z Sept. 5th as a pre event can, 2:45Z Sept 6th in the middle of the event, and 7:30Z Sept 6, for post event.

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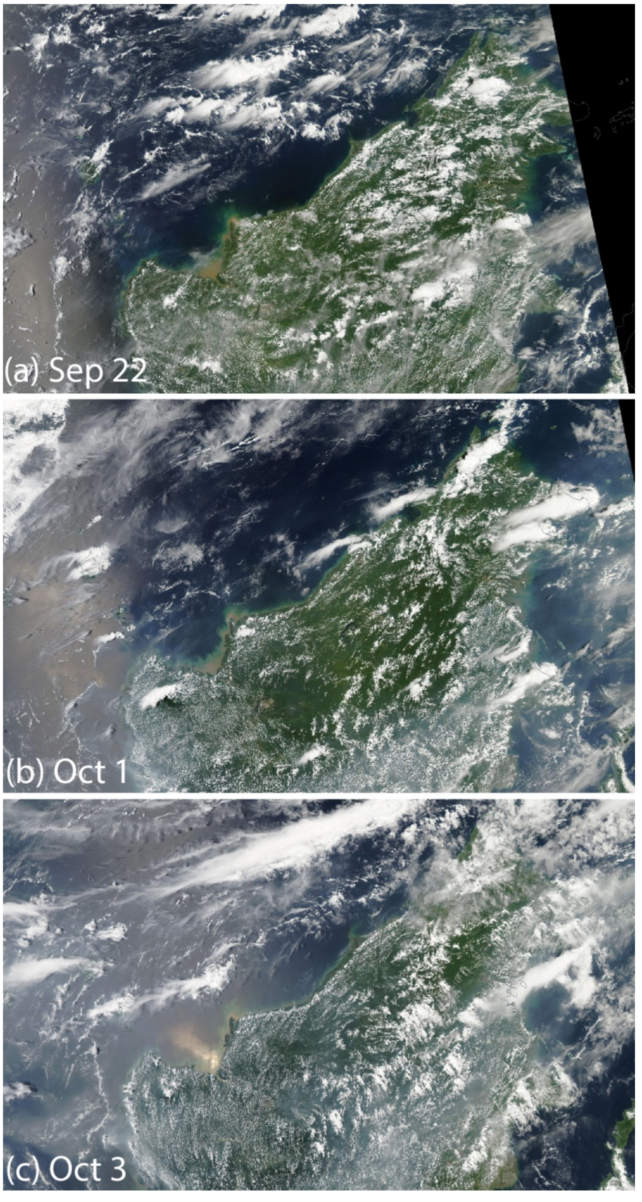

Figure 12. NASA Terra MODIS images of the northern Borneo coast off of Sarawak Malaysia demonstrating land breeze induced ejection events. Such events are identified by clear areas just offshore of the island with a leading edge of clouds. Occasionally embedded in this line of clouds are thunderstorms.