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Importance of transboundary transport of biomass burning emissions to regional air quality in Southeast Asia during a high fire event

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Abstract. Smoke from biomass and peat burning has a notable impact on ambient air quality and climate in the Southeast Asia (SEA) region. We modeled a large fire-induced haze episode in 2006 stemming mostly from Indonesia using the Weather Research and Forecasting model coupled with chemistry (WRF-Chem). We focused on the evolution of the fire plume composition and its interaction with the urbanized area of the city state of Singapore, and on comparisons of modeled and measured aerosol and carbon monoxide (CO) concentrations. Two simulations were run with WRF-Chem using the complex volatility basis set (VBS) scheme to reproduce primary and secondary aerosol evolution and concentration. The first simulation referred to as WRF-FIRE included anthropogenic, biogenic and biomass burning emissions from the Global Fire Emissions Database (GFED3) while the second simulation referred to as WRF-NOFIRE was run without emissions from biomass burning. To test model performance, we used three independent data sets for comparison including airborne measurements of particulate matter (PM) with a diameter of 10 µm or less (PM_{10}) in Singapore, CO measurements in Sumatra, and aerosol optical depth (AOD) column observations from four satellite-based sensors. We found reasonable agreement between the model runs and both ground-based measurements of CO and PM_{10}. The comparison with AOD was less favorable and indicated the model underestimated AOD, although the degree of mismatch varied between different satellite data sets. During our study period, forest and peat fires in Sumatra were the main cause of enhanced aerosol concentrations from regional transport over Singapore. Analysis of the biomass burning plume showed high concentrations of primary organic aerosols (POA) with values up to 600 µg m^{-3} over the fire locations. The concentration of POA remained quite stable within the plume between the main burning region and Singapore while the secondary organic aerosol (SOA) concentration slightly increased. However, the absolute concentrations of SOA (up to 20 µg m^{-3}) were much lower than those from POA, indicating a minor role of SOA in these biomass burning plumes. Our results show that about 21 % of the total mass loading of ambient PM_{10} during the July–October study period in Singapore was due to biomass and peat burning in Sumatra, but this contribution increased during high burning periods. In total, our model results indicated that during 35 days aerosol concentrations in Singapore were above the threshold of 50 µg m^{-3} day^{-1} indicating poor air quality. During 17 days this was due to fires, based on the difference between the simulations with and without fires. Local pollution in combination with recirculation of air masses was probably the main cause of poor air quality during the other 18 days, although fires from Sumatra and probably also from Kalimantan (Indonesian part of the island of Borneo) added to the enhanced PM_{10} concentrations. The model versus measurement comparisons highlighted that for our study period and region the GFED3 biomass burning aerosol emissions were more in line with observations than found in other studies. This indicates that care should be taken when us-

ing AOD to constrain emissions or estimate ground-level air quality. This study also shows the need for relatively high resolution modeling to accurately reproduce the advection of air masses necessary to quantify the impacts and feedbacks on regional air quality.

1 Introduction

Biomass burning plays an important role in atmospheric composition and chemistry (Crutzen and Andreae, 1990; Lamarque et al., 2010). Fires occurring close to populated areas severely impact air quality affecting millions of inhabitants (Johnston et al., 2012; Marlier et al., 2013). Governments and international organizations such as the World Health Organization (WHO) have produced pollution guidelines in the last decade (WHO, 2006), but the contribution of biomass burning emissions to local air quality is neither well understood nor quantified.

Southeast Asia (SEA), especially Indonesia, has high biomass burning fuel consumption (up to 20 kg C per m^{-2} burned) due to fires burning in the peatlands (Page et al., 2002; van der Werf et al., 2010). This, in combination with frequent fire activity ensures that the region has the highest density of fire emissions globally. Fire activity is highly modulated by the El Niño–Southern Oscillation (ENSO) and the Indian Ocean Dipole (IOD) (Hong et al., 2008; Field et al., 2009; Reid et al., 2013). Densely populated areas such as Java and the city of Singapore are located relatively close to large fires mainly in Sumatra and Kalimantan and regularly show high particulate pollution levels which are often related to emissions from forest, agriculture and peat fires (Hyer and Chew, 2010; Salinas et al., 2013a, b; Wang et al., 2013). Models that accurately simulate biomass burning plumes and their air quality impacts in this complex orographic and meteorological region are necessary to better understand the transport and evolution of smoke plumes.

Air pollution caused by aerosol particles is of concern because of reduction in visibility and adverse environmental and health impacts (Mauderly and Chow, 2008). Depending on their size and chemical composition, aerosol particles can penetrate into the respiratory system and increase throat and lung infections (Karthikeyan et al., 2006; Pavagadhi et al., 2013). In addition, aerosols also increase the risk of developing lung cancers (Abba et al., 2012). Fires emit high concentrations of particles of small sizes as well as volatile and semi-volatile organic compounds which may act as precursors in the formation of secondary aerosols (See et al., 2006, 2007; Keywood et al., 2003; He et al., 2010; Yee et al., 2013). In this study, we focus on transboundary particulate pollution levels affecting the Republic of Singapore (population of over 5 million) due to the release of aerosol particles from biomass burning in Indonesia. We used WRF-Chem to (1) advect the aerosol and gaseous precursor concentrations emitted by biomass burning, (2) represent the evolution of the aerosol plume dynamics and chemistry and (3) evaluate the interactions between this transported and aged air mass from fires with freshly emitted urban pollution in Singapore.

2 WRF-Chem setup and evaluation

2.1 Model setup

We used the online-coupled regional Weather Research and Forecasting model with chemistry (WRF-Chem) (Grell et al., 2005) v3.4 to simulate meteorology and atmospheric composition at a regional scale. The fully coupled model WRF-Chem computes at each time step the dynamic processes including advection as well as the microphysics and the atmospheric chemistry and aerosol processes.

The simulation was done for a domain with 100 × 100 grid points, each with a 15 km × 15 km horizontal resolution. The domain included Sumatra (Indonesia), the Republic of Singapore and the southern part of the Malaysian peninsula (see Fig. 1). The model had 30 vertical levels from ground level up to 23 km height with a stretching resolution from 60 m to 1.6 km for the bottom and top level, respectively. The simulation was run from 1 July to 31 October 2006 (a 4-month period) including a high fire episode in Sumatra occurring in October. The temporal resolution of the simulation was 90 s.

The domain was initialized by the National Centers for Environmental Prediction FiNaL reanalysis (NCEP-FNL) data for the meteorological variables (NCEP-FNL, 2000) and by the MOZART4-NCEP model output for the chemical gases and primary aerosols initialization (Emmons et al., 2010). The boundaries of the domain were also forced by the NCEP-FNL and MOZART4-NCEP re-analyses model outputs which were called for input every 6 h. The WRF-Chem configuration used the volatility basis set (VBS) scheme for aerosol chemistry (Ahmadov et al., 2012), the MADE (modal aerosol dynamics model for europe) module for the aerosol dynamic processes and the RACM (regional atmospheric chemistry modeling) (Stockwell et al., 1997) reaction scheme for the gaseous chemistry reactions. The aerosol particle population was described by three modes (Aitken, accumulation and coarse), each of them following a lognormal distribution. Each aerosol mode was composed of primary particles (primary organic carbon, black carbon, dust and sea salt) and secondary particles (sulfate, nitrate, ammonium, 4 classes of anthropogenic secondary organic aerosol, 4 classes of biogenic secondary aerosols and resulting water). Dust, sea salt and biogenic particles showed concentration values lower than 1 % of the total aerosol concentrations and are therefore not discussed in the rest of this study, but were included in the model runs. The simulation included anthropogenic, biogenic and biomass burning emissions prepared by the PREP-CHEM-SRC software tool (Freitas et al., 2011).

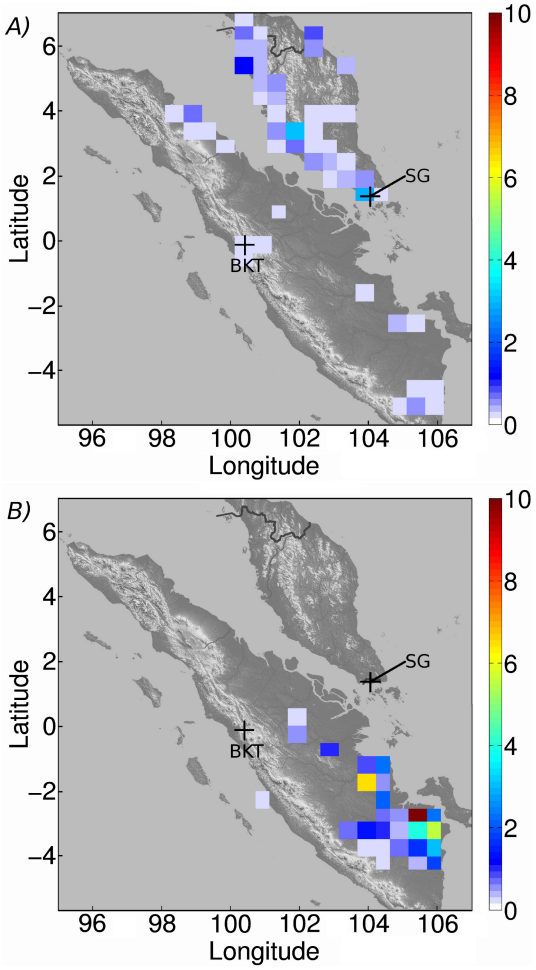

Figure 1. Monthly averaged emissions in $\mu g\,m^{-2}\,s^{-1}$ of primary organic carbon from anthropogenic (top) and biomass burning (bottom) sources for October 2006. SG is short for Singapore and BKT indicates the CO measurement station Bukit Kototabang.

The anthropogenic emissions were derived from the EDGARv4 (EDGAR, 2009) and RETRO (Pulles et al., 2005) inventories. The biogenic emissions were computed by the MEGAN model v2.1 (Guenther et al., 2012). The daily biomass burning emissions were taken from Global Fire Emissions Database (GFED3) (van der Werf et al., 2010; Mu et al., 2011) and the emission factors for the volatile organic compounds (VOCs) as well as for the primary aerosol particles are deduced from Akagi et al. (2011). As mentioned by Hyer et al. (2013), the methodology used in GFED3 based on burned area may fail to detect relatively small fires such as those burning in agricultural areas (Randerson et al., 2012). On the other hand, GFED3 burned area in deforestation regions received a boost based on fire persistence partly to account for missing these fires (van der Werf et al., 2010). Preliminary GFED4 emissions estimates, which are corrected for small fires 2.1 and subsequently do not receive the boost anymore, indate that these two factors were of similar mag-

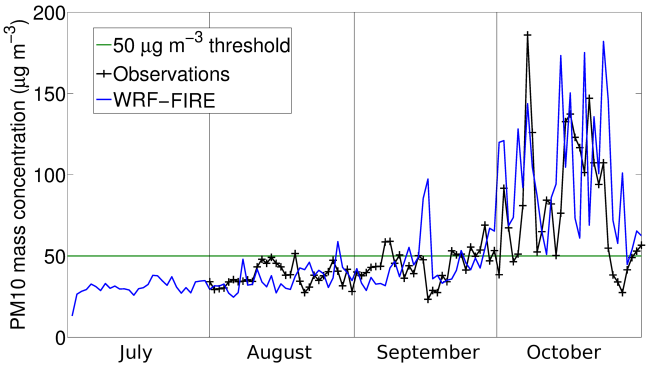

Figure 2. A total of 24 h averaged aerosol mass concentration observed (in black crosses) and modeled (in blue line) over Singapore for our study period. The $50\,\mu g\,m^{-3}$ indicates the WHO definition of polluted air.

Table 1. Comparison of aerosols emission factors (in gram per kilogram of dry matter) used in GFED3 and from Akagi et al. (2011) as used in the simulations. The relative differences in percentage are given in parenthesis.

	OCp	BC	PM_{10}
GFED	4.49	0.55	5.04
Akagi et al. (2011)	6.23 (+38.6 %)	0.20 (−165 %)	6.43 (+27.7 %)

nitude. Table 1 shows the emission factors of aerosol species as used in the GFED3 database and deduced from the newer emission factor compilation from Akagi et al. (2011) as used in our simulations. Table 1 shows that for the 4 months of interest, the aerosol particle emissions from biomass burning used for the simulation are 27.7 % higher than in the GFED3 database. Emissions of primary organic carbon from anthropogenic sources and biomass burning sources are shown in Fig. 1.

2.2 Comparison with observations

We compared the model outputs with observations to gain confidence in our model setup. The observations used include ground-based measurements (PM_{10} and carbon monoxide, CO) as well as a set of various satellite sensors measuring AOD. The PM_{10} observations were the averaged values of five urban ambient air quality stations located in different parts of Singapore and monitored by Singapore's National Environment Agency. The CO observations were located on the Bukit Kototabang site on the island of Sumatra; see Fig. 1 (Zellweger et al., 2007). The AOD observations at 550 nm used in this study are 2-week averaged observations of a $1° \times 1°$ area centered over Singapore from the Moderate Resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging SpectroRadiometer (MISR), Ozone Monitoring Instrument (OMI) and Sea-viewing Wide Field-of-view Sensor (SeaWiFS) sensors.

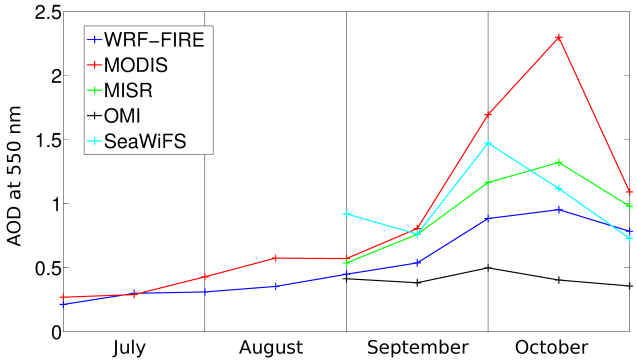

Figure 3. WRF-Chem modeled aerosol optical depth (AOD) and AOD observed by MODIS, MISR, OMI and SeaWiFS. AOD values are averaged over a $1° \times 1°$ area centered over Singapore.

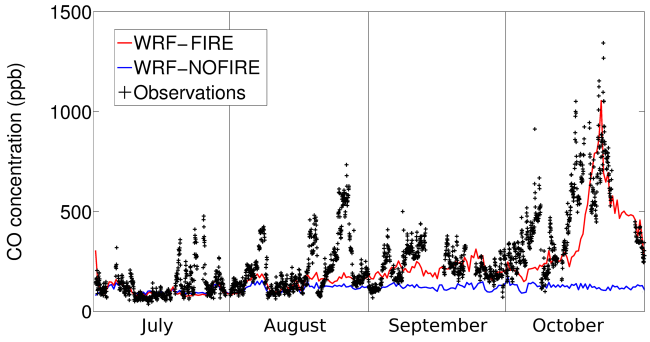

Figure 4. CO concentration observed (in black crosses) and modeled with WRF-Chem either with fires (red) or without (blue) over Bukit Kototabang for our study period.

The model results indicated that there were three distinct time periods with regard to aerosol concentrations in Singapore (Fig. 2). The first period lasted from July to the end of September, and the 24 h averaged aerosol concentrations in Singapore were relatively low and almost never exceeded the value of 50 $\mu g\,m^{-3}$ for PM_{10}, indicated by the World Health Organization (WHO, 2006) as the threshold for classifying the ambient air quality as polluted. The averaged value for this period was 35 $\mu g\,m^{-3}$ representing urban background concentrations in Singapore. During this time period, only small fires occurred in Sumatra and the wind regime did not advect the resulting plumes in the direction of Singapore. During the second period, from the end of September until the middle of October, the aerosol concentrations (PM_{10}) were high (values reaching 160 $\mu g\,m^{-3}$) and were coupled with relatively steady southeasterly winds with a surface mean velocity values of 7 $m\,s^{-1}$. The third period ran from the middle of October until the end of October, and the aerosol concentrations remained high (values reaching 160 $\mu g\,m^{-3}$). The wind regime over Singapore showed relatively low velocities (4 $m\,s^{-1}$) and directions varied between day and night, indicating that the main wind component in Singapore during this period was the thermal wind regime between land and sea. Fires also occurred in Sumatra during this latter period, but the wind regime did not advect the resulting plumes to Singapore according to our model.

In Fig. 2, the 24 h modeled average values of aerosol mass concentrations in Singapore at ground level and the 50 $\mu g\,m^{-3}$ threshold as used by the WHO to define polluted air are shown. The modeled results agreed reasonably well with surface observations. Figure 2 shows that the WRF-Chem model managed to reproduce the evolution of the aerosol mass concentration in Singapore both for background aerosol concentrations and during the haze period, characterized by elevated aerosol concentrations occurring in October. The correlation coefficient (R) between field observations and model results for the whole period was 0.62.

Besides the local aerosol concentration at ground level, we also compared our modeled results to aerosol optical depth (AOD) as measured by various satellite instruments. Figure 3 shows 2-weekly average AOD modeled at the wavelength of 550 nm as observed by different satellite sensors. The data are shown as 2-week moving averages, this was done to present a consistent comparison of the various sensor measurements and minimize the error and noise due to the different overpass times from the various satellites and as a result of cloud contaminated pixels. For the first period (July–September) with low values, the different observations and model results were in relatively good agreement with AOD. However, Fig. 3 also shows that the modeled AOD was low during the month of October compared to observations from MODIS, MISR and SeaWiFS. The quantitative disagreement varies between the different sensors but is largest when compared to MODIS observations with up to a factor of 2.5 in the middle of October. In addition, Reid et al. (2013) showed that AOD measurements in this region are often underestimated by up to 50 % which would deteriorate the comparison even further. There is, however, agreement on the temporal trend in aerosol concentrations with most of the observations. The discrepancy with measured AOD can probably be explained by an elevated aerosol layer observed over Singapore as described by Campbell et al. (2013) and Chew et al. (2013). This pollution layer appears to come from outside the domain and is represented in the boundary conditions entering the domain from the east. After entering the domain, the model located this advected pollution layer south of Singapore and therefore it is not represented in the simulated AOD over Singapore. Due to the height of the transported pollution layer (2500 m) it does not affect our results which focus on the lower atmosphere. Another explanation of the model AOD underestimation may be contamination of the observed AOD due to tropical cirrus and opaque clouds as described by several studies (Huang et al., 2011, 2012; Chew et al., 2011).

In addition to these comparisons with aerosol observations, we compared our results with one station in Sumatra with continuous carbon monoxide (CO) observations (Zellweger et al., 2007). The CO is measured by a TEI48C TL instrument installed in 2001 and the data set can be accessed through the Global Atmosphere Watch network (http://gaw.empa.ch/). Figure 4 shows the evolution of the CO concentrations during our 4-month study period at the Bukit Kototabang station (BKT, see Fig. 1). The model results are drawn in blue and red lines and indicate the simulations excluding biomass burning emissions (referred to as WRF-NOFIRE later on this document) and including biomass burning emissions (referred to as WRF-FIRE), respectively. The model managed to correctly represent the background concentrations as well as the high level of CO concentrations (up to 1300 ppb) in October due to biomass burning, inducing that both model transport and CO emissions from the GFED3 database are correctly represented in this study. One can also note, however, that several smaller fire episodes were not well captured by either WRF or GFED3, especially in August.

3 Aerosol plume analyses: composition and distribution

The comparison of model outputs with observations shows that the WRF-Chem model setup is capable of representing quite accurately the evolution of the total aerosol mass concentrations for the 4 months of simulation. While the PM_{10} comparison indicated the model was able to reproduce the measurements, we cannot conclusively state that the model managed to reproduce the aerosol chemical composition because no measurement information on the exact aerosol composition was available. However, given our efforts to accurately take into consideration the partitioning of emissions (including various VOCs) as well as the use of one of the most accurate aerosol–chemistry reaction schemes available at the present time (VBS scheme), the good match between the total aerosol mass concentrations modeled and observed yields some confidence in these results. We now turn our focus on the composition of aerosol particles at the biomass burning emission location, along the plume, and in Singapore. Figure 5 shows the horizontal cross section of primary aerosol mass concentration on the left and SOA mass concentration on the right, at the surface level on 3 October 2006 at 12:00 LT (local time). Although being a snapshot, it is a representative one involving the interaction between remotely emitted biomass burning aerosols and freshly emitted urban aerosols in Singapore.

Figure 5 illustrates that primary aerosols were highly concentrated over emission sources and reached values of 350 $\mu g\,m^{-3}$ at the main biomass burning location (marked as point A) and 180 $\mu g\,m^{-3}$ in Singapore (marked as point B). Those high concentrations of primary aerosols thus rapidly

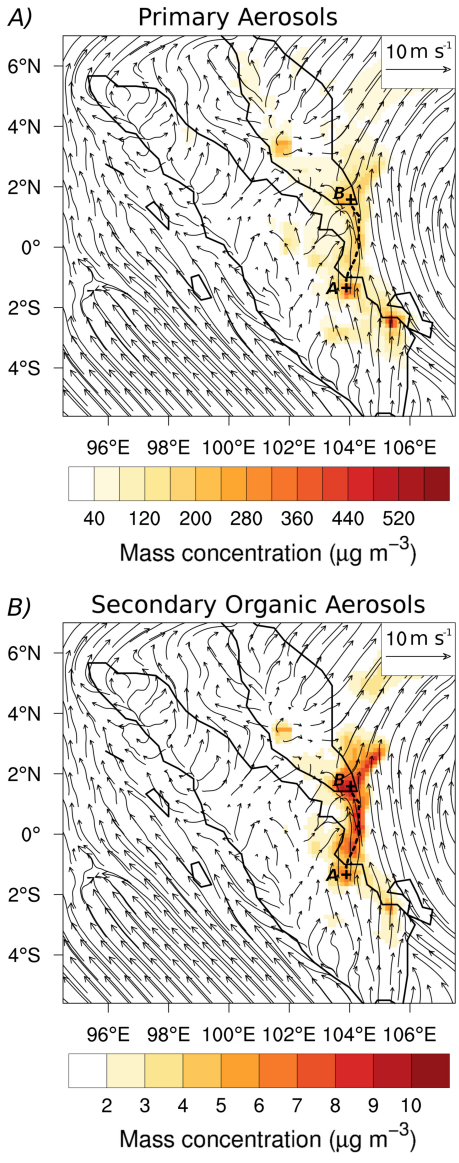

Figure 5. Primary aerosol (secondary organic aerosol) mass concentrations on the top (bottom) with values higher than 10 (1) $\mu g\,m^{-3}$ at the surface level on 3 October 2006 at 12:00 LT. The wind speed vectors are overlaid in black arrows.

decreased away from the emission sources. On the other hand, SOA reached high concentrations a few kilometers away from the emissions sources. While the amplitude and the variability were much lower than for the primary aerosols (from 1 to 10 $\mu g\,m^{-3}$ compared with 20 to 600 $\mu g\,m^{-3}$), Fig. 5b shows that SOA were formed remotely from the biomass burning emissions along the plume and were mixed with freshly formed secondary organic aerosols from fast chemical reactions in Singapore.

Not only does the aerosol concentration change rapidly along the plume, its chemical composition also shows substantial fluctuations, as seen in Fig. 6. The figure shows a

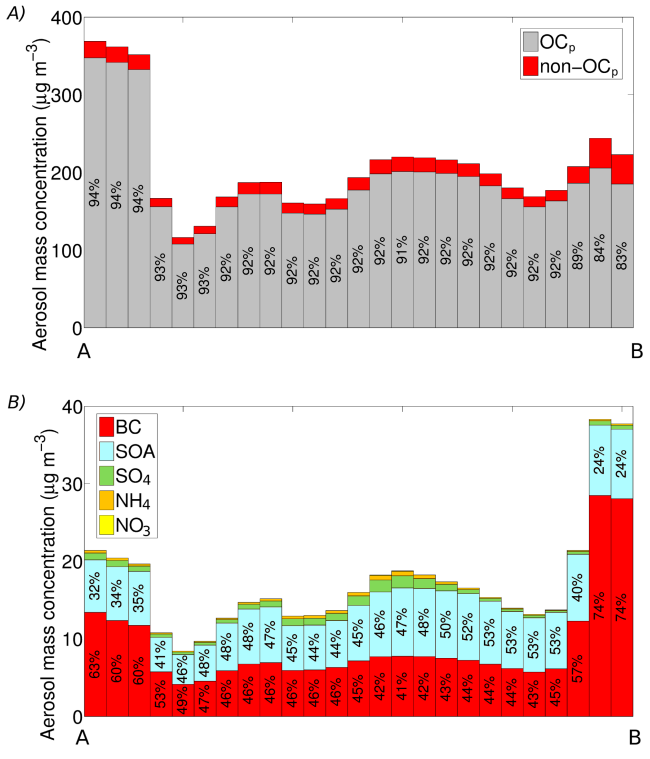

Figure 6. Aerosol mass composition evolution from point A to point B in Fig. 5 with **(a)** showing the primary organic carbon versus non-primary organic carbon speciation, and **(b)** details the non-primary organic aerosol composition. Each bar is spaced by 15 km along the transect drawn in Fig. 5. The numbers in the bars indicate the relative contribution.

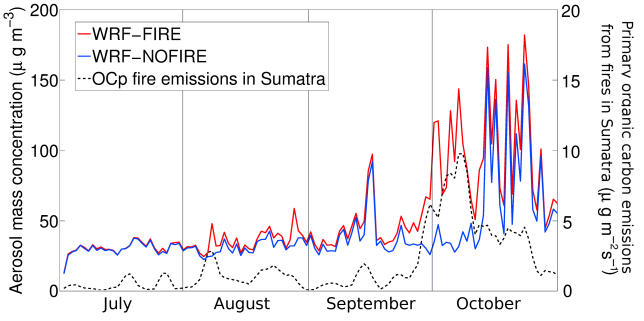

Figure 7. Aerosol mass concentrations from the simulations WRF-FIRE (red line) and WRF-NOFIRE (blue line) in Singapore for our study period in 2006. The fire emissions of primary organic carbon aerosols in Sumatra are drawn as a dashed black line.

transect from the source in Sumatra marked by A in Fig. 5 to the city of Singapore marked with a B. The main message from Fig. 6 is that the total aerosol population, in the A to B transect, was largely dominated by primary organic aerosols (POA) representing 83 to 95 % of the total aerosol mass concentration. The contribution of POA varied along the plume with the highest values at the biomass burning location. It sharply decreased about 75 km away from the biomass burning location, but then slightly increased again along the plume. This initial decrease was due to deposition of bigger particles close to the fire location while the smallest ones kept being advected toward the Malaysian peninsula. The contribution of POA to total aerosol concentration was relatively stable along the plume around 92 %, but dropped to 83 % close to the city of Singapore largely due to increased non-POA concentrations. While the percentage of POA may appear high compared to other recent studies (See et al., 2006, 2007), it remains consistent for this intense fire episode with the emission ratios reported in Akagi et al. (2011). Moreover, the results in this study show a significantly lower SOA / POA ratio in the plume than the ratio reported by several studies mainly focused over North America (Vakkari et al., 2014; Yokelson et al., 2009; Akagi et al., 2012; Cubison et al., 2011; Hennigan et al., 2011). This difference may be related to the high density of fire emissions leading to very large emissions of both primary particles and precursory gases responsible for the formation of secondary organic aerosols. However, the formation of secondary organic aerosols is a strongly non-linear process which depends on numerous and complex processes (such are the VOC concentrations, ozone concentrations, NO_x concentrations, water vapor, aerosol internal mixing rate, etc.) (Seinfeld and Pandis, 1997; Ng et al., 2007). Therefore, its formation can quickly reach its saturation mixing ratio or a threshold due to a limiting factor, while the primary particles are linearly emitted as a function of the burned fuel. In our case we believe that the partitioning between the vapor and aerosol phase has quickly reached a saturation point due to the NO_x and ozone conditions. So even though the VOC concentrations needed for the formation of SOA were still high other factors became limiting, suggesting that the SOA / POA ratio varies between different fire types and intensities. It should be mentioned though that due to the complexity involved in the chemical reactions, almost every numerical model tends to underestimate the secondary aerosol formation (Seinfeld and Pandis, 1997).

The non-POA aerosol concentration (represented in Fig. 6b) shows first relatively high values at the biomass burning location dominated for 63 % by black carbon (BC). The non-POA fraction sharply decreased away from the source to about half its original value. The absolute concentration of non-POA aerosol mass concentration increased slightly along the plume due to an increase of the SOA formation. Finally in Singapore, the local anthropogenic emissions of BC dominated the non-POA aerosol concentrations while the SOA concentration remained stable. The differences in the contribution of primary aerosols between BC and POA in the biomass burning location and in Singapore were due to the difference in the emission factors for peat fires and combustion (EPA, 2010; Akagi et al., 2011).

Table 2. Comparison of speciated averaged aerosol mass concentrations in $\mu g\,m^{-3}$ over Singapore for the FIRE simulations (columns 2 and 4) and the NOFIRE simulations (columns 3 and 5) for the 4-month period (columns 2–3) and for the high fire period (columns 4–5). The relative differences between the two runs are given in columns 3 and 5 in parentheses.

	4-month period (Jul–Oct)		28 Sep–13 Oct	
	WRF-FIRE	WRF-NOFIRE	WRF-FIRE	WRF-NOFIRE
Total aerosol	53.3	42.1 (−21 %)	97.4	50.5 (−48 %)
Black carbon	10.7	10.1 (−6 %)	14.1	12.3 (−13 %)
Organic carbon	40.7	30.0 (−26 %)	81.0	36.4 (−54 %)
Secondary organic carbon	1.5	1.4 (−7 %)	3.3	2.0 (−39 %)
Inorganic aerosols	0.4	0.4 (0 %)	0.6	0.4 (−67 %)

4 Relative and absolute contribution of aerosols from biomass burning to pollution level in Singapore

In order to identify and quantify the impact of biomass burning on aerosol pollution levels in Singapore, we ran two different simulations to isolate the impact of fires on the region. The first one included the biomass burning emissions and is referred to as WRF-FIRE. The second one only included anthropogenic and biogenic emissions and is referred to as WRF-NOFIRE.

The results for both simulations with regard to aerosol mass concentration in Singapore are shown in Fig. 7. From July to the end of September the two simulations varied marginally. From early October until the middle of October large fires in Sumatra induced big differences between the two simulations in Singapore. The maximum difference was found on 10 October with values of 40 and 140 $\mu g\,m^{-3}$ for the WRF-NOFIRE and WRF-FIRE, respectively. Somewhat surprisingly, the second half of the month of October shows high values of aerosol concentrations but no major differences between the two simulations. During this period, 12.7 % of the aerosol concentration was coming from outside the domain and was probably due to advected fire plumes emitted in southern Kalimantan as shown by Engling et al. (2014) and Wang et al. (2013), and 14.4 % was due to fires occurring in Sumatra. Thus, the model indicated that during the second half of October, 73 % of the aerosol concentration was due to anthropogenic emissions within the domain. The high aerosol concentration during this period can be explained by the fact that from 13 October to the end of the simulation the wind regime showed quite low intensities and a recirculation of the wind pattern, resulting in an accumulation of anthropogenic pollution over Singapore. Although the results from this modeling study showed a relatively good match with observations and indicate that the high aerosol concentrations for the second half of October 2006 are dominated by local pollution, it should be noted that other studies attribute this high pollution levels to biomass burning occurring in southern Kalimantan (Engling et al., 2014; Wang et al., 2013). While it appears that transport from Kalimantan to Singapore occurred, our model indicated that the emitted particles were not in the lower layers of the atmosphere when reaching Singapore but were mostly concentrated at higher altitude (between 3 and 5 km above ground level). HYbrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) simulation (see Supplement) shows the ground concentrations (0–100 m a.g.l.) evolution from an idealized emitted mass at the location of the fires in Kalimantan. This simulation supports the notion that the emitted particles from fires in Kalimantan did not reach the surface in Singapore.

To characterize the aerosol pollution levels in Singapore, we compared the aerosol composition for the two simulations and calculated the number of days for which the 24 h averaged aerosol mass concentration was above the threshold of 50 $\mu g\,m^{-3}$, also for both simulations.

Table 2 shows the average mass concentrations for total aerosol, POA, black carbon, SOA and inorganic particles for the two simulations. Those values are presented both for the total 4-month study period and for the 2-week period when Singapore was affected most by biomass burning. The relative difference (as a percentage) between the WRF-FIRE and WRF-NOFIRE simulations is also reported for each aerosol component. For the 4 months of simulation, 21 % of the total aerosol particles in Singapore were due to fires in Sumatra. This increase of particles from biomass burning is largely dominated by primary organic carbon. On the other hand black carbon, inorganics and SOA concentrations in Singapore showed less than 7 % increase due to fires in Sumatra.

Focusing on the 28 September–13 October period during which fires in Sumatra had the highest impact on Singapore, Table 2 shows that almost half of the total aerosol particles in Singapore were due to fires. Again, this pollution was highly dominated by primary organic carbon particles (54 %). SOA showed low absolute concentrations but the relative increase due to fires was substantial (39 % increase).

Finally, the number of days when 24 h averages of aerosol mass concentration in Singapore was above the threshold of 50 $\mu g\,m^{-3}$ shows that while observations indicated 37 days with such values, WRF-FIRE and WRF-NOFIRE showed 35 and 17 days, respectively. These results indicate once more the importance of biomass burning in affecting local and re-

gional air quality. However, they also highlight the importance of properly accounting for regional meteorology.

In the past, GFED estimates have been found too low to properly model AOD (e.g., Petrenko et al., 2012; Marlier et al., 2013). Our results initially support this notion; while we boosted aerosol emissions by 28 % by applying new emission factors we still underestimated AOD. However, we were able to reproduce ground-level concentrations of PM_{10} and CO. It is important to note here that our increase of 28 % is substantially lower than Petrenko et al. (2012) who showed an underestimation up to 300 % of biomass burning aerosol emissions in Indonesia, or in Marlier et al. (2013) who increased the aerosol emissions from fires with 226 %. Clearly, if we had boosted our emissions that much we had overestimated the ground observations to a large degree. In our study region, coarse scale inverse model setups constrained by AOD would probably boost fire emissions to account for lower than observed AOD, while in reality the discrepancy in AOD may also be related to other factors including grid cell size and the use of simplified aerosol chemistry modules in models which may have difficulty calculating all optical properties correctly. Although just a case study, our results highlight the complexity of the various processes involved in the evolution of the regional and long-range transported aerosol particles and indicate that more work is needed to reconcile the differences in emissions strength required to match AOD versus ground observations.

5 Conclusions

We used the atmospheric model WRF-Chem with VBS configuration to simulate the aerosol evolution during 4 months over Sumatra and Singapore. The main objectives were to estimate, simulate and analyze the aerosol particle emission and evolution due to biomass burning in Sumatra. We focused on the year 2006, the highest fire year in the last decade in the region. The comparison with observations of PM_{10} and CO showed that the WRF-Chem model managed to reproduce quite accurately the surface concentrations. However, we underestimated AOD possibly related to regionally transported elevated particle layers misrepresented in the simulation, or tropical cirrus clouds affecting the AOD measurements. This mismatch is of concern and was also found in other studies. However, here we focused on air quality for which matching surface observations is more relevant than matching column concentrations For this simulation, we used new emission factors which were 28 % above those used in GFED3. This increase is much smaller than suggested by several other studies, yet it resulted in a good match with surface observations.

The analysis of the biomass burning plume composition mixing with the freshly emitted urban aerosol population in Singapore highlighted the very high concentrations of primary organic carbon with maximum values of 600 $\mu g\,m^{-3}$ at the fire source. SOA were formed within the plume but with much lower values of up to 20 $\mu g\,m^{-3}$. Black carbon concentrations were highest in Singapore where combustion processes from anthropogenic sources such as traffic with high black carbon emission factors are dominating. The analysis of the differences between two simulations, including and omitting fire emissions, allowed us to isolate and quantify the impact of biomass burning on aerosol pollution levels in Singapore. We showed that 21 % of the total aerosol concentration was due to biomass burning occurring in Sumatra during the 4-month period of the simulation, and 48 % when focusing on a 2-week period in October when smoke reaching Singapore was most intense. This contribution of fires resulted in 18 days when the 50 $\mu g\,m^{-3}$ threshold was exceeded, in addition to 17 days due to a mixture of mainly local anthropogenic pollution and smaller contributions from fires in Sumatra and probably Kalimantan.

Accurate quantification of the contribution from biomass burning to particulate pollution levels in highly populated cities such as Singapore, Kuala Lumpur and Jakarta may help to develop strategies to either control the amount and timing of biomass and peat burning depending on the meteorology and the urban pollution levels, or apply more effective urban air pollution reduction plans when fire plumes significantly impact the air pollution levels in populated areas.

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