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# Importance of transboundary transport of biomass burning emissions to regional air quality in Southeast Asia

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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 the largest fire-induced haze episode in the past decade (2006) in Indonesia using the Weather Research and

- <sup>5</sup> Forecasting model coupled with Chemistry (WRF-Chem). We focused mainly 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 CO concentrations. Two simulations were run with the model using the complex Volatility Basis Set (VBS) scheme to reproduce primary and secondary aerosol evolution and
- <sup>10</sup> 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 datasets for comparison including airborne measurements of Particulate Matter
- with a diameter of 10 μm or less (PM<sub>10</sub>) in Singapore, CO measurements in Sumatra, and Aerosol Optical Depth (AOD) column observations from 4 satellite-based sensors. We found reasonable agreement of the model runs with both ground-based measurements of CO and PM<sub>10</sub>. The comparison with AOD was less favorable and indicated the model underestimated AOD, although the degree of mismatch varied between dif-
- ferent 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 µgm<sup>-3</sup> over the fire locations. The concentration of POA remained quite stable within the plume between the main burning
- <sup>25</sup> region and Singapore while secondary organic aerosol (SOA) concentration slightly increased. The absolute values of SOA (up to 20 µgm<sup>-3</sup>) were much lower than those from POA, indicating a minor role of SOA in biomass burning plumes. Our results show that about 21 % of the total mass loading of ambient PM<sub>10</sub> 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 μgm<sup>-3</sup> day<sup>-1</sup> 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 Borneo added to the enhanced PM<sub>10</sub> concentrations. The model vs. 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 using 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 air quality.

#### 15 **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 not well understood nor quantified.

SEA, especially Indonesia, has the highest concentration of fire emissions globally due to the intensive burning in areas with high fuel loads including peatlands (Page et al., 2002; van der Werf et al., 2010). 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 hot spots (intense fires) mainly in Sumatra and Borneo and regularly show high particulate pollution levels which may be impacted by emissions from forest, agriculture, and peat fires (Hyer and Chew, 2010; Salinas et al., 2013a, b; Wang et al., 2013). Models that accurately simulate
<sup>5</sup> 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). De-<sup>10</sup> pending 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 precursors concentrations emitted by biomass
- <sup>20</sup> 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 set-up and evaluation

# 2.1 Model set-up

<sup>25</sup> 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 as well as the microphysics and the atmospheric chemistry and aerosol processes.

The simulation was done for a domain with 100 by 100 grid points, each with a 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 2006 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 Fi-NaL reanalysis 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

- the NCEP-FNL and MOZART4-NCEP re-analyses model outputs which were called for input every 6 h. The WRF-Chem configuration used the 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 chem-
- istry reactions. The aerosol particle population was described by 3 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 tool (Freitas et al., 2011).





The anthropogenic emissions were derived from the EDGARv4 (EDGAR, 2009) and RETRO (Pulles et al., 2005) inventory. The biogenic emissions were computed by the MEGAN model v2.1 (Guenther et al., 2012). The daily biomass burning emissions were taken from GFED3 (van der Werf et al., 2010; Mu et al., 2011) and the emission factors for the volatile organic compounds as well as for the primary aerosol particles are

- for the volatile organic compounds as well as for the primary aerosol particles are deduced from Akagi et al. (2011). Table 1 shows the emission factors of aerosol species as used in the GFED3 database and deduced from the newer emission factors from Akagi et al. (2011) as used in the simulations. Table 1 shows that for the 4 months of interest, the aerosol particle emissions from biomass burning as input for the simulation
- <sup>10</sup> 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 set-up. The model results indicate that there were 3 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<sub>10</sub>, 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  $35 \,\mu g \,m^{-3}$  represent-

- ing 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<sub>10</sub>) were high (values reaching 160 µgm<sup>-3</sup>) and were coupled with relatively steady Southeasterly winds with a surface mean velocity values of 7 m s<sup>-1</sup>. 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<sup>-1</sup>) 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 period, but the wind regime did not advect the resulting plumes to Singapore.

In Fig. 2, the 24 h modeled average values of aerosol mass concentrations in Sin gapore at ground level and the 50 μgm<sup>-3</sup> threshold as used by the WHO to define polluted air are shown. The modeled results agreed reasonably well with surface observations. The observations are the averaged values of 5 ambient air quality stations located in different part of Singapore measuring PM<sub>10</sub> mass concentrations. Figure 2 shows that the WRF-Chem model manages to reproduce the evolution of the aerosol mass concentration in Singapore both for background aerosol concentrations and during the smoke haze period, characterized by elevated aerosol concentration 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. This comparison was done for a 1° × 1° area centered over Singapore. Figure 3 shows 2 weekly average AOD modelled at the wavelength of 550 nm and observed by different satellite sensors. 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 modelled AOD was low during the month of October compared to observations from the Moderate Resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging SpectroRadiometer (MISR) and Sea viewing Wide Field of view Sensor (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. There is, however, agreement on the temporal trend in aerosol concentrations with most of observations. The discrepancy of AOD can probably be explained by an elevated aerosol layer observed over Singapore as described by Campbell et al. (2013); 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 locates this advected pollution layer south of Singapore and therefore is not represented in the simulated AOD over Singapore. Due to the height of the tranported pollution layer (2500 m) it does not affect our results which focus on the lower atmophere. Another explanation of

the model AOD underestimation may be a contamination of the observed AOD due to tropical cirrus cloud and opaque cloud as described by several studies (Huang et al., 2012, 2011; 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). 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 standing for simulation 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 man-
- ages to correctly represent the background concentrations as well as the high level of CO concentrations (up to 1300 ppb) 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 are not well captured by either WRF or GFED3.

# 20 3 Aerosol plume analyses: composition and distribution

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The comparison of model outputs with observations shows that the WRF-Chem model set-up is capable of representing quite accurately the evolution of the aerosol concentrations for the 4 months of simulation. This part of the study focuses 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 noon 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 μgm<sup>-3</sup> at biomass burning location (marked as point A) and 180 μgm<sup>-3</sup> in Singapore (marked as point B). Those high concentration of primary aerosols rapidly decreased away from the emission sources. On the other hand, SOA reached high concentrations few kilometers away from the emissions sources. While the amplitude of the variability is much lower than for the primary aerosols (from 1 to 10 μgm<sup>-3</sup> compared with 20 to 600 μgm<sup>-3</sup>), Fig. 5b shows that
SOA are formed remotely from the biomass burning emissions along the plume and are mixed with freshly formed secondary organic aerosols from fast chemical reactions in Singapore. The modelled SOA concentrations appear relatively low, but are based on the VBS module which includes a semi-volatility approach to compute the SOA concentrations and has been tested to be one of the most accurate and computation-

<sup>15</sup> ally efficient scheme at present (Matsui et al., 2014; Ahmadov et al., 2012). In addition, these results appear consistent with recent studies confirming that the net SOA to POA ratio for biomass burning emissions is far lower than for urban emissions (Ortega et al., 2013).

Not only does the aerosol concentration change rapidly along the plume, its chemical composition also shows substantial fluctuations, as seen Fig. 6. The figure shows a 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, is largely dominated by primary organic carbon (OC<sub>p</sub>) representing 83 to 95 % of the total aerosol mass concentration. This high contribution of OC<sub>p</sub>

<sup>25</sup> varies along the plume with highest values at the biomass burning location. This  $OC_p^p$  concentration sharply decreases about 75 km away from the biomass burning location, but then slightly increases along the plume. This initial decrease is due to a fluctuation of the wind regime leading to an alternating pattern between steady winds from south and the associated plume along the ocean, and low wind regime associated with



accumulation of particles as for the local maximum in Sumatra. The contribution of  $OC_p$  to total aerosol concentration is relatively stable along the plume around 92%, but drops to 83% close to the city of Singapore largely due to increased non- $OC_p$  concentrations. While the percentage of  $OC_p$  may appear high in regard 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).

The non-OC<sub>p</sub> aerosols (represented in Fig. 6b) show first relatively high values at the biomass burning location and is dominated for 63 % by black carbon (BC). The non-OC<sub>p</sub> fraction then sharply decreases by a factor of 2. The absolute values of non-OC<sub>p</sub> aerosol mass concentration increases slightly along the plume due to an increase of the secondary organic aerosol formation. Finally in Singapore, the local anthropogenic emissions of black carbon dominate the non-OC<sub>p</sub> aerosol concentrations while the SOA concentration remains stable. The differences in the contribution of primary aerosols between BC and OC<sub>p</sub> in the biomass burning location and in Sin-

(EPA, 2010; Akagi et al., 2011)

# 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 level 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 includes 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 vary 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 concentrations

- <sup>5</sup> tration was coming from outside the domain and was probably due to advected fire plumes emitted in southern Borneo as shown by Engling et al. (2014) and Wang et al. (2013), and 14.4% was due to fires occuring in Sumatra. Thus, the model indicates that during the second half of October, 73% of the aerosol concentration was due to anthropogenic emissions occuring within the domain. This 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 modelling study show 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
- <sup>15</sup> be noted that other studies attribute this high pollution levels to biomass burning occuring in southern Borneo (Engling et al., 2014; Wang et al., 2013). In order to study the aerosol pollution levels in Singapore, aerosol composition for the two simulations is compared as well as the number of day for which the 24 h averaged aerosol mass concentration was above the threshold of  $50 \,\mu g \,m^{-3}$ .
- 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 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 are 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 show 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 is highly dominated by primary organic carbon particles (54%). SOA showed low absolute concentrations but the relative increase due to fires is substantial (39% increase).

Finally, the number of days when 24 h averages of aerosol mass concentration in Singapore were above the threshold of  $50 \,\mu g m^{-3}$  shows that while observations indicates 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 regional air quality. However, they also highlight the importance of

- affecting local and regional 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 support this notion because the aerosol estimates we used were 27.7 % higher than standard GFED3 emissions due to applying new emission factors. How-
- ever, this increase is substantially lower that in Petrenko et al. (2012) where they show an underestimation up to 300 % of biomass burning aerosol emissions in Indonesia, or in Marlier et al. (2013) where they increase the aerosol emissions from fires by a scaling factor up to 226 %. In our study region, coarse scale inverse model setups would probably boost fire emissions to account for lower than observed AOD, while in reality
- the discrepancy in AOD may come from various other causes than an underestimation of the biomass burning aerosol emissions, such as a too rough representation of the aerosol size and chemistry leading to wrong optical properties. 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.

#### 25 5 Conclusions

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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  $PM_{10}$ , AOD and CO showed that the WRF-Chem model managed to reproduce quite accurately the aerosol

- concentrations in Singapore. This agreement confirms that relatively high spatial resolution is required when studying regional air quality cases. However we underestimated AOD probably due to regionally transported elevated particle layer misrepresented in the simulation, or tropical cirrus clouds affecting the AOD measurements. Nevertheless AOD is not a variable affecting directly air quality. 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 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 µgm<sup>-3</sup> at the fire source. SOA were formed within the plume but with much lower values of up to 20 µgm<sup>-3</sup>. Black carbon concentrations were highest in Singapore where combustion processes from anthropogenic sources such as traffic are dominating with high black carbon emission factors. The analysis of the differences between two simulations, including and omit-

ting fire emissions, allowed us to isolate and quantify the impact of biomass burning to 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 µgm<sup>-3</sup> 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 Borneo. This study has revealed the impact of the high contribution of

biomass burning smoke in Indonesia on aerosol pollution levels in Singapore.





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 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 impact the air pollution levels in populated areas significantly.

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### References

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- Abba, E. J., Unnikrishnan, S., Kumar, R., Yeole, B., and Chowdhury, Z.: Fine aerosol and PAH carcinogenicity estimation in outdoor environment of Mumbai City, India, Int. J. Environ. Heal. R., 22, 134–149, doi:10.1080/09603123.2011.613112, 2012. 11224
- Ahmadov, R., McKeen, S. A., Robinson, A., Bahreini, R., Middlebrook, A., de Gouw, J., Meagher, J., Hsie, E., Edgerton, E., Shaw, S., and Trainer, M.: A volatility basis set model for summertime secondary organic aerosols over the eastern United States in 2006, J. Geo-
- phys. Res., 117, D06301, doi:10.1029/2011JD016831, 2012. 11225, 11229
   Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, doi:10.5194/acp-11-4039-2011, 2011. 11226, 11230
- <sup>25</sup> Campbell, J. R., Reid, J. S., Westphal, D. L., Zhang, J., Tackett, J. L., Chew, B. N., Welton, E. J., Shimizu, A., Sugimoto, N., Aoki, K., and Winker, D. M.: Characterizing the vertical profile of aerosol particle extinction and linear depolarization over Southeast Asia and the Maritime Continent: the 2007-2009 view from CALIOP, Atmos. Res., 122, 520–543, 2013. 11227





Chew, B. N., Campbell, J. R., Reid, J. S., Giles, D. M., Welton, E. J., Salinas, S. V., and Liew, S. C.: Tropical cirrus cloud contamination in sun photometer data, Atmos. Environ., 45, 6724–6731, 2011. 11228

Chew, B. N., Campbell, J. R., Salinas, S. V., Chang, C. W., Reid, J. S., Welton, E. J., Hol-

- ben, B. N., and Liew, S. C.: Aerosol particle vertical distributions and optical properties over Singapore, Atmos. Environ., 79, 599–613, doi:10.1016/j.atmosenv.2013.06.026, 2013. 11227
  - Crutzen, P. J. and Andreae, M. O.: Biomass burning in the tropics: impact on atmospheric chemistry and biogeochemical cycles, Science, 250, 1669–1678, doi:10.1126/science.250.4988.1669. 1990. 11223
- EDGAR: Emission Database for Global Atmospheric Research (EDGAR), release version 4.0, available at: http://edgar.jrc.ec.europa.eu (last access: 1 May 2014), 2009. 11226 Emmons, L. K., Walters, S., Hess, P. G., Lamargue, J.-F., Pfister, G. G., Fillmore, D., Granier, C.,

- Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C.,
- Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010. 11225
  - Engling, G., He, J., Betha, R., and Balasubramanian, R.: Assessing the regional impact of Indonesian biomass burning emissions based on organic molecular tracers and chemical
- <sup>20</sup> mass balance modeling, Atmos. Chem. Phys. Discuss., 14, 2773–2798, doi:10.5194/acpd-14-2773-2014, 2014. 11231
  - EPA: Report to Congress on Black Carbon, Tech. rep., Department of the Interior Environment and Related Agencies, availavle at: http://www.epa.gov/blackcarbon/2012report/fullreport. pdf (last access: 1 May 2014), 2010. 11230
- Field, R. D., van der Werf, G. R., and Shen, S. S. P.: Human amplification of drought-induced biomass burning in Indonesia since 1960, Nat. Geosci., 2, 185–188, doi:10.1038/NGEO443, 2009. 11223
  - Freitas, S. R., Longo, K. M., Alonso, M. F., Pirre, M., Marecal, V., Grell, G., Stockler, R., Mello, R. F., and Sánchez Gácita, M.: PREP-CHEM-SRC 1.0: a preprocessor of trace gas
- and aerosol emission fields for regional and global atmospheric chemistry models, Geosci. Model Dev., 4, 419–433, doi:10.5194/gmd-4-419-2011, 2011. 11225
  - Grell, G., Peckham, S., Schmitz, R., McKeen, S., Frost, G., Skamarock, W., and Eder, B.: Fully coupled "online" chemistry in the WRF model, Atmos. Environ., 39, 6957–6976, 2005. 11224





- Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012. 11226
- <sup>5</sup> He, J., Zielinska, B., and Balasubramanian, R.: Composition of semi-volatile organic compounds in the urban atmosphere of Singapore: influence of biomass burning, Atmos. Chem. Phys., 10, 11401–11413, doi:10.5194/acp-10-11401-2010, 2010. 11224
  - Hong, C.-C., Lu, M.-M., and Kanamitsu, M.: Temporal and spatial characteristics of positive and negative Indian Ocean dipole with and without ENSO, J. Geophys. Res., 113, D08107, doi:10.1029/2007JD009151. 2008. 11223
- Huang, J., Hsu, N. C., Tsay, S., Jeong, M., Holben, B. N., Berkoff, T. A., and Welton, E. J.: Susceptibility of aerosol optical thickness retrievals to thin cirrus contamination during the BASE-ASIA campaign, J. Geophys. Res., 116, D08214, doi:10.1029/2010JD014910, 2011. 11228

- <sup>15</sup> Huang, J., Hsu, N. C., Tsay, S., Holben, B. N., Welton, E. J., Smirnov, A., Jeong, M.-J., Hansell, R. A., Berkoff, T. A., Liu, Z., Liu, G.-R., Campbell, J. R., Liew, S. C., and Barnes, J. E.: Evaluations of cirrus contamination and screening in ground aerosol observations using collocated lidar systems, J. Geophys. Res., 117, D15204, doi:10.1029/2012JD017757, 2012. 11228
- Hyer, E. J. and Chew, B. N.: Aerosol transport model evaluation of an extreme smoke episode in Southeast Asia, Atmos. Environ., 44, 1422–1427, doi:10.1016/j.atmosenv.2010.01.043, 2010. 11224
  - Johnston, F. H., Henderson, B., Chen, Y., Randerson, J. T., Marlier, M., DeFries, R. S., Kinney, P., Bowman, D. M. J. S., and Brauer, M.: Estimated global mortality attributable to smoke
- <sup>25</sup> from landscape fires, Environ. Health Persp., 120, 695–701, doi:10.1289/ehp.1104422, 2012. 11223
  - Karthikeyan, S., Balasubramanianab, R., and Iouri, K.: Particulate air pollution from bushfires: human exposure and possible health effects, J. Toxicol. Env. Heal. A, 69, 1895–1908, doi:10.1080/15287390600751264, 2006. 11224
- 30 Keywood, M. D., Ayers, G. P., Gras, J. L., Boers, C. P., and Leong: Haze in the Klang Valley of Malaysia, Atmos. Chem. Phys., 3, 591–605, doi:10.5194/acp-3-591-2003, 2003. 11224
  - Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aar-





denne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010. 11223

Marlier, M. E., DeFries, S. R., Voulgarakis, A., Kinney, P. L., Randerson, J. T., Shindell, D. T., Chen, Y., and Faluvegi, G.: El Nino and health risks from landscape fire emissions in southeast Asia, Nature Climate Change, 3, 131–136, doi:10.1038/nclimate1658, 2013. 11223, 11232

Matsui, H., Koike, M., Kondo, Y., Takami, A., Fast, J. D., Kanaya, Y., and Takigawa, M.: Volatility

basis-set approach simulation of organic aerosol formation in East Asia: implications for anthropogenic-biogenic interaction and controllable amounts, Atmos. Chem. Phys. Discuss., 14, 6203–6260, doi:10.5194/acpd-14-6203-2014, 2014. 11229

Mauderly, J. L. and Chow, J. C.: Health effects of organic aerosols, Inhal. Toxicol., 20, 257–288, doi:10.1080/08958370701866008, 2008. 11224

- <sup>15</sup> Mu, M., Randerson, J. T., van der Werf, G. R., Giglio, L., Kasibhatla, P., Morton, D., Collatz, G. J., DeFries, R. S., Hyer, E. J., Prins, E. M., Griffith, D. W. T., Wunch, D., Toon, G. C., Sherlock, V., and Wennberg, P. O.: Daily and 3-hourly variability in global fire emissions and consequences for atmospheric model predictions of carbon monoxide, J. Geophys. Res., 116, D24304, doi:10.1029/2011JD016245, 2011. 11226
- NCEP-FNL: NCEP FNL Operational Model Global Tropospheric Analyses, continuing from July 1999. Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems, available at: http://rda.ucar.edu/datasets/ds083.2/ (last access: 1 May 2014), 2000. 11225

Ortega, A. M., Day, D. A., Cubison, M. J., Brune, W. H., Bon, D., de Gouw, J. A., and

- Jimenez, J. L.: Secondary organic aerosol formation and primary organic aerosol oxidation from biomass burning smoke in a flow reactor during FLAME-3, Atmos. Chem. Phys. Discuss., 13, 13799–13851, doi:10.5194/acpd-13-13799-2013, 2013. 11229
  - Page, S. E., Siegert, F., Rieley, J. O., Boehm, H. V., Jaya, A., and Limin, S.: The amount of carbon released from peat and forest fires in Indonesia during 1997, Nature, 420, 61–65, doi:10.1038/nature01131, 2002. 11223
  - Pavagadhi, S., Betha, R., Venkatesan, S., Balasubramanian, R., and Hande, M. P.: Physicochemical and toxicological characteristics of urban aerosols during a recent Indonesian





biomass burning episode, Environ. Sci. Pollut. R., 20, 2569–2578, doi:10.1007/s11356-012-1157-9, 2013. 11224

- Petrenko, M., Kahn, R., Chin, M., Soja, A., Kucsera, T., and Harshvardhan, N.: The use of satellite-measured aerosol optical depth to constrain biomass burning emis-
- sions source strength in the global model GOCART, J. Geophys. Res., 117, D1821, doi:10.1029/2012JD017870, 2012. 11232
  - Pulles, T., Brand, R., van het Bolscher, M., Sørgard, Ø., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A., Berglen, T. F., Gravir, G., and Schultz, M.: RETRO Emission Inventory: anthropogenic emission database, available at: ftp://ftp.retro.enes.org (last access: 1 May 2014), 2005. 11226
  - Reid, J. S., Hyer, E. J., Johnson, R. S., Holben, B. N., Yokelson, R. J., Zhang, J., Campbell, J. R., Christopher, S. A., Girolamo, L. D., Giglio, L., Holz, R. E., Kearney, C., Miettinen, J., Reid, E. A., Turk, F. J., Wang, J., Xian, P., Zhao, G., Balasubramanian, R., Chew, B. N., Janjai, S., Lagrosas, N., Lestari, P., Lin, N.-H., Mahmud, M., Nguyen, A. X., Norris, B.,

10

<sup>15</sup> Oanh, N. T. K., Oo, M., Salinas, S. V., Welton, E. J., and Liew, S. C.: Observing and understanding the Southeast Asian aerosol system by remote sensing: an initial review and analysis for the Seven Southeast Asian Studies (7SEAS) program, Atmos. Res., 122, 403– 468, 2013. 11223

Salinas, S. V., Chew, B. N., Miettinen, J., Campbell, J. R., Welton, E. J., Reid, J. S., Yu, L. E.,

- and Liew, S. C.: Physical and optical characteristics of the October 2010 haze event over Singapore: a photometric and lidar analysis, Atmos. Res., 122, 555–570, 2013a. 11224 Salinas, S. V., Chew, B. N., Mohamad, M., Mahmud, M., and Liew, S. C.: First measurements of aerosol optical depth and Angstrom exponent number from AERONET's Kuching site, Atmos. Environ., 78, 231–241, doi:10.1016/j.atmosenv.2013.02.016, 2013b. 11224
- See, S. W., Balasubramanian, R., and Wang, W.: A study of the physical, chemical, and optical properties of ambient aerosol particles in Southeast Asia during hazy and nonhazy days, J. Geophys. Res., 111, D10S08, doi:10.1029/2005JD006180, 2006. 11224, 11230
  - See, S. W., Balasubramanian, R., Rianawati, E., Karthikeyan, S., and Streets, D. G.: Characterization and source apportionment of particulate matter < or = 2.5 micrometer in Suma-
- tra, Indonesia, during a recent peat fire episode, Environ. Sci. Technol., 41, 3488–3494, doi:10.1021/es061943k, 2007. 11224, 11230
  - Stockwell, W., Kirchner, F., Kuhn, M., and Seefeld, S.: A new mechanism for regional atmospheric chemistry modeling, J. Geophys. Res., 102, 847–825, 879, 1997. 11225





- van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010. 11223, 11226
- <sup>5</sup> Wang, J., Ge, C., Yang, Z., Hyer, E. J., Reid, J. S., Chew, B. N., Mahmud, M., Zhang, Y., and Zhang, M.: Mesoscale modeling of smoke transport over the Southeast Asian Maritime Continent: interplay of sea breeze, trade wind, typhoon, and topography, Atmos. Res., 122, 486–503, doi:10.1016/j.atmosres.2012.05.009, 2013. 11224, 11231
- WHO: World Health Organization Air Quality Guidelines for Particulate Matter, Ozone,
   Nitrogen Dioxide and Sulfur Dioxide, Tech. rep., World Health Organization, available at: http://whqlibdoc.who.int/hq/2006/WHO\_SDE\_PHE\_OEH\_06.02\_eng.pdf (last access: 1 May 2014), 2006. 11223, 11226
  - Yee, L. D., Kautzman, K. E., Loza, C. L., Schilling, K. A., Coggon, M. M., Chhabra, P. S., Chan, M. N., Chan, A. W. H., Hersey, S. P., Crounse, J. D., Wennberg, P. O., Flagan, R. C.,
- and Seinfeld, J. H.: Secondary organic aerosol formation from biomass burning intermediates: phenol and methoxyphenols, Atmos. Chem. Phys., 13, 8019–8043, doi:10.5194/acp-13-8019-2013, 2013. 11224
  - Zellweger, C., Klausen, J., and Buchmann, B.: System and performance audit of surface ozone and carbon monoxide at the global GAW station Bukit Koto Tabang Indonesia, February
- 20 2007, Tech. rep., WMO World Calibration Centre for Surface Ozone, Carbon Monoxide and Methane, Empa Dübendorf, Switzerland, 2007. 11228



Table 1. Comparison of aerosols emission factors (in g per kg dry matter) from GFED 3 and
from Akagi et al. (2011) as used in the simulations. The relative differences in percentage are
given in parenthesis.

	OCp	BC	PM <sub>10</sub>
GFED	4.49	0.55	5.04
Akagi et al. (2011)	6.23 (+38.6%)	0.20 (-165%)	6.43 (+27.7%)

<b>ACPD</b> 14, 11221–11248, 2014				
Relative contribution from fire emissions to regional air quality in Southeast Asia				
B. Aouizerats et al.				
Title Page				
Abstract	Introduction			
Conclusions	References			
Tables	Figures			
I	۶I			
•				
Back	Close			
Full Screen / Esc				
Printer-friendly Version				
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Table 2. Comparison of speciated averaged aerosol mass concentrations 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 1 month October 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%)



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**Fig. 2.** 24 h averaged aerosol mass concentration observed (in black crosses) and modelled (in blue line) over Singapore for the year 2006. The  $50 \,\mu g m^{-3}$  indicates the WHO definition of polluted air.



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**Fig. 3.** WRF-Chem modelled Aerosol Optical Depth (AOD) and AOD observed by MODIS, MISR, OMI and SEAWIFS. AOD values are averaged over the  $1^{\circ} \times 1^{\circ}$  area centered over Singapore for the year 2006.



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**Fig. 4.** CO concentration observed (in black crosses) and modelled with WRF-Chem either with fires (red) or without (blue) over Bukit Kototabang for the year 2006.





**Fig. 5. (A)** Primary aerosol mass concentrations with values higher than  $10 \mu gm^{-3}$  and **(B)** secondary organic aerosol mass concentrations with values higher than  $1 \mu gm^{-3}$  at the surface level on 3 October 2006 at 12:00 LT. The wind speed vectors are overlaid in white arrows.







**Fig. 6.** Aerosol mass composition evolution from point A to point B in Fig. 5 with **(A)** showing the primary organic carbon vs. non primary organic carbon speciation, and **(B)** details the non primary organic aerosol composition.







**Fig. 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 also drawn in dashed black line.

