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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 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 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 $10\ \mu\text{m}$ or less (PM_{10}) 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_{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 $600\ \mu\text{g m}^{-3}$ over the fire locations. The concentration of POA remained quite stable within the plume between the main burning region and Singapore while secondary organic aerosol (SOA) concentration slightly increased. The absolute values of SOA (up to $20\ \mu\text{g m}^{-3}$) 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_{10} during the July–October

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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 Fv3L2 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 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 chemistry 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).

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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 Singapore at ground level and the $50\mu\text{g m}^{-3}$ 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_{10} 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 $1^\circ \times 1^\circ$ 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

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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 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 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 manages to correctly represent the background concentrations as well as the high level of CO concentrations (up to 1300 ppb) due to biomass burning, indicating 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.

3 Aerosol plume analyses: composition and distribution

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 $350 \mu\text{g m}^{-3}$ at biomass burning location (marked as point A) and $180 \mu\text{g m}^{-3}$ 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 $10 \mu\text{g m}^{-3}$ compared with 20 to $600 \mu\text{g m}^{-3}$), 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 computationally 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_p) representing 83 to 95% of the total aerosol mass concentration. This high contribution of OC_p varies along the plume with highest values at the biomass burning location. This OC_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

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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_p 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_p fraction then sharply decreases by a factor of 2. The absolute values of non- OC_p 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_p aerosol concentrations while the SOA concentration remains stable. The differences in the contribution of primary aerosols between BC and OC_p in the biomass burning location and in Singapore are due to the difference in the emission factors for peat fires and combustion. (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

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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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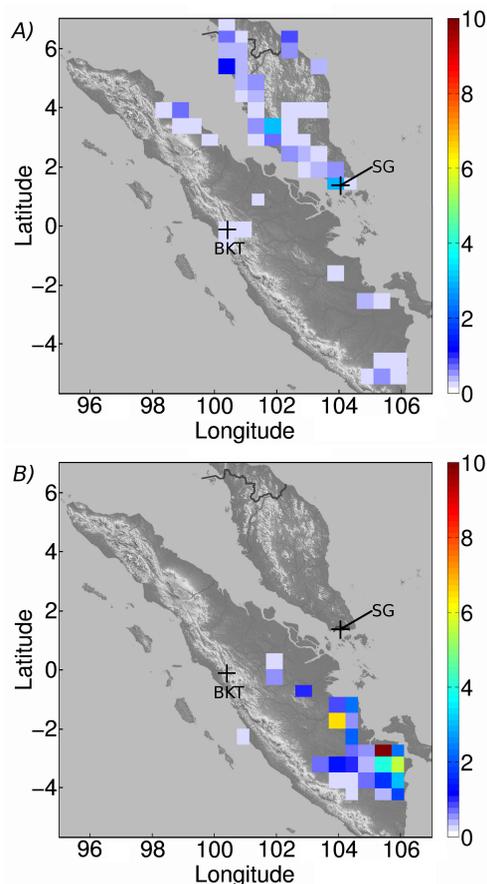


Fig. 1. Monthly averaged emissions in $\mu\text{g m}^{-2} \text{s}^{-1}$ of primary organic carbon from anthropogenic **(A)** and biomass burning **(B)** sources for October 2006. Singapore is marked with a black cross and the symbol SG and the CO measurement station Bukit Kototabang with the symbol BKT.

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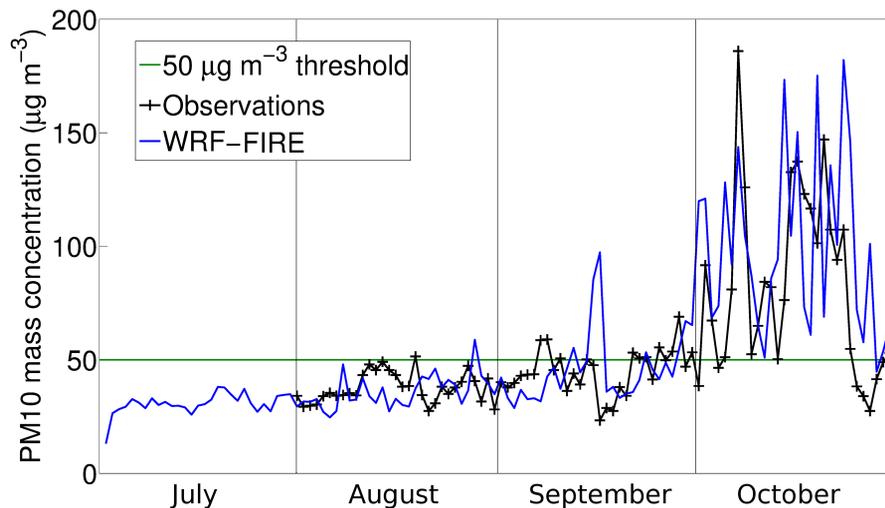


Fig. 2. 24 h averaged aerosol mass concentration observed (in black crosses) and modelled (in blue line) over Singapore for the year 2006. The $50 \mu\text{g m}^{-3}$ indicates the WHO definition of polluted air.

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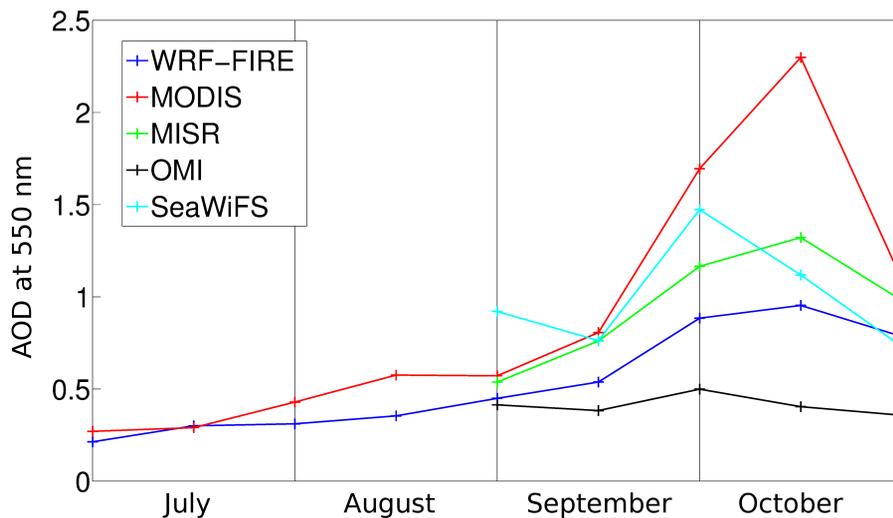


Fig. 3. WRF-Chem modelled Aerosol Optical Depth (AOD) and AOD observed by MODIS, MISR, OMI and SEAWIFS. AOD values are averaged over the $1^\circ \times 1^\circ$ area centered over Singapore for the year 2006.

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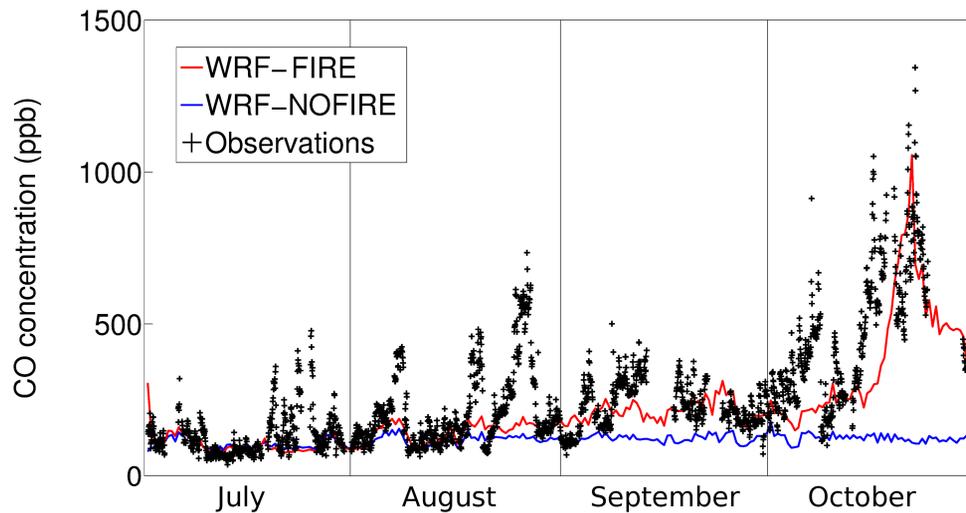


Fig. 4. CO concentration observed (in black crosses) and modelled with WRF-Chem either with fires (red) or without (blue) over Bukit Kototabang for the year 2006.

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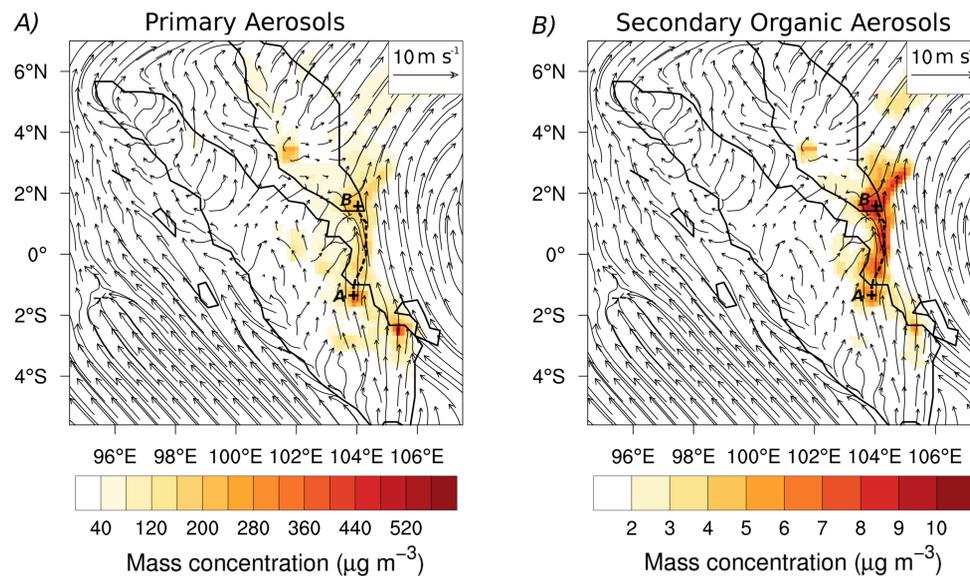


Fig. 5. (A) Primary aerosol mass concentrations with values higher than $10 \mu\text{g m}^{-3}$ and (B) secondary organic aerosol mass concentrations with values higher than $1 \mu\text{g m}^{-3}$ at the surface level on 3 October 2006 at 12:00 LT. The wind speed vectors are overlaid in white arrows.

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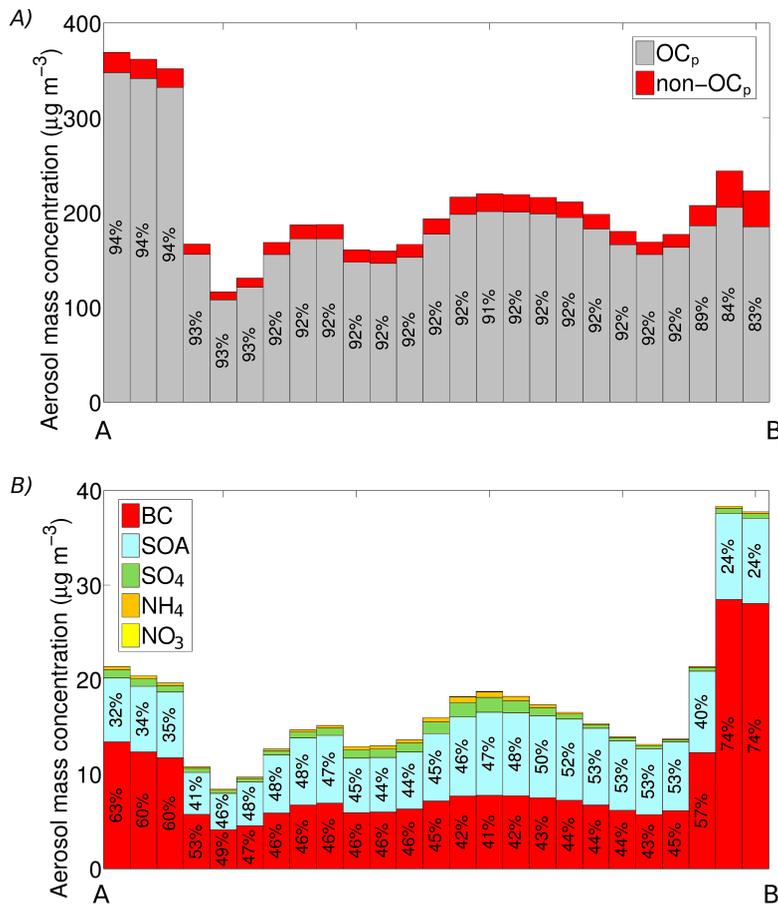


Fig. 6. Aerosol mass composition evolution from point A to point B in Fig. 5 with **(A)** showing the primary organic carbon vs. non primary organic carbon speciation, and **(B)** details the non primary organic aerosol composition.

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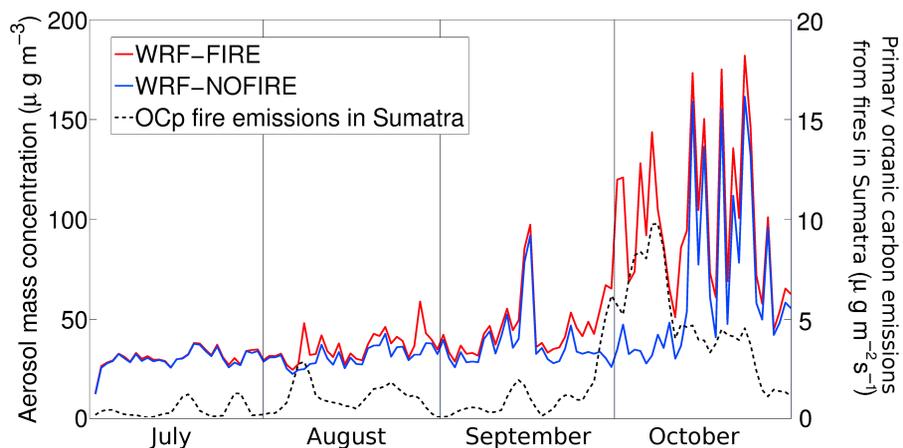


Fig. 7. Aerosol mass concentrations from the simulations WRF-FIRE (red line) and WRF-NOFIRE (blue line) in Singapore for our study period in 2006. The fire emissions of primary organic carbon aerosols in Sumatra are also drawn in dashed black line.

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