Fast secondary aerosol formation in southern African biomass burning smoke

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Biomass burning (BB) is one of the largest sources of reactive trace gases and fine particles in the Earth's atmosphere, significantly affecting the global radiation balance (e.g. Bond *et al*, 2013). However, there is large uncertainty in the climate effect of the BB emissions, mostly because of the reactive species co-emitted with black carbon (BC) particles (Bond *et al*, 2013).

We investigated the temporal evolution of BB smoke in southern Africa utilising continuous measurements at Welgegund, South Africa (26.57°S, 26.94°E, 1480 m above sea level) from 20 May 2010 to 31 December 2014. Of the measurements performed at Welgegund (welgegund.org), we used aerosol particle number size distribution, carbon monoxide (CO), BC with a MAAP, local wind and solar radiation. Submicron particulate mass (PM_1) was estimated from the number size distribution using a constant density of 1.5 g cm^{-3}.

Using the methodology of Vakkari *et al* (2014) we could identify 118 BB plumes over the nearly five year period. For 83 plumes the location of the fire could be identified from MODIS burnt area and/or SEVIRI fire radiative power observations. For these episodes – a total of 250 hours of in-plume sampling – we estimated the plume age based on local wind speed and direction (c.f. Vakkari *et al*, 2014). For each plume the excess CO, BC and PM_1 (denoted with a Δ) were calculated by subtracting respective background concentrations, with ΔCO, ΔBC and ΔPM_1 given as μg m^{-3} at STP.

In order to estimate the ratio of flaming to smouldering combustion, we calculated the ΔBC to ΔCO ratio for each 10-minute ΔPM_1 sample: low ΔBC/ΔCO indicates smouldering combustion and high ΔBC/ΔCO designates flaming combustion. According to plume transport time we could also estimate the time each 10-minute sample had been transported in daylight, i.e. when global radiation > 100 W m^{-2} at Welgegund. Combination of these parameters allowed us to investigate the effects of burning conditions and atmospheric transport on PM_1 mass in BB plumes, both in daylight and in the dark.

Smouldering BB smoke (Figure 1a) exhibits fast secondary aerosol formation in daylight: up to a factor of 4 increase in $\Delta PM_1/\Delta CO$ in less than 5 hours. Previously, aged BB plumes at Welgegund have been shown to be dominated by organic aerosol (OA) (Vakkari *et al*, 2014), suggesting that the increase in $\Delta PM_1/\Delta CO$ is mostly due to formation of secondary organic aerosol (SOA). As samples with higher and higher ΔBC/ΔCO are considered, the increase in $\Delta PM_1/\Delta CO$ becomes slower and even decrease for the most flaming cases (Figure 1b).

In the dark both increase and decrease rates in $\Delta PM_1/\Delta CO$ are an order of magnitude slower than in daylight, suggesting that the rapid daytime changes are driven by photochemical reactions: SOA formation in smouldering BB smoke and fragmentation of primary OA in flaming BB smoke.

To conclude, we have shown that during daytime secondary aerosol formation can increase the PM_1 mass in BB smoke by up to a factor of 4 in a few hours, but the increase depends critically on whether the fire was flaming or smouldering.

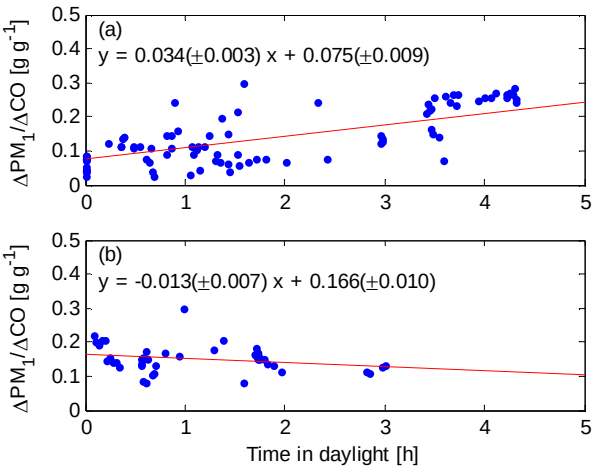

Figure 1. Daytime $\Delta PM_1/\Delta CO$ time evolution for (a) smouldering cases, ΔBC/ΔCO < 0.01 g g^{-1} and (b) flaming cases, ΔBC/ΔCO > 0.03 g g^{-1}. A bivariate linear fit with standard error is included in both (a) and (b).

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