Biomass burning at Cape Grim: using modeling to explore a possible urban influence on plume photochemistry and composition

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Robbins Island fire gives unique opportunity for plume characterisation



(trace gas emission ratios to CO calculated)

Wind speed ~60km hour: plume from Robbins Island takes ~20-30 min to reach Cape Grim



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Measurements – P2P campaign



| Measurement | Instrument | | | | |
|--|--|--|--|--|--|
| Particle size distribution (14 – 700 nm) | TSI SMPS | | | | |
| Particle number >10 nm | TSI 3010 CN Counter | | | | |
| Particle number > 3 nm | TSI 3025a UCN Counter | | | | |
| Black Carbon | Aethalometer | | | | |
| Cloud Condensation Nuclei (CCN) | DMT CCN counter | | | | |
| VOCs (10 minute) | Proton Transfer Reaction –Mass Spectrometer | | | | |
| Ozone | TECO ozone analyser | | | | |
| CH_4 | AGAGE GC FID | | | | |
| CO, H ₂ | AGAGE GC-MRD | | | | |
| CO ₂ | LoFIo NDIR | | | | |
| N ₂ O CHCl ₃ , CH ₃ CCl ₃ , CCl ₄ | AGAGE GC-ECD system | | | | |
| Ethane, methyl halides | AGAGE Medusa GCMS | | | | |
| | | | | | |
| No aerosol chemical composition measurements except for BC | | | | | |

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Different composition of fresh, aged plumes





CCN / CN>80nm (%)

What is driving these changes in chemcial composition?
Fire emissions and/or emissions from Melbourne/mainland?



Can modelling help determine the contribution of different sources to photochemistry, plume age?

- TAPM-CTM (meteorology)
- 20km, 12km, 3km, 1km, 400m domains
- Carbon Bond 5, includes inorganic aerosol, EC, SOA
- Emissions from mainland,Tasmania (incl. fire), ocean (determine contribution from different sources)
- Fire scar used to estimate area burned
- We modified
 - boundary concentrations, diurnal emissions and plume rise
- We explored affect of
 - Changing trace gas and aerosol fire emission factors, corresponding to modified combustion efficiency (MCE) of 0.89 (lower), 0.92 (best estimate), 0.95 (upper).
 - spatial variability (1km each side of Cape Grim)





Black carbon – modelled concentrations sensitive to emission factors



Modelled ozone – sensitive to emission factors



Mainland and fire emission are driving ozone production (MCE 0.89)



What are relative contributions to ozone production?





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What is driving enhanced CCN ratio in aged plume?



Enhanced SOA and inorganics in aged plume likely responsible for increased CCN activity (mainly from mainland emissions)

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What is driving particle growth event?



Mainland urban emissions and fire emissions make equal contribution to particle growth event

.....(size resolved number and composition GLOMAP model runs to come)



Conclusions



- Chemical transport model outputs are very sensitive to emission factors
- Modelling suggests that both the fire and urban emissions make a contribution to the formation of secondary pollutants and aerosol chemical composition
- Model predicts that the aged plumes are ~ 2 days old
- GLObal Model of Aerosol Processes (GLOMAP) will provide more specific information about composition of growing particles, CCN



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- All sources RI = mainland influence
- All sources mainland = fire influence
- This gives contribution of each source above background







GLOMAP GLObal Model of Aerosol Processes





Developed in Leeds since 2003 to simulate global aerosol with size-resolved number and composition. Resolves processes that grow aerosol from nm to CCN sizes and beyond

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Ozone and oxygenated VOCs enhanced in aged plume





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| Species | This study | Temperate south eastern Australia | | Tropical Savar | Temperate Northern Hemisphere | | | |
|--|---|--------------------------------------|-----------------------------|----------------------------|-------------------------------|---------------------|------------------------------------|--|
| | g kg ⁻¹ (calculated using Agaki 2011 CO EF of 89 g kg ⁻¹) | Paton Walsh et al (2005, 2008) | Paton Walsh et al (2010) | Hurst (1994a, 1994b) | Meyer et al (2012) | Shirai et al (2003) | Agaki et al Temperate (2011) | Yokelson et al Semi arid shrubland (2013) |
| Hydrogen (H ₂) | 0.64 | n/a | n/a | n/a | n/a | n/a | 2.03 (1.79) | n/a |
| Methane (CH ₄) | 2.49 | n/a | n/a | 2.26-2.33 | 2.03 (0.13) | 2.20 (0.32) | 3.92 (2.39) | 3.69 (1.36) |
| Ethane (C_2H_6) | 0.30 | 0.26 (0.11) | 0.13 (0.04) | 0.11-0.60 | n/a | 0.53 (0.02) | 1.12 (0.67) | 0.48 (0.61) |
| Hydrogen cyanide (HCN) | 0.49 | 0.43 (0.22) | 0.11 (0.04) | 0.024-0.035 | n/a | n/a | 0.73 (0.19) | 0.75 (0.26) |
| Acetonitrile (CH ₃ CN) | 0.17 | n/a | n/a | 0.11 | n/a | n/a | n/a | 0.15 (0.07) |
| Acetaldehyde (C ₂ H ₄ O) | 0.62 | n/a | n/a | 0.55-1.0 | n/a | n/a | n/a | 0.56 (0.40) |
| Phenol (C ₆ H ₅ OH) | 0.24 | n/a | n/a | n/a | n/a | n/a | 0.33 (0.38) | 0.45 (1.9) |
| Acetic acid (CH ₃ COOH) | 0.52 | n/a | n/a | n/a | n/a | n/a | 1.97 (1.66) | 1.91 (0.94) |
| Methanol (CH ₃ OH) | 1.37 | 2.3 (0.8) | n/a | n/a | n/a | n/a | 1.93 (1.38) | 1.35 (0.4) |
| Benzene (C ₆ H ₆) | 0.47 | n/a | n/a | 0.29 - 0.42 | n/a | 0.21 (0.02) | n/a | 0.45 (0.29) |
| Toluene (C ₇ H ₈) | 0.20 | n/a | n/a | n/a | n/a | n/a | n/a | 0.17 (0.13) |
| Methyl chloride (CH ₃ Cl) | 0.2082 | n/a | n/a | n/a | n/a | 0.0605 (0.0072) | 0.059* | n/a |
| Methyl bromide (CH ₃ Br) | 0.0148 | n/a | n/a | n/a | n/a | 0.0018 (0.0003) | 0.0036* | n/a |
| Methyl iodide(CH ₃ I) | 0.0019 | n/a | n/a | n/a | n/a | 0.0013 (0.0002) | 0.0008* | n/a |



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Ozone enhancement suggests plume age of several days



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Exploring model uncertainty- fire emissions and plume transport



- Systematically explore sensitivity of model to varying inputs
- 1.Influence of different modified combustion efficiencies (MCE) and Emission Factors (EF)?
 - EF corresponding to MCE of 0.89, (lower) 0.92 (best estimate), 0.95 (upper).

2.Spatial influence?

- Investigated modelled concs 1km north, east, south west of Cape Grim
- 3.Influence of different meteorology? (TAPM-CTM & CCAM)





Enhancements of OVOCs



Spatial variability – CO in direct plume (MCE 0.89)



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Ozone formation in background air (MCE 0.89) – significant spatial variability



•Significant spatial variability at higher ozone concentrations (>20 ppb)







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Thank you to Cape Grim Staff for their support during the Precursors to Particles campaign.



MCE and EF have large influence on modelled concentrations



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Particle hygroscopicity changes with aging



Spatial variability – CO in direct plume (MCE 0.89)



Significant spatial variability in CO concs between different modelled locations



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Modified Combustion Efficiencies





No mainland influence

Period 2 Minor mainland influence Period 3 Substantial mainland influence

Air Map Origin Source: UK Met Office (www.metoffice.com)



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Cape Grim Baseline Station, NW Tasmania







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