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Measurements and modelling of emissions from biomass burning in Australia

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Measurements and Modelling of Emissions from Biomass Burning in Australia

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by

Clare Murphy, (née Paton-Walsh), BSc, MSc

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CERTIFICATION

I, Clare Murphy, (née Clare Paton-Walsh) declare that this thesis, submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the Department of Chemistry, University of Wollongong, is wholly my own work unless otherwise referenced or acknowledged. The document has not been submitted for qualifications at any other academic institution.

Clare Murphy

Date:

Abstract

This thesis describes work aimed at improving our knowledge of emissions to the atmosphere from Australian vegetation fires. The thesis contains three main parts. First there is a study to characterise the emissions from forest fires in southeast Australia. This uses ground-based Fourier transform infrared solar absorption spectroscopy, coupled with ultra-violet/visible spectroscopy, to explore the properties of smoke plumes from Australian forest fires that passed over the observation site at Wollongong, in New South Wales, Australia (34.4°S, 150.9°E). The particulate loading in the smoke plumes is characterised by the aerosol optical depth, measured at visible wavelengths. Vertically integrated amounts of a several emitted trace gases are also determined, (limited to those detectable by solar absorption spectroscopy in the infrared). Enhanced trace gas amounts of carbon monoxide, hydrogen cyanide, formaldehyde, ammonia, acetylene, ethylene, ethane, formic acid and methanol were measured in the smoke plumes and quantified via the use of emission ratios. The emission ratios determined in this study indicate that emissions from fires in southeastern Australian forests (which are predominantly eucalypts) are broadly similar to those from other geographical regions except for comparatively low emissions of ethane.

The second part of this thesis describes a new method of making estimates of gaseous emissions from fires. Strong correlations between trace gases and aerosol optical depth (AOD) in smoke plumes are used in conjunction with satellite-based measurements of AOD to estimate the total amounts of carbon monoxide and other gases emitted from the Canberra fires of 2003. There are significant difficulties with the new method, in particular the interruption of the satellite record due to clouds or technical problems with the satellite. Nevertheless the estimated emissions of carbon monoxide from the Canberra fires (4.9 – 9.6 Tg), is in agreement with an estimate made by existing techniques. The addition of another tool for making estimates of gaseous emissions from biomass burning is useful for corroborating existing techniques, especially since the sources of

uncertainties inherent in the different techniques are largely independent of one another.

The third part of the thesis is a study to characterise the emissions from savanna fires in the tropical north of Australia. Again ground-based Fourier transform infrared solar absorption spectroscopy is used with automated measurements in the near infrared from a site in Darwin, Northern Territory, Australia, (12.4°S, 130.9°E). Alternatively measurements in the mid infrared can be made by overriding the automated system, and this has been done when there is evidence of significant smoke plumes in the area. Total column amounts of carbon monoxide from Darwin from 2005-2008 show a very clear annual cycle, with evidence of transported pollution from Indonesian fires in 2006. The time series agrees within the expected uncertainties with measurements of carbon monoxide derived from the MOPITT satellite instrument, giving greater confidence to MOPITT retrievals in the tropics. Mid infrared spectra have been recorded through smoke plumes over Darwin on 20 separate days, yielding column amounts of carbon monoxide, formaldehyde, acetylene, ethane and hydrogen cyanide and emission ratios with respect to carbon monoxide for the four latter gases from tropical north Australian savanna fires. Emission ratios for acetylene and ethane from this work are in broad agreement with other literature values, whilst emission ratios for formaldehyde and hydrogen cyanide are significantly higher than the only previous field measurements from Australian savannas (but in agreement with laboratory studies) suggesting storage losses in the earlier study.

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I thank Arndt Meier for coming into work over Christmas and New Year in 2001/2002 and running the spectrometer at Wollongong to collect the first of the smoke affected spectra upon which this work is based. I also acknowledge Nicholas Jones and all other colleagues with the Centre of Atmospheric Chemistry who have run the spectrometer in Wollongong and captured smoke plumes as a result.

The spectrometer operating at Darwin is designed to operate automatically for the collection of near-infrared data. The system was designed and installed by the efforts of Paul Wennberg, Jean-Francois Blavier, Geoff Toon, Yael Yavin, Rebecca Washenfelder, Gretchen Keppel-Aleks, Nicholas Deutscher, David Griffith, Glenn Bryant, Brian Connor, Charles Miller and Ross Salawitch. The automated collection of near infrared data at Darwin is kept running, despite frequent glitches, by the efforts of Nicholas Deutscher. Nicholas occasionally has also taken data for me in the mid infrared and has been tirelessly patient in providing advice on how to run the system at Darwin. Technical assistance on site in Darwin (including cooling of the detector with liquid nitrogen) has been provided by John Glowacki, Rex Pearson, Troy Culgan, Mike Alsop, Krzysztof Krzton and Maciej Ryczek, many of whom have also alerted me when there were signs of smoke in Darwin. On other occasions the very useful Geosciences Australia "Sentinel" website (http://sentinel.ga.gov.au/acres/sentinel/) was used in conjunction with wind forecasts for the Darwin region from the Bureaux of Meteorology as a crude fire alert system. Aerosol Optical Depth measurements from Darwin were provided by Bruce Forgan.

The TAPM model was developed at CSIRO and purchased by the University of Wollongong for use in this and other projects. Advice was offered for its correct use by Martin Cope, Peter Hurley and Bronwyn Dunse. The programs to derive aerosol optical depth from measurements at Wollongong were written by Stephen Wilson. Some aspects of the uncertainty in retrieving total column amounts using the SFIT2 algorithm were estimated using Matlab scripts written by Vanessa Haverd.

Satellite image courtesy of MODIS Rapid Response Project at NASA/GSFC was used in many of the maps showing the location of fires. Some of the maps produced in the software ArcMap used Spatial Analysis Laboratories products OSDM_GMA2001_coastl and OSDM_GMA2001_builtupp that incorporate data which is Copyright Commonwealth of Australia 2001. Back trajectories were generated by the European Centre for Medium-Range Weather Forecasts, (ECMWF) Trajectories, via the website http://badc.nerc.ac.uk/data/ecmwf-trj/ run by the British Atmospheric Data Centre, 2006-2008.

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List of Acronyms used in this Thesis

AOD = aerosol optical depth

VMR = volume mixing ratio

InSb = indium antimonide

CO = carbon monoxide

H_2CO = formaldehyde

HCN = hydrogen cyanide

NH_3 = ammonia

HCOOH = formic acid

C_2H_2 = acetylene

C_2H_4 = ethylene

C_2H_6 = ethane

CH_3OH = methanol

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