Investigating greenhouse gases in Australia using atmospheric measurements with Fourier transform spectrometry and atmospheric modeling

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INVESTIGATING GREENHOUSE GASES IN AUSTRALIA USING ATMOSPHERIC MEASUREMENTS WITH FOURIER TRANSFORM SPECTROMETRY AND ATMOSPHERIC MODELING

A thesis submitted in fulfilment of the requirements

for the award of the degree

DOCTOR OF PHILOSOPHY

from

UNIVERSITY OF WOLLONGONG

by

Nicholas M. Deutscher, BSc(Hons), BMaths

School of Chemistry

2009
DECLARATION

I, Nicholas M. Deutscher, declare that this thesis, submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the School of Chemistry, University of Wollongong, is wholly my own work unless otherwise referenced or acknowledged below. The document has not been submitted for qualifications at any other academic institution.

Nicholas M. Deutscher

1/12/2009
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The following are publications that have arisen from the work presented here or in work related to this project:

ABSTRACT

Anthropogenic emissions of greenhouse gases CO$_2$, CH$_4$ and N$_2$O are important drivers of changes in radiative forcing and consequent climate changes. Despite their importance, estimates of the source and sink distributions of these gases to and from the atmosphere remain uncertain, largely because more measurements are needed to adequately constrain the problem. Because of the infrared signatures of these gases, Fourier Transform (InfraRed) Spectroscopy (FTS) provides an ideal method for simultaneous measurements.

This thesis investigates the potential of new Australian measurements to constrain estimates of CO$_2$ fluxes in the Australian region. Initially, we use a pseudodata study to determine the utility of adding North-South transect measurements from Darwin to Adelaide on the Ghan train platform, as well as continuous measurements at a number of fixed sites. We see that even in the case of a transect occurring once per month, considerable constraint on the flux estimates is possible for several Australian regions, but that the train measurements fail to provide any detail about the diurnal variability of the fluxes.

The setup and calibration of a high resolution solar absorption FT spectrometer in Darwin is detailed. The instrument is shown to have high precision in measurements of column-average dry-air mole fractions, of the order of 0.1% for $X_{\text{CO}_2}$ within clear days. A calibration factor of 0.990 ± 0.003 is necessary to bring the measurements onto the WMO global standard CO$_2$ scale.

FT spectroscopy is also used in an instrument measuring surface in situ concentrations. The instrument is described, along with a comparison to existing recognised instrumentation at Cape Grim Baseline Air Pollution Station, which shows that it is capable of continuous, high-precision simultaneous measurements of CO$_2$, CH$_4$, N$_2$O, CO and $\delta^{13}$CO$_2$. Time series of measurements made with these instruments at Darwin, Lauder and Cape Grim are examined. We also look at a case study where one of the instruments is deployed on the Ghan railway running from Adelaide to Darwin, and used, along with a modelling study, to infer methane emissions from tropical savannah wetlands.
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<td>Emission ratios calculated from air measured at Cape Grim on January 6, 2009, when a depletion of CH₄ is observed, and back trajectory analyses indicate a westerly ocean source.</td>
</tr>
<tr>
<td>7.4</td>
<td>Fit parameters to Lauder trace gas concentration time series. $Y = a*\sin(2\pi(x+b)) + c<em>x + d + e</em>\sin(4\pi(x+f))$, where $x =$ years since 2005. Values given as fit ± 95% confidence interval.</td>
</tr>
<tr>
<td>7.5</td>
<td>Fit parameters to Darwin trace gas concentration time series. $Y = a*\sin(2\pi(x+b)) + c<em>x + d + e</em>\sin(4\pi(x+f))$. Values given as fit ± 95% confidence interval.</td>
</tr>
<tr>
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</tr>
<tr>
<td>10.2</td>
<td>List of sites included in GLOBALVIEW-2007, along with their latitude, longitude and altitude, whether they are a marine boundary layer site, and the laboratory responsible for the measurements.</td>
</tr>
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