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incorporating the

Cape Grim Annual Science Meeting 2014

Abstracts

12-13 November 2014

hosted by

CSIRO Oceans and Atmosphere Flagship

Priestley Lecture Theatre
Aspendale, Victoria



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Day 1: Wednesday 12 November 2014

09:00	**Convenors**	**Welcome & general information on arrangements etc**
	Helen Cleugh	**Opening of meeting**
Chair	**Paul Krummel**	**Oral Session 1**
09:05	Nicholas Deutscher	Drivers of column-average CO_2 variability at Southern Hemispheric Total Carbon Column Observing Network sites
09:25	Roger Francey	How and when will anthropogenic slowing of atmospheric CO_2 growth become evident?
09:45	Chris Lu	Interannual variability of simulated global land carbon sink and the contribution to inter-hemisphere differences of CO_2 concentration
10:05	Rachel Law	Transport modelling and inversions for the interpretation of greenhouse gas measurements
10:25	Tilo Ziehn	Analysis of ACCESS-ESM1 results: progress and future plans
10:45		Morning Tea provided - Aspendale canteen
Chair	**Melita Keywood**	**Oral Session 2**
11:15	Galbally/Lawson	**The Cape Grim Reactive Gases Program Summary 2008 - 2014**
12:15	Wayne Kirstine	The global distribution of atmospheric acetonitrile away from the influence of biomass burning
12:35	Sarah Lawson	First *in situ* measurements of glyoxal and methylglyoxal over the remote temperate oceans
12:55		Lunch Break provided - Aspendale canteen
Chair	**Paul Fraser**	**Oral Session 3**
14:00	Kathryn Emmerson	MEGAN: Online & Fabulous
14:20	Elise-Andree Guerette	Update on the MUMBA Campaign: Measurements of urban, marine and biogenic air
14:40	Ray Langenfelds	Multi-species interannual variability from GASLAB flask network data
15:00	Jenny Fisher	Seasonal changes in the tropospheric carbon monoxide profile over the remote Southern Hemisphere
15:20		Afternoon Tea provided - Aspendale canteen
Chair	**Sam Cleland**	**Oral Session 4**
15:50	Chulkyu Lee	GAW Activities in Korea
16:10	Marcel van der Schoot	Progress of enhanced atmospheric composition measurement capability at Cape Grim BAPS and the expanding CSIRO atmospheric observation network
16:30	Haeyoung Lee	WCC-SF_6 activities at Anmyeondo station, Korea; Various analytical methods to monitor SF_6 using GC-μECD
16:50	Brad Hall	Recent developments in NOAA calibration activities
17:10	Paul Krummel	Comparisons of *in situ* carbon dioxide and methane measurements at Cape Grim – an update; and an overview of GCWerks software for in situ data processing
18:30	**Dinner**	**McNaughton Room, Level 1, Mordialloc Sporting Club – Mordialloc**

Day 2: Thursday 13 November 2014

Chair	Alastair Williams	Oral Session 5
09:05	Stephen Wilson	Unifying the AOD picture
09:25	Michael Harvey	Surface Ocean Aerosol Production (SOAP)
09:45	Luke Cravigan	Observation of internally mixed volatile and non-volatile organic sea spray from the Chatham Rise, New Zealand
10:05	Clare Paton-Walsh	Emissions of volatile organic compounds and greenhouse gases from fires in the CSIRO pyrotron
10:25	Melita Keywood	Aerosol composition at Cape Grim: the long term record
10:45	Morning Tea	provided - Aspendale canteen
Chair	**Stephen Wilson**	**Oral Session 6**
11:15	Martin Cope	Short-term forecasting of fine particle exposure from prescribed burns and wild fires
11:35	Paul Selleck	Savannah early dry season biomass burning – Campaign overview and a first look at aerosol chemical and microphysical properties from Gunn Point June 2014
12:55	Maximilien Desservettaz	Fourier transform spectrometer results from the Gunn Point campaign, June 2014, including emission factors of greenhouse gases from savanna fires
12:15	Zoran Ristovski	Volatility-separated measurement of aerosol chemical composition and hygroscopicity from the Gunn Point campaign, June 2014
12:35	Holly Winton	Aerosol iron solubility: comparison between the Australian subtropics and Southern Ocean
12:55	Lunch Break	provided - Aspendale canteen
Chair	**Ian Galbally**	**Oral Session 7**
14:00	Alastair Williams	Radon vertical profiles during the morning transition period
14:20	Scott Chambers	A radon-only technique for characterising baseline constituent concentrations at Cape Grim
14:40	David Griffith	Open path Fourier transform spectroscopy measurements of greenhouse gases in Heidelberg
15:00	Christopher Caldow	Water-atmosphere exchange of greenhouse gases (CO2, CH4 and N2O) measured on the Murray River, Australia
15:20	**Afternoon Tea**	**Poster Session** - All poster presenters to give a 2 - 3 minute poster introduction
Maximilien Desservettaz		Open-Path FTIR Measurements of Trace Gas Emissions from Australian Eucalypt Fires in the CSIRO Pyrotron
Jesse Greenslade		Australian Biogenic HCHO Observations from the OMI and GOME-2 Satellites
Ruhi Humphries		New particle formation events over Antarctic sea-ice
Alexander Norton		How productive is Earth's terrestrial biosphere? Novel satellite measurements and methods to understand global primary productivity
Ritu Taneja		Constraining greenhouse gas emissions in the Latrobe Valley
Matthew Woodhouse		The ACCESS-UKCA chemistry-climate model: exploring long-term trends in atmospheric composition
Bin Yang		Conventionally-heated microfurnace for graphitisation of microgram-sized carbon samples
Chair	**Sarah Lawson**	**Oral Session 8**
16:10	David Griffith	Continuous measurements of $\delta^{18}O$-CO_2 in air with a Fourier Transform InfraRed spectrometer
16:30	Blagoj Mitrevski	New gas chromatographic technologies for GASLAB: pulsed discharge helium ionisation detector for analysis of trace gases
16:50	Paul Fraser	New insights into global perfluorocarbon emissions
17:10	Paul Steele	Signatures of human actions recorded in the atmosphere
17:30		Closing remarks

Table of Contents

Day 1: Wednesday 12 November 2014

Drivers of column-average CO_2 variability at Southern Hemispheric Total Carbon Column Observing Network sites..... 1
N. M. Deutscher, V. Sherlock, S. E. Mikaloff Fletcher, D. W. T. Griffith, J. Notholt, R. Macatangay, B. J. Connor, J. Robinson, H. Shiona, V. A. Velazco, Y. Wang, P. O. Wennberg and D. Wunch

How and when will anthropogenic slowing of atmospheric CO_2 growth become evident? 2
R. J. Francey et al.

Interannual variability of simulated global land carbon sink and the contribution to inter-hemisphere differences of CO_2 concentration 3
X. Lu, Y.-P. Wang and R. M. Law

Transport modelling and inversions for the interpretation of greenhouse gas measurements 5
R. M. Law, Z. M. Loh, T. Ziehn, K. D. Haynes, P. B. Krummel, L. P. Steele, P. J. Fraser, S. D. Chambers and A. G. Williams

Analysis of ACCESS-ESM1 results: Progress and future plans..... 6
T. Ziehn, R. M. Law, R. Matear, M. Chamberlain, A. Lenton, Y.-P. Wang and L. E. Stevens

The Cape Grim Reactive Gases Program Summary 2008 - 2014 7
I. E. Galbally and S. J. Lawson

The global distribution of atmospheric acetonitrile away from the influence of biomass burning 9
W. V. Kirstine, I. E. Galbally, E. Dunne, S. J. Lawson, S. Molloy E. Zardin, S. Saunders, É.-A. Guerette and C. Murphy

First *in situ* measurements of glyoxal and methylglyoxal over the remote temperate oceans 10
S. J. Lawson, P. W. Selleck, I. E. Galbally, M. D. Keywood, M. J. Harvey, C. Lerot, D. Helmig and Z. Ristovski

MEGAN: Online & Fabulous..... 11
K. Emmerson

Update on the MUMBA Campaign: Measurements of urban, marine and biogenic air 12
C. Paton-Walsh, É.-A. Guérette, G. Rea, D. Kubistin, R. Humphries, S. R. Wilson, D. W. T. Griffith, R. Buchholz, V. Velazco, X. Shi, I. E. Galbally, M. D. Keywood, S. J. Lawson, P. W. Selleck, M. Cheng, S. Molloy, M. Bhujel, A. D. Griffiths, S. D. Chambers and P. Davy

Multi-species interannual variability from GASLAB flask network data..... 13
R. L. Langenfelds, L. P. Steele, P. B. Krummel and R. J. Francey

Seasonal changes in the tropospheric carbon monoxide profile over the remote Southern Hemisphere..... 14
J. A. Fisher, S. R. Wilson, G. Zeng, J. E. Williams, L. K. Emmons, R. L. Langenfelds, P. B. Krummel and L. P. Steele

GAW Activities in Korea..... 15
C. Lee, B.-H. Heo, H. Lee, H. Lee, H. Yoo, S. Park, J. Yu, Y. Yoo, H.-J. Lee, D.-B. Yu, T. Jeong, H.-C. Lim, H. Choi, E. Kim, S. Moon and M. Ko

Progress of Enhanced Atmospheric Composition Measurement Capability at Cape Grim BAPS and the Expanding CSIRO Atmospheric Observation Network..... 16
M. V. van der Schoot, Z. M. Loh, M. D. Keywood, J. P. Ward, C. P. Meyer, S. Molloy, I. E.Galbally, S. J. Lawson, D. A. Spencer, J. Ward, N. T. Somerville, P. B. Krummel, L. P. Steele, R. L. Langenfelds, D. P. Thornton, S. A. Coram and R. L. Gregory

WCC-SF_6 activities at Anmyeondo station, Korea; Various analytical methods to monitor SF_6 using GC-μECD 17
H. Lee, H. Yoo, J. S. Lee, J.-S. Lim, C. Lee and B.-H. Heo

Recent developments in NOAA calibration activities..... 18
B. D. Hall, E. J. Dlugokencky, P. P. Tans, G. S. Dutton, J. D. Nance and J. W. Elkins

Comparisons of *in situ* carbon dioxide and methane measurements at Cape Grim – an update; and an overview of GCWerks software for *in situ* data processing..... 19
P. B. Krummel, M. V. van der Schoot, L. P. Steele, Z. M. Loh, D. A. Spencer, P. J. Fraser, R. L. Langenfelds, N. Somerville, J. Ward and P. K. Salameh

Day 2: Thursday 13 November 2014

Unifying the AOD picture 20
S. R. Wilson

Surface Ocean Aerosol Production (SOAP) 21
M. J. Harvey, C. S. Law, M. J. Smith, C. Walker, T. Bell, S. J. Lawson and SOAP team

Observation of internally mixed volatile and non-volatile organic sea spray from the Chatham Rise, New Zealand 23
L. Cravigan, M. Mallet, M. Harvey, C. Law, G. Olivares and Z. Ristovski

Emissions of Volatile Organic Compounds and Greenhouse Gases from fires in the CSIRO Pyrotron 24
É.-A. Guérette, C. Paton-Walsh, M. Desservettaz, N. Surawski, C. P. Meyer, A. Sullivan, F. Reisen, C. Weston, L. Volkova, G. Edwards and D. Howard

Aerosol Composition at Cape Grim: the long-term record 25
M. D. Keywood, P. W. Selleck, R. W. Gillett and J. P. Ward

Short-term forecasting of fine particle exposure from prescribed burns and wild fires 26
M. Cope, S. Lee, C. P. Meyer, A. Wain and B. Ebert

Savannah early dry season biomass burning – Campaign overview and a first look at aerosol chemical and microphysical properties from Gunn Point June 2014 28
P. W. Selleck, M. D. Keywood, S. J. Lawson, J. P. Ward, J. N. Harnwell, M. Cheng, R. W. Gillett, M. V. van der schoot, Z. Ristovski, A. Milic, J. Alroe, B. Miljevic, M. Mallet, L. Cravigan, M. Desservettaz, C. Paton-Walsh, D. W. T. Griffith, S. R. Wilson, G. Kettlewell, D. Howard, G. Edwards, L. Williams, H. Winton and B. Atkinson

Fourier transform spectrometer results from the Gunn Point campaign, June 2014, including emission factors of greenhouse gases from savanna fires 29
M. Desservettaz, C. Paton-Walsh, D. W. T. Griffith, S. R. Wilson, G. Kettlewell, M. D. Keywood, M. V. van der Schoot, P. W. Selleck, S. J. Lawson, J. P. Ward, J. N. Harnwell, M. Cheng, R. W. Gillett, Z. Ristovski, B. Miljevic, M. Mallet, L. Cravigan, D. Howard, G. Edwards, L. Williams, H. Winton and B. Atkinson

Volatility-separated measurement of aerosol chemical composition and hygroscopicity from the Gunn Point campaign, June 2014 30
Z. Ristovski, A. Milic, J. Alroe, B. Miljevic, M. Mallet, L. Cravigan, R. Jayaratne, M. Desservettaz, C. Paton-Walsh, D. W. T. Griffith, S. R. Wilson, G. Kettlewell, M. D. Keywood, M. V. van der Schoot, P. W. Selleck, S. J. Lawson, J. P. Ward, J. N. Harnwell, M. Cheng, R. W. Gillett, D. Howard, G. Edwards, L. Williams, H. Winton and B. Atkinson

Aerosol iron solubility: comparison between the Australian subtropics and Southern Ocean 31
H. Winton, R. Edwards, A. Bowie, S. D. Chambers, M. D. Keywood, S. Wercynski and A. G. Williams

Radon vertical profiles during the morning transition period 32
A. G. Williams, S. D. Chambers, J. Crawford and A. D. Griffiths

A radon-only technique for characterising baseline constituent concentrations at Cape Grim 33
S. D. Chambers, A. G. Williams, J. Crawford, A. D. Griffiths, P. B. Krummel, L. P. Steele, I. E. Galbally S. Molloy and M. V. van der Schoot

Open path Fourier transform spectroscopy measurements of greenhouse gases in Heidelberg 34
D. W. T. Griffith, S. Hammer, S. N. Vardag, I. Levin, S. Schmitt, D. Poehler and U. Platt

Water-atmosphere exchange of greenhouse gases (CO_2, CH_4 and N_2O) measured on the Murray River, Australia 35
C. Caldow, D. Kubistin, M. Desservettaz, D. Müller, T. Warneke, C. Murphy and D. W. T. Griffith

Continuous measurements of $\delta^{18}O$-CO_2 in air with a Fourier Transform InfraRed spectrometer 37
S. N. Vardag, D. W. T. Griffith, S. Hammer, M. Sabasch and I. Levin

New gas chromatographic technologies for GASLAB: pulsed discharge helium ionisation detector for analysis of trace gases 38
B. Mitrevski, L. P. Steele, P. B. Krummel, D. A. Spencer and P. K. Salameh

New insights into global perfluorocarbon emissions 39
P. J. Fraser, P. B. Krummel, L. P. Steele, J. Kim and D. Wong

Signatures of human actions recorded in the atmosphere 41
L. P. Steele, R. L. Langenfelds, P. B. Krummel, C. M. Trudinger and P. J. Fraser

Posters

Open-Path FTIR Measurements of Trace Gas Emissions from Australian Eucalypt Fires in the CSIRO Pyrotron 42
M. Desservettaz, É.-A. Guérette, N. Surawski, C. P. Meyer, A. Sullivan, F. Reisen, M. Cope, C. Weston, L. Volkova, G. Edwards, D. Howard and C. Paton-Walsh

Australian Biogenic HCHO Observations from the OMI and GOME-2 Satellites 44
J. Greenslade, J. A. Fisher, N. Jones, R. Buchholz, L. Zhu and E. A. Marais

New particle formation events over Antarctic sea-ice 45
R. Humphries, R. Schofield, M. D. Keywood, J. P. Ward, S. R. Wilson, C. Gionfriddo, M. Tate, D. Krabbenhoft, I. E. Galbally, K. Kreher, P. Johnston, A. Thomas, A. Robinson and N. Harris

Abstract Title: How productive is Earth's terrestrial biosphere? Novel satellite measurements and methods to understand global primary productivity 46
A. J. Norton and P. J. Rayner

Constraining greenhouse gas emissions in the Latrobe Valley 48
R. Taneja, P. J. Rayner and S. Utembe

The ACCESS-UKCA chemistry-climate model: exploring long-term trends in atmospheric composition 49
M. T. Woodhouse, A. K. Luhar, P. F. Uhe, M. Thatcher, I. E. Galbally, S. Molloy, R. L. Langenfelds, L. P. Steele, P. B. Krummel, M. V. van der Schoot, D. M. Etheridge, C. M. Trudinger and P. J. Fraser

Conventionally-heated microfurnace for graphitisation of microgram-sized carbon samples 50
B. Yang and A. M. Smith

Contents

Drivers of column-average CO_2 variability at Southern Hemispheric Total Carbon Column Observing Network sites

N. M. Deutscher[1,2*], V. Sherlock[3^], S. E. Mikaloff Fletcher[3], D. W. T. Griffith[1], J. Notholt[2], R. Macatangay[1#], B. J. Connor[4], J. Robinson[5], H. Shiona[5], V. A. Velazco[1], Y. Wang[2], P. O. Wennberg[6] and D. Wunch[6]

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Abstract

We investigate factors that drive the variability in total column CO_2 at the Total Carbon Column Observing Network sites in the Southern Hemisphere using fluxes tagged by process and by source region from the CarbonTracker analysed product as well as the Simple Biosphere model. We show that the terrestrial biosphere is the largest driver of variability in the Southern Hemisphere column CO_2. However, it does not dominate in the same fashion as in the Northern Hemisphere. Local- and hemispheric-scale biomass burning can also play an important role, particularly at the tropical site, Darwin. The magnitude of seasonal variability in the column-average dry-air mole fraction of CO_2, X_{CO2}, is also much smaller in the Southern Hemisphere and comparable in magnitude to the annual increase. Comparison of measurements to the model simulations highlights that there is some discrepancy between the two time series, especially in the early part of the Darwin data record. We show that this mismatch is most likely due to erroneously estimated local fluxes in the Australian tropical region, which are associated with enhanced photosynthesis caused by early rainfall during the tropical monsoon season.

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How and when will anthropogenic slowing of atmospheric CO_2 growth become evident?

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Abstract

The increasing international efforts to avoid adverse environmental consequences of the fossil energy industry will be verified if slowing CO_2 growth in the global atmosphere can be unambiguously attributed to reductions in the anthropogenic emissions.

The difficulty in linking a future emissions slowdown to slowing atmospheric growth was highlighted by evidence for wide-scale slowing CO_2 growth in the atmosphere from 2002-2008 at a time of unprecedented increased growth in reported emissions [Francey *et al*., 2013]. They suggested that a 7% underestimation of fossil emissions around 1995-2005 period would provide the simplest resolution. However most global carbon budget modelling studies retain reported emissions and argue for an unusually sustained terrestrial biota uptake that closely offsets the emission changes in space and time [Le Quéré *et al*., 2014].

An update of the atmospheric records, along with an examination of the evolution of measurement methodologies, now provide further insight into this puzzle and into subsequent unusual events, like the proposed prominent global role of Australia's savannahs in 2011 [Poulter *et al*., 2014].

Overly simplistic estimates of uncertainty in the two largest budget terms, fossil fuel emissions and atmospheric CO_2 growth may obscure detection of recent global atmospheric CO_2 changes attributable to human behaviour. Improvements in ways that atmospheric data are collected and utilised for this purpose are identified.

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Interannual variability of simulated global land carbon sink and the contribution to inter-hemisphere differences of CO_2 concentration

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Abstract

Interannual variability of terrestrial carbon sink and anthropogenic fossil-fuel emission are two major drivers to the variation of atmospheric CO_2 concentration. Recently, an extremely large land carbon sink in 2010-2011 is reported by Le Quéré *et al*. [2013]. Analysis from Poulter *et al*. [2014] suggests that the anomaly of this global carbon sink is mainly contributed by semi-arid ecosystems in the Southern Hemisphere, especially in the Australian region. To explore the regional contribution of the interannual variability of carbon sink, we conduct a carbon-nutrient cycle simulation using CABLE forced with TRENDY forcing from 1981 to 2013. Our CABLE simulation results show that global terrestrial carbon sink peaks at 4.3 GtC/year in 2011 (Figure 1a), which is well above the average carbon uptake by global land biosphere from 2003 to 2012. Over that period, Carbon sink from the Southern Hemisphere in 2011 increased by about two times, and the carbon sink from the Northern Hemisphere increased by 50% in 2011, as compared to their respective averages from 2003 to 2011 (Figure 1b). Our result agrees with Poulter's [2014] conclusion that the Southern Hemisphere contributes most of the carbon sink anomaly in 2011. However, the increased carbon uptake by land ecosystems from eastern and southeast Australia also makes a very significant contribution in 2011.

To investigate how the interannual variability of CABLE biosphere fluxes affects the interhemispheric difference (IHD) in atmospheric CO_2, we have used CO_2 response functions calculated from an atmospheric model (CCAM) to reconstruct atmospheric CO_2 concentrations at Mauna Loa and Cape Grim. We find that the large increase in observed IHD between 2009 and 2010 [Francey *et al*., 2013] is also seen for CO_2 derived from CABLE fluxes.

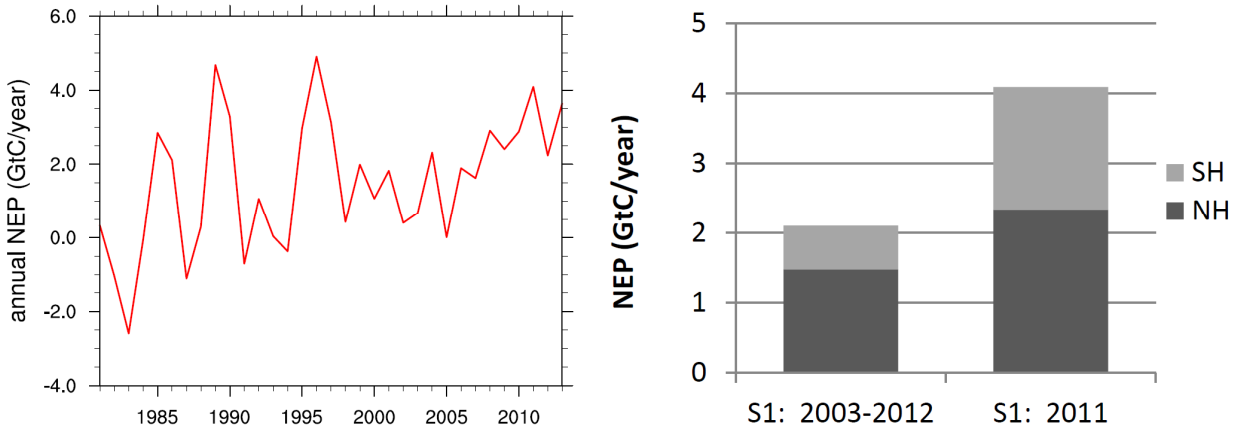


Figure 1. Results from global carbon cycle simulation using CABLE and TRENDY forcing from 1981 to 2013. a) Global land annual NEP from 1981 to 2013. Positive indicates carbon sink. b) Comparison of terrestrial carbon flux in 2011 to a period average from 2003 to 2012 at hemisphere scale.

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Transport modelling and inversions for the interpretation of greenhouse gas measurements

R. M. Law[1*], Z. M. Loh[1], T. Ziehn[1], K. D. Haynes[2], P. B. Krummel[1], L. P. Steele[1], P. J. Fraser[1], S. D. Chambers[3] and A. G. Williams[3]

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Abstract

The interpretation of greenhouse gas measurements can be aided by forward transport modelling while greenhouse gas fluxes can be estimated using atmospheric inversions. Here we (a) provide an update on a study of methane model simulations at Cape Grim and their use for determining methane fluxes from SE Australia and (b) show results from some recent CO_2 inversions.

Observed and model simulated non-baseline methane concentrations at Cape Grim have been compared [Loh *et al*., 2014]. Two atmospheric models (CCAM and ACCESS) and six different methane emission scenarios are used. To minimise the influence of transport model errors on the analysis, deviations of Cape Grim methane concentration above baseline have been compared to coincident radon measurements. This methane to radon ratio shows a clear seasonal signal implying seasonal variations in methane emissions from SE Australia relative to a more temporally uniform radon flux. The ability of the model simulations to match the observed seasonality is dependent on the choice of methane emission scenario but all scenarios underestimate the observed methane to radon ratio in spring. We find that the most likely explanation for the discrepancy is wetland emissions that are too small in some emission scenarios or at the wrong time of year in other scenarios.

CO_2 inversions have been run recently for two purposes. The first is an international comparison of greenhouse gas inversions focussed on South, East and South East Asia. We have submitted a CCAM inversion for 1993-2012 using a fixed year of winds and expect to submit a second inversion with interannually varying winds. The second purpose is to use a CO_2 inversion to estimate the magnitude of regional fluxes that are required to fit the larger difference in annual mean CO_2 concentration between Mauna Loa and Cape Grim over recent years.

References

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Analysis of ACCESS-ESM1 results: Progress and future plans

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Abstract

Although the number of climate models has increased rapidly in recent years, only a small number of institutes worldwide have been developing earth system models (ESMs). However, it is well recognised that the carbon cycle and the carbon-climate feedback in particular have a large impact on the climate and only ESMs are able to simulate both physical and biogeochemical processes, including potential feedbacks. The Australian Community Climate and Earth System Simulator (ACCESS) has been recently extended to include land and ocean carbon cycle components in its ACCESS-ESM1 version. ACCESS-ESM1 is based on ACCESS-1.4 which uses the Community Atmosphere Biosphere Land Exchange (CABLE) model as its land surface model. The biogeochemistry component of CABLE includes the coupling of carbon, nitrogen and phosphorus cycles. Ocean carbon fluxes are calculated in WOMBAT (World Ocean Model of Biogeochemistry And Trophic-dynamics) which consists of a two component plankton model.

Here we focus on the analysis of the results from ACCESS-ESM1 simulations and the validation of the land and ocean carbon cycle components against independent estimates. Currently all model runs have been set up as concentration driven simulations, which means that atmospheric CO_2 concentrations are prescribed as an input to ACCESS-ESM1 and changes in the land and ocean carbon pools do not feedback on to atmospheric CO_2 concentrations. However two passive tracers in the atmosphere calculate the atmospheric CO_2 concentrations separately for the modelled land and ocean fluxes, enabling simulated features of atmospheric CO_2, such as the seasonal cycle, to be compared to observations. Results are mainly presented from the pre-industrial control run and from a small ensemble of historical simulations. Preliminary results are shown for future projections, which have been carried out using representative concentration pathways (RCPs).

The Cape Grim Reactive Gases Program Summary 2008 - 2014

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Abstract

A report is presented of the activities of the Cape Grim Reactive Gases Program since its last review in 2007.

The report briefly covers: (a) the Reactive Gases program goals; (b) instrumentation; (c) the monitoring of surface ozone, NO_x, VOCs and ammonia; (d) documentation and data archiving; (e) discoveries and achievements and (f) plans for the future. The full details are presented elsewhere in the reports and publications and these are listed.

The achievements related to surface ozone (in many cases in collaboration with other scientists) are:

- Four instrument replacements (two analysers, one calibrator, one zero and span gas switching unit) have been achieved without interruption to the quality of the observations. New SOPs have been prepared and training conducted.
- Development of the "Guidelines for Continuous Measurement of Ozone in the Troposphere, WMO GAW Report No 209" led by the Cape Grim Reactive Gases Lead Scientist.

WCC-EMPA staff conducted an Audit of the Surface Ozone Measurement at Cape Grim in March 2010. The result of the comparison was that the Cape Grim standard and the travelling WMO standard were statistically indistinguishable in their measurements.

- Cape Grim surface ozone data from 2008 to 2013 are archived with the WDCGG.
- Determination of the long term surface ozone increase in concentration at Cape Grim 1982 – 2014 and its relation to increases at other sites in the extra-tropical Southern Hemisphere.
- Determination of the contribution of a local biomass burning event to ozone formation at Cape Grim using chemical transport modelling

The achievements in relation to nitrogen oxides include:

- Commissioning of a commercial high sensitivity NO_{xy} analyser at Cape Grim. Development of protocols and SOPs for NO_{xy} monitor checks and maintenance suitable for baseline conditions in the southern hemisphere.
- Successful NO_x measurements in baseline air at Cape Grim. The design of algorithms and decision trees suitable for processing the NOxy data on an ongoing basis for Cape Grim.

The achievements in relation to volatile organic compounds (VOCs) include:

- Installation of an in situ VOC monitoring system
- Highlighting an unknown source of climatically important oxygenated VOCs in Baseline air, incorporating data from several Cape Grim Programs, NOAA and INSTAAR VOC flask data, and satellite observations
- Integration of VOC data with Particulate Program data to infer a new particle formation event in Baseline air
- Determination of biomass burning emission factors for a variety of VOCs the first time in Australian conditions (incorporating data from several other Programs)

- Identification of a significant discrepancy in the global atmospheric budget of ethanol from atmospheric observations
- Quantification of possible interferences with the measurement of acetonitrile by PTR-MS.
- Development of understanding of the role and usefulness of background concentrations of acetonitrile in the atmosphere.

Ammonia measurements commenced in late 2014 at Cape Grim.

Throughout the review period there has been an active use of Cape Grim data and also advocacy of global atmospheric composition monitoring. Ten journal papers and ten peer reviewed reports and conference papers and one DSc and one PhD thesis have been produced by the Program during 2008-2014. Approximately 70 presentations at international science meetings, Cape Grim annual science meetings and national conferences have been made during this period.

Plans for the activities within the Reactive Gases program for the next three years are presented.

The global distribution of atmospheric acetonitrile away from the influence of biomass burning

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Abstract

New measurements of acetonitrile from eleven campaigns in the Southern Hemisphere are presented. These increase the available Southern Hemisphere acetonitrile observations made away from biomass burning plumes, that is, in the background troposphere from 36 days to over 500 days coverage. For the first time a global view of acetonitrile in the background atmosphere is available.

The mean mixing ratio of acetonitrile in the unpolluted troposphere of the Southern Hemisphere is less than half of that in the Northern Hemisphere, and may be lower still due to the absence of observations in the polar regions. In both hemispheres there is a decrease in acetonitrile moving from the tropics to temperate latitudes. Both these features are consistent with the major source of acetonitrile being biomass burning in the tropics, mainly in the Northern Hemisphere.

The observations in both hemispheres in urban areas show a higher acetonitrile concentrations in some cities compared with the background atmosphere. This appears to be due to local biomass burning.

Comparisons conducted with a two-hemisphere atmospheric box model indicate that there is now sufficient information now to understand the global atmospheric background cycle of acetonitrile. The source is biomass burning and net removal is both at the terrestrial surface and in the oceans.

Acetonitrile is not only a good immediate tracer of biomass burning, but also in the background atmosphere can indicate the history of biomass burning in the one to two immediately preceding years. This may provide a useful new constraint on global biomass burning estimates.

First *in situ* measurements of glyoxal and methylglyoxal over the remote temperate oceans

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Abstract

Dicarbonyls glyoxal and methylglyoxal are short-lived trace gases, produced by oxidation of parent compounds such as isoprene and aromatic hydrocarbons, and are important precursors of secondary organic aerosol (SOA). The widespread presence of glyoxal in the remote marine boundary layer (MBL) has been confirmed in recent years by ground-based and satellite observations, however there is suggestion that glyoxal mixing ratios observed are in far in excess of the yields expected from oxidation of its parent compounds. There are currently insufficient methylglyoxal observations in the MBL to confirm its presence and importance to SOA formation over the remote oceans.

We present observations of glyoxal and methylglyoxal, measured with 2,4-dinitrophenylhydrazine (2,4-DNPH) cartridges and high performance liquid chromatography (HPLC), over the temperate Southern Hemisphere oceans. Samples were collected in summer (Feb-Mar) over the Chatham Rise in the South West Pacific Ocean during the Surface Ocean Aerosol Production (SOAP) voyage in 2012, and at Cape Grim Baseline Air Pollution Station in late winter (Aug-Sep) 2011. Measurements of a range of dicarbonyl precursors were made in parallel. We compare these measurements to other available observations of dicarbonyls over remote oceans, including observations made by MAX-DOAS. We use precursor VOC mixing ratios to calculate the expected yield of glyoxal and methylglyoxal and find that a large fraction of the dicarbonyls observed are unaccounted for, highlighting a substantial unknown source. Finally, surface glyoxal mixing ratios are compared to satellite vertical column densities retrieved from GOME-2, highlighting a large discrepancy; reasons for this are discussed.

This study provides further evidence of an as yet unidentified source of both glyoxal and also methylglyoxal over the remote oceans.

References

Lawson, S. J., P. W. Selleck, I. E. Galbally, M. D. Keywood, M. J. Harvey, C. Lerot, D. Helmig and Z. Ristovski, Seasonal *in situ* observations of glyoxal and methylglyoxal over the temperate oceans of the Southern Hemisphere, *Atmos. Chem. Phys. Discuss., 14*, 21659-21708, doi:10.5194/acpd-14-21659-2014, 2014.

MEGAN: Online & Fabulous

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Abstract

Guenther *et. al*.'s [1995] Model of Emissions of Gases and Aerosols from Nature (MEGAN) has been included as a module online within our Chemical Transport Model. This means it can now be used in air quality studies at high resolution in our nested model framework system. Validation studies have been undertaken using data from the Sydney particle study [Cope *et al*., 2013]. This talk will demonstrate how the system works and what data is needed to use it. Results highlight the need for good Australian vegetation maps.

References

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Update on the MUMBA Campaign: Measurements of urban, marine and biogenic air

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Abstract

The Measurements of Urban, Marine and Biogenic Air campaign (MUMBA) took place in Wollongong, NSW (a city approximately 80 km south of Sydney, Australia) from 21 December 2012 to 15 February 2013. Instruments were deployed during MUMBA to measure the gaseous and aerosol composition of the atmosphere with the aim of providing a detailed characterisation of the complex ocean/forest/urban interface that could be used to test the skill of atmospheric models.

In this presentation we will provide an update on the analysis and interpretation of the results including first comparisons with the regional atmospheric model CHIMERE [*Menut et al.,* 2013]. We will also present a more detailed characterisation of the clean air signature, including concentrations of a number of VOCs measured using the PTR-MS.

References

Menut, L., B. Bessagnet, D. Khvorostyanov, M. Beekmann, N. Blond, A. Colette, I. Coll, G. Curci, G. Foret, A. Hodzic, S. Mailler, F. Meleux, J.-L. Monge, I. Pison, G. Siour, S. Turquety, M. Valari, R. Vautard and M. G. Vivanco, CHIMERE 2013: a model for regional atmospheric composition modelling, *Geosci. Model Dev*., 6, 981-1028, doi:10.5194/gmd-6-981-2013, 2013.

Multi-species interannual variability from GASLAB flask network data

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Abstract

Since 1992, CSIRO GASLAB has maintained a global flask sampling network, with samples routinely analysed for multiple trace gas species that include CO_2 and its $\delta^{13}C$, CH_4, H_2 and CO. An earlier study reported correlated interannual variability (IAV) among these species between 1992 and 2000 that was attributed to biomass burning [Langenfelds *et al*., 2002]. This talk will explore the IAV observed in these species from more than a decade of additional data.

References

Langenfelds, R.L., R.J. Francey, B.C. Pak, L.P. Steele, J. Lloyd, C.M. Trudinger and C.E. Allison, Interannual growth rate variations of atmospheric CO_2 and its $\delta^{13}C$, H_2, CH_4 and CO between 1992 and 1999 linked to biomass burning, *Global Biogeochem. Cycles*, 16(3), 1048, doi:10.1029/2001GB001466, 2002.

Seasonal changes in the tropospheric carbon monoxide profile over the remote Southern Hemisphere

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Abstract

We use aircraft observations from the 1991-2000 Cape Grim Overflight Program and the 2009- 2011 HIAPER Pole-to-Pole Observations (HIPPO), together with output from four chemical transport and chemistry-climate models, to better understand the vertical distribution of carbon monoxide (CO) in the remote Southern Hemisphere. Observed CO vertical gradients at Cape Grim vary from 1.6 ppbv km^{-1} in austral autumn to 2.2 ppbv km^{-1} in austral spring. CO vertical profiles from Cape Grim are remarkably consistent with those observed over the southern mid- latitudes Pacific during HIPPO, despite major differences in time periods, flight locations, and sampling strategies between the two datasets. Using multi-model simulations from the Southern Hemisphere Model Intercomparison Project (SHMIP), we find that observed CO vertical gradients in austral winter-spring are well-represented in models and can be attributed to primary CO emissions from biomass burning. In austral summer-autumn, inter-model variability in simulated gradients is much larger, and two of the four SHMIP models significantly underestimate the Cape Grim observations. Sensitivity simulations show that CO vertical gradients at this time of year are driven by long-range transport of secondary CO of biogenic origin, implying a large sensitivity of the remote Southern Hemisphere troposphere to biogenic emissions and chemistry. Inter-model variability in summer-autumn gradients can be explained by differences in both the chemical mechanisms that drive secondary production of CO from biogenic sources and the vertical transport that redistributes this CO throughout the Southern Hemisphere. This suggests that the CO vertical gradient in the remote Southern Hemisphere provides a sensitive test of the chemistry and transport processes that define the chemical state of the background atmosphere.

GAW Activities in Korea

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Abstract

Korea Meteorological Administration (KMA) operates three main measurement stations, which are located in the west (at Anmyeon), south (at Gosan), and east (at Ulleungdo) of Korea, in aim of monitoring of transportation of the atmospheric substances and variation in the atmospheric composition over the Korean Peninsula. It also has the auxiliary stations at Seoul, Pohang, Gwangju, Mokpo, Uljin, Gangneung, and Jeju to collect more specific data, e.g. radioactivity (Radon), ozone sonde, and UV. From the measurement stations, KMA collects and provides reliable scientific data and information on the chemical composition of the atmosphere, its natural and anthropogenic change to help improve the understanding on climate change.

KMA collects the atmospheric observation data of 37 components in the fields of greenhouse gases, aerosols, reactive gases, ozone, atmospheric radiation including ultraviolet (UV) radiation, and precipitation chemistry in the Korean Peninsula, in accordance with the measurement recommendations of the Global Atmosphere Watch (GAW) program. In the fields of greenhouse gases, the abundance of carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), chlorofluorocarbons (CFCs), and sulfur hexafluoride (SF_6) has been measured with high accuracy and precision. Physical, chemical, and optical characteristics of aerosols, widely acknowledged as one of the most significant and uncertain aspects of climate change projections, have been measured with in-situ instruments, including their vertical profiles retrieved using a ground-based LIDAR. The reactive gases such as carbon monoxide (CO), surface ozone (O_3), nitrogen oxides (NOx), and sulfur dioxide (SO_2) have been measured because the compounds play a role in the chemistry of the atmosphere and the formation of aerosols. Regarding to the stratospheric ozone, total column ozone is measured using the Brewer spectrometers, together with UV and solar and terrestrial radiation. Precipitation chemistry observations focus on the acidity, conductivity, and major 9 ions.

New parameters and methodologies are also secured and applied, e.g. greenhouse gas isotopes, aerosol chemical species, and volatile organic compounds (VOCs). To enhance the effectiveness and application of the long-term measurements within GAW, KMA cooperates with the atmospheric measurement networks worldwide along with focusing on the quality assurance and control.

World Calibration Center for SF6 (WCC-SF_6) was designated to be established in KMA in 2012, and has been operated since 2013. WCC-SF_6 conducts the missions for the traceability and compatibility of the SF_6 measurement for the GAW stations. The Asia- Pacific GAW Workshop on Greenhouse Gases has annually been held by KMA since 2009. It provides a good opportunity to share our knowledge on greenhouse gases measurements in the Asia and Pacific region.

Progress of Enhanced Atmospheric Composition Measurement Capability at Cape Grim BAPS and the Expanding CSIRO Atmospheric Observation Network

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Abstract

The 'Earth Health' program in CSIRO's Ocean & Atmosphere Flagship is continuing to expand its atmospheric composition measurement capability at Cape Grim BAPS and CSIRO's Australian atmospheric observation network in general. In particular, at the developing Australian pilot tropical atmospheric observation site established at Gunn Point in Australia's Northern Territory (12.249°S, 131.045°E, elevation 25 metres).

In addition to an expanding ground-based observing network, CSIRO now has a new Marine National Facility research vessel, the 'R.V. Investigator', with a purpose-built atmospheric research capability, previously not available to the Australian research community. This new capability offers a unique opportunity for collaborative atmospheric-focussed research and campaigns to be undertaken in relation to the Southern Hemisphere oceans, sea-ice zones and coastal and tropical regions.

This presentation will provide an update of the current state of the development of instrumentation at the Australian network of GAW stations (Cape Grim, Gunn Pt. and including the R.V. Investigator), and future plans, as well as the key operational, instrumental and calibration strategies for this network. In addition, some preliminary results from the current state of instrumental evaluation (for greenhouse gases).

WCC-SF_6 activities at Anmyeondo station, Korea; Various analytical methods to monitor SF_6 using GC-μECD

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Abstract

The main activity of World Calibration Center is focusing on the quality assurance through organizing comparison and training session, developing the measurement guidelines, providing training and workshops, and assessing the quality of data residing at the World Data Center.

Amyeondo (AMY) station (36°32' N, 126°19'E, 45.7 m), one of regional stations in Korea, started monitoring SF_6 from 2007. Samples are collected from 40 m tower with decabon tube and passed through dehumidification system, pre-concentrator and analysed by using GC-μECD with an Activated Alumina F1 column. Various types of tube, column and two conditions with and without a pre-concentrator to analyse SF_6 were compared to demonstrate its system performance and to improve its analytical method. The air sample concentrations had been determined by one point standard gas having traceability to KRISS but it has been changed from KRISS to Central Calibration Lab (CCL, NOAA) since 2013. For data continuity an inter-comparison experiment between both of them was implemented and the offset was 0.022 ppt indicating that it satisfied the extended goal, 0.05 ppt. And also for its quality assurance, the calibration methods with multi points, two points and one point were compared. The differences between those of two methods, multi - one point and multi - two points were 0.009 ppt (0.12%) and 0.005 ppt (0.06%) respectively indicating one point calibration is reasonable while standard gas is focusing on background level or targeting level.

For the stations that have their own method to monitor background SF_6 concentrations and the stations that prepare for monitoring it, WCC-SF_6 has a plan to suggest how to guarantee quality assurance with the guideline including various methods which are applicable back to in situ stations of SF_6 and to provide technical education courses based on it from this year.

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Recent developments in NOAA calibration activities

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Abstract

The NOAA Global Monitoring Division serves as the WMO/GAW Central Calibration Laboratory for the greenhouse gases CO_2, CH_4, N_2O, and SF_6. In order to provide WMO/GAW partners with pertinent information regarding calibration standards, we are re-evaluating uncertainties associated with calibration scales and their transfer. We will soon be reporting total uncertainty (including all aspects of the calibration scale), as well as reproducibility, which is relevant to scale transfer. We are currently investigating different methods to evaluate reproducibility, including examining results from standards analysed more than once in our laboratory (Figure 1).

Recent developments with respect to calibration scales, comparisons, and GMD time series data will be presented.

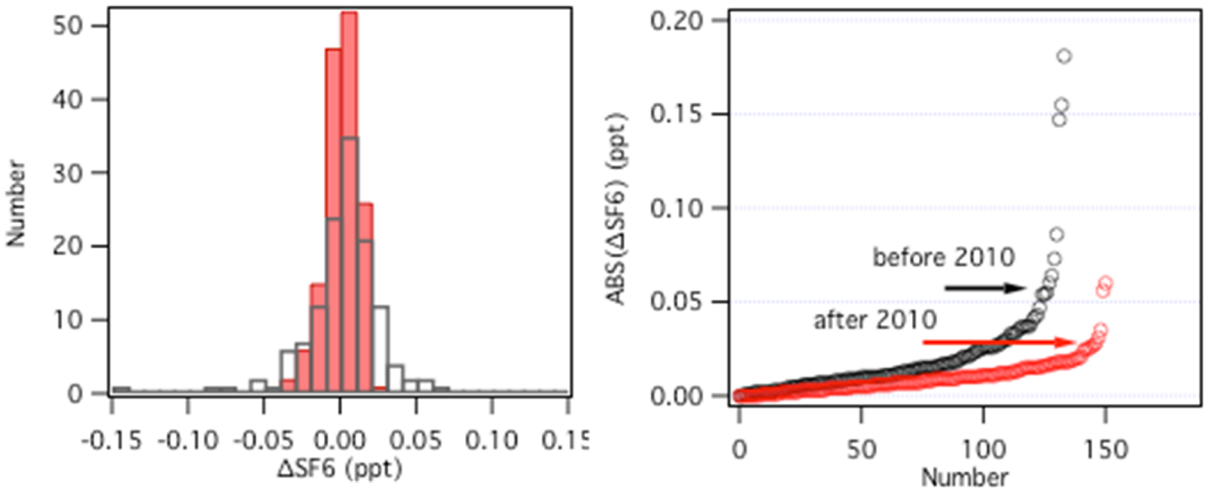


Figure 2. Reproducibility of SF_6 calibrations represented as a histogram (left) and cumulative plot (right). Reproducibility, estimated from the 95%ile, was 0.055 ppt before 2010 (black), and 0.025 ppt after 2010 (red).

Comparisons of *in situ* carbon dioxide and methane measurements at Cape Grim – an update; and an overview of GCWerks software for *in situ* data processing

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Abstract

Loh *et al.* [2013] presented a first look at the Picarro cavity ringdown spectrometer (CRDS) record for continuous measurement of atmospheric CO_2, CH_4 (and water vapour) from the instrument that was installed at Cape Grim in February 2012. The Picarro record provides a more truly continuous time coverage than the LoFlo NDIR (non-dispersive infra-red) CO_2 record (which normally measures ambient air for 44 minutes in every hour) and the AGAGE GC-FID (gas chromatograph – flame ionisation detector) CH_4 record which consists of a discrete air sample measurement every 40 minutes. In this presentation, an update of how the Picarro record compares to the longer-term CO_2 and CH_4 records at Cape Grim will be given. In addition, comparisons to the CSIRO GASLAB flask record for CO_2 and CH_4 will also be shown. Major differences in the techniques will be highlighted.

In the second part of the presentation, an overview of the GCWerks software package for processing of *in situ* data will be given. The GCWerks package was originally developed by Peter Salameh for the Advanced Global Atmospheric Gases Experiment (AGAGE) program for instrument control and processing of GC & GC/MS (gas chromatograph/mass spectrometer) instruments, an overview was presented in Krummel and Salameh [2008]. More recently, GCWerks has been extended for spectroscopic instruments such as CRDS. GCWerks is now available commercially, see http://gcwerks.com/ for details. A demonstration of the various GCWerks components will be given, including the GCcompare package that was originally developed to help streamline the data review procedures at the six-monthly AGAGE meetings.

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Unifying the AOD picture

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Abstract

Aerosol optical depth has been measured at Cape Grim from the early days of the station. In 1986 the manual measurements were replaced with an automated system, an essential change given the cloudiness of the site, and the low optical depths often observed.

Since then a number of different instruments have been used, and a variety of data processing approaches. In order to make the data truly comparable, it has been necessary to reformat and reanalyse the data, which has now been completed for the period 1/ 1986 – 6 / 2014. In addition, the data has been filtered to reduce the impact of contamination of the instrument. This is therefore a much longer period and more homogeneous dataset than that presented earlier [*Wilson and Forgan*, 2002].

Consistent with previous studies, a clear annual cycle is observed, with a minimum in winter and two maxima, one in September and the second in December/ January. These three periods can be contrasted, to see if there is evidence in the optical depth record that helps to characterize the aerosol sources. In particular, the daily cycle in optical depth appears very different between spring and summer, consistent with a difference between long-range transport and local production.

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Surface Ocean Aerosol Production (SOAP)

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Abstract

Biologically-active regions of the surface ocean support production of a range of volatile compounds that influence aerosol particle production, composition and properties in the overlying marine boundary layer (MBL). In February 2012 the SOAP (Surface Ocean Aerosol Production) voyage examined biotic influences on aerosol production to the east of New Zealand, by targeting phytoplankton blooms along the Sub-Tropical Front. The research included characterisation of a) surface ocean biogeochemistry b) DMS, CO_2 and aerosol exchange, and c) aerosol type, number and composition, with a collective aim to link spatial variation in flux and aerosol characteristics to variation in phytoplankton biomass and community composition. Particular areas of interest included the role of the surface microlayer, comparison of on-board micrometeorological (eddy covariance) flux estimates with gas exchange parameterisations, primary and secondary organic aerosol formation in the MBL and in bubble burst chamber experiments.

SOAP characterised emissions associated with three 'bloom areas' and a wide range of environmental conditions (Table 1).

Table 1. SOAP blooms

	Bloom 1	**Bloom 2**	**Bloom 3**
Composition	Dinoflagellate	Coccolithophores	Dinoflagellate/Coccolithophores
Max Chl-a (µg/L)	3.5	1.0	2.5
Mean DMSd (nmol/L)	16.8 ±1.5	9.1±2.9	5.9±1.5
Mean Windspeed (u10 m/s)	5.6	9.6	10.1
Insolation (MJ/m^2/d)	20	17.8	17.9

For the sulfur system, 'Bloom 1' had the highest mixed layer concentration of chlorophyll and DMS. It was during fair weather conditions of 'Bloom 1' with light winds and strong insolation, that there was evidence of significant enrichment of DMS in the sea surface microlayer (SSM); this coincided with enhanced emission of the gas and occasions where the measured (eddy covariance) fluxes were larger than would be implied by bulk water concentrations. Conversely there was no clear evidence for impact on emission of a SSM deficient in DMS. There is some evidence that this high sulfur environment influenced the ubiquitous accumulation mode and during bloom 1 the ammonium sulphate fraction had the greatest number and mass with both super and sub-micron components that were lower under stronger wind conditions with elevated sea-salt aerosol.

Instances of particle nucleation were observed in the SOAP experiments with nucleation (10 nm and 15 nm) and Aitken mode sized particles (50 nm) typically including a significant (i.e. up to volume fraction of 50%) secondary organic component during sunny conditions in biologically active regions.

References

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Observation of internally mixed volatile and non-volatile organic sea spray from the Chatham Rise, New Zealand

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Abstract

Recent modelling has highlighted the importance of pristine environments to the uncertainty in cloud condensation nuclei (CCN) concentrations [Carslaw *et al*., 2013]. A key component to this uncertainty is the emission of sea spray aerosol (SSA) from the surface ocean, particularly in the Southern Ocean [Lee *et al*., 2012]. The composition and concentration of sub-200 nm SSA remains uncertain, however it provides a significant contribution to global CCN concentrations. SSA composition inferred from hygroscopitity and volatility measurements of laboratory generated nascent SSA will be considered in this presentation. These measurements were undertaken at the Chatham Rise (East of New Zealand; latitude 42°S-44°S, longitude 174°E-177°E), a region characterized by intensive summer phytoplankton blooms. These observations were undertaken during the IGBP-SOLAS Surface Ocean Aerosol Production (SOAP) voyages in February 2011 and February/March 2012 on board the research vessel RV-Tangaroa (NIWA, Wellington, New Zealand).

Internally mixed volatile and non-volatile organic SSA components were observed for laboratory generated SSA from phytoplankton rich waters. The volatile organic component evaporated at below 200°C and was consistently present at volume fractions of up to 18%, while the non-volatile organic component was observed in volume fractions of up to 34%. Sub-150 nm SSA produced from water samples taken from the Chatham Rise in 2011 showed distinct size dependence, with larger particles containing an increasing organic fraction. Similar results were observed for hygroscopicity and volatility measurements of ambient SSA from the Cape Grim baseline sector. Characterisation of SSA composition down to sub-100 nm sizes is rare and these results are distinct from that observed in other ocean basins [Quinn *et al*., 2014]. These results indicate that existing primary marine aerosol emission schemes could provide a significant overestimation of the organic SSA flux and therefore could significantly underestimate the number of potential SSA CCN.

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Emissions of Volatile Organic Compounds and Greenhouse Gases from fires in the CSIRO Pyrotron

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Abstract

In February and March 2014 a measurement campaign took place at the CSIRO Pyrotron to measure the emissions of trace gases and particulates from eucalypt fires. The fuel was collected from the Wombat State Forest in Victoria, as part of the DEPI Smoke Emissions and Transport Project.

The fires were designed to mimic conditions typical of hazard reduction burning in terms of 'Byram fireline Intensity'. Fuel moisture, fuel load and wind speed were carefully selected to achieve a Byram fireline intensity of around 500 kW/m. Fuel contiguity is also an important factor in enabling a fire to spread, so the fuels used were sieved to remove fragmented material. The campaign originally aimed to determine the dependence of both the coarse fuel load and the mode of fire spread (heading or backing) on the resulting emission factors. Unfortunately, there were problems with backing fires failing to propagate well, and so the campaign concentrated on a detailed study of whether the emission factors (primarily for heading fires) were sensitive to the fraction of coarse fuel in the total fuel burned.

Greenhouse and trace gases, mercury, volatile organic compounds and particulates were monitored during each fire. This presentation will present results from open-path FTIR measurements and grab samples analysed for CO_2, CO, CH_4 and volatile organic compounds.

Aerosol Composition at Cape Grim: the long-term record

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Abstract

A major goal of the Global Atmosphere Watch (GAW) Programme is to ensure long-term measurements of atmospheric constituents in order to detect and explain trends. The Scientific Advisory Group for aerosols recommends that GAW stations measure the concentration of major chemical components in PM10 and PM2.5 aerosols. The core activity of the Multilphase Atmospheric Chemistry Program at Cape Grim has been the generation of a long term record of aerosol mass and composition for the background atmosphere. While samples have been collected since 1976, the first reliable PM10 samples were collected in 1989 and the PM2.5 record extends back to 2002.

We will present a preliminary assessment of the annual and seasonal trends in sea salt measured in PM10, the predominant source of aerosol at Cape Grim under baseline conditions. We will utilise the shorter PM2.5 record to investigate trends present in secondary aerosol products, particularly nssSO4 and MSA. Combining the PM10 and PM2.5 records also provides valuable information about the particles between 10 and 2.5 µm in diameter which contribute significantly to PM10 mass in the marine atmosphere.

This work builds on almost 40 years of investment into aerosol composition measurements at Cape Grim.

Short-term forecasting of fine particle exposure from prescribed burns and wild fires

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Abstract

Wild fires in Southern Australian forests can have catastrophic effects on the land, property and people. A Royal Commission into the 2009 Black Saturday fires (which took the lives of 173 people and devastated over 450,000 ha of land), concluded that one of the contributing factors was the heavy fuel load present in the areas burnt, and consequently recommended that the annual rate of fuel reduction burning should increase to 5% of total State managed forest area.

Fuel reduction burns are designed to be of low intensity and to be well contained within a designated burn zone. As such, the program of prescribed burning is generally limited to light winds, minimal convection, low temperature, and moderate humidity. However, such conditions are also characterised by low levels of atmospheric ventilation and the build-up of air pollution. This can be further exacerbated if the burning season extends into the early winter when the smoke emissions add to those of domestic wood heaters and other near-surface sources of air pollution. The principal pollutant of concern for health generated by wood combustion is fine (less than 2.5 um) particles which have a well-established risk for mortality. There is also an attendant risk of smoke damage to some agricultural sectors (e.g. vineyards where smoke taint can be of particular concern).

Because fuel burns can only occur under conditions that may maximize air pollution, there is a need to select conditions in which the build-up of existing particle pollution is low, and in which smoke plumes from the fuel reduction process have a low probably of impacting populated areas. For situations in which the latter is not possible then fire managers need to provide advance warning to potentially effected populations, thus enabling at-risk individual to take steps to minimise their exposure to the smoke particles.

In this presentation we will introduce a prognostic numerical forecasting system which is being developed by CSIRO, the Bureau of Meteorology (BOM) and key university partners to provide fire managers with a robust tool for forecasting smoke exposure from fuel reduction burns. Variants of the system are being tested in Victoria as part of a Department of Environment and Primary Industry (DEPI) Smoke Emissions and Transport Project; and in NSW for the Rural Fire Service (RFS) and Office and Environment and Heritage (OEH) as part of BOM's forecast demonstration project (FDP).

BOM's ACCESS operational weather forecasts drive the system- providing ensemble forecasts out to 10 days at global scale (~50 km grid spacing); single 72 hour forecasts at the Australian scale (~15 km); 24 hour forecasts at the city scale (~3 km). For the FDP, high resolution (~1.5 km grid spacing) forecasts will be provided on a rapid update cycle (down to hourly) with full assimilation of radar precipitation fields. The transport and chemical transformation of smoke and secondary particle precursors is modelling using CSIRO's chemical transport model (CTM), covering scales ranging from continental to a few kilometres. Anthropogenic emissions for the CTM will be provided by the OEH and Victorian EPA air emissions inventories. Smoke emissions are currently provided by ECMWF at the continental scale, and by the fire agencies at the State

scale. In this mode the CTM provides the background primary and secondary particle loading. The resultant particle field is then used to initialise HYSPLIT, a fast hybrid particle/puff model which will be available for use by fire managers in real time to aid in planning fuel reduction burns.

Examples of the systems operation, including performance results for the NSW October 2013 fires will be provided in the presentation.

Savannah early dry season biomass burning – Campaign overview and a first look at aerosol chemical and microphysical properties from Gunn Point June 2014

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Abstract

Global climate models under predict aerosol optical depth (AOD) over regions impacted by fire, relative to satellite observations of AOD [Kaiser *et al*., 2012]. This may be due to parameterisations in global climate models assuming a diameter and chemical composition of the aerosol that more closely represents fresh smoke.

In June 2014 a sampling campaign was carried out in the Northern Territory at Gunn Point during the savannah early dry season biomass burning period to investigate the chemical and physical properties of aerosols and precursor gases in fresh and aged smoke plumes. Measurements included aerosol microphysical properties, aerosol chemical composition and reactive gases including volatile organic compounds. This was a collaboration between CSIRO, Bureau of Meteorology, University of Wollongong, Queensland University of Technology, Macquarie University, Curtin University, Aerodyne and ANSTO.

CSIROs new Time of flight Aerosol Chemical Speciation Monitor (ToF-ACSM) was deployed for the first time during this campaign. The ToF-ACSM enables near real time measurement of the chemical composition of non refractory aerosols, including organic components. We will present an overview of the campaign, a description of the ToF-ACSM and some preliminary results on the aerosol chemistry and microphysics.

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Fourier transform spectrometer results from the Gunn Point campaign, June 2014, including emission factors of greenhouse gases from savanna fires

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Abstract

In June 2014 an intensive measurement campaign took place at Gunn Point in the Northern Territory primarily aimed at characterising the emissions from early dry season savanna fires. A Fourier transform spectrometer was deployed and from this the concentration of CO_2, CO, N_2O, CH_4 and $^{13}CO_2$ over the measurement campaign was determined. Other measurements included aerosol microphysical properties, aerosol chemical composition, concentrations of reactive gases including volatile organic compounds, ozone and oxides of nitrogen.

This presentation will present results of CO_2, CO, N_2O, CH_4 and $^{13}CO_2$ from the Fourier transform spectrometer throughout the campaign. In particular individual fire events have been identified and emission factors calculated for CO_2, CO, CH_4 and N_2O. In the future, these measurements will enable the calculation of emission factors for many other species that have been measured simultaneously with some of the other instruments deployed.

Volatility-separated measurement of aerosol chemical composition and hygroscopicity from the Gunn Point campaign, June 2014

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Abstract

Queensland University of Technology (QUT) participated in the Gunn Point Field Savannah Campaign, held at the Australian Tropical Atmosphere Research Station at Gunn Point, Northern Territory between 28 May and 28 June 2014. The campaign was conducted in collaboration with the CSIRO, Bureau of Meteorology, University of Wollongong, Macquarie University and Aerodyne Research. Measurements were performed on aerosol primarily emitted from prescribed biomass burning events. Other sources of sampled aerosol may include continental dust and marine emissions, due to the coastal location. QUT contributed to the campaign through measurements performed with the Neutral cluster and Air Ion Spectrometer (NAIS), Aerodyne compact Time of Flight Aerosol Mass Spectrometer (c-ToF-AMS), and Volatility and Hygroscopicity Tandem Differential Mobility Analyser (VH-TDMA). The latter two instruments were deployed in a novel configuration to provide simultaneous volatility-separated measurement of aerosol chemical composition and hygroscopicity, respectively. The presentation will cover the analysis of the preliminary results as well as discussion on possible directions of future work.

Aerosol iron solubility: comparison between the Australian subtropics and Southern Ocean

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Abstract

Past changes in the atmospheric deposition of soluble, or bioavailable, trace metals to high nutrient low chlorophyll (HNLC) and nitrogen replete tropical waters have been shown to modulate primary production, atmospheric CO_2, and global climate. The deposition of soluble trace metals can also trigger toxic algal blooms, which impact Australia's fisheries and coral reefs. An understanding of the sources (e.g. mineral dust and biomass emissions) and geochemistry of soluble trace metals in atmospheric aerosols is critical for determining the impact of trace metal deposition on ocean fertility in the past and the future. However, to date no trace metal solubility data exists for biomass emissions from Australian fires and there are very few estimates of soluble trace metal aerosols entering the Southern Ocean.

Trace metal clean aerosols were collected during the early-late dry season experiment at Gunn Point, Northern Territory to investigate the trace metal aerosol solubility associated with biomass burning. Previous studies have suggested that mineral dust is the dominant source of trace metal aerosol. However, mineral dust is relatively insoluble and a significant fraction of soluble trace metals in the atmosphere could originate from biomass burning rather than mineral dust. Here we use the combination of soluble aerosol chemistry, back trajectories and diurnal and advective radon components to identify trace metal source regions throughout the campaign duration. We compare aerosol iron solubility at Gunn Point in the subtropics, where biomass burning can dominate the aerosol load in the dry season, to iron solubility in baseline air at Cape Grim which is representative of the Southern Hemisphere background. In doing this we highlight the importance of aerosol source at different latitudes for the solubility and bioavailability of trace metals.

Radon vertical profiles during the morning transition period

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Abstract

Vertical profiles and suitably-conditioned surface time histories of the natural radioactive noble gas radon-222 (radon) have long been demonstrated to be useful as quantitative indicators of diurnal- to synoptic-scale mixing processes within the continental lower troposphere. Radon's well-characterised and slowly-varying source function over (ice-free) terrestrial surfaces, together with its short half-life of 3.8 days, makes it a particularly suitable passive scalar for the evaluation of boundary layer and convective mixing parameterisation schemes in a range of regional and global climate and pollution transport models.

We provide a brief overview of ANSTO measurement programs using radon to characterise vertical mixing in the lower atmosphere, together with examples of their applications in modelling and pollution studies. We then present preliminary results from recent field campaigns collecting high resolution vertical radon profiles in the terrestrial boundary layer over rural New South Wales, using a radon sampler mounted on an instrumented motor-glider. The flights were conducted in the lowest 1000m of the atmosphere and, together with simultaneous ground-based and tower measurements, document the dispersion of radon emissions accumulated below the nocturnal stable inversion into the developing daytime convective boundary layer during the important morning transition period.



Figure 1. *Airborne Research Australia*'s "Eco-Dimona" aircraft engaged in early morning radon sampling over rural NSW

A radon-only technique for characterising baseline constituent concentrations at Cape Grim

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Abstract

Nine years (2004-2013) of hourly Radon-222, carbon dioxide and ozone concentration observations at Cape Grim are used to assess the residual terrestrial influence on air masses with radon concentrations below the 100 mBq m^{-3} threshold traditionally used for 'baseline' delineation (Figure 1a). Subsequently, a two-step radon-only approach for estimating 'baseline' constituent concentrations on monthly timescales is proposed. Based on a stringent 40 mBq m^{-3} radon threshold followed by a simple 10^{th}/90^{th} percentile constituent outlier removal, the technique is completely independent of meteorological or aerosol observations. An initial evaluation of the technique using hourly carbon dioxide and ozone records yielded monthly 'baseline' concentration estimates more consistent with expectations of minimally perturbed Southern Ocean air masses than existing baseline selection techniques (Figure 1c). This work builds upon prior studies that have identified radon as a valuable baseline criteria [e.g. Gras and Whittlestone, 1992; Molloy and Galbally, 2014].

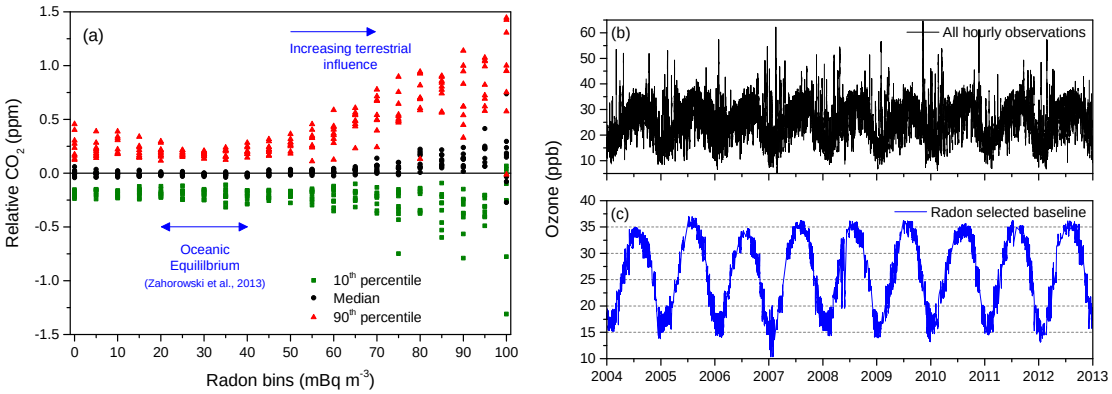


Figure 3. (a) Distributions of hourly CO_2 concentrations (monthly means have been removed) arranged into 5 mBq m^{-3} radon bins, for air masses containing less than 100 mBq m^{-3} of radon (each symbol represents one year of data); (b) all hourly ozone observations for 2004-2013; and (c) 'baseline' ozone concentrations as selected by the radon-based method.

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Open path Fourier transform spectroscopy measurements of greenhouse gases in Heidelberg

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Abstract

How representative are in situ measurements of greenhouse gases at a point in an inhomogeneous urban environment? Open path FTIR measurements potentially offer spatial averaging and continuous measurements of several trace gases (CO_2, CH_4, CO, N_2O) without sampling artefacts. Spatial averaging over kilometre scales is a better fit to the finest scale atmospheric models becoming available. With what precision, accuracy and reliability can such measurements be made?

Building on our pooled experience in open path Fourier transform spectroscopy and TCCON in the infrared (Wollongong) and long path DOAS techniques in the UV-visible (Heidelberg), we have set up an open path FTIR measurement system working in the near infrared across a 1.5 km path in urban Heidelberg, Germany. At one end of the path, an in situ FTIR analyser collects well calibrated measurements which can be compared to the path-integrated measurements. The system has run continuously since June 2014 to the present – we introduce the measurement system and present a preliminary analysis of the results to date.

Water-atmosphere exchange of greenhouse gases (CO_2, CH_4 and N_2O) measured on the Murray River, Australia

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Abstract

Global greenhouse gas (CO_2, CH_4 and N_2O) emissions from inland waters are significant, yet estimates remain highly varied and contain relatively large uncertainty [Regnier *et al*., 2013; Raymond *et al*., 2013]. For example, recent CO_2 estimates are comparable in magnitude to the net global uptake of CO_2 by the land or ocean sinks, yet these estimates range from 0.6 – 3.3 Pg C yr^{-1} [Regnier *et al*., 2013; Raymond *et al*., 2013, Richey, 2004; Tranvik, 2009; Cole *et al*., 2007; Battin *et al*., 2009; Aufdenkampe *et al*., 20011; Butman and Raymond, 2011; Bastviken *et al*., 2011]. To better constrain these estimates, and the carbon cycle as a whole, there is an urgent need for further ground-based measurements, particularly for CH_4 and N_2O, which have received far less attention than CO_2 [Bastviken *et al*., 2011; Beaulieu *et al*., 2011].

Here we present the *in situ* measurement and investigation of inland water-atmosphere greenhouse gas exchange as examined on the Murray River, Australia. Instrumentation was installed aboard a series of houseboats and measurements were made continuously, whilst cruising the river, for about one month during Nov-Dec 2013. Spatial variation was captured over several hundred kilometres on the Murray River with the inclusion of multiple reservoirs, major tributaries (e.g. the Darling River) and adjoining lakes and wetlands.

Gas analysis was performed using a Fourier-Transform InfraRed (FTIR) Trace Gas Analyser, which allowed simultaneous and continuous measurement of CO_2, CH_4, N_2O, CO and $\delta^{13}C$ in CO_2. The analyser was coupled to an equilibrator and a floating chamber, which facilitated either continuous measurement of gas partial pressures within the waters or the direct measurement of water-atmosphere gas exchange. Overall, we provide an overview of the results from the Murray River as well as the employed measurement techniques and their capabilities.

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Continuous measurements of $\delta^{18}O$-CO_2 in air with a Fourier Transform InfraRed spectrometer

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Abstract

The continuous in-situ measurement of $\delta^{18}O$ in atmospheric CO_2 opens a new door to differentiating between CO_2 source and sink components with high temporal resolution. Continuous ^{13}C-CO_2 measurement systems have already been commercially available for some time, but until now, only few instruments have been able to provide a continuous measurement of the oxygen isotope ratio in CO_2. Besides precise $^{13}C/^{12}C$ observations, the Fourier Transform InfraRed (FTIR) spectrometer also measures the $^{18}O/^{16}O$ ratio in CO_2, but the precision and accuracy of the measurements have not yet been evaluated. Here we present a first analysis of $\delta^{18}O$-CO_2 (and $\delta^{13}C$-CO_2) measurements with the FTIR analyser in Heidelberg. We used Allan deviation to determine the repeatability of $\delta^{18}O$-CO_2 measurements to be 0.25‰ for ten minute averages reducing to about 0.1 ‰ after two hours. We evaluated the repeatability of measurements over a 10 month period to be around 0.3‰ for 10 minute measurements. FTIR and IRMS measurements of a series of air samples do not differ significantly within measurement errors.

We discuss two weekly episodes of ambient air measurements in Heidelberg, one in winter and one in summer, for an initial assessment of the potential insights and challenges that combined, highly resolved $\delta^{13}C$-CO_2 and $\delta^{18}O$-CO_2 records may provide in terms of better understanding regional scale continental carbon exchange processes.

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New gas chromatographic technologies for GASLAB: pulsed discharge helium ionisation detector for analysis of trace gases

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Abstract

Since the establishment of GASLAB (Global Atmospheric Sampling LABoratory) in 1990, five important atmospheric trace gases (CO_2, CH_4, N_2O, CO, and H_2) have been measured in thousands of air samples, using the technique of gas chromatography (GC) with packed columns. Over this 24-year period, only three gas chromatographic instruments have been used: a Carle Series 400 with flame ionisation detector (FID) to measure CH_4 and CO_2; a Shimadzu GC-8A with electron capture detector (ECD) to measure N_2O; and a Trace Analytical RGA-3 with a heated mercuric oxide detector to measure H_2 and CO. While these systems have been able to deliver consistent and high performance over this long period, they are due for replacement by more modern instruments.

The GC detector which has a range of attractive features for our purpose is the Valco pulsed discharge helium ionisation detector. When used with pure helium as the discharge gas (that is, without any addition of trace amounts of noble gases), the ionisation energies available in the detector are high enough for effective detection of all 5 species we seek. Use of this detector would allow the elimination of the need for the use of hydrogen gas (fuel gas for the FID, and reagent gas for the nickel catalyst); it would also allow the elimination of radioactive nickel (^{63}Ni) in the ECD, and mercuric oxide in the RGA-3 detector. Carbon dioxide could be detected without the need to first convert it to CH_4 with a heated nickel catalyst. With a judicious choice of operating conditions, this detector is also likely to have linear response over the range of concentrations likely to be encountered for most air samples. But a universal detector such as this Valco detector also brings a range of challenges which must be overcome. The chromatographic separations must be exceedingly good. For example, with this detector it is necessary to achieve complete column separation of the required analytes from the massive oxygen/nitrogen peak, as well as separation between the analytes themselves. Leaks of air at column connections and other parts of the 'plumbing' need to be rigorously eliminated because the detector responds strongly to any unwanted source of oxygen or nitrogen, introducing noise and high background.

Here we present initial results for H_2, CH_4, CO_2, and N_2O, as well as for two noble gases (neon and krypton). Carbon monoxide remains elusive so far, since the CO peak is not separated from the large air peak.

Part of the development of these new GC techniques involves the software. It has been decided to use the GCWerks package, since it provides a powerful, integrated platform for instrument control, peak integration, data management and data processing. Some of these features will be highlighted.

New insights into global perfluorocarbon emissions

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Abstract

Perfluorocarbons (PFCs) are very persistent, potent greenhouse gases (GHGs) largely emitted to the atmosphere from aluminium smelting and the production of microchips for the semiconductor industry. Small quantities of PFCs are used/emitted as speciality refrigerants and possibly from the smelting of rare earths minerals. Perfluoromethane (CF_4) accounts for 92% (molar) of PFCs in the atmosphere, with about half from natural sources (fluorite minerals), perfluoroethane (C_2F_6) accounts for 4.9%, perfluorocyclobutane 1.5% (c-C_4F_8) and perfluoropropane (C_3F_8) 0.7%.

Industrial PFC emissions are estimated by the aluminium and semiconductor industries, using Intergovernmental Panel on Climate Change (IPCC) protocols, and are reported as national aggregates to the United Nations Framework Convention on Climate Change (UNFCCC). To date, only developed nations are required to report PFC emissions, so the current global UNFCCC PFC emissions inventory significantly underestimates actual global PFC emissions.

Advanced Global Atmospheric Gases Experiment (AGAGE) established a global network of PFC monitoring stations in 2004 and, using *in situ* and air archive (Cape Grim, La Jolla) PFC measurements, has accurately estimated global PFC emissions back to the mid-1970s [Mühle *et al*., 2010]. Global PFC emissions estimated from atmospheric observations are more than a factor of 2 higher than those in the UNFCCC inventory.

In situ PFC measurements at Gosan, South Korea, at Cape Grim, Tasmania, and at Aspendale, Victoria, have identified unique C_2F_6/CF_4 emissions ratios for the aluminium smelting and the semiconductor industries [Kim *et al*., 2014]. The global semiconductor industry is the major source of these 'missing' PFC emissions. The global PFC emissions from the aluminium industry outside of China are largely consistent with atmospheric observations, but once aluminium PFC emissions from China are included, then the global aluminium industry is also a significant source of these 'missing' PFC emissions.

Part of the under-reporting of PFC emissions by the aluminium industry, in particular in China, may be because the emissions estimation protocols only account for episodic PFC emissions above a defined threshold. Increasingly accurate PFC measurements at individual smelters are suggesting that significant PFC emissions are occurring that are quasi-continuous and 'sub-threshold' [Wong *et al*., 2015]. The IPCC methodologies for estimating PFC emissions need to be revised to account for these 'sub-threshold' emissions.

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Signatures of human actions recorded in the atmosphere

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Abstract

Systematic, well-calibrated measurements of atmospheric mixing ratios of trace gases can be used to derive estimates of their global emissions. A recent summary of such results for 25 of the long-lived synthetic greenhouse gases (SGHGs) measured as part of AGAGE (Advanced Global Atmospheric Gases Experiment) has been published by Rigby *et al*. [2014]. In Figure 1, the estimated annual global emissions for four of these trace species are shown as per capita annual emissions, using estimated human population in each year as the divisor. The first vertical dashed line is drawn for the year 1974, the year of publication in Nature of the famous paper by Molina and Rowland [1974]. The second vertical dashed line is drawn for the year 1989, when the Montreal Protocol on Substances that Deplete the Ozone Layer entered into force. Features in the figure will be discussed, and such results for several other trace species will be presented.

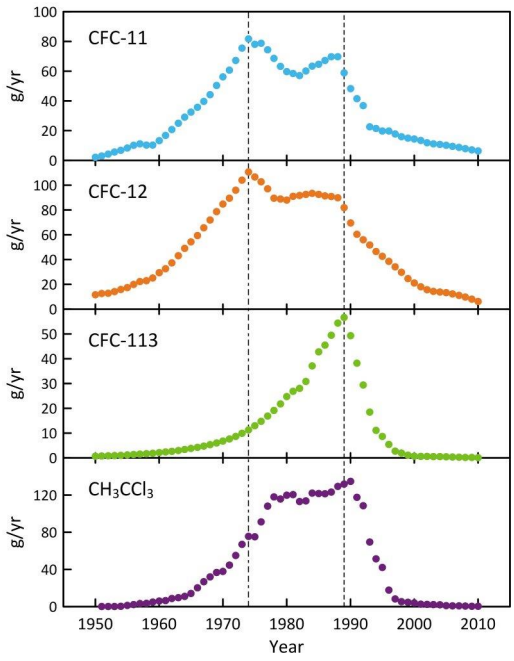


Figure 1. Estimated per capita annual emissions of 4 man-made trace gases

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Open-Path FTIR Measurements of Trace Gas Emissions from Australian Eucalypt Fires in the CSIRO Pyrotron

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Abstract

Australia's hot, dry and drought-prone climate triggers frequent wildfires with the most dangerous period in summer and autumn for its southern part. In the period 2001-10, wildfires burnt a total of 12 Mha of southern Australian forests releasing large emissions of toxics (H_2CO, Hg, etc.) and greenhouse gases (CO_2, CH_4, etc.) into the atmosphere. Australian gross annual emissions of total carbon from fires are similar to emissions from burning of fossil fuels. Safe ways for the scientific community in Australia to study those bush fires in detail are prescribed fires and laboratory facilities (such as the CSIRO Pyrotron in Canberra) designed to study fire combustion.

In this poster, we report Open-Path FTIR measurements (see Figure 1) for online characterisation and quantification of trace gases in the smoke from Australian eucalypt fires.



Figure 1. The Open-Path FTIR measuring across the CSIRO Pyrotron exit.

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Australian Biogenic HCHO Observations from the OMI and GOME-2 Satellites

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Abstract

Biogenic VOCs and in particular isoprene are major contributors to ozone and aerosols. Models show high isoprene emissions over Australia however there are few in situ measurements available to test these values. The sparseness of measurements and uncertainties about precursor sources could explain why models over northern Australia are not good at reproducing observed data [Stavrakou *et al*., 2009]. Models show a hotspot over SE Australia but this is yet to be tested. HCHO is an intermediate chemical product and can be used as a proxy for determining biogenic isoprene emissions. Isoprene and HCHO can be related via linear regression, and this has been used successfully in several countries including North America [Palmer *et al*., 2003], South America [Barkley *et al*., 2013], and Africa [Marais et al., 2012].

A 12 year record of satellite measurements of HCHO is available from the GOME, GOME-2, OMI, and SCIAMACHY instruments. Here we use these data to show seasonal, spatial and interannual variability in HCHO over Australia. There is a seasonal pattern in Australian HCHO emissions with a summer peak and winter minimum, likely driven by biogenic sources. OMI and GOME-2 show similar seasonal variability, but with some differences in spatial distribution. We also present some preliminary comparisons to two different versions of the GEOS-Chem model, both run with isoprene emissions from the Model of Emissions of Gases and Aerosols from Nature(MEGAN). Both versions show large positive biases in HCHO which we infer to reflect biases in Australian isoprene emissions in the MEGAN model. In future work we will use these data to provide quantitative constraints to Australian isoprene emissions.

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New particle formation events over Antarctic sea-ice

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Abstract

Observations of atmospheric aerosols in the Antarctic sea-ice are rare. Here we present a case-study of a new particle formation event from data collected in the East Antarctic region (~120°E, ~65°S) as part of the second Sea Ice Physics and Ecosystems eXperiment (SIPEXII). A single air-mass encompassed the measurement site for a period of about 14 hours, during which solar radiation was the highest it had been during the 32 day cruise. Approximately 2 hours after widespread clouds cleared, number concentrations of particles larger than 3 nm began to rise rapidly from background levels of around 400 cm^{-3} to over 8500 cm^{-3} within a few hours. Just 1.25 hours after the first aerosol signal of the event, number concentrations in the >10 nm size range began to rise, giving a growth rate of around 5.8 $nm.hr^{-1}$. A period of decline, caused by coagulational losses occurred after sunset, with a rapid return to background values resulting from an air-mass change observed in numerous parameters.

Investigation of precursor identity using modelling studies of known chemistry and concurrent measurements rules out sulfur and halogen species as part of the nucleation chemistry. Time-lagged negative-correlations of aerosol with Hg^0, combined with real-time positive correlations beginning when Hg^0 recovery began, suggest nucleation may be catalyzed by Hg, after which growth occurs via sulfur chemistry. This is supported by simple box modelling studies. This chemistry is likely only possible in the unique environment of polar regions where pre-existing aerosol surface area is low, preventing scavenging of Hg oxidation products.

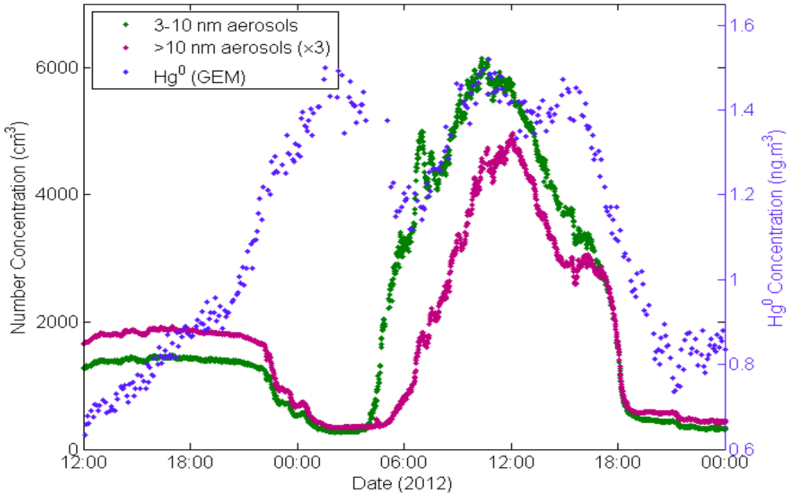


Figure 1. Ultrafine aerosol number concentrations during the new particle formation event on the 18th October, 2012, along with Hg0 concentrations.

How productive is Earth's terrestrial biosphere? Novel satellite measurements and methods to understand global primary productivity

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Abstract

New satellite observations of solar-induced plant chlorophyll fluorescence (SIF) offer a promising constraint on photosynthesis and gross primary productivity (GPP) of terrestrial ecosystems globally [Frankenberg *et al.*, 2011]. GPP is the largest global surface-to-atmosphere carbon flux, yet current estimation methods show significant uncertainties [Beer *et al.*, 2010]. This project will investigate the benefit of combining these new measurements of SIF, more closely linked with photosynthetic processes than traditional remote sensing indices [Porcar-Castell *et al.*, 2014], with ancillary prior and biosphere model information in mapping the spatiotemporal distribution of GPP. A data assimilation approach will be used for SIF measurements with two biosphere models, BETHY and CABLE, to be performed sequentially. A model SCOPE already includes a module for simulating canopy SIF, which will be coupled to the two biosphere models as the SIF observation operator. A carbon cycle data assimilation system already exists for BETHY [Rayner *et al.*, 2005], while one will have to be built around CABLE to enable the assimilation of SIF measurements and investigation of their utility in estimation of GPP.

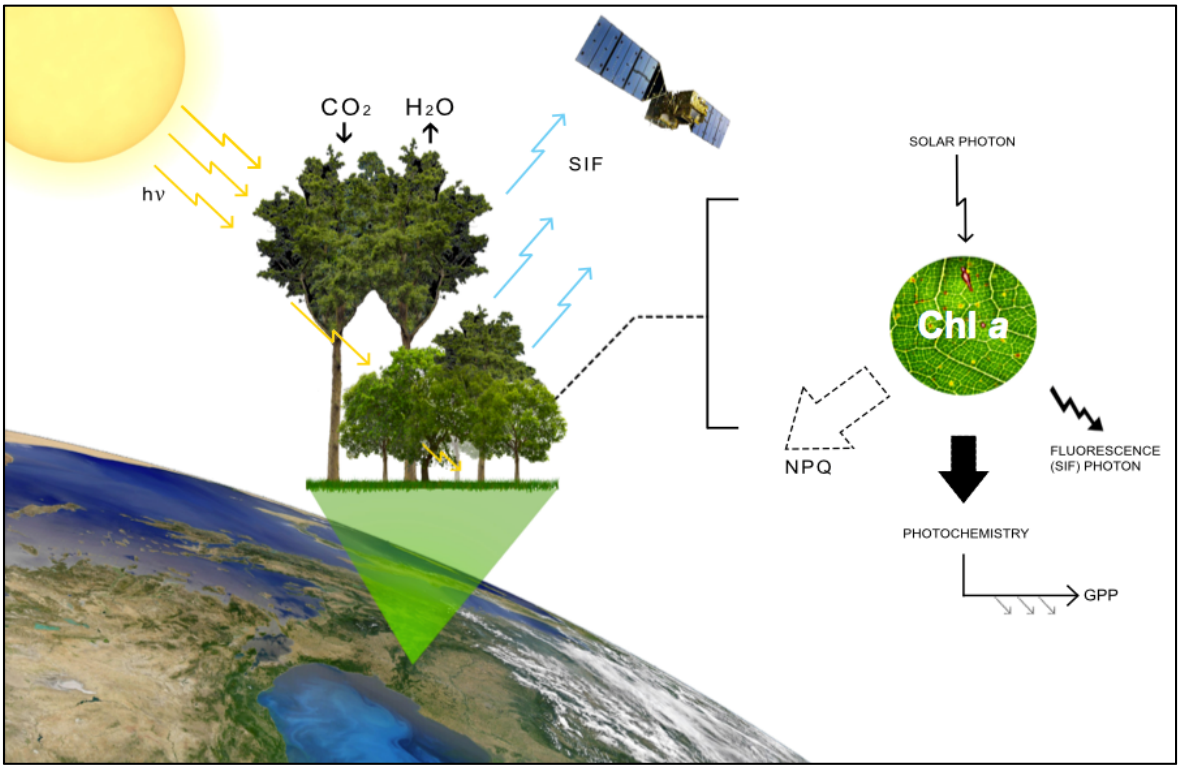


Figure 1. Solar-induced chlorophyll fluorescence (SIF) emission at the canopy and molecular level, retrieved by satellites. Chl a = chlorophyll a; NPQ = non-photochemical quenching of SIF.

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Constraining greenhouse gas emissions in the Latrobe Valley

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Abstract

Motivation for the project: Anthropogenic emissions of trace gases especially CO_2 from burning fossil fuel has major influence on climate change. Regional inversions are extremely useful tool for detecting local changes in climate due to land use changes, anthropogenic emissions and biospheric emissions. Estimating carbon exchange at regional scales is vital for understanding feedbacks between climate and the carbon cycle, but also to verifying climate change mitigation such as emission reductions. High-resolution spatial and temporal regional inversion provides a mean to reduce uncertainties and variability especially for continental land surface exchanges that are highly variable in time and space and closely related to land use and climate variability.

Objective

- Constrain the emissions of greenhouse gases in the Latrobe Valley
- Provide baseline atmospheric CO_2 concentration for Carbon Capture and Storage project (CCS)

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The ACCESS-UKCA chemistry-climate model: exploring long-term trends in atmospheric composition

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Abstract

ACCESS is the Australian Community Climate Earth-System Simulator [Bi *et al*., 2013]. ACCESS includes the United Kingdom Chemistry and Aerosol scheme (UKCA) [O'Connor *et al*., 2013], which can be coupled to the radiation scheme to simulate chemistry-climate feedbacks. Recent developments to ACCESS-UKCA include the use of a more comprehensive tropospheric chemistry scheme (including isoprene chemistry), and the possibility to simulate stratospheric chemistry and whole-atmosphere aerosol.

ACCESS-UKCA is used here to simulate global tropospheric chemistry over recent decades. The simulations are nudged to ECMWF ERA-interim reanalysis meteorology, conducted at 1.875 degrees x 1.25 degrees resolution, with 85 vertical levels extending to 85 km in height, with prescribed sea-surface temperatures. Anthropogenic emissions are time-varying, derived from the ACCMIP dataset.

In this work, we compare long term observations of atmospheric composition (including ozone and carbon monoxide taken at remote sites including Antarctica, Cape Grim and Greenland over multiple decades) to modelled concentrations. The comparisons build on previous work, and give further confidence in the ACCESS-UKCA modelling system.

Testing is under way of the enhanced coupled tropospheric-stratospheric chemistry scheme. This latest configuration (including aerosol) will soon be available to conduct coupled-chemistry climate simulations, and represents a world-leading chemistry-climate model.

We acknowledge the long term commitment of the Australian Antarctic Division and the Bureau of Meteorology in obtaining the Antarctic samples and the support of CSIRO and the Bureau of Meteorology for Cape Grim. The Antarctic measurements were supported in part by the Australian Climate Change Science Program.

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Conventionally-heated microfurnace for graphitisation of microgram-sized carbon samples

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Abstract

We present recent developments in design, construction and testing of a novel, externally-heated graphitisation reaction vessel we named the 'Micro Conventional Furnace' (MCF). Heating is by means of an electrically-heated 'tube furnace' inside which the reaction volume is placed. Carbon dioxide sample gas and hydrogen are introduced into the reaction volume, which contains about one milligram of an iron catalyst, and the Bosch reaction proceeds when this is heated to about 600°C. Based on our systematic studies aimed at optimising the performance of the first laser heated microfurnace (mark-I MF) [Yang *et al*., 2013; Smith *et al*., 2007, 2010], we have designed both the MCF and the second generation of laser heated microfurnace (LHF). The MCF is a cost-effective solution for producing graphite from carbon dioxide sample gas. We have made two types of MCF: the type 1 furnace, with an integral stainless steel cold finger and the type 2 furnace, with changeable glass tube cold finger. The MCF have the following features: 1) it has a small volume of 0.9 ml (type 1) and adjustable volume from 1.3 ml to 5 ml (type 2) allowing us to completely graphitise carbon dioxide samples containing from 5 micrograms to 2000 micrograms of carbon, 2) it can operate over a large pressure range between 0 and 3 bar; 3) it is compact, with three valves integrated into the MCF body, 4) these valves can be controlled electronically, enabling automation in the future of the process through the LabVIEW™ interface, 5) it is compatible with the new version laser heated microfurnace and shares a common vacuum system. During graphitisation, the reaction pressure, cold finger temperature and other parameters are logged by LabVIEW™. Experimental results with the MCF are presented and discussed.



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