Atmospheric Composition Observations and Modelling Conference

incorporating the

Cape Grim Annual Science Meeting 2012

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at

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South Durras, NSW



Edited by: C. Paton-Walsh, N. Derek & P. B. Krummel

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Day 1

Wednesday 28 November 2012

09:00	**Opening**	**Welcome from Organisers, University of Wollongong & Cape Grim Program**

Chair	**Clare Paton-Walsh**	**Oral Session 1**
09:10	Melita Keywood	Persistent Organic Pollutants at Cape Grim: an update
09:30	Paul Fraser	Australian carbon tetrachloride (CCl_4) emissions: a paradigm for a 'missing' CCl_4 source of possible global significance? PFC emissions from Australian aluminium smelters and the Kurri Kurri experiment SE Australian fumigant emissions
10:10	Xue Shi	Ensemble empirical mode decomposition of monthly CO_2 measurements and GEOS-Chem simulations

10:30	**Morning Tea**	**provided - poolside**

Chair	**David Griffith**	**Oral Session 2**
11:00	Stephen Wilson	**Spectral Solar Radiation 1986 - 2011 - Review**
11:40	Joseph Michalsky	Four Years of field measurements of surface albedo in the western United States
12:00	Paul Krummel	The 2012 Ozone hole and EESC calculations
12:20	John Ivey	Carbondisulfide measurements at Cape Grim, marine, terrestrial and anthropogenic contributions (mining the tailings of the Sulphur Program)

12:40	**Lunch Break**	**provided - Poolside**

Chair	**Paul Krummel**	**Oral Session 3**
14:10	Chris Caldow	Greenhouse gas exchange between inland waters and the atmosphere: a case study from the Elbe River, Germany
14:30	Ruhi Humphries	Atmospheric measurements over East Antarctic melting spring sea ice
14:50	Ann Stavert	The Southern Ocean CO_2 Project: observations and modelling
15:10	Dagmar Kubistin	Measurements of greenhouse gases and ozone in the Southern Pacific Ocean

15:30	**Afternoon Tea**	**provided - Poolside**

Chair	**Marcel van der Schoot**	**Oral Session 4**
16:00	Colin Allison	The MURL SF6 release and what happened next
16:20	Andrew Smith	Early results from the ANSTO/NIWA ^{14}C of atmospheric methane program
16:40	Tilo Ziehn	Designing a greenhouse gas network for Australia
17:00	CG Working Group	**Cape Grim Working Group Meeting**

19:00	**Dinner**	**provided – Buffet in the Bistro**

Day 2

Thursday 29 November 2012

Chair	Stephen Wilson	Oral Session 5
09:00	Sarah Lawson	Composition of fresh and aged biomass burning plumes from the 2006 Robbins Island fire – Cape Grim
09:20	Clare Paton-Walsh	Measurement of trace gas emissions from Australian hazard reduction burns using open-path Fourier transform infrared spectroscopy
09:40	Elise-Andree Guerette	Measurement of trace gas emissions from Australian hazard reduction burns using Selected Ion Flow Tube Mass Spectrometry and Fourier Transform Infrared Spectrometry
10:00	Fabienne Reisen	Smoke impacts on the fire ground
10:20	Jenny Fisher	Identifying sources and transport pathways of carbon monoxide over Australasia using a Chemical Transport Model constrained by surface-based and satellite data
10:40	**Morning Tea**	**provided - Poolside**
Chair	**Melita Keywood**	**Oral Session 6**
11:10	David Griffith	An ^{15}N tracer study of N_2O emissions from soil
11:30	Peter Rayner	Constraining emissions at high-resolution using measurements from geostationary orbit: A preliminary Observing system Simulation Experiment
11:50	David Etheridge	Revised records of greenhouse gases and the variability in the carbon cycle over the past 2000 years
12:10	Poster Presenters	All poster presenters to give a 2 - 3 minute poster introduction
12:40	**Lunch Break**	**provided - Poolside**
14:20		**Poster Session**
Rebecca Buccholz		Total Column CO and H_2CO at three Southern Hemisphere sites, investigated with FTIR measurements, and the models GEOS-Chem and ACCESS
Min Cheng		Volatile Organic Compounds in typical Australian dwellings
Ulla Heikkilä		Seasonal variability of solar proxy Beryllium-10 in Law Dome snow, Antarctica, and its connection to stratosphere-troposphere exchange
Ashok Luhar		A new global chemical emissions database and meteorological nudging for ACCESS
Suzie Molloy		Identification of a suitable baseline definition of tropospheric ozone at Cape Grim, Tasmania
Kay SteinKamp		Footprinting Australasian CO_2 measurement stations using high-resolution Lagrangian dispersion modelling
Voltaire Velazco		The Total Carbon Column Observing Network (TCCON): Australia's continuing contribution
15:30	**Afternoon Tea**	**provided – Poolside**

Day 2 continued…….

Thursday 29 November 2012

Chair	Nicholas Jones	Oral Session 7
16:00	Ian Galbally	Tropospheric ozone distribution and trends at Cape Grim and in the extra-tropical Southern Hemisphere
16:20	Sam Cleland	Cape Grim wind and baseline climatology
16:40	Scott Chambers	Identifying baseline air masses in the lower troposphere at Mauna Loa Observatory between 2004 and 2010 using Radon-222 and back trajectories
17:00	Perry Davy	The influence of marine aerosol concentrations on New Zealand urban air quality and source region determination
19:00	**Dinner**	**provided – Beach Bistro**

Day 3

Friday 30 November 2012

Chair	Clare Paton-Walsh	Oral Session 8
09:00	Nicholas Jones	High precision CO_2 measurements using a small low resolution spectrometer
09:20	Steve Utembe	Modeling *in situ* and column integrated CO_2 observed during the Greenhouse Gas Monitor experiment in Port Augusta in May 2012
09:40	Kathryn Emmerson	Lauder gas and aerosol observations, modelled with the new Southern Hemisphere version of UKCA
10:00	Ashok Luhar	An inverse system for locating and quantifying greenhouse gas leakage, and its testing using Otway data
10:20	Jocelyn Turnbull	Quantification of fossil fuel CO_2 emissions at the urban scale
10:40	**Morning Tea**	**provided - Poolside**

Chair	Ian Galbally	Oral Session 9
11:10	Lingxi Zhou	Atmospheric Greenhouse Gases Observations at the Chinese GAW stations and Modelling Attempt
11:40	Alastair Williams	Bulk Richardson Number, surface decoupling and the mixing of passive scalars in the nocturnal stable boundary layer over land
12:00	Sarah Lawson	Surface Ocean Aerosol Production (SOAP) in NZ waters
12:20	Paul Steele	Trend and seasonal cycle of atmospheric carbon monoxide at Cape Grim
12:40		Closing remarks
12:40	**Lunch Break**	**provided – Poolside**

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Persistent Organic Pollutants at Cape Grim: An update

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Abstract

Persistent Organic Pollutants (POPs) are compounds that resist degradation and once in the environment, can cycle through environmental media, travel long distances from its source and bio-accumulate in humans and other biota, resulting in adverse effects on human and ecosystem health.

As a Party to the Stockholm Convention on Persistent Organic Pollutants, Australia has committed to eliminating or restricting the production and use of certain POPs. As part of this commitment, Parties need to assess the effectiveness of the Convention by monitoring changes in the concentrations of POPs over time in the environment. To this end a Pilot Air Monitoring Project has been established across Australia by CSIRO and the Department of Sustainability, Environment, Water, Population and Communities.

This network will operate between January 2011 and December 2014. During 2011 and 2012, the network has comprised of 3 sites around Australia (Cape Grim, Darwin and Aspendale in Melbourne, Victoria, 38.025°S 145.102°E). The network will be extended to seven additional locations in 2013. POPS measured include number of chemicals falling into five groups of compounds, polychlorinated dibenzo-p-dioxins and furans (PCDD/F), polychlorinated biphenyls (PCB), organochlorine pesticides (OC), brominated diphenyl ethers (BDE), and perfluoro compounds (PFC).

In this presentation, data from the first 18 months of the Pilot Monitoring Program with a focus on the Cape Grim site will be reported. The data collected during this time will be compared with data collected during earlier studies of POPs in Australia. The design for the extended network will also be presented.

Australian carbon tetrachloride (CCl_4) emissions: a paradigm for a 'missing' CCl_4 source of possible global significance?

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Abstract

In Chapter 1 of the Scientific Assessment of Ozone Depletion: 2010 (Montzka & Reimann, 2011), 'bottom-up' estimates of global carbon tetrachloride (CCl_4) emissions, based on fugitive emissions from the production, use and destruction of CCl_4, as recorded by UNEP (with some adjustments and additions), fall well short (currently by about 50 Gg/yr) of 'top-down' estimates of global emissions derived from AGAGE and NOAA global atmospheric observations.

Australian production of CCl_4 ceased in the 1980s and Australian consumption of CCl_4 effectively ceased in the early 1990s, when imports were severely restricted, following Australia's signing (1990s) of the Montreal Protocol. However the long-term AGAGE CCl_4 record at Cape Grim (1978-2012; Xiao *et al*., 2010; Krummel *et al*., 2011) shows significant, but relatively small, CCl_4 emissions from SE Australian urban and industrial centres (Dunse *et al*., 2005; Figure 1).

Australia's contribution to the global fugitive emissions described above is essentially zero, so where do the Australian emissions come from? This paper will report an update of current CCl_4 emissions from the Melbourne/Port Phillip/Latrobe Valley region of SE Australia, based on Cape Grim in situ GC-ECD CCl_4 data, using inter-species correlation (Dunse *et al*., 2005) and regional transport modelling (NAME; Manning *et al*., 2011). We attempt to identify the location and nature of these sources within the Melbourne/Port Phillip region, using in situ GC-MSD measurements of CCl_4 at CSIRO, Aspendale. The possible global significance of these emissions will be discussed.

Some previously unidentified CCl_4 sources will be discussed.

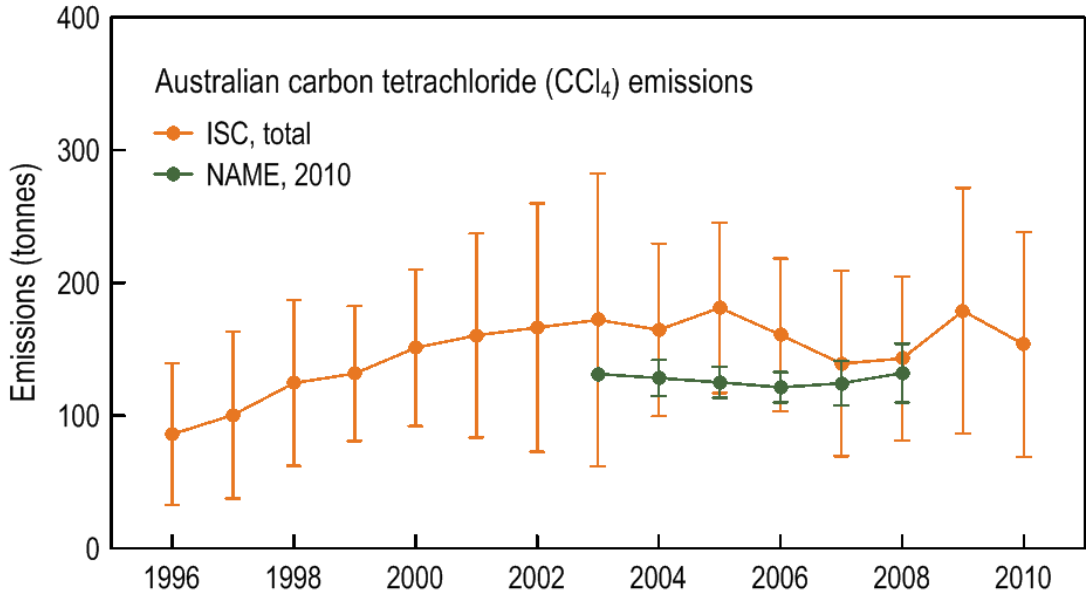


Figure 1. Australian CCl_4 emissions obtained from AGAGE GC-ECD CCl_4 observations at Cape Grim, Tasmania (1995-2011), using interspecies correlation (ISC, Dunse *et al*., 2005) and inverse modeling via the Lagrangian particle dispersion model NAME (Manning *et al*., 2011).

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PFC emissions from Australian aluminium smelters and the Kurri Kurri experiment

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Abstract

In situ PFC measurements at Cape Grim, Tasmania, and Aspendale, Victoria, have been used to estimate emissions and emission factors for CF_4 and C_2F_6 from Australian aluminium smelters. The data show declining PFC emissions with time (2004-2011), consistent with the 'bottom-up' estimates of Australian PFC emissions contained in the National Greenhouse Gas Accounts published annually by DCCEE. Both these 'top-down' and 'bottom-up' estimates of Australian PFC emissions have been reported to the UNFCCC. The recent emissions and emission factors from atmospheric data are higher than those contained in the Greenhouse Gas Accounts, although their relative uncertainties likely overlap.

More accurate estimates of PFC emissions and emission factors have been made from a stack-sampling experiment at the Kurri Kurri aluminium smelter, in the Hunter Valley, in 2009. The Kurri Kurri results are compared to those obtained remotely at Cape Grim for three SE Australian smelters (Portland, Port Henry, Bell Bay). The overall agreement between the two approaches is encouraging, with the Kurri Kurri results producing, as expected, a more precise estimate of emissions and emission factors from an individual smelter.

SE Australian fumigant emissions

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Abstract

Recent results from Cape Grim, Tasmania, show a significant increase in the SE Australian emissions of the grain fumigant sulfuryl fluoride (SO_2F_2), which is being used to replace phosphine (PH_3) in regional grain storage. SO_2F_2 emissions have been estimated by inter-species correlation (ISC), with carbon monoxide (CO) as the reference species, and by inverse techniques using NAME/INTEM. SE Australian methyl bromide emissions (CH_3Br) are now largely from quarantine/pre-shipment (QPS) use in grain storage on Australian docks, prior to export. SE Australian CH_3Br emissions have been estimated by ISC and are compared to emissions from a simple UNEP 'bottom-up' usage model that incorporates emissions factors for both QPS and non-QPS CH_3Br use.

Ensemble Empirical Mode Decomposition of monthly CO_2 measurements and GEOS-Chem simulations

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Abstract

With the increasing availability of new sources of atmospheric data, time series analysis has been playing a more prominent role in atmospheric and climate studies. Time series analysis of greenhouse gas concentrations in the atmosphere reveals different patterns of their variability (e.g. diurnal, synoptic, seasonal, annual, and other concealed time scales). Atmospheric and climatic phenomena are highly non-stationary and nonlinear, and traditional spectral analysis is generally based on certain assumptions of the data, such as its linearity and stationary nature. Decomposing a nonlinear time series, will often produces physically meaningless harmonics. In this study, we used Ensemble Empirical Mode Decomposition (EEMD; Wu, 2009) to accommodate the variety of data generated by nonlinear and non-stationary processes, which can decompose a time series adaptively into time-frequency space and produces a set of independent modes called Intrinsic Mode Functions (IMFs).

CO_2 is the single most important anthropogenic climate-relevant greenhouse gas in the Earth's atmosphere, being the one that controls climate change (Lacis, 2010). In addition to the trend, different time scale oscillations have been found which may relate to biospheric influence or the atmospheric circulation variability. In this study, we focus on the surface CO_2 time series from baseline stations in Cape Grim, Australia and Barrow, Alaska and use EEMD to investigate the climate relevant surface CO_2 oscillation. To determine the possible cause or driving force of the oscillations, and understand the mismatching in model and observation, we use the chemistry transport model GEOS-Chem (Nassar, 2010) to simulate the surface emissions, and compare them with the IMFs decomposed by EEMD.

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Spectral Solar Radiation 1986 - 2011

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Abstract

Solar radiation measurements have been an integral part of the Cape Grim measurement program since the 1980s. The Solar Radiation program is being reviewed as part of the cycle of program reviews for Cape Grim. This talk will summarize both the measurement/calibration process and some key results.

The spectral measurements to be discussed here began with the deployment of a sunphotometer on an active solar tracker in 1986. This instrument, designed to quantify aerosol optical depth at several wavelengths, has been replaced with several other instruments over the intervening period. The analysis of the record up to 1999 (Wilson & Forgan, 2002) has been extended to 2011 with a clearer picture of the annual cycle emerging.

UV measurements were added to the program, and six years of measurements of UV-B have produced estimates of O (^{1}D) photolysis rates which demonstrate a clear dependence on the total ozone column and quantify the impact of clouds on the photolysis rates in this cloudy environment.

These measurements will soon be extended with the use of DOAS (Direct Sun and Multi-Axis) based techniques for determining atmospheric composition. This will be demonstrated with measurements from trials at Wollongong measuring column formaldehyde and some of the characteristics of the measurements at Wollongong will be summarised. While the measurements are automated, the analysis remains labour intensive for us, a bottleneck that will need to be resolved before automated measurements can successfully be deployed.

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Four years of field measurements of surface albedo in the western United States

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Abstract

Quasi-continuous measurements of spectral surface reflectivity (albedo) have been made for over four years at an undisturbed site on the western U.S. prairie near Boulder, Colorado USA. Analyses of these data reveal some interesting cases where biases, if ignored, lead to wrong conclusions about the character and magnitude of the albedo. Two of the six filters of the multi-filter radiometer (MFR) are used to define the Normalized Difference Vegetative Index (NDVI) that can be used to measure the state of the vegetation at the site. Examples of the wavelength dependence and solar-zenith angle dependence for stages of vegetative growth and senescence are illustrated. Some examples of wavelength and solar-zenith angle dependence for fresh snow that do not agree with prior measurements are also discussed. Finally, plans for extending the wavelength coverage and adding more sites are outlined.

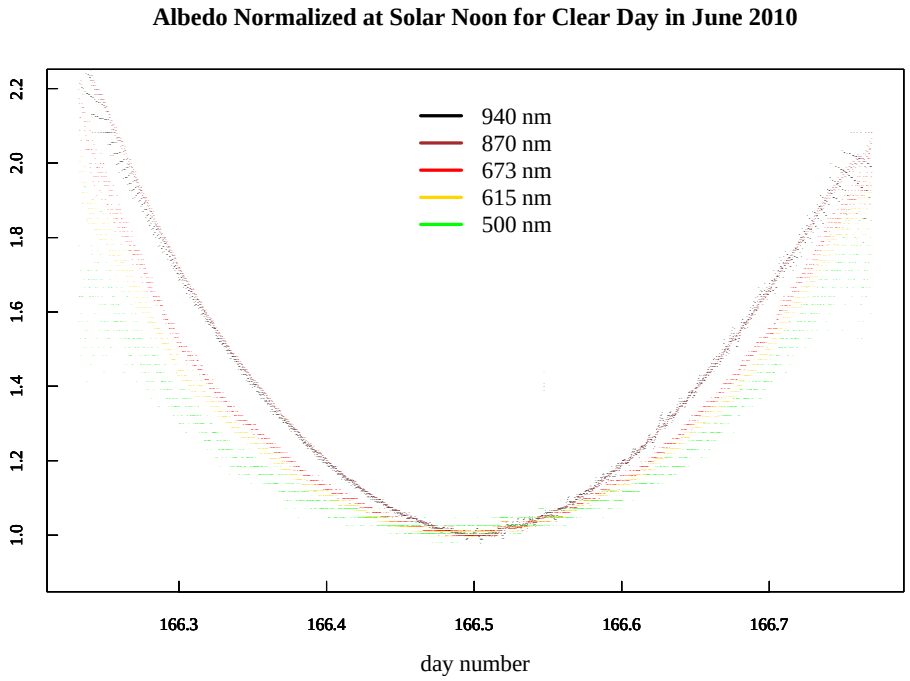


Figure 1. Early summer spectral albedos on the western U.S. prairie normalized at solar noon. The end points are at a solar-zenith angle of 80° and the noon solar-zenith angle is 18º. The higher incident angle enhancements are asymmetrical, similar at each wavelength, and about a factor of two higher compared to solar noon.

Reference

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The 2012 Ozone Hole and updated Equivalent Effective Stratospheric Chlorine data

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Abstract

The 2012 ozone hole was one of the smallest since the late 1980's due to a warmer than normal mid-lower stratosphere over the Antarctic region this year. The development and recovery of the 2012 ozone hole will be shown via a set of metrics based on the Ozone Monitoring Instrument (OMI) on board the Earth Observing Satellite (EOS) Aura. Other observational results will be shown to complement these satellite data. The 2012 ozone hole metrics will be compared to the historical metrics, and these will be linearly regressed against Equivalent Effective Stratospheric Chlorine (EESC) data. The EESC data are based entirely on Cape Grim Air Archive, Antarctic firn and AGAGE global measurements of ozone depleting substances, and are calculated using the new formulation outlined in Newman *et al*. (2006) and Newman *et al*. (2007). A brief overview of the EESC calculation method and the trace gas observational data used in the calculation will be given.

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Carbondisulfide measurements at Cape Grim, marine, terrestrial and anthropogenic contributions (Mining the tailings of the Sulphur Program)

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Abstract

This work reports our revisit to the on line sulfur gas data record gathered between 1993 and 1996 as a component of the Sulfur Program. The original data gathering was primarily focused on the dimethylsulfide (DMS) flux and contribution via oxidation products to tropospheric sulfur particle production. These acting as precursors for cloud condensation nuclei (CCN) either, discretely, or synergistically on sub micron sea salt.

Carbondisulfide (CS_2) was concurrently measured with DMS. We have examined the data, and contrasted the findings with those of DMS. Additionally the diurnal concentration variation of DMS has been extended to include winter periods.

The data has been segmented to address annual and diurnal cycles as well as episodic 'pollution' events.

In marine air streams DMS and CS_2, poorly co-varied both seasonally and diurnally. The association is modified by the magnitude of the seasonality in DMS concentration and the lower destruction rate of CS_2 compared to DMS. From the limited data, an influence is found for mixed marine/ terrestrial air streams in that, CS_2 concentration is enhanced through contact with coastal margins and population centres.

Introduction

Sulphur gases in the marine atmosphere at Cape Grim, include DMS, CS_2, carbonylsulfide (COS) and sulfurdioxide (SO_2). CS_2 and COS are of mixed marine and terrestrial origin (Bates, Lamb *et al*., 1992). DMS is an artefact of phytoplankton metabolism and is a source via SO_2 and methanesulfonate (MSA), of CCN after oxidation (Ayers & Gillett 2000). The relative importance of this process in terms of climate has been recently reviewed (Quinn & Bates 2011).

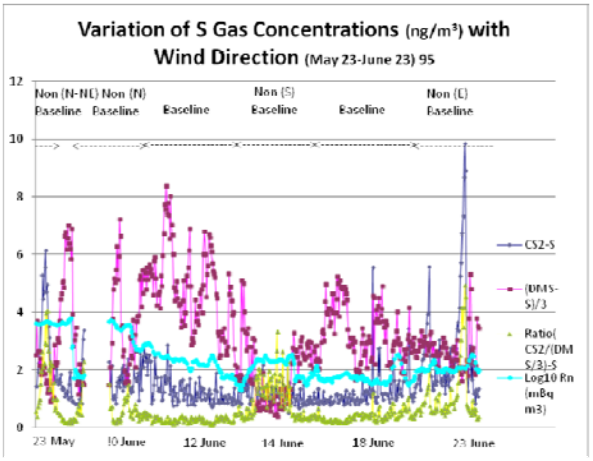
The Cape Grim data record is a chronology to be revisited by reason of insight or recent data. This is an example of the former. During the collection of the DMS data (Ayers *et al*., 1995) using the automated gas chromatograph (1993-96), CS_2 concentrations were concurrently measured.

The methodology for the data collection has been previously reported (Ivey & Swan 1995, Swan & Ivey, 1999).

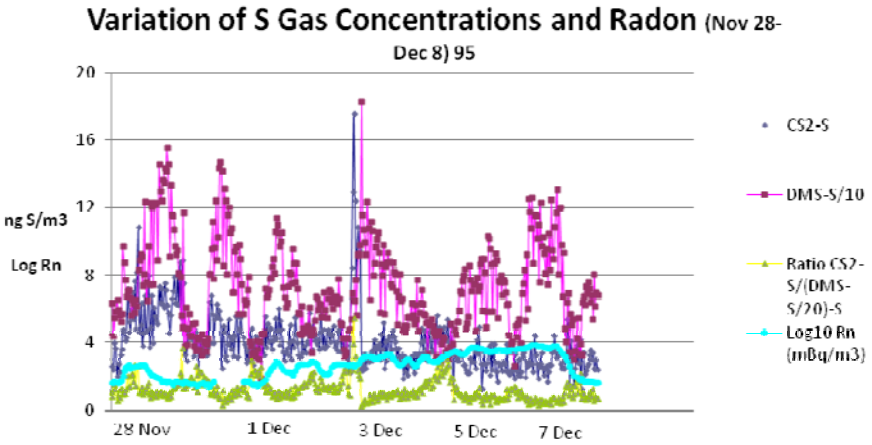
In general terms, CS_2 has an atmospheric concentration of one half, to one twentieth (ng S/m^3) of DMS, varying seasonally. CS_2 measurement hence carries an enhanced analytical uncertainty. In addition, an inert sample line was inadvertently replaced with a rubber one for a period in 1994. This required the editing of suspect data. As DMS concentration can vary significantly within hourly time scales we have restricted our data set to matched pairings of DMS and CS_2, i.e. results in the same chromatogram. Fortunately there are extensive periods to draw upon.

Results and Discussion

With some exceptions, previous interpretations of the DMS data were not 'baseline' selected unlike the data to which they were compared. For the purposes of this abstract we include the figures below, the methodology of which we will extend to the full data set. In the paper by Gillett *et al*. (1992), Gillett speculated that CS_2 was the origin of the lack of seasonal variation in the expected ratio of MSA to non-sea salt sulphate (nss-sulphate) seasonally. This work contributes to that speculation and related papers.



Figures 1. The variation of sulphur gas concentration with radon and wind direction for the period 23 May to 23 June 1995.



Figures 2. The variation of sulphur gas concentration with radon from the 23 Nov. to 8 Dec. 1995.

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Greenhouse gas exchange between inland waters and the atmosphere: a case study from the Elbe River, Germany

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Abstract

The emissions of greenhouse gases from inland waters (e.g. rivers, streams, reservoirs, lakes, ponds, wetlands) are significant and comparable in quantity to the net uptake of anthropogenic carbon dioxide emissions by the terrestrial biosphere or the oceans (Bastviken *et al*., 2011; Cole *et al*., 2007; Tranvik *et al*., 2009). These emissions, however, have been neglected in terms of their inclusion in the recent 'conventional' carbon cycle (e.g. Intergovernmental Panel on Climate Change (IPCC) Assessment Report Four (AR4); Denman *et al*., 2007). Furthermore, emission estimates from inland waters are not adequately constrained by measurements and are therefore often quoted as conservative values, particularly for CH_4 and N_2O which have received far less attention than CO_2.

The focus of this research is the in-situ measurement of greenhouse gas exchange between inland waters and the atmosphere. This has been investigated in a recent measurement campaign which involved a series of three cruises on the Elbe River in Germany and the Czech Republic. During these cruises the water-atmosphere exchange was determined from model derived estimates of the gas transfer velocity (k) combined with measured gas partial pressures in the surface water and overlying atmosphere. CO_2, CH_4 , N_2O, CO and $\delta^{13}C$ (^{13}C isotopic form of CO_2) were measured using Fourier-Transform InfraRed (FTIR) Spectroscopy.

The presentation will provide an overview of the current understanding of greenhouse gas exchange between inland waters and the atmosphere. The results of the Elbe River campaign will be presented and discussed in terms of quantification of the exchange, the assessment of factors controlling the flux (e.g. turbulence, DOC) and the significance of the exchange compared to other inland waters and ecosystems.

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Atmospheric measurements over East Antarctic melting spring sea ice

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Abstract

Measurements of a number of atmospheric components will be undertaken over 1 year-old sea ice off East Antarctica as part of the Sea Ice Physics EXperiment (SIPEX II) during September and October this year (2012). SIPEX II is an integrated and multi-disciplinary study of physical and ecological sea ice processes on small to regional scales which aims to enhance our understanding of sea ice in Antarctic climate and ecosystem processes. A number of atmospheric measurements will be made, including mercury (total in-situ), ozone (in-situ and column), halocarbons (in-situ using GC-ECD), particle number (in-situ) and aerosol and trace gas (e.g. BrO, IO) concentration profiles (using UV-Vis Multi-AXis Differential Optical Absorption Spectrometry (MAX-DOAS)). Presented here are preliminary results from the campaign which aims to develop our understanding of a number of atmospheric and environmental phenomena including the reactive chemistry and biological impact of mercury; the chemistry of new particle formation; and the physics and presence of column aerosol and its local climate implications.

Southern Ocean CO_2 sink project: Observations and inversions

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Abstract

The Southern Ocean (south of 44°S) is a major sink of atmospheric CO_2, however studies (Le Quéré *et al*., 2007) have suggested that the efficiency of this sink may be decreasing, which would have serious implications for the global carbon cycle. Our ability to constrain the mean Southern Ocean carbon flux, seasonality and interannual variability has been limited by the small number of Southern Ocean pCO_2 measurements, especially in winter. We are investigating whether the magnitude of and variations in the Southern Ocean carbon flux can be inferred using a high precision network of continuous *in situ* atmospheric CO_2 measurements. Key to this project is the combination of new continuous *in situ* measurements, forward modelling and inversions.

This presentation will outline the methods used to address possible instrumental and site biases and to link the data sets to a common, derived calibration scale. We will present the results obtained from the cylinder intercomparison program (ICP) and use these as a 'best case' estimate of the uncertainty inherent in the data collected in the Southern Ocean region. Although these uncertainty estimates (0.01 to 0.2 ppm CO_2) are smaller than the uncertainties typically assigned to concentrations used in inversions (> 0.3 ppm CO_2) we found that our inversions often fit the data to better than 0.1 ppm. This suggests that inversions may be able to 'over fit' the data.

Source and sink estimates made using an inversion approach can be influenced not only by the observations used in the inversion but by the number and structure of the regions used, the network of sites chosen, the priors selected and the use of repeating or interannually varying winds. Some preliminary work investigating the sensitivity of Southern Ocean CO_2 sink estimates to these factors will also be presented.

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Measurements of greenhouse gases and ozone in the Southern Pacific Ocean

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Abstract

Climate change is one of the most pressing global environmental issues of our time. It is driven by atmospheric change, and in particular by the large growth in greenhouse gases. There have been a great number of measurement campaigns focused on the Northern Hemisphere, however the data coverage in the Australasian region is sparse.

During three ship cruises in the Southern Pacific (April till June 2012) measurements of the key greenhouse gases CH_4, CO_2, N_2O, O_3 as well as CO and $\delta^{13}C(CO_2)$ were performed in the marine boundary layer by a continuous in-situ FTIR system and an UV-absorption ozone monitor aboard the research vessel *Southern Surveyor*. Here we present the observation of CH_4, CO_2, CO and O_3 across the latitudinal and longitudinal transect. During the first transect from Hobart (42.5°S, 147.2°E) to Brisbane (27.3°S, 153.1°E) in April 2012 air influenced by terrestrial sources from Australia, such as pollution plumes originating from biomass burning or large metropolitan areas, were sampled. Southern Hemisphere marine air over the Pacific Ocean were characterised during the second and third transect from Brisbane to Fiji (17.6°S, 177.5°E) and Fiji to Hobart in May and June 2012.

The MURL SF_6 release and what happened next

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Abstract

In September 2011 the Victorian Government Department of Transport released sulfur hexafluoride (SF_6) into the Melbourne Underground Rail Loop (MURL) in an attempt to '*know how oxygen and gases disperse in the tunnels in the event of a fire, gas leak or terrorist attack*' (The Age, 2011). Upon learning of the release, and noting that ''*The whole purpose of the trial is to study where the sulfur hexafluoride travels to*'', we collected samples of air downwind of the MURL on both days of the release to determine if we could detect the released SF6 in the atmosphere. Samples were collected 10 km downwind on day one and 40 km downwind on day two. To assist with the interpretation of our results, we used TAPM (The Air Pollution Model; Hurley, 2008) to model the SF_6 release over the greater Melbourne region.

We will present results from this study and discuss where the study led.

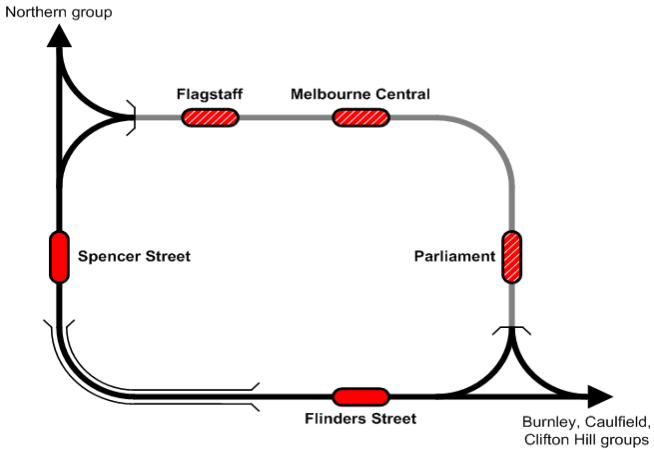


Figure 1. A schematic of the MURL, the SF_6 release occurred at the Melbourne Central station.

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Early results from the ANSTO/NIWA ^{14}C of atmospheric methane program

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Abstract

Following development and proving of the laser heated microfurnace (Yang 2011, Smith 2010 a, b), designed for the efficient graphitisation of micro-samples, we have used it to prepare 149 samples containing ~17 μg of carbon as CO_2. Most of these were derived from atmospheric methane, frozen back into glass breakseals following measurement for CH_4 concentration and $\delta^{13}C$ of CH_4 at NIWA, NZ. There are three sample sets: 62 from Baring Head, NZ (BHD), collected each ~20 days between March 2009 and May 2012, 18 from Arrival Heights, Antarctica (SCT), collected each ~37 days between February 2008 and November 2009, plus 66 samples taken along Pacific Ocean voyages from Nelson (NZ) – Osaka (Japan) in 2008, 2009 and 2011 (FTW). The remaining three samples were prepared to establish the procedural background correction for the overall extraction and measurement process.

We present early results from these sample sets, which demonstrate the capability of the laser heated microfurnace for the preparation of micro-samples and of the ANTARES facility for their radiocarbon measurement. Graphitisation efficiencies approached 100% and all samples were measured to better than 1% precision, sufficient to reveal the $^{14}CH_4$ signal. The BHD set shows significant temporal variation in ^{14}C for baseline air passing over the Southern Ocean, whereas the SCT set shows a lesser variation for Antarctic air. The FTW set covers a S-N transect across the Pacific Ocean, showing the influence of the ITCZ (5°-10° N) and different meteorological conditions on the concentration, $\delta^{13}C$ and $\Delta^{14}C$ of CH_4 and demonstrates that CH_4 is not well mixed.

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Designing a greenhouse gas network for Australia

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Abstract

Carbon Dioxide (CO_2) and Methane (CH_4) are major green house gases (GHGs), which are measured at a number of ground based stations worldwide. Monitoring GHGs is important, not only for the continuous observation of atmospheric concentrations in order to detect global trends, but also for deriving GHG fluxes and their uncertainties on a local scale. Only in situ, flask and total column measurements are currently able to provide the accuracy that is needed to retrieve fluxes via an inversion scheme. Here, we aim to improve the methodology used in Law *et al*. (2004) in order to assess how the current network of GHG ground based measurement stations in Australia can be extended to reduce the uncertainties on Australian GHG flux estimates. This is done by optimising the location of proposed stations for CO_2 and CH_4 under consideration of logistic constraints such as availability of supporting infrastructure and accessibility and maintenance of the site.

The Lagrangian Particle Dispersion Model (LPDM) is used for the inversion of atmospheric transport and a genetic algorithm is applied for the optimisation of the network. Driving fields are provided using the high resolution (12 km) operational version of the Australian Community Climate and Earth-System Simulator for the Australian region (ACCESS-R). Prior estimates of CO_2 fluxes for the Australian biosphere are obtained from high resolution model simulations from the REgional Carbon Cycle Assessment and Processes (RECCAP) study. Various scenarios are taken into account for the optimisation of the network, starting from existing stations. Preliminary results will be presented which may include the assessment of the benefits of individual stations if added to or removed from the current network and the assessment of the benefits of existing infrastructure such as telecommunication towers or wind farms if used as potential sites.

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Composition of fresh and aged biomass burning plumes from the 2006 Robbins Island fire – Cape Grim

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Abstract

Biomass burning (BB) is globally the second largest source of trace gases (second only to biogenic sources), and the largest source of primary fine carbonaceous particles (Akagi *et al*., 2011). Reactive gases (for example ozone) and particles in BB plumes are short lived climate forcers, and may have a significant impact on global climate. However, the processes that govern ozone formation and influence particle number and properties are poorly understood, hampered by difficulties in characterising the complex chemical composition of fresh smoke, and the chemical transformations that occur as the smoke ages. The effect of plume particle transformation on cloud condensation nuclei (CCN) and cloud properties is not well understood, and global climate models do not currently include particle transformations in aging of BB plumes (Hennigan *et al*., 2012). Modelling ozone production in BB plumes is challenging, and is limited by an incomplete understanding of oxygenated emissions and their photochemical reactions, and scarcity of time resolved data sets of ozone production in BB plumes (Jaffe & Wigder, 2012). With a number of climate projection studies predicting increased fire severity and frequency as a result of climate change (as summarised in Keywood *et al*., 2011) it is essential to better understand the likely impacts of BB plumes on climate.

BB emissions were opportunistically measured at Cape Grim during the 2006 Precursors to Particles campaign (Cainey *et al*., 2007), when a fire burned through coastal vegetation on Robbins Island for two weeks. An easterly wind advected the plume over the station on two occasions, giving an opportunity to measure the composition of the fresh (~20 minute old) plumes. Subsequently when the wind direction changed from easterly to westerly, the plumes were diluted and recirculated over the ocean and mainland Australia, allowing the changing composition of the plumes to be observed.

We provide an update on the work from the Robbins Island fire presented at the 2011 Cape Grim Science meeting with a focus on comparing our observations to other studies, and using a chemical transport model to explore the photochemistry of the plume. Emission factors have been calculated for a range of gas and aerosol species in the fresh plume and these are compared to BB emission factors from other local and international studies. Enhanced concentrations of ozone and oxygenated volatile organic compounds (OVOCs) were observed in the diluted plume, suggesting photochemical processing was occurring as the plume aged. Enhancements of ozone and OVOCs normalised to ΔCO are compared to observations of aging biomass burning plumes elsewhere in the world. The contribution of the fire, marine, terrestrial and urban sources to the enhancements in ozone and OVOCs will be explored using chemical transport modelling. Modelling will also be used to estimate the age of the recirculated plume observed at Cape Grim.

The ability of particles to act as CCN was investigated both in fresh and aged plumes, by comparing the ratio of ΔCCN number to ΔCN number, for particles > 80nm (CN80). Because all CN particles > 80nm are at a size capable of acting as CCN, the observed difference in the CCN/CN80 ratio may be attributed to different chemical composition of fresh and aged particles. The ratio of ΔCCN/ΔCN80 in aged plumes with

ozone production was almost twice as high as the ratio in the fresh plumes. Reasons for this difference will be discussed.

This work provides much needed trace gas and particle emission factors for Southern Australia and demonstrates the substantial changes in BB plume composition which can occur with dilution and aging.

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Measurement of trace gas emissions from Australian hazard reduction burns using open-path Fourier Transform Infrared spectroscopy

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Abstract

Biomass burning releases trace gases and aerosol particles that significantly affect the composition and chemistry of the atmosphere. Australia contributes approximately 7-8 percent of global carbon emissions from biomass burning, yet little is known about the emissions from Australian forest fires. This paper describes the results of field measurements of trace gases emitted during hazard reduction burns in Australian temperate forests using open-path Fourier transform infrared spectroscopy. Measurements were made at a total of six separate hazard reduction burns during the controlled burns seasons of 2010 and 2012. Unusually wet conditions prevented most hazard reduction burns planned for 2011.

Spectra recorded at the burns were later analysed for a number of trace gases using the MALT software (Griffith, 1996) and emission factors have been obtained for a number of emitted gases including carbon dioxide, carbon monoxide, methane, methanol, ethylene, formic acid, acetic acid and ammonia. Methods of deriving fire averaged emission factors will be discussed including those described by (Burling et al., 2010).

These open-path measurements were complemented by grab sample measurements for later analysis by SIFT-MS and FTIR spectrometry coupled to a White cell (described in Guerette *et al*. 2012, this publication).

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Measurement of trace gas emissions from Australian hazard reduction burns using Selected Ion Flow Tube Mass Spectrometry and Fourier Transform Infrared Spectrometry

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Abstract

Biomass burning releases trace gases and aerosol particles that significantly affect the composition and chemistry of the atmosphere. Australia contributes approximately 7-8 percent of global carbon emissions from biomass burning, yet little is known about the emissions from Australian forest fires.

This paper describes the results of laboratory measurements performed on smoke samples collected at hazard reduction burns in Australian temperate forest during the 2012 controlled burns season. Grab samples of smoke were collected at four separate fires and analysed for a variety of trace gases using Selected Ion Flow Tube Mass Spectrometry and Fourier Transform Infrared spectrometry coupled to a White cell. Emission factors were derived for a number of trace gases including carbon dioxide, carbon monoxide, methane, acetonitrile, benzene, toluene, methanol, acetylene and acetone. When possible, these emission factors are compared to those derived using Open-Path FTIR at the same fires (described in Paton-Walsh *et al*. 2012, this publication) and to values listed in Akagi *et al*. (2011).

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Smoke impacts on the fire ground

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Abstract

Firefighters are exposed to many health-damaging air pollutants present in bushfire smoke and poorly managed exposure can result in serious health issues. A better understanding of exposure levels and the major factors influencing exposures is crucial for the development of mitigation strategies to minimise exposure risks and adverse health impacts.

A range of air toxics were monitored within the breathing zone of firefighters at prescribed burns and at wildfires in Australia. The exposure of firefighters to air toxics and the consequent risk for health were evaluated in relation to established occupational exposure standards. The pollutants identified to be of primary concern were carbon monoxide, respirable particles and formaldehyde. On occasions these pollutants were measured at high concentrations and therefore have the potential to impact firefighter's health. Exposure levels varied widely with work task and burn conditions thus providing opportunities for mitigation strategies. It was concluded that risks could be minimised by revised firefighter work routines and fire ignition patterns that limit the duration of firefighter exposure to high smoke conditions.

Identifying sources and transport pathways of carbon monoxide over Australasia using a Chemical Transport Model Constrained by surface-based and satellite data

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Abstract

Carbon monoxide (CO) plays a critical role in the atmosphere, both regulating atmospheric composition through its role as a sink for OH radicals (the main tropospheric oxidant) and influencing climate as an indirect greenhouse gas. CO serves as an important tracer of atmospheric pollution due to its intermediate-length lifetime and can also be used to constrain emissions of a variety of trace gas pollutants (Kopacz *et al*., 2010). Over Australasia, CO concentrations are affected by anthropogenic and wildfire emissions from both local and distant sources, including Africa, South America, and the Northern Hemisphere; however, the relative importance of these different sources is not well understood. Here, we use a global chemical transport model (GEOS-Chem) to identify the sources of pollution over Australia and New Zealand. The model is constrained by surface-based remote sensing spectrometers at Darwin (Australia), Wollongong (Australia), and Lauder (New Zealand). Using GEOS-Chem, we quantify the relative contributions of emissions from different regions and source types (fossil fuel combustion, wildfires) to CO levels and seasonal variability. Case study analyses of individual pollution events are used to identify pathways of pollution transport from major source regions. Events identified using the surface-based spectrometers are explored using the GEOS-Chem simulation combined with satellite CO observations from MOPITT. Possible synoptic drivers of these transport events and their implications for pollution variability over Australasia will be discussed.

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^{15}N tracer study of N_2O emissions from soil following N-addition

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Abstract

Estimates of greenhouse gas emissions can be improved by understanding how nitrogen transformations in the soil affect fluxes of nitrous oxide (N_2O). Soil nitrogen transformations and their impact on N_2O emission are frequently studied using ^{15}N labelling techniques, but continuous measurements of the total amount of added ^{15}N transformed to N_2O are rarely achieved in the field. Here we study the short-term (2-4 week) fate of applied nitrogen using a mobile Fourier Transform Infrared (FTIR) spectrometer to measure ^{15}N isotopic signatures of N_2O emitted from an agricultural soil after ^{15}N addition as nitrate or urea. The experiment site was on a coastal floodplain site managed for silage production near Nowra, NSW, in SE Australia. We deployed five 0.25 m^2 automated chambers connected to a multi-pass cell and low resolution FTIR spectrometer to measure mole fractions and fluxes of all N_2O isotopologues every three hours. Root mean square uncertainties for all isotopologue mole fractions were less than 0.3 nmol mol^{-1} for 1 minute average measurements, and minimum detectable fluxes for each isotopologue were <0.1 ng N m^{-2} s^{-1}. We applied a light dose of ^{15}N-labeled substrate (0.4-0.8 g ^{15}N m^{-2}) as potassium nitrate (KNO_3) or urea [$CO(NH_2)_2$]. Flux measurements were made for 1-2 weeks before and after application of the labelled nitrogen.

Figure 1 illustrates results from one period before and following ^{15}N addition. Emissions of all isotopologues were evident within three hours following ^{15}N addition. $^{15}N^{15}NO$ emission subsided in less than 7 days, and emissions of $^{14}N^{15}NO$ and $^{15}N^{14}NO$ isotopomers after 10 days. The chamber soils were destructively sampled after 2 weeks, so that ^{15}N could be analysed in soils and plant material using isotope ratio mass spectrometry. Emissions of $^{14}N^{14}NO$ were evident throughout the entire experiment. Cumulative ^{15}N-N_2O fluxes (sum of the three isotopologues) per chamber for the 14 days following ^{15}N addition ranged from 8 to 20 mg ^{15}N-N_2O m^{-2}, representing 1 to 3% of the applied N.

The results show that FTIR spectroscopy provides a powerful isotopic labelling technique for study of N_2O emissions under field conditions.

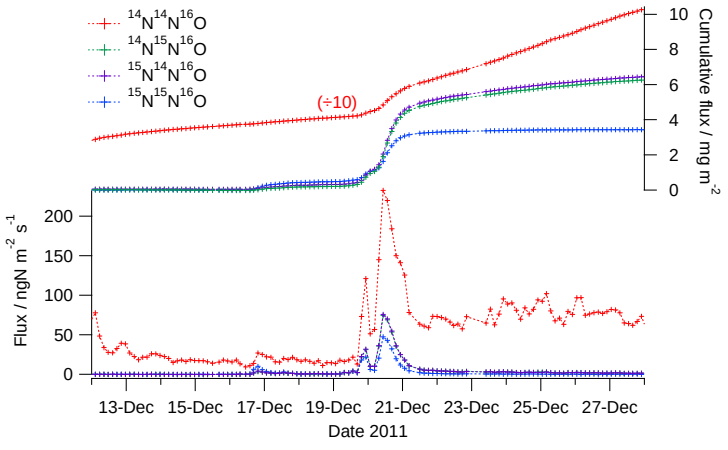


Figure 1. Fluxes and cumulative fluxes of N_2O and its isotopologues following addition of ^{15}N as potassium nitrate to the soil. The ^{15}N was added on 16 December 2011, and emissions of $^{15}N_2O$ observed almost immediately afterwards.

Constraining emissions at high-resolution using measurements from geostationary orbit: A preliminary Observing system Simulation Experiment

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Abstract

We use a simplified, column-integrated plume model to assess the impact of high-resolution measurements of CO_2 and CO on the estimation of fluxes at high-resolution. The test domain is a 200 x 200 km region based around Indianapolis in the US. The study period is one week in early November 2010. The hypothetical instrument is capable of measurements at a resolution of 3x3km during sunlit conditions. Analysis of CALIPSO cloud and aerosol statistics suggests measurements are available for 15% of pixels during daylight hours. the instrument is capable of retrieving column-integrated CO_2 with a precision of 3ppm and CO with a precision of 7 ppb. We model the relationship between CO and CO_2 emissions using uncertain emission factors.

We find that using CO_2 only we can reduce the uncertainty on 3x3km pixels within the urban domain by about 10-20% with one week of measurements for the daytime component of the fluxes. We achieve little uncertainty reduction for the biospheric background, pointing out the importance of measurements immediately adjacent to the fluxes. We reduce the uncertainty of the power-plant emission by 50% with one week of measurements suggesting this technique is a good candidate for remote detection of long-term emission changes.

Adding CO measurements produces the strongest effect on the urban fluxes where uncertainty reductions become as strong as for the power-station. this isolated emission is not improved by the addition of CO measurement. this apparent paradox can be understood by analysis of simple inversions using two component systems. Overall the experiment suggests that such an instrument may be extremely useful in constraining high-resolution emissions.

We also show that methane emission estimates from rice agriculture are substantially improved at high resolution by measurements with 1% precision.

Revised records of greenhouse gases and the variability in the carbon cycle over the past 2000 years

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Abstract

Measurements of greenhouse gas concentrations and isotopes in ice cores and firn air provide the best available evidence of changes in atmospheric composition over the industrial period and the previous millennia. Records from Law Dome benefit from high age resolution and overlap with recent direct atmospheric measurements. They are widely used as forcings in climate model simulations (eg. IPCC AR5) and in biogeochemical model interpretations of trace gas budgets and carbon-climate feedbacks. In particular, the pre-industrial record reveals natural changes in these gases that are attributed to climate variations, biomass burning, carbon cycle dynamics and internal unforced variability. As such, it is essential that the records represent the original atmospheric composition as precisely as possible.

We have revised our Law Dome records of CO_2, CH_4, N_2O and $\delta^{13}C$-CO_2, including new measurements of ice cores and firn air. We include a new ice core from Law Dome and firn air from South Pole. Previous measurements (spanning 2 decades of measurement campaigns) were reselected using a consistent rule-based system and calibrations to most recent scales. Dating has also been improved with new ice chronology and firn air modelling. The main features and decadal variability of the Law Dome records are confirmed by these revisions and by new data. When compared with the records from the Dronning Maud Land and South Pole ice cores and with new records emerging from the West Antarctic Ice Sheet Divide core, most of the differences can be explained by the narrower age spread of the air enclosed in Law Dome ice. However, some CO_2 variations are observed only in the Law Dome record and are at the limit of carbon model interpretations. We review the palaeo evidence that helps confirm that climate and biomass burning changes were behind some of the significant CO_2 and CH_4 variations over the past 2000 years.

Tropospheric ozone distribution and trends at Cape Grim and in the extra-tropical Southern Hemisphere

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Abstract

Tropospheric ozone is a greenhouse gas, ranked third in anthropogenic radiative forcing after CO_2 and CH_4. Ozone in the troposphere makes up an important fraction of the atmospheric shield against ultraviolet radiation particularly when stratospheric ozone depletion events occur. Tropospheric ozone drives tropospheric chemistry being the major precursor of the OH radical which is the sink for gases such as methane. In the boundary layer increases in background ozone have other impacts through urban and regional photochemical smog. These increases affect human health, crop productivity and the sequestration of carbon into the biosphere.

There are few long-term monitoring stations and few process studies of tropospheric ozone in the Southern Hemisphere. Since 1976 there have been measurements of ozone in surface air at Cape Grim and one of the early purposes identified for these measurements was to detect long term trends in tropospheric ozone. In fact it took until 1982 to establish a reliable monitoring system that has continued into the present. Part of this record will be presented here along with observations from elsewhere in the extra-tropical southern hemisphere. Some of our previous work on these issues includes Elsworth & Galbally (1984), Elsworth, Douglas & Galbally (1985), Galbally *et al*. (2011) and Oltmans *et al*. (1998, 2006, 2012).

In an international collaboration, analyses are presented of two decades of observations from seven background surface ozone stations spanning 34°S to 90°S and four long-term ozonesonde stations spanning 38°S to 69°S. Special consideration is taken to consider only those parts of the ozone record that are free of local influences of ozone production and destruction. Halogen based ozone destruction and local ozone production are evident in lower tropospheric air at some Antarctic stations.

The ozone distribution measurements reflect a NO_x depleted environment where there is net ozone loss at most oceanic boundary layer and free troposphere locations. There is little destruction of ozone at the water and snow surfaces. The seasonal signature of stratospheric-tropospheric exchange is evident in upper tropospheric ozone records in the mid-latitudes but not in the polar regions.

Over the last two decades there has been a growth in ozone abundance in the lower troposphere. This growth is stronger over Antarctica compared with the mid-latitudes and regional differences are evident. As well, there is inter-annual variability that appears to be correlated over long distances. Possible drivers of both the growth and inter-annual variability will be discussed.

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Cape Grim 10-m wind climatology

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Abstract

A wind climatology has been developed from Cape Grim using the anemometers located on the 10-m air intake mast. A 15-year data-set of hourly values from September 1997 – August 2012 was used to create a monthly and annual climatology, to be published in the 2009-2010 edition of *Baseline*. A distinct bipolar regime was evident, with westerly and easterly peaks. Nearly 50% of hourly wind observations from the 15-year period were from the station's 'baseline' sector (190-280°), similar to the values recorded in the individual years of 2009 and 2010. A comparison is made between data from this location, with instruments sited on the co-located Telstra tower at levels of 30, 50 and 70 m, as well as the new AWS site to the north, northeast of the station, A comparison is also made with statistics of full baseline conditions (which includes a particle count criteria)

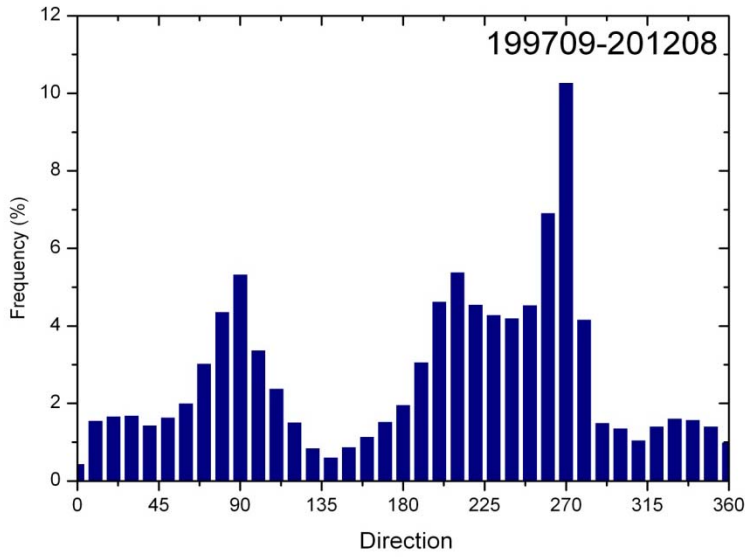


Figure 1. Climatological wind-direction frequency plot from Cape Grim Station, (Bureau site number 91245, 10-m wind) based on hourly observations. Data from September 1997 to August 2012. Vertical axis is percentage frequency of occurrence. Horizontal axis is wind direction (degrees from), in 10° bins. Zero represents calm conditions, 360° represents northerly wind.

Identifying baseline air masses in the lower troposphere at Mauna Loa Observatory between 2004 and 2010 using Radon-222 and back trajectories

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Abstract

The Mauna Loa Observatory (MLO) is distinct from many other World Meteorological Organisation Global Atmosphere Watch Baseline stations, since, by virtue of its elevation, sampling is typically conducted within the lower troposphere instead of the atmospheric boundary layer. Here we use hourly MLO radon observations, together with HYSPLIT v4.0 back trajectories, to identify baseline air masses at the station over the 2004 – 2010 period. We also characterise the diurnal-to-interannual variability in MLO radon concentration based on measurements from an improved 1500 L detector commissioned in December 2003, which differ significantly from previously published results. The composite diurnal radon cycle (amplitude 49 mBq m^{-3}) is discussed with reference to the influences of local flow features affecting the site, and a 3-hour diurnal sampling window (0730-1030 HST) is proposed for observing the least terrestrially influenced tropospheric air masses.

The mean annual radon concentration at MLO, based on all hourly data, was 102 mBq m^{-3} (range: 84 to 115 mBq m^{-3}); compared with 79.6 mBq m^{-3} (range: 58.6 to 97.3 mBq m^{-3}) for observations conducted only within the 0730-1030 HST sampling window. The amplitude of the annual MLO radon cycle, based on monthly means, was 98 mBq m-3 (39 - 137 mBq m^{-3}); with maximum values in February (90th percentile 330 mBq m^{-3}) and minimum in August (10th percentile 8.1 mBq m^{-3}). A set of 763 hourly baseline events was selected over the 7-year period, using the proposed sampling window, radon concentration distributions, and back-trajectory information. This dataset – which can be provided on request – represents a valuable resource for the selection of baseline events at MLO for use with a range of other trace species observations. A reduced set of 196 'deep-baseline' events (occurring in July – September), is also discussed. The distribution (10th/50th/90th percentile) of radon in deep-baseline events (8.7/29.2/66.1 mBq m^{-3}), was considerably lower than that for the overall set of 763 baseline events (12.3/40.8/104.1 mBq m^{-3}).

Results from a simple budget calculation, using sonde-derived mixing depths and literature-based estimates of oceanic radon flux and radon concentrations in the marine boundary layer, indicate that the main source of residual radon in the lower troposphere under baseline conditions at MLO is downwards mixing from aged terrestrial air masses in the upper troposphere.

The influence of marine aerosol concentrations on New Zealand urban air quality and source region determination

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Abstract

New Zealand's mid-latitude oceanic location in the 'roaring-40's' places it squarely in the path of the southern hemisphere atmospheric circulation with a fetch of many thousands of kilometres across the sea surface. This not only has a profound effect on New Zealand climate and meteorology but also on air pollution and aerosol concentrations in particular. This paper presents the results of aerosol sampling and receptor modelling research at three main cities in New Zealand with a focus on the contributions of marine aerosol to urban air particulate matter concentrations.

More than 8000 samples of size-resolved (PM2.5, PM10-2.5, PM10) have been collected in the cities of Auckland, Wellington and Dunedin. Elemental concentrations in the particulate matter were determined by Ion Beam Analysis and black carbon (BC) by light reflectance at the New Zealand ion Beam Analysis Facility (Trompetter *et al*., 2005). The elemental and BC concentrations provided the basis for multivariate analysis using positive matrix factorisation receptor modelling software (Paatero, 1997, Hopke *et al*., 1999) to apportion particulate matter mass to the sources of urban air pollution.

Marine aerosol was found to have the highest concentrations during summer and contributions to PM10-2.5 (and therefore PM10) were significantly higher than for PM2.5 indicating that the marine aerosol was largely composed of coarse particles. The range of marine aerosol concentrations within samples was considerable with maxima in the order of 35 μg m^{-3} (24-hour average) (Figure 1). The high marine aerosol concentrations has significant implications for air quality management as the New Zealand National Environmental Standards for Air Quality stipulate a maximum PM10 concentration of 50 μg m^{-3} such that, at times, the standard may be breached due to the contribution of marine aerosol coupled with more local sources of particulate matter.

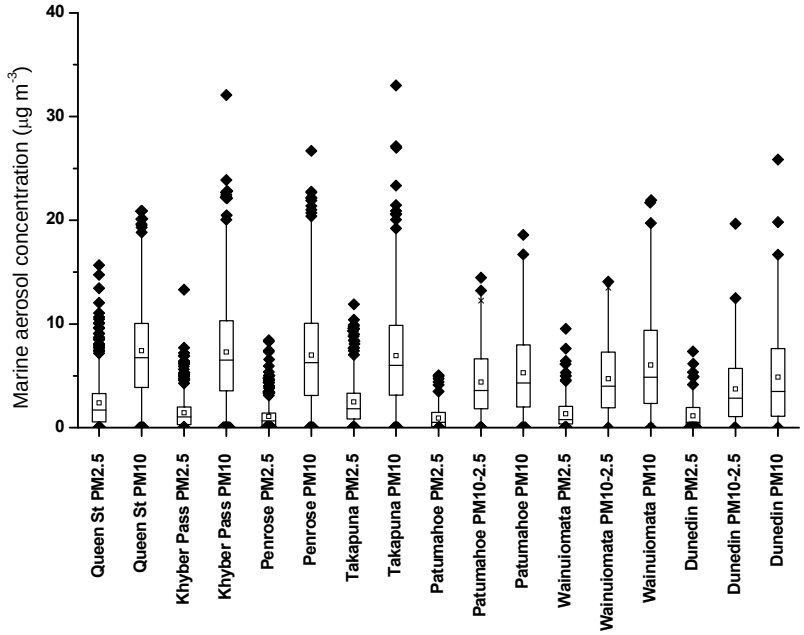


Figure 1. Marine aerosol concentrations in various size fractions sampled at NZ urban areas

An analysis of marine aerosol concentrations across multiple sites in Auckland shows a strong correlation (r = 0.84-0.91) indicating that concentrations were homogenous in the regional air mass. Marine aerosol source regions were calculated using Potential Source Contribution Function analysis (Polissar *et al*., 2001) which demonstrated source regions (Figure 2) consistent with southern hemisphere circulation, long oceanic fetch and regions of high average wind-field strength. The results pave the way to providing good estimates of marine aerosol fluxes to atmosphere for use in global aerosol models.

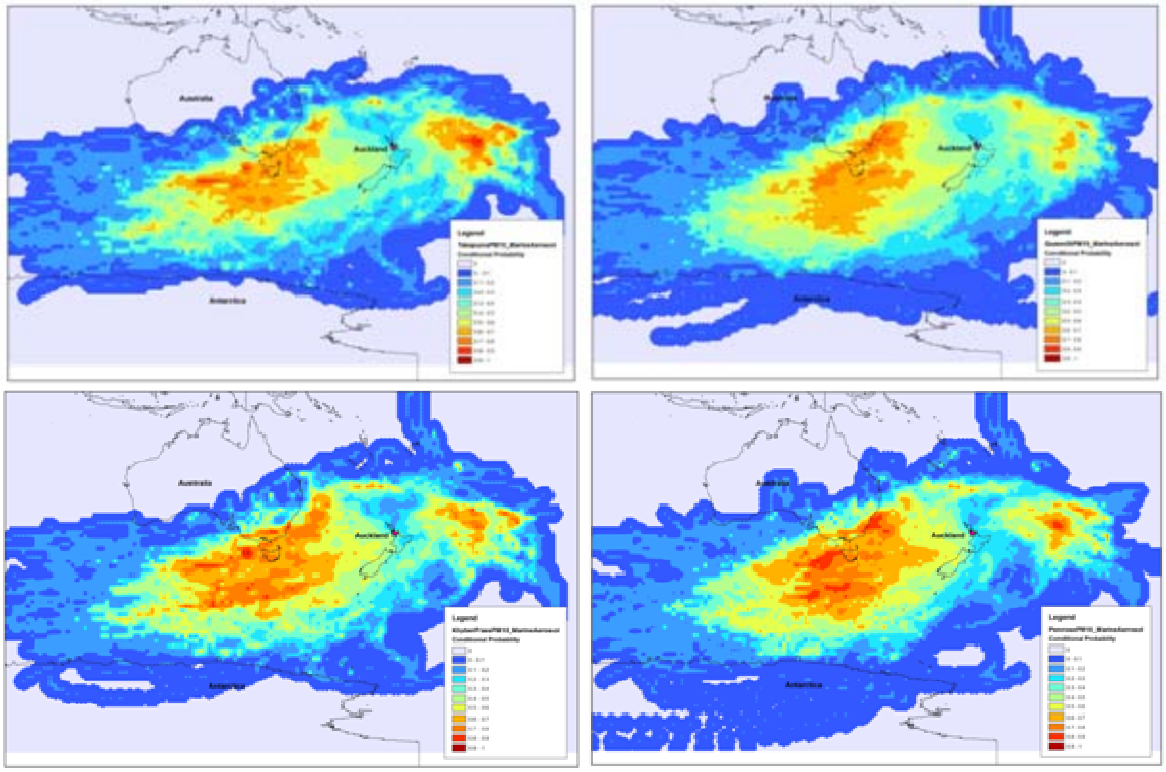


Figure 2. Marine aerosol PSCF source regions calculated for various Auckland sampling sites

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Total Column CO and H_2CO at three Southern Hemisphere sites, investigated with FTIR measurements, and the models GEOS-Chem and ACCESS

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Abstract

Tropospheric pollutants carbon monoxide (CO) and formaldehyde (H_2CO) have a major interaction with hydroxyl radicals, the main cleansing species of the atmosphere. Increased concentrations of tropospheric pollutants can also have detrimental effects on human health and vegetation. Understanding the concentration of these trace gases is therefore important for understanding evolution of atmospheric composition and related health impacts. Further study of these gases is particularly important in the Southern Hemisphere, where there is still much uncertainty in the behaviour and distribution of tropospheric pollutants.

Here, CO and H_2CO are studied with measurements and model simulations over three sites in the Australasian region: Wollongong, NSW, Australia (34.4°S, 150.9°E), Darwin, NT, Australia (12.4°S, 130.9°E) and Lauder, New Zealand (45.0°S, 169.7°E). For this comparison, remote sensing total column retrievals from ground based Fourier Transform Infrared Spectrophotometers (FTIR) for CO and H_2CO are used. The global Chemical Transport Model (CTM) GEOS-Chem has been used to simulate atmospheric trace gases the time period 2004-2009. Additionally, simulation results are used from the Australian Community Climate Earth-System Simulator (ACCESS), being developed in Australia as a joint project between the Commonwealth Scientific and Industrial Research Organisation (CSIRO) and the Australian Bureau of Meteorology. This study focuses on preliminary results from the chemistry module of ACCESS.

Results indicate GEOS-Chem generally reproduces the magnitude and seasonal cycle of total column measurements. In contrast, ACCESS consistently underestimates the magnitude of total column values and also misrepresents several annual cycle peaks, suggesting emission inventory inconsistencies. Furthermore, a lack of isoprene and related chemistry in ACCESS is likely to impact total column values for both trace gases. Future simulations of ACCESS will include updated emission inventories and isoprene chemistry.

Comparison of results from a developing Earth-System model with a well established CTM and measurements gives insight into modelling strengths, indicating processes accurately captured by each of the models. Model processes identified as weaknesses brings focus to areas requiring further development. Overall, the study increases our knowledge of atmospheric trace gases in the Southern Hemisphere, including seasonal variability of target trace gases in the Australasian region.

Volatile Organic Compounds in Typical Australian Dwellings

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Abstract

Volatile organic compounds (VOCs) are of increasing concern with regard to indoor air quality. Research shows that indoor VOCs are associated with the sick building syndrome (SBS) and chronic and acute health effects at high concentrations. Until recently there has been limited information about indoor VOCs concentrations in typical Australian dwellings (Brown, 2002; Environment Australia, 2001). We have added to this knowledge by conducting a study of indoor air pollutants including VOCs in 40 typical dwellings within the temperate zone in Australia in Winter/Spring of 2008 and Summer/Autumn of 2009. During each sampling period, household surveys and diaries were administered to characterize homes and personal activities that might impact on the level of VOCs in the home. Here we present the findings of VOCs levels in these dwellings.

More than 80 VOCs including formaldehyde were identified in the study. Aromatic and aliphatic hydrocarbons and aldehydes were the most common classes of VOCs detected. Other classes of VOCs including ketones, alcohols, terpenes, esters, halocarbons and carboxylic acids were also found in the dwellings. The indoor concentrations of all VOCs detected were higher than outdoor concentrations. The possible indoor sources of VOCs were investigated through analysis of the relationships of indoor VOCs concentrations to the characteristics of homes and personal activities in the dwellings. Seasonal effects of VOCs concentrations were explored. The indoor VOCs concentrations from this study were also compared with other Australian or overseas studies. 11 of the VOCs identified in Australian dwellings are not present in overseas dwellings in the studies reviewed.

Overall, apart from building age and ventilation, the factors that control these VOCs inside Australian dwellings are not well established. The dominant VOCs detected indoors are consistent with those occurring as propellants in aerosol sprays, in emissions from fragranced personal care products, household cleaning products, solvents, composite wood materials and combustion appliances.

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Seasonal variability of solar proxy Beryllium-10 in Law Dome snow, Antarctica, and its connection to stratosphere-troposphere exchange

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Abstract

The interpretation of ice core Be-10 as a solar activity proxy depends on the climate impact on Be-10 deposition into the ice sheet. Precipitation rate and stratosphere to troposphere transport (STT) are the main drivers of Be-10 deposition. Because the majority of Be-10 is produced in the stratosphere the location and timing of STT bringing the Be-10 atoms into the troposphere is essential in understanding the seasonal variations of Be-10 in deposition. Moreover, the solar and geomagnetic modulation of the production rate of Be-10 are stronger in the stratosphere. A potential direct exchange between surface and upper levels could then lead to an amplification of the solar signal in ice. Indications of such direct coupling have been found in Antarctica, especially at Law Dome. We therefore investigate the seasonal variability of the STT at Law Dome and its potential connection to enhanced Be-10 snow concentrations.

The monthly resolved Be-10 record (2000-2010) from Law Dome exhibits a maximum during late summer - autumn (FMAM). This maximum has been found to coincide with minimum tropopause height. To investigate a possible connection with STT events we apply a new STT climatology based on the ERA-Interim reanalysis (1979-2011) and the potential vorticity (2pvu) definition of the tropopause. These data show maximum stratospheric mass flux into Law Dome air at 600 hPa during MAM, when tropopause is lowest. These events are among the strongest on global scale as well. These findings suggest a more direct coupling of Antarctic air with upper levels, especially near the coast, than in other parts of the world.

A new global chemical emissions database and meteorological nudging for ACCESS

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Abstract

Global atmospheric chemistry and aerosols are modelled in ACCESS (Australian Community Climate and Earth-System Simulator, see http://www.accessimulator.org.au) using the UKCA (United Kingdom Chemistry and Aerosol) model. UKCA is used to simulate various reactive chemical gases and aerosol species, as well as the radiative feedbacks of ozone and methane within the coupled climate system. A number of chemical scheme options are available for UKCA. The base scheme is the standard tropospheric chemistry scheme, which is designed for climate studies using a backward-Euler solver. An extended tropospheric scheme (with isoprene) includes a simple SOA sub-model. The UKCA aerosol component is a simplified version of the GLOMAP model called MODE. Our focus here is on the base standard tropospheric chemistry scheme without aerosol-formation chemistry. It describes essential background inorganic chemistry involving O_x- NO_x-HO_x-CO cycles, together with near-explicit degradation of methane, ethane and propane. CO_2, H_2, N_2, O_2, and CH_4 are used as global constants.

The standard tropospheric configuration involves 8 emitted species, 26 advected tracers, 13 non-advected tracers, a total of 46 species and 129 reactions (102 gas phase and 27 photolysis reactions). Interactive dry and wet deposition processes for relevant species are included. Surface and aircraft NO_x emissions are included, and so are online lightening NO_x emissions. The eight reactive gases for which the surface emissions need to be specified are: NO_x (nitrogen oxides), CH_4 (methane), CO (carbon monoxide), HCHO (formaldehyde), C_2H_6 (ethane), C_3H_8 (propane), Me2CO (acetone), and MeCHO (acetaldehyde). Emission ancillaries can either hold data as a timeseries (covering the whole simulation period) or as a cyclic/periodic set (e.g. 12 monthly fields).

Our version of UKCA uses a supplied emission file for the year 2000, which is thought to include an anthropogenic emission component that predates AR5. We have developed a methodology for deriving a new emission database based on IPCC AR5 anthropogenic emissions (which also includes biomass-burning and shipping emissions). Significant quantities of some species are also emitted from natural sources (e.g., soil NO_x, methane from wetlands and CO from isoprene oxidation), and these are accounted for based on available information (e.g. http://data.giss.nasa.gov/ch4_fung, Global Emissions InitiAtive (GEIA) Project, http://www.geiacenter.org). Three-dimensional emission fields due to aircraft NO_x reported by IPCC-AR5 are also incorporated. The emissions database is currently for the year 2000, but can be extended to previous decades. One aim is to assess and refine the Australian emission component based on the data available from national inventories.

A parameterised isoprene chemistry scheme is optionally included in UKCA, which allows detailed studies of land-surface-climate interactions. Isoprene emissions were also derived using the GEIA data climatological data. In the model, these are scaled to be diurnally varying.

We will describe the above new emissions database and its use in ACCESS (configuration: N96L38, resolution 1.25 lat. x 1.875 long., 38 levels, extending to a height of 40 km).

We have also incorporated meteorological nudging in ACCESS, which is used to constrain the model to follow some prescribed synoptic behaviour, for example, ECMWF's ERA-Interim reanalysis data. This allows us to compare time-paired statistics of species concentrations and other parameters from the simulation and observations. Of particular value is the ability of nudging schemes to suppress transport errors in the simulation, thereby allowing us to isolate the relative contributions of process errors and transport errors to the performance of UKCA.

Two different nudging schemes have been implemented in the ACCESS model. The first is a standard relaxation scheme based on the UK Met Office code. In this case the winds and air temperature of the ACCESS model are 'relaxed' towards the host dataset (i.e. ERA-Interim) by perturbing these fields at each time step in the simulation with a six-hourly e-folding time. We have also implemented a scale-selective filter (sometimes referred to as a spectral filter), which only perturbs large wavelengths in the simulation towards the host. Scale-selective filters are more computationally expensive, but have the advantage of leaving the finer spatial length scales unperturbed, enabling us to run the ACCESS simulation at much finer resolution than the ERA-Interim reanalyses without damping small scale structure. Some initial results from the nudging work will also be described.

Acknowledgements

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Identification of a suitable baseline definition of tropospheric ozone at Cape Grim, Tasmania

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Abstract

Tropospheric zone concentrations, like numerous other atmospheric constituents, are a result of a complex mix of local, regional and global sources and sinks. Identifying the contribution of each potential component can be a difficult task but is essential to understanding the processes which affect the atmospheric species in question. An understanding of the contribution of different sources and sinks enables a more accurate incorporation of the species into a range of models, which in turn can be used for a variety of purposes including forecasting.

A fundamental component of the concentration of a given atmospheric species is the baseline or background concentration. This can be considered the underlying concentration of the species in the absence of all other local and regional effects; the starting point upon which the sources and sinks then act on to produce the final concentration which is observed. This baseline concentration is not static, as there are many factors that affect it depending on the atmospheric species.

With regard to ozone, a knowledge and understanding of this baseline or background concentration provides essential input for three related objectives:

- To provide advice concerning the contribution of background ozone concentrations to ozone pollution events that may exceed environmental standards (Fiore *et al*., 2002);
- To investigate the variations and trends in tropospheric ozone and the physical/chemical processes regulating tropospheric ozone;
- To provide appropriate data for model-observation comparisons.

To separate baseline from non-baseline data, a variety of variables can be used depending on the atmospheric species. For tropospheric ozone, selection of baseline data depends on the particular circumstances under which the concentrations are measured and the data that is available. In this study, we build on previous work by Zahorowski *et al*., (1996) and trial a set of baseline definitions for use with ozone concentrations measured in the marine boundary layer at the Cape Grim Baseline Air Pollution Station (CGBAPS). These baseline definitions are based on wind speed and direction, radon concentrations and condensation nuclei counts and applied to a 10 year ozone data set from 1992 to 2001. The resulting baseline data sets are analysed for long term trends and the differences between the trends are examined.

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Footprinting Australasian CO_2 measurement stations using high-resolution Lagrangian dispersion modelling

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Abstract

Since 1990, New Zealand's CO_2 equivalent greenhouse gas emissions increased by about 20%. Yet, the New Zealand Ministry for the Environment estimates that its net greenhouse gas budget remained at 1990 levels due to a compensating increase in CO_2 uptake by forests. This terrestrial uptake remains poorly quantified despite its key role in the contemporary and future carbon cycle. Global studies suggest that this sink is sensitive to changes in climate and could already have become less efficient, with strong impacts on net carbon release.

One powerful tool to link CO_2 observations at stations directly with atmospheric transport and regional CO_2 uptake/release is Lagrangian modelling. In such a model, trace particles are released at specified locations, and their trajectory is computed while they are transported and mixed within the atmosphere until they reach a receptor, i.e. a CO_2 observing station. In backward mode, regions of CO_2 release or uptake that contributed to the measurements made at a specified receptor can be identified. At NIWA, we are currently implementing two regional high-resolution Lagrangian dispersion models, NAME and FlexPart, which have the potential to give detailed insight into the terrestrial carbon sink in New Zealand and serve as a bridge between global modelling efforts and local CO_2 research already established at NIWA. The Lagrangian approach will allow us to cluster large numbers of back trajectories over long time periods into a discrete set of transport regimes and relate them to climate drivers and CO_2 observations at Australasian stations. The models are interfaced with meteorological output from the regional NZLAM and WRF models at resolutions of 9 to 12 km over the New Zealand and Tasman region.

Here, we present preliminary results characterising the catchment areas for NIWA and CSIRO's flagship monitoring stations: Baring Head, New Zealand and Cape Grim, Australia. The region sampled by each station, or the station's 'footprint', will be shown and the main transport pathways will be identified using cluster analysis.

The Total Carbon Column Observing Network (TCCON): Australia's continuing contribution

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Abstract

TCCON is a network dedicated to the accurate and precise measurements of two main anthropogenic greenhouse gases: Carbon dioxide (CO_2) and Methane (CH_4) as well as other atmospheric constituents including N_2O, HF, CO, H_2O, and HDO. Here we show results and ongoing efforts from two Australian TCCON sites managed by the University of Wollongong. The sites are located in Darwin and Wollongong.

High precision CO_2 measurments using a small low resolution spectrometer

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Abstract

Understanding how the release of carbon, mainly in the form of carbon dioxide, is cycled through the earth's various complex systems is a major challenge. The residual amount of CO_2 left in the atmosphere (after various source and sink mechanisms come into play), and the subsequent heating that CO_2, a greenhouse gas, imposes on the atmosphere, is a major driving force in the earth's climate system. Since this is a global phenomenon (the largest temperature effects will occur at the poles) the best method of approach to quantify, and qualify the amount of CO_2 is through space based measurements. Missions are currently operating (Greenhouse gas Observing SATellite GOSAT) or are in the planning stage (Orbiting Carbon Observatory, OCO2, CarbonSat), to globally map CO_2 concentrations. These satellite data products however are only useful if they are directly tied to an equivalent ground based measurement of sufficient precision and spatial density, a process known as ground-truthing. The Total Column Carbon Observing Network (TCCON) is a collection of ground based scientific measurement sites setup by the Californian Institute for Technology, designed specifically for the purpose of ground-truthing CO_2 satellite measurements. The Centre for Atmospheric Chemistry (CAC), at the University of Wollongong, in partnership with CalTech, operates two of these sites, one on the University of Wollongong Campus, the other site near the Darwin airport.

The instrument of choice for the TCCON network is the Bruker 125HR, a high resolution Fourier Transform Spectrometer (FTS). For TCCON measurements (in the near infrared), the demonstrated precision is better than 0.5% in the column averaged dry-air-mole fraction. The precision is directly related to quality of the spectra produced by the instrument. The Bruker HR125 is a very stable instrument, producing spectra of the highest quality. Because of this, the cost to setup a TCCON site is very high, limiting the density of any worldwide network. In this presentation, measurements from a much lower resolution (and therefore less expensive) instrument, a 0.5 cm^{-1} Bruker cube FTS will be discussed. The Bruker cube has been run along-side the Bruker HR125 during 2012. These data will be presented in terms of the comparative precision and accuracy from the two instruments.

Modeling *in situ* and column integrated CO_2 observed during the Greenhouse Gas Monitor experiment in Port Augusta in May 2012

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Abstract

The Greenhouse Gas Monitor (GGM) Program, which is funded by VIPAC and the Australian Space Research Program (ASRP) has developed a novel, terrestrial instrument to autonomously measure carbon dioxide (CO_2) and other greenhouse gases in the atmosphere. The Program also is using advanced regional models to model the evolution of the measured greenhouse gases and attribute their sources and sinks in an inversion framework.

Here we present the results of a CO_2 measurement campaign in Port Augusta in May 2012 where near-perfect weather conditions enabled the observation and measurement of in-situ and column integrated concentrations of CO_2 as sourced from a near-by coal-fired power plant. With CO_2 as a passive tracer, we model its evolution in the atmosphere by using the Weather Research and Forecasting Chemistry (WRF-CHEM) model. We utilise the CO_2 measurements and simulations in an atmospheric inverse framework to estimate CO_2 fluxes in Port Augusta. Preliminary results show good timing of observed and simulated plumes at the measurement site but that WRF-CHEM is too dispersive in the near field. We will test this with parallel runs of TAPM with an embedded Lagrangian plume model.

Lauder gas and aerosol observations, modelled with the new Southern Hemisphere version of UKCA

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Abstract

The atmospheric measurements made at Lauder in New Zealand are an important archive of composition change spanning more than 30 years. Lidar and balloon-borne instruments yield aerosol data, with some size resolution, and gas phase measurements up to 35 km, and thus can show atmospheric change with height as well as time. Atmospheric change is never more rapid than after a large volcanic injection of gas and aerosol, such as that from Pinatubo in the Philippines in June 1991.The volcanic material was carried high into the stratosphere and transported around the globe. Lauder measurements were able to capture this change.

In a collaboration between CSIRO and NIWA, we have brought our chemistry and size-resolved aerosol versions of the UKCA code together. For the first time, we will use this southern hemisphere version of UKCA to model the Pinatubo period. We present results from a 5-year coupled stratospheric chemistry-stratospheric aerosol integration.

The future purpose for our model is to investigate the increasing background aerosol loading in the stratosphere. This increase is certainly apparent in the Lauder measurements at a rate of 3.8% per year for a decade, but currently we can only speculate as to why it is, or was, happening.

An inverse system for locating and quantifying greenhouse gas leakage, and its testing using Otway Data

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Abstract

The CO2CRC Otway Project in south-western Victoria (http://www.co2crc.com.au/otway) is Australia's first, and one of world's largest demonstrations of geosequestration, where about 65,000 tonnes of fluid (approximate molar composition 80% CO_2 and 20% CH_4) derived from a natural geological source have been injected into a depleted natural gas reservoir at a depth of approximately 2 km during March 2008 – August 2009. A monitoring program that involves both subsurface and atmospheric measurements of CO_2, CH_4 and other gases has been operating since 2007, well before the injection. Atmospheric measurements, in conjunction with numerical modelling of dispersion, have the potential to detect, locate and quantify leakage of the stored fluid into the atmosphere.

As part of the Otway Project, we have developed an inverse modelling system based on the Bayesian probabilistic inferential framework, which provides a natural means for incorporating ambient concentration measurements, model and observational errors, and available prior source information to enable the determination of the emission rates and locations of leakage sources. The required source-receptor relationship is determined using a backward Lagrangian particle dispersion model for the diabatic surface layer. The input meteorological information is derived from the data collected from the local flux station. Because of the high dimensionality of the posterior source probability density function (PDF), its direct calculation and subsequent integration throughout the source parameter space can be computationally very expensive, or even impossible. We address this problem by sampling the posterior PDF using a Markov chain Monte Carlo (MCMC) method, which generates a Markov chain of sample in proportion to the value of the posterior PDF so that there is no waste of computational time in exploring less important parts of the parameter space which have low contributions to the PDF. The posterior PDF is then integrated to obtain marginalised PDFs, which in turn are used to determine summary statistics related to each source parameter (e.g., the mean emission rate and standard deviation). We use the slice sampling MCMC method.

Control experiments involving near-surface releases of injected fluid (CO_2 and CH_4) conducted at Otway during August–September 2011 in neutral to weakly-convective atmospheric conditions provided an opportunity to further develop and evaluate the inverse modelling system. Two Picarro cavity ringdown spectrometers (continuously measuring CO_2, CH_4 and H_2O concentrations) were installed almost north and south of the release points, thus providing simultaneous upwind and downwind concentration measurements, depending on whether the flow was northerly or southerly. This configuration also provided the background concentration and its spatial variability/uncertainty, which are fed into the modelling. We applied the inverse modelling system in two manners: one in which the source location is given and the hourly emission rates are predicted, and the other in which both location and emission rate are predicted using data from a series of hourly periods assuming that the emission rate is constant with time. The predicted emission parameters compare favourably with the measured values. Here, we describe the inverse modelling system and various results pertaining to source estimation at Otway and their potential application to monitoring fugitive emissions from other geosequestration and coal methane activities.

Quantification of fossil fuel CO_2 emissions at the urban scale

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Abstract

Quantification of the magnitude of fossil fuel CO_2 emissions (CO_2ff) is vital to improving our understanding of the global and regional carbon cycle, and independent evaluation of reported emissions is essential to the success of any emission reduction efforts. The urban scale is of particular interest, because ~75% CO_2ff is emitted from urban regions, and cities are leading the way in attempts to reduce emissions.

Measurements of $^{14}CO_2$ can be used to determine CO_2ff, yet existing ^{14}C measurement techniques require laborious laboratory analysis and there are often insufficient measurements to infer an urban emission flux. This presentation will focus on how $^{14}CO_2$ measurements can be combined with those of more easily measured ancillary tracers to obtain high resolution CO_2ff mixing ratio estimates and then infer the emission flux. A pilot study over Sacramento, California showed strong correlations between CO_2ff and carbon monoxide (CO) and demonstrated an ability to quantify the urban flux, albeit with large uncertainties.

The Indianapolis Flux Project (INFLUX) aims to develop and assess methods to quantify urban greenhouse gas emissions. Indianapolis was chosen as an ideal test case because it has relatively straightforward meteorology; a contained, isolated, urban region; and substantial and well-known CO_2ff emissions. INFLUX incorporates atmospheric measurements of a suite of gases and isotopes including ^{14}C from light aircraft and from a network of tall towers surrounding the Indianapolis urban area. The recently added CO_2ff content is calculated from measurements of ^{14}C in CO_2, and then convolved with atmospheric transport models and ancillary data to estimate the urban CO_2ff emission flux. Significant innovations in sample collection include: collection of hourly averaged samples to remove short term atmospheric variability; and direct measurement of the background signal from towers immediately upwind of the urban area and from the boundary layer.

We find that CO_2ff and other anthropogenic trace gases are consistently enhanced at tower sites downwind of the city. Measurements made directly over or very close to the urban area show only weak correlations between CO_2ff and trace gases associated with combustion, likely because emissions from the different fossil fuel sectors are not yet well mixed. In winter, total CO_2 is also consistently enhanced, and the enhancement is slightly higher than the fossil fuel CO_2 enhancement, in agreement with Indiana's requirement for 10% bioethanol use in gasoline. This result implies that the enhancement in total CO_2 can be used to infer CO_2ff emissions for Indianapolis during winter. We therefore use the high resolution in situ total CO_2 measurements in a simple mass balance model to estimate the urban CO_2ff emissions. An initial comparison shows a ~20% difference between the top-down and bottom-up methods.

Atmospheric greenhouse gases observations at the Chinese GAW stations and modelling attempt

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Abstract

Atmospheric greenhouse gases (GHGs) observations (ground based / tall tower / aircraft, etc.), remote sensing and inverse modelling techniques are currently not sufficiently developed to provide a comprehensive routine and verification at the desired accuracy and compatibility, especially on a smaller scale. Progress is to be obtained from working together, to improve network coverage, robust measurement and verification techniques as well as a better understanding of inventory estimates. The CMA represents the World Meteorological Organization (WMO)'s Commission for Atmospheric Sciences in China and is deeply involved in the Global Atmospheric Watch (GAW). The CMA has strong working relationship with the global observation community within the GAW and especially has long-term bilateral cooperation with the National Meteorological and Hydrological Services (NMHSs). A GHGs & Tracers Lab was initiated in Beijing and a number of GAW stations were developed in China. For example, Mt. Waliguan (one of the 28 GAW baseline stations) and the >20years long-term GHGs record contribute to the World Data Centre for Greenhouse Gases (WDCGG), Greenhouse Gases Bulletin, Globalview data products, IPCC and WMO/UNEP assessment and so on. In-situ high accuracy observations of atmospheric GHGs by custom designed systems has been added at the five background stations (Waliguan, Shangdianzi, Linan, longfengshan, Xianggelila) in typical regions of China since 2009. To serve the needs of the expending Chinese GHGs observation and application communities and better contribute to the global network, one of the CMA's capability and effort is to firm a National Central Calibration Lab (CCL China) with tight linkage to the WMO/CCLs (refer to the CIPM Mutual Recognition Arrangement, signed to the WMO in April 2010 for CO2, CH4, N2O, SF6 and CO with designated institute the NOAA/ESRL). The system requires well calibration relative to international standard scale, which is essential to an integrated network and database.

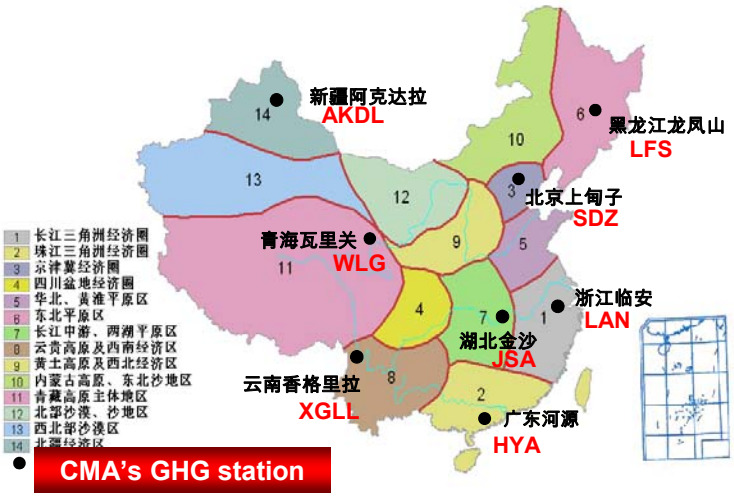


Figure 1. Map of stations in China conduct accurate & compatible observations of atmospheric GHGs

Bulk Richardson Number, surface decoupling and the mixing of passive scalars in the nocturnal stable boundary layer over land

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Abstract

The relationship between the Bulk Richardson Number (R_{ib}) and the degree of decoupling of the nocturnal stable boundary layer (SBL) from the overlying residual layer is investigated for a range of stability conditions at a complex land surface site near Sydney, Australia.

A mass-based decoupling index (D_{rb}) is formulated for the layer spanning the lowest 50 m above ground level (agl), based on the bulk gradient of the ubiquitous noble gas Radon-222:

$$D_{rb} = \frac{\Delta C}{C_2 - C_b}$$

where $\Delta C = C_2 - C_{50}$ is the vertical radon gradient, C_2 and C_{50} are the radon concentrations at 2 m and 50 m, respectively, and C_b is the radon 'background' concentration in the residual layer (approximated as C_{50} in the previous afternoon). Radon has a well-characterised, slowly-varying surface source function and an internal sink (radioactive decay) that is negligible on turbulent time scales.

It is verified that D_{rb} unambiguously expresses the degree of decoupling between the surface-based shear-driven mixing layer and the atmosphere above. This simple radon-based index is then employed together with the Bulk Richardson Number (R_{ib}) to interpret eddy correlation measurements of turbulence characteristics at 10 m and 50 m agl.

As expected from previous studies, larger values of R_{ib} tend to be associated with more decoupled conditions (exhibiting reduced turbulence intensities and fluxes) and D_{rb} changes most steeply in the vicinity of the critical Richardson Number (R_{ib} ~0.2). However, D_{rb} exhibits a large variability for any given value of R_{ib}, indicating that the relationship between bulk stability and mixing in the SBL is more complex than suggested by this simple description. We turn to R_{ib}–D_{rb} bivariate distributions in an effort to understand this variability, and a clearer picture of SBL turbulence regimes and their interactions is revealed.

Examination of bivariate relationships reveals that significant differences in turbulence behaviour and the degree of decoupling can be associated with similar values of the Bulk Richardson Number. This finding re-emphasizes the important role known to be played by non-local external influences (e.g. breaking gravity waves, mesoscale circulations) in exciting local turbulence and mixing in the stable boundary layer. For the characterisation of vertically-integrated SBL behaviour, our approach which combines a bulk stability parameter (R_{ib}) with an independent measure of decoupling (D_{rb}) provides a valuable framework allowing new insights into the conditions under which we might expect to find various classes of turbulence behaviour.

An example of the insights to be gained from this approach is shown in Figure 1, which plots the bivariate distribution of 50 m−2 m temperature gradients. At high stabilities (R_{ib} >1), it is found that the most decoupled conditions (D_{rb} ~1) are encountered when temperature (and wind) gradients are proportionally smaller for a given value of Rib (mark 'A' in Figure 1). Other distributions indicate that these conditions correspond to the smallest turbulence intensities encountered at this site, and tend to be associated with times of high surface pressure (subsidence) and relatively small net radiation (partial cloud or light fog). Conversely, for the same Rib, conditions of higher temperature gradients (and correspondingly higher winds) tend to be associated with lower values of the decoupling index (D_{rb}~0.5) (mark 'B' in Figure 1).

Other distributions reveal that relatively large turbulence intensities and momentum fluxes are found at 50 m under these conditions, and temperature variances and heat-flux-based intermittency factors are large at 10 m. Increases in mesoscale variability, together with these high Brunt-Väisälä frequencies, point to the presence of gravity waves, and we infer a top-down mixing regime operating in this part of the distribution, associated with rapid surface cooling on clear-sky nights.

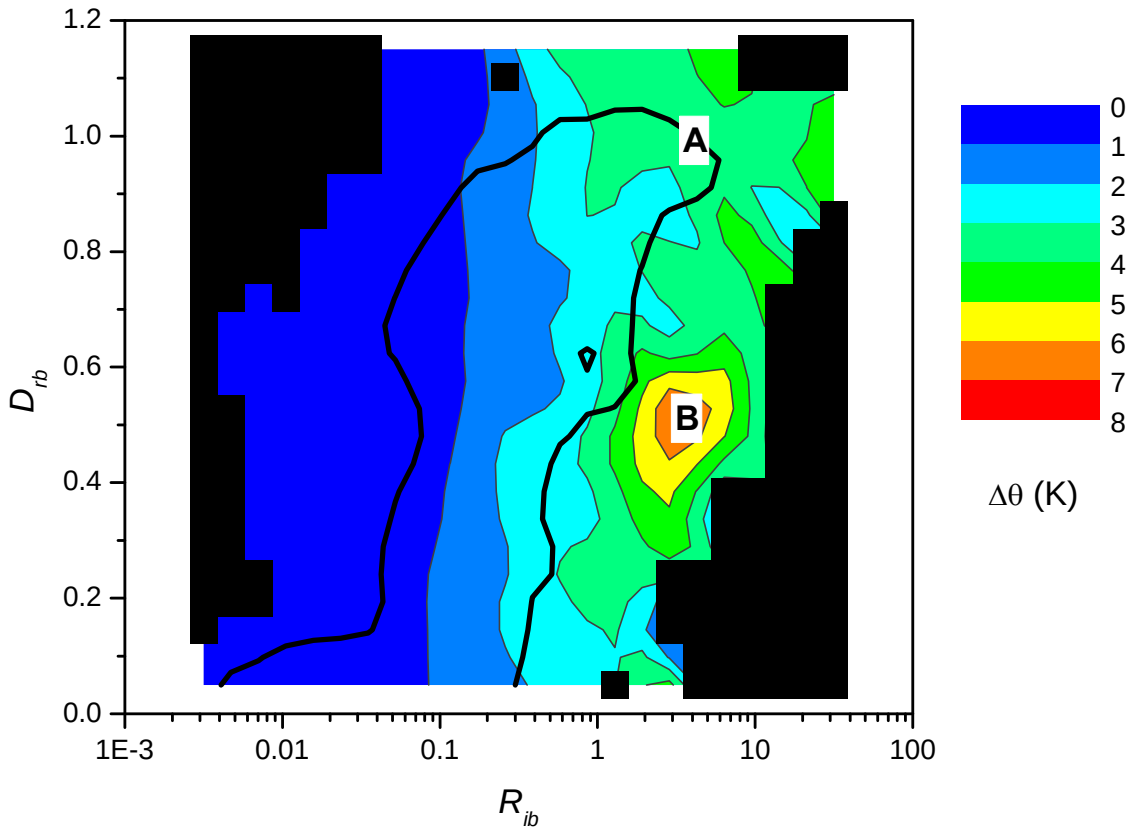


Figure 1. R_{ib}–D_{rb} bivariate distribution (24x24 grid points) of 50 m−2 m temperature gradients. Grid squares containing less than 4 data points are masked out in black. The thick black contour line surrounds the central section of the R_{ib}–D_{rb} histogram (>1% per grid square). See text for meaning of annotations and description of other details.

Surface Ocean Aerosol Production (SOAP) in NZ waters

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Abstract

Biologically-active regions of the surface ocean support production of a range of 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 eddy covariance flux estimates with gradient flux estimates on a floating catamaran, and primary & secondary organic aerosol formation in the MBL and in bubble burst chamber experiments. An overview and preliminary results from the SOAP voyage will be presented.

With an interest in increased vessel-based observation, there were a number of successful first-time sea-going instrument system deployments. In this category, continuous measurements of VOCs were made over the entire voyage by CSIRO PTR-MS coupled to an air inlet sector switch on R.V. Tangaroa for avoidance of contamination from local ship combustion sources. Discrete samples of DMS in air and water were analysed with sulfur chemiluminescence detection which ran for the majority of the voyage. Limited instrument comparisons were made.

References

Contacts and background:

http://www.niwa.co.nz/atmosphere/projects/soap

http://www.solas-int.org/science/researchendorsements/resendprojects/endorsedprojects.html#soap

[^]'The SOAP team' includes the following collaborations:

Queensland University of Technology: Luke Cravigan, Marc Mallet, Zoran Ristovski
CSIRO Marine and Atmosphere: Sarah Lawson, Jason Ward, Melita Keywood
University of Eastern Finland: Petri Vaattovaara
Department Earth System Science, UCI: Saltzman Research team and associates: Tom Bell, Warren de Bruyn, Christa Marandino, Cyril McCormick, Eric Saltzman
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Trend and seasonal cycle of atmospheric carbon monoxide at Cape Grim

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Abstract

Atmospheric carbon monoxide (CO) has been measured at Cape Grim for the past 27 years. Since 1985 flask air samples have been collected at Cape Grim and sent to CSIRO laboratories in Aspendale for analysis by gas chromatography. Since 1993, CO has also been measured by in situ gas chromatography at Cape Grim, every 40 minutes, as part of the AGAGE program.

Recently, improvements have been made to the treatment of the strongly non-linear response of the AGAGE CO instrument. The revised CO data will be presented, and compared to the CO flask data.

Features of the Cape Grim CO data will also be compared to partial column (from the ground to 12 km height) CO derived from ground-based Fourier Transform Spectroscopy at Lauder, New Zealand (Zeng *et al*., 2012).

References

Zeng, G., S. W. Wood, O. Morgenstern, N. B. Jones, J. Robinson & D. Smale, Trends and variations in CO, C_2H_6, and HCN in the Southern Hemisphere point to the declining anthropogenic emissions of CO and C_2H_6, *Atmos. Chem. Phys*., 12, 7543-7555, 2012.