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incorporating the

Cape Grim Annual Science Meeting 2016

Abstracts

16-18 November 2016

hosted by

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Stanley Town Hall
Stanley, Tasmania, Australia



Edited by: N. Derek and P. B. Krummel

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Day 1: Wednesday, 16 November 2016

09:00	**Convenors**	**Welcome & general information on arrangements etc**
Chair	**Paul Krummel**	**Session 1 – Oral Presentations**
09:10	Paul Fraser	Global 500 ppm equivalent carbon dioxide (CO_2-e) – when will it occur?
09:30	Roger Francey	Milestones in global carbon budget research – Tasmanian contributions
09:50	David Etheridge	Carbon monoxide concentrations in the southern hemisphere during the past century: reconstructed record and model simulations
10:10	Cathy Trudinger	Past atmospheric composition inferred from air in polar firn and ice: Latest results
10:30	**Morning Tea**	
Chair	**Melita Keywood**	**Session 2 – Oral Presentations**
11:00	Paul Selleck	Fine aerosol chemical composition over the Southern Ocean
11:20	John Ivey	A summary of seasonal atmospheric observations at Mackay: Implications for the Great Barrier Reef
11:40	Scott Chambers	Radon: a universal baseline indicator at sites with contrasting physical settings
12:00	Alastair Williams	Radon as an atmospheric tracer in urban environments
12:20	David Griffith	Radon tracer flux measurements of CO_2, N_2O and CH_4 at Wagga Wagga: OASIS revisited?
12:40	**Lunch**	
Chair	**Simon Alexander**	**Session 3 – Oral Presentations**
13:40	Stephen Montzka	On the uneven decline of atmospheric CFC-11: Bumps in the road to ozone recovery or variations in atmospheric transport and/or loss?
14:00	Paul Krummel	The 2016 Antarctic ozone hole
14:20	Jesse Greenslade	Stratosphere to troposphere ozone transport using ozonesondes at Davis, Macquarie Island, and Melbourne
14:40	Bruce Forgan	Revisiting deriving Total Column Ozone from solar spectral transmission in the Chappuis band (450-800 nm) from a 100 station-year record
15:00	Matthew Tully	Melbourne Total Ozone 1972-2016
15:20	Robyn Schofield	Reanalysis of Australian ozone profiles from Dobson observations
15:40	**Afternoon Tea**	
Chair	**David Griffith**	**Session 4 – Oral Presentations**
16:10	Martin Schultz	Comprehensive analysis of global surface ozone observations in the first Tropospheric Ozone Assessment Report
16:30	Ian Galbally	The semi-annual cycle in ozone in the marine boundary layer
16:50	Ashok Luhar	Improving dry deposition of ozone to seawater in a global composition model
17:10	Dagmar Kubistin	More than 15 years long-term monitoring of hydroxyl radicals at the GAW station Hohenpeissenberg
17:30		**End of Day 1**

Day 2: Thursday, 17 November 2016

Chair	Clare Paton-Walsh	Session 5 – Oral Presentations
09:00	Melita Keywood	Gunn Point: from Infant to Preppie
09:20	Christoph Zellweger	Traceability of measurements within the Global Atmosphere Watch Programme: Results from the World Calibration Centre WCC-Empa
09:40	Ann Stavert	The impact and importance of intercalibration and intercomparisons within the UK GAUGE greenhouse gas observational network
10:00	Zoë Loh	Using and improving the Australian Greenhouse Gas Observation Network
10:20	Beata Bukosa	GEOS-Chem simulations of greenhouse gas measurements (CO_2, CH_4 and CO) from moving platforms in and around Australia
10:40	**Morning Tea**	
Chair	**Stephen Wilson**	**Session 6 – Oral Presentations**
11:10	Peter Sperlich	Carbon isotope ratios suggest no additional methane from boreal wetlands during the rapid Greenland Interstadial 21.2
11:20	Lingxi Zhou	Study on variation of atmospheric CO_2 and source/sink characteristics based on stable carbon isotopes at SDZ in Beijing and LAN in Zhejiang
11:50	Simon Alexander	Cloud structure and phase at Cape Grim, Macquarie Island and over the Southern Ocean
12:10	Matthew Woodhouse	Modelling and observations of organic carbon in the marine atmosphere
12:30	Alicia Gressent	Optimal estimation of sulfuryl fluoride emissions on regional and global scales using advanced 3D inverse modeling and AGAGE observations
12:50	**Lunch**	
Chair	**Ruhi Humphries**	**Session 7 – Oral Presentations**
14:00	Christopher Roulston	Emissions of particulates from tropical peat burning in Malaysia
14:20	Clare Paton-Walsh	Long term trends in tropospheric composition from Australian ground-based remote sensing measurements
14:40	Nicholas Jones	Long term trends in stratospheric composition from Australian ground-based remote sensing measurements
15:00	Nicholas Deutscher	Total Carbon Column Observing Network activities in Australia: moving to portable technologies to complement primary sites
15:20	Voltaire Velazco	Southeast Asian & Australian TCCON operations-readying for Ibuki 2-validation (SATORI): The TCCON-Philippines Story

Day 2: Thursday, 17 November 2016 – Poster Session

15:40 Afternoon Tea	**Each presenter to give a 1-2 minute, 1-slide poster summary**
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Maximilien Desservettaz	Preliminary comparison of biomass burning inventories processed by GEOS-Chem with surface, total column and maybe satellite measurements in Australia
James Elkins	Using box models to quantify zonal distributions and surface emissions of halocarbons in the background atmosphere
Sonya Fiddes	Does the Great Barrier Reef have an impact on local climate?
Dean Howard	Four years of gaseous elemental mercury measurements at Cape Grim station
Ruhi Humphries	Measurements of aerosol-cloud properties in the Southern Ocean
Wenwen Huo	Using atmospheric radiocarbon to estimate fossil fuel sources: A test case with a known source
Sarah Lawson	Update from the Cape Grim VOC monitoring program
Jeongsoon Lee	Impact of background gas composition on CO_2 measurement by cavity ringdown spectroscopy
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Jennifer Powell	Trends in chlorobenzenes in air at marine background, urban and biomass burning-impacted sites in Australia
Fabienne Reisen	The impact of the 2016 Tasmanian fires on air quality
Mauro Rubino	Sensitivity of terrestrial biosphere CO_2 to temperature derived from pre-industrial CO_2, $\delta^{13}CO_2$ and COS
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Jason Ward	Aerosol measurements at Gunn Point: an update
Joel Wilson	Using skydiving and AirCore for convenient atmospheric profile sampling
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11:20	Jenny Fisher	Impacts of C1-C3 alkyl nitrates on the NOx budget in remote regions
11:40	Sarah Lawson	Atmospheric composition in a region of intensive coal seam gas production
12:00	Doreena Dominick	Observation of new particle formation in an urban marine coastal environment
12:20	Élise-Andrée Guérette	Evaluation of regional air quality models over Sydney, Australia
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Global 500 ppm equivalent carbon dioxide (CO_2-e) – when will it occur?

P. J. Fraser*, P. B. Krummel, L. P. Steele, M. V. van der Schoot, Z. M. Loh, B. Mitrevski,
R. L. Langenfelds and N. Derek

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Abstract

Using data from the CSIRO/BoM and AGAGE (Advanced Global Atmospheric Gases Experiment) global networks for measuring carbon dioxide (CO_2) and non-CO_2 greenhouse gases (GHGs: methane - CH_4, nitrous oxide - N_2O, chlorocarbons - e,g. carbon tetrachloride - CCl_4, chlorofluorocarbons - CFCs, hydrochlorofluoro-carbons - HCFCs, hydrofluorocarbons - HFCs, perfluorocarbons - PFCs, sulfur hexafluoride - SF_6, and others) we are able to derive key Kyoto and Montreal Protocol policy-relevant parameters such as global CO_2 levels, equivalent-CO_2 (CO_2-e) levels and Effective Equivalent Stratospheric Chlorine (EESC).

Global CO_2 levels exceeded 400 ppm in 2015 and Southern Hemispheric CO_2 in 2016. Global CO_2 first exceeded 400 ppm in March 2015, but dropped below from June-October, then rose again in November 2015 where it has remained ever since. Southern Hemisphere CO_2 passed 400 ppm in March 2016, and has/will remain above this from now on. May 2016 saw the 400 ppm milestone reached at Cape Grim, for both monthly mean and hourly mean (on the 10th) CO_2. Global CO_2-e levels are fast approaching 500 ppm. Just as global 400 ppm CO_2 was a significant psychological milestone that resonated in the public and policy domains, it is likely that 500 ppm CO_2-e will be so in the future. When will this occur? We make some predictions about when this will occur – 2020 may be a significant year for both marking anticipated progress in GHG emissions reductions versus national commitments and also for GHG levels in the atmosphere.

EESC levels continue to decline, but furthering this task, with the aim ultimately to close the Antarctic ozone 'hole', is becoming difficult. We have already reaped the benefit of falling levels of relatively short-lived ozone depleting substances (ODSs), such as methyl chloroform (CH_3CCl_3) and methyl bromide (CH_3Br). Unexplained emissions of CCl_4 persist and the anticipated significant decline in HCFCs is yet to occur. An update of global EESC levels will be presented.

We will present a brief review the status of all GHG (including ODS) measurements at Cape Grim.

Milestones in global carbon budget research – Tasmanian contributions

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Abstract

In the 40-years of Cape Grim measurements there have been several major revisions of the global carbon budget. Most have involved air (but also tree) samples collected in Tasmania and measured at CSIRO's GASLAB in Aspendale. Selected Tasmanian air samples can demonstrate larger spatial representation and higher precision in CO_2 and $\delta^{13}CO_2$ multi-decadal measurements compared to any other surface location. The $\delta^{13}CO_2$ track the anthropogenic terms on multi-year timescales and distinguish the ocean and terrestrial exchange on shorter timeframes. The Cape Grim data are providing new constraints on the global carbon budget that are yet to be fully appreciated.

The annual global carbon budget (influencing ocean acidification and climate change) can be represented by

$$\mathrm{dC/dt} = \mathrm{F_{(ff+luc)}} - \mathrm{F_{ocean}} - \mathrm{F_{terr}}$$

The annual global carbon budget (influencing ocean acidification and climate change) can be represented by where $\mathrm{dC/dt}$ = Annual increase in atmospheric CO_2; $\mathrm{F_{(ff+luc)}}$= Annual (anthropogenic) emissions from fossil fuel and land-use change; $\mathrm{F_{ocean}}$= Annual net CO_2 uptake by oceans, and $\mathrm{F_{terr}}$ = Annual net CO_2 uptake by land plants. Higher time and space resolution inversion models that track the link between anthropogenic emission changes and atmospheric CO_2 levels require extra information to achieve this budget, namely models of atmospheric transport that describe atmospheric exchanges between elements of a surface grid (typically 100x100km).

In the early 1980s, reports in Science of major contributions of soil carbon (Peng et al., 1982) and deforestation [Woodwell *et al*., 1983] to the global budget. CSIRO $\delta^{13}CO_2$ measurements on tree-rings [Francey and Farquhar, 1982] and ice cores [Francey *et al*., 1999] ruled out the soil carbon suggestion, while deforestation estimates rapidly declined in the face of early global budget evidence (summarised in Francey and Enting, [1990]). This still left a search for a 'missing sink' throughout following decades.

By 1990 a major disagreement had emerged on the inter-annual variability of the ocean and terrestrial exchange, based on $\delta^{13}CO_2$ measurements from Cape grim versus the pioneering Scripps Institution of Oceanography program [Francey e*t al*., 1995]. Only the Cape Grim record exhibits stable behaviour (more consistent with ocean carbon modelling) over four decades.

In a highly cited 1995 *Science* paper [Ciais *et al*., 1995, ~1000 citations], CSIRO isotopic data contributed to the location of the major terrestrial sink at mid Northern Hemisphere latitudes, large enough to balance the global budget for the first time. The absence of subsequent ground-based biospheric evidence, and a decadal perspective on the early 1990s Pinatubo-influenced atmospheric data used, question this result.

Emphasising the doubt, in 2007 Cape Grim aircraft data anchored a Science study claiming to resolve the 'missing sink' issue by locating the major terrestrial sink in the tropics [Stephens *et al*., 2007]. Widely used inversion models, relying on inventory estimates of anthropogenic emissions and atmospheric transport parameterisations, appear unable to resolve this issue.

Recent detailed examination of Cape Grim data has focussed on assumptions used in the models.

In a 2010 *Nature* paper [Francey *et al*., 2010], Cape Grim CO_2 growth was shown to be incompatible with the reported anthropogenic emissions growth spurt from around 2000, attributed mainly to development in China. The reported emissions are usually prescribed in inversion models. An underestimate of Asian anthropogenic estimates in the late 1990s remains the simplest explanation for this growth rate behaviour. (Note: If the Tasmanian data are given equal weight to data from other (~90) sites in the Global Atmospheric Watch program in models assimilating all available data, this result is lost in the uncertainties.)

In 2016, anomalous behaviour in the annual average CO_2 and $\delta^{13}CO_2$ difference (interhemispheric difference) using CSIRO Cape Grim and Mauna Loa measurements, was shown to coincide with anomalies in upper troposphere equatorial winds over the Pacific Ocean. The Cape Grim $\delta^{13}CO_2$ record is particularly sensitive to such exchange.

A follow-up study on seasonal timescales is suggesting that episodic interhemispheric transport through this region is the main inter-hemispheric exchange process for CO_2 [Francey, Frederiksen and Allison, in preparation]. This process is not captured with current global carbon budget models using diffusive inter-hemispheric exchange, which continue to favour terrestrial source/sink behaviour to explain changes the measured spatial gradients.

This result has relevance for other long-lived trace gas species based on transport modelling.

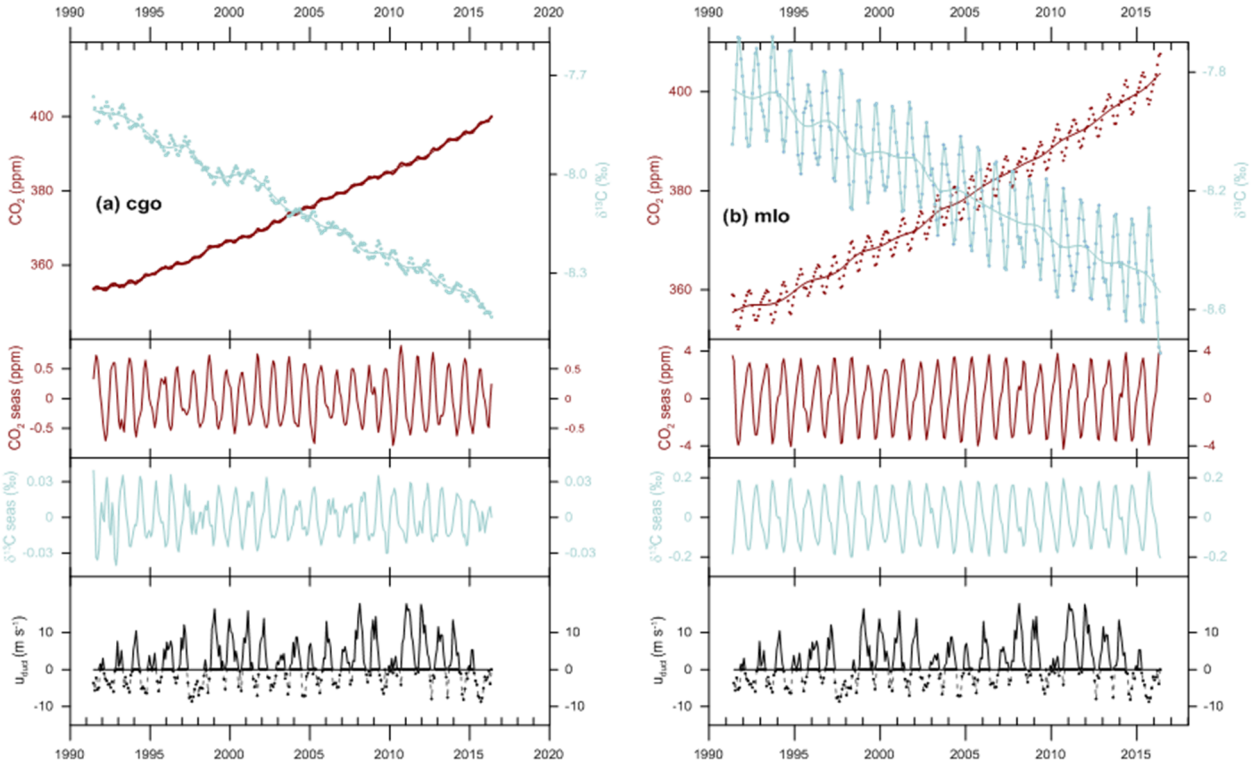


Figure: For each of (a) cgo and (b) mlo, the top panels show mean monthly baseline CO_2 (red) and $\delta^{13}C$ (blue) and respective trend curves, the second and third panels shows CO_2 (red) and $\delta^{13}C$ (blue) seasonality, and the bottom panel shows wind strengths in the upper troposphere equatorial duct.

Acknowledgements

GASLAB, Cape Grim and NOAA personnel for sampling and analyses; Paul Krummel for data processing; Jorgen Frederiksen and Colin Allison for recent unpublished work.

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Carbon monoxide concentrations in the Southern Hemisphere during the past century: reconstructed record and model simulations

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Abstract

Carbon monoxide (CO) has significant roles in tropospheric chemistry, including being a major sink for the hydroxyl radical which in turn controls the lifetimes of reactive greenhouse gases and ozone depleting gases. Significant changes in CO concentration probably occurred over the past century but atmospheric measurements began only in 1979. The high quality Antarctic firn air measurements published so far extend the atmospheric record back in time by only about 10 years and are available from just one site.

We reconstructed CO concentrations in the high latitude southern hemisphere over the past century from measurements of firn air, ice core air and atmospheric observations. Measurements of firn air from multiple Antarctic sites (Law Dome, South Pole, EPICA Dome C, Berkner Island, Fletcher Promontory and Aurora Basin North), with different characteristics help exclude the possibility of artefacts from in situ processes and sampling methods. The Law Dome samples provide high air age resolution and preserve decadal atmospheric variations. Firn air models are used to derive the atmospheric CO concentrations that are consistent with the measurements from these sites. The firn air reconstruction compares well with the atmospheric records over recent decades from nearby Casey and Mawson stations. CO increased about 30% with most of the increase occurring between about 1945 and the early 1990s.

The CO measurements are compared to simulations from the ACCESS-UKCA Earth System model. The causes of the CO increase from the main sources inferred from the model simulations and the reconstructed record will be discussed.

Past atmospheric composition inferred from air in polar firn and ice: Latest results

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Abstract

Air extracted from firn (the layer of unconsolidated snow overlying an ice sheet) or bubbles in polar ice provides a reliable way to reconstruct the atmospheric abundance of trace gases prior to direct atmospheric measurements. Here we discuss how we reconstruct the past atmospheric history of trace gases, taking account of the processes in firn and ice that affect the enclosed air. We present our latest reconstructions of perfluorocarbons [Trudinger *et al*., 2016; Mühle *et al*., in preparation], halons [Vollmer *et al*., 2016], carbon monoxide (Etheridge et al., in prep), minor CFCs [Vollmer *et al*., in preparation] and SO_2F_2 [Mühle *et al*., in preparation] using firn and ice core measurements from combinations of Antarctic and Greenland sites, specifically DE08, DE08-2 and DSSW20K from Law Dome, South Pole 2001, NEEM 2008, EDML, Megadunes, Summit 2013 and ABN. We also derive global emissions for some of these trace gases. Our reconstructions provide information on background/pre-anthropogenic levels. We demonstrate the advantage of locations with narrow air-age resolution – for example, we are able to resolve small peaks in emissions of perfluorocarbons CF_4 and C_2F_6 due to aluminium production during World War II from the high-resolution DE08 and DE08-2 ice core measurements.

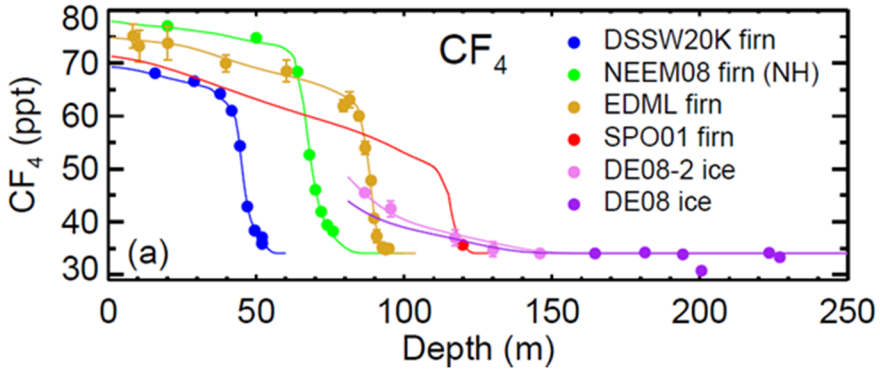


Figure: Depth profiles of CF_4 mole fraction in the firn and ice at 6 different locations. Symbols show measurements with 1σ uncertainties; lines show model results corresponding to emissions inferred using all of these observations plus archived and *in situ* atmospheric measurements simultaneously.

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Fine aerosol chemical composition over the Southern Ocean

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Abstract

Aerosols have direct effects on the climate by scattering and absorbing light or indirectly by modifying cloud properties. Clouds over the Southern Ocean are poorly simulated in global climate models and present-day reanalysis products [Naud *et al*., 2014] resulting in large radiation biases. The processes that influence cloud-forming aerosol properties in the remote marine atmosphere such as the Southern Ocean are poorly understood [Quine and Bates, 2011] and the atmosphere over the Southern Ocean is one of the most poorly studied regions on the globe due primarily to the harsh conditions and remote location. Climatically important marine aerosols may be comprised of non sea-salt sulfate, sea-salt and organic compounds.

Weekly samples of Southern Ocean fine ($PM_{2.5}$) aerosol have been collected at Cape Grim since 2002. The samples are collected during baseline conditions onto Teflon filters and then weighed and analysed for water soluble ions by suppressed ion chromatography. The $PM_{2.5}$ samples show a clear seasonal cycle with a summer maximum and winter minimum for methane sulfonate, non sea-salt (nss) sulfate, oxalate and ammonium. Sea-salt is the dominant component year round making up 78±3% of the mass. The major sea salt components of sodium and chloride do not show a seasonal cycle, however a seasonal cycle is observed in the chloride/sodium mass ratio, with a maximum in winter of 1.76 close to the seawater ratio of 1.80 and a minimum in summer of 1.60. This indicates a loss of chloride in the summer months which could be due to displacement by sulphuric and nitric acid.

During February 2015, CSIRO's research vessel the RV Investigator undertook a cold water trial (CWT) to the ice edge in the Southern Ocean, during which online submicron aerosol chemical composition measurements where made using a time of flight aerosol chemical speciation monitor (ToF-ACSM). Mass analysis by a time of flight mass spectrometer allows high resolution fits to the data enabling the separation of isobaric peaks. The major species measured were sea-salt with an average concentration of 0.20 μg m^{-3}, nss sulphate with an average concentration of 0.10 μg m^{-3} and total organic compounds with an average concentration of 0.03 μg m^{-3}.

The concentration of nss sulfate and ammonium measured in $PM_{2.5}$ samples collected at Cape Grim at the same time as the CWT voyage was 0.45 μg m^{-3} and 0.11 μg m^{-3} respectively; the nss sulfate/ammonium ratio of 4, was similar to the long term average of 4.2±0.9 indicating that these species may be a mixture of ammonium bisulfate and ammonium sulfate. The average concentrations of nss sulfate and ammonium measured during the CWT was 0.10 μg m^{-3} and 0.01 μg m^{-3} and the nss sulfate/ammonium ratio was 10 suggesting that these species may be a mixture of ammonium bisulfate and sulfuric acid. The ratio of sulfate ion fragments $H_2SO_4^+/SO^+$, was higher during periods of low ammonium and low sea salt concentrations indicating that nss sulfate exists in the form of sulfuric acid. During periods of higher sea salt concentrations, lower $H_2SO_4^+/SO^+$ ratios indicate that the nss sulfate may have reacted with the alkaline sea salt aerosol.

This initial comparison of the two data sets show that the aerosol chemistry of some species measured at Cape Grim may not represent the rest of the Southern Ocean or that artefacts may have changed some of the components on the filters during sampling. This highlights the need for online chemical composition measurements to be made at Cape Grim alongside the filter sampling.

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A summary of seasonal atmospheric observations at Mackay: Implications for the Great Barrier Reef

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Abstract

The Cape Grim Baseline Air Pollution Station (CGBAPS) has provided critical insight into many fields of atmospheric studies of global and regional importance. One such field of study has been the natural sulfur cycle and the potential for anthropogenic perturbation. The Mackay (lat. -21.14°, long. 149.18°) atmospheric study is designed to address principally, but not exclusively, the relative contributions of the natural marine sulphur cycle, the anthropogenic marine emissions from shipping, and the deposition of their oxidation artefacts to the Great Barrier Reef (GBR). The shipping artefacts include nitrate, sulfate, and acidity.

By contrasting the marine biogenic oxidation artefact, methane sulphonic acid (MSA) with the anthropogenic artefact nitrate we hope to proportion the contribution of both sources to the deposition of acidity, nitrate and non-sea-salt sulfate to the GBR. Although sulfur dioxide (SO_2) emissions have been modelled [Goldsworthy and Goldsworthy, 2014; Golsworthy and Galbally, 2011] and marine shipping emissions measured [Blasco *et al*., 2014], no confirming observational studies for the GBR have been undertaken.

The sampling design utilises two sites, one at the Mt. Bassett Bureau of Meteorology (BoM) Station north of the Mackay Central Business District (CBD) where a site sharing agreement is in place, and the second south of the CBD adjacent to the Mackay airport which also has a BoM meteorological station. The Mackay location in relation to the GBR and shipping corridors is shown in the Figure below. Observations include precipitation, aerosol collections, sulphur dioxide, nitrogen dioxide, particle size, radon (^{222}Rn) and meteorological parameters over a seasonal cycle. Our preliminary interpretation of results based on precipitation and some aerosol collections imply an enhancement of nitrate and sulphate above the background expected by the MSA concentration and comparison with Cape Grim and remote Pacific measurements [Savoie *et al*., 1989]. The precipitation acidity (pH) was in close association with the nitrate concentration averaging below 5.

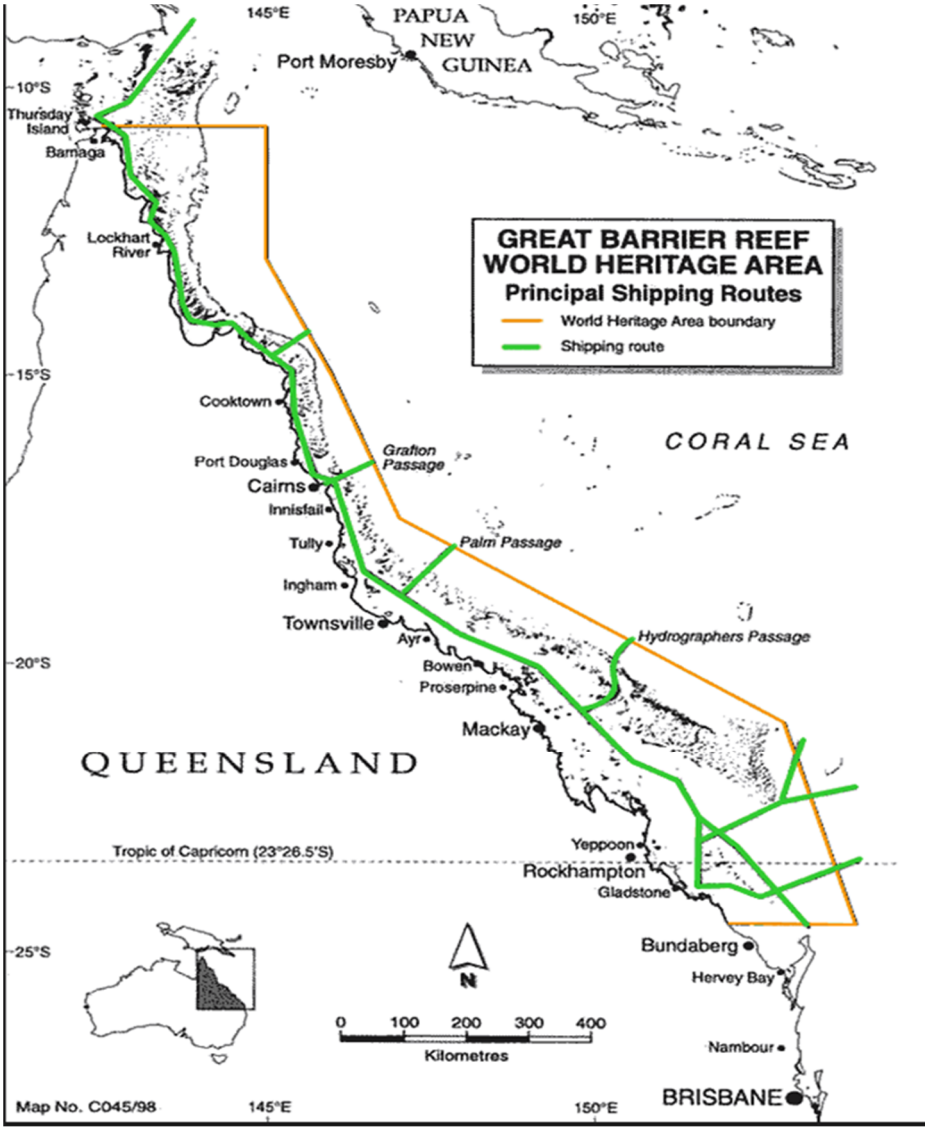


Figure: Mackay and its proximity to the Great Barrier Reef, shipping routes and the World Heritage Area.

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Radon: a universal baseline indicator at sites with contrasting physical settings

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Abstract

The primary goal of World Meteorological Organisation Global Atmosphere Watch (WMO-GAW) baseline stations is systematic global monitoring of chemical composition of the atmosphere, requiring a reliable, consistent and unambiguous approach for the identification of baseline air. Premier stations in the GAW baseline network span a broad range of physical settings, from remote marine to high-altitude continental sites, necessitating carefully tailored site-specific requirements for baseline sampling, data selection, and analysis. Radon-222 is a versatile and unambiguous terrestrial tracer, widely-used in transport and mixing studies. Since the majority of anthropogenic pollution sources also have terrestrial origins, radon has become a popular addition to the 'baseline selection toolkit' at numerous GAW stations as a proxy for 'pollution potential'. In the past, detector performance and post-processing methods necessitated the adoption of a relaxed (e.g. 100 mBq m^{-3}) radon threshold for minimal terrestrial influence, intended to be used in conjunction with other baseline criteria and analysis procedures, including wind speed, wind direction, particle number, outlier rejection and filtering. However, recent improvements in detector sensitivity, stability and post-processing procedures have reduced detection limits below 10 mBq m^{-3} at Cape Grim and to ~25 mBq m^{-3} at other baseline stations. Consequently, for suitably sensitive instruments (such as the ANSTO designed and built two-filter dual-flow-loop detectors), radon concentrations alone can be used to unambiguously identify air masses that have been removed from terrestrial sources (at altitude or over ice), or in equilibrium with the ocean surface, for periods of >2-3 weeks (radon ≤ 40 mBq m^{-3}). Potentially, radon observations alone can thus provide a consistent and universal (site independent) means for baseline identification. Furthermore, for continental sites with complex topography and meteorology, where true 'baseline' conditions may never occur, radon can be used to indicate the least terrestrially-perturbed air masses, and provide a means by which to apply limits to the level of 'acceptable terrestrial influence' for a given application. We demonstrate the efficacy of the radon-based selection at a range of sites in contrasting physical settings, including: Cape Grim (Tasmania), Cape Point (South Africa), Mauna Loa (Hawaii), Jungfraujoch (Switzerland) and Schneefernerhaus (Germany).

Radon as an atmospheric tracer in urban environments

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Abstract

We demonstrate the multiple uses of radon as a tracer to explain observed diurnal characteristics of air pollution levels in a small inland European city (Bern, Switzerland), and to separate the influences of: local emissions within the city area; vertical dilution by boundary layer mixing; and horizontal advection of cleaner air from outside the city. This is accomplished by: (i) characterising the temporal variability in traffic density, radon, benzene and CO emissions at Bern city centre on diurnal and seasonal timescales; (ii) adapting a radon-based stability technique developed by Chambers et al. (2015) to make it independent of seasonal changes in day length and radon source function; (iii) characterising the combined influence of traffic density and meteorological conditions on benzene and CO concentrations in Bern; and (iv) utilising the diurnal radon signal, together with an idealised box model incorporating a simple advection term, to remove the combined effects of local horizontal advection and atmospheric dilution. This allows us to seek an accurate relationship between traffic density and pollutant emissions in this compact inland city.

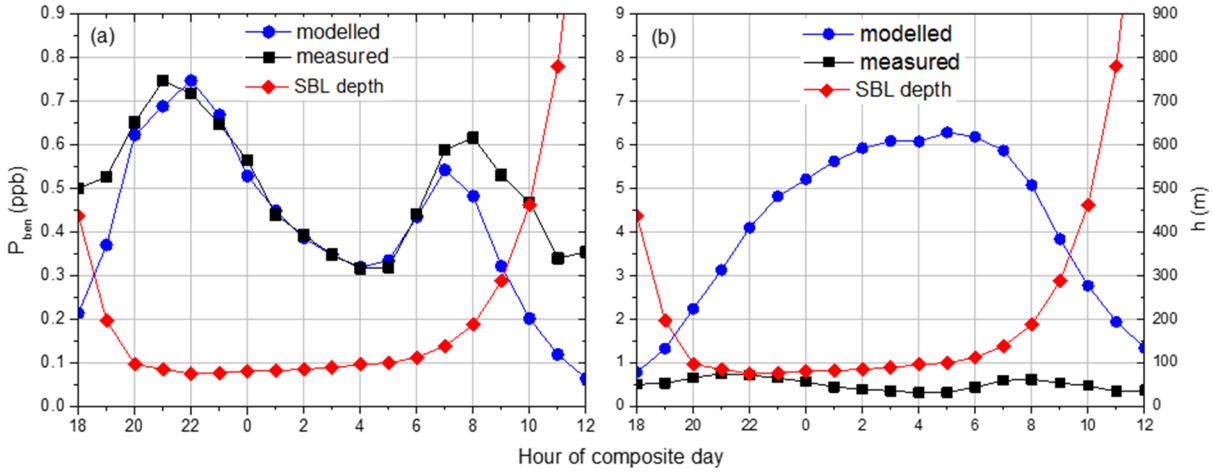


Figure: Comparison of measured diurnal benzene concentrations in Bern city centre on strongly stable nights in springtime (black) with results from a radon-calibrated box model (blue): (a) with; and (b) without a simple horizontal advection term. Radon-based nocturnal stable boundary layer depths (h) are shown in red (right-hand axis). Adapted from Williams *et al*. [2016].

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Radon tracer flux measurements of CO_2, N_2O and CH_4 at Wagga Wagga: OASIS revisited?

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Abstract

Estimating regional scale emissions of nitrous oxide from agricultural environments to refine greenhouse gas budgets and inventories remains a difficult task because of the low atmospheric concentrations, small fluxes and small concentration gradients of N_2O. In this work we revisit the site of the OASIS flux campaigns in 1994 and 1995 near the Charles Sturt University (CSU) campus at Wagga Wagga, a mixed-farming region typical of large areas of NSW. We combine continuous flux measurements of tracer species Radon and CO_2 (by chamber and eddy covariance, respectively) with concentration measurements of these gases as well as N_2O and CH_4 on a 10 m tower in a grassed paddock. We utilise correlations between the trace gases and tracers as they build up in the nocturnal boundary layer (NBL). Under favourable conditions, the NBL is capped by a low inversion and acts as a km-scale 'megachamber'; if the flux of tracer and ratio of concentrations of trace gas and tracer are measured, the unknown nocturnal flux of the trace gas can be calculated assuming similar source distributions of the unknown and tracer. This allows the measurement of regionally averaged soil N_2O emission fluxes at the low levels typical of low-nitrogen Australian soils.

The measurement campaign is currently running, from August – December 2016, using radon flux and concentration measurements from ANSTO, FTIR and LGR analysis of trace gases from University of Wollongong (UoW) and CSU, and local support and infrastructure from CSU. We will present the measurements to date with preliminary results and calculations of fluxes.



[#] Ray Leuning (1948-2016) was a colleague and friend to many of us; sadly he passed away in February 2016. Ray was involved in the modelling and development of nocturnal boundary layer (NBL) integrating methods which led, in part, to this research.

On the uneven decline of atmospheric CFC-11: Bumps in the road to ozone recovery or variations in atmospheric transport and/or loss?

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Abstract

Long-term, high-quality atmospheric measurements, such as those made at the Cape Grim Baseline Air Pollution Station, allow important policy-relevant science questions to be addressed. An example relates to tracking atmospheric concentrations of ozone-depleting substances to gauge the effectiveness of the Montreal Protocol to heal the stratospheric ozone layer and to understand the persistence of the atmosphere's ability to cleanse itself of pollutants. We have measured mole fractions of the ozone-depleting and greenhouse gas CFC-11 at the CGBAPS and other remote stations around the globe since the mid-1990s. The concentration of CFC-11 has decreased since the early 1990s because of controls on global production associated with the fully adjusted and amended Montreal Protocol on Substances that Deplete the Ozone Layer. The rate of atmospheric decline accelerated gradually over the next decade, and from 2002 to 2012 CFC-11 mole fractions decreased at a near-constant rate of 2.2 ± 0.2 ppt/yr in both the northern and southern hemispheres. Since 2012, however, a substantial slow-down in the atmospheric decline of CFC-11 has been observed by three quasi-independent techniques at NOAA/GMD (the 2013 to 2015 rate was –1.3 ± 0.1 ppt/yr), with the slow-down being most prominent in the northern hemisphere. Given that global production of CFC-11 has been essentially zero since 2007, it seems improbable that this anomaly is due to increased emissions. Here we will explore this possibility, as well as the possibility that variations in transport (or in loss rates as captured by surface observations) might explain the slower decline. Preliminary analyses with an idealized model suggest that the mass flux of CFC-11 from the stratosphere to the troposphere was anomalously low during 2014. Does this transport-related anomaly explain the slowdown in the CFC-11 decline in 2014 and does it imply changes in the atmosphere's ability to cleanse itself of ozone-depleting gases? Or do the observations provide irrefutable evidence of a significant increase in global CFC-11 emissions since 2013?

The 2016 Antarctic ozone hole

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Abstract

The 2016 ozone hole developed in the first week of August and reached its maximum size in late September. Overall, the 2016 ozone hole was in the 'middle of the pack' compared to the last 37 holes for which we have satellite data since 1979, ranking between 17th and 20th for a range of metrics. The polar vortex in 2016 appears to be affected by wave activity that results in the ozone hole oscillating between periods of distortion and relative stability.

The development and recovery of the 2016 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, and the OMPS (Ozone Mapping and Profiler Suite) instrument on the Suomi National Polar-orbiting Partnership satellite (Suomi NPP). Other observational results will be shown to complement these satellite data. The 2016 ozone hole metrics will be compared to the historical metrics, and these will be linearly regressed against Equivalent Effective Stratospheric Chlorine (EESC) data that are based entirely on Cape Grim Air Archive, Antarctic firn and AGAGE global measurements of ozone depleting substances (ODS). The year-to-year variability of the ozone hole does not reflect variations in the level of ODSs in the Antarctic stratosphere, which, based on surface observations, have fallen approximately 10% since their peak around 2000, or have fallen by approximately 19% since their peak around 2000 relative to 1980 levels of ODSs. The decadal variations in the ozone hole do reflect the long-term changes of ODSs in the Antarctic stratosphere and the results shown here suggest that ozone hole recovery has commenced.

Stratosphere to troposphere ozone transport using ozonesondes at Davis, Macquarie Island and Melbourne

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Abstract

We develop a quantitative method to identify Stratosphere to Troposphere Transport events (STTs) from ozonesonde profiles. Using this method we estimate the seasonality and quantity of ozone transported across the tropopause over Melbourne (38°S), Macquarie Island (54°S), and Davis (69°S). Using the multi-year average from the ozonesonde dataset we show a distinct summer maximum and winter minimum for each of our sites, although this is less pronounced at Davis.

The meteorological cause of these events is examined qualitatively using ERA-I pressure and wind datasets. Synoptic scale low pressure fronts are the main cause of STTs, with around half of the detected events near or under a coincident front. Most of the impact is in the upper troposphere with the majority ozone peaks occurring within 3~km of the tropopause at Melbourne and Macquarie Island, and within 2~km of the tropopause at Davis.

Using the AIRS CO dataset, all detected tropospheric enhancements which are co-located with enhanced CO plumes are flagged as possibly influenced by biomass burning smoke. Almost all of these flagged events occur in the austral winter, during the southern Africa and South America burning seasons.

Using 7-9 years of ozonesonde data, we calculate a conservative estimate of the fraction of tropospheric ozone attributed to STT events of 2 – 4% at each site. We use the GEOS-Chem [Bey *et al*., 2001] model and extrapolate southern oceanic tropospheric ozone with STT likelihood and impact. The GEOS-Chem model is run with active stratospheric chemistry [Eastham *et al*., 2014], but is too coarsely resolved in the vertical dimension to determine STTs.

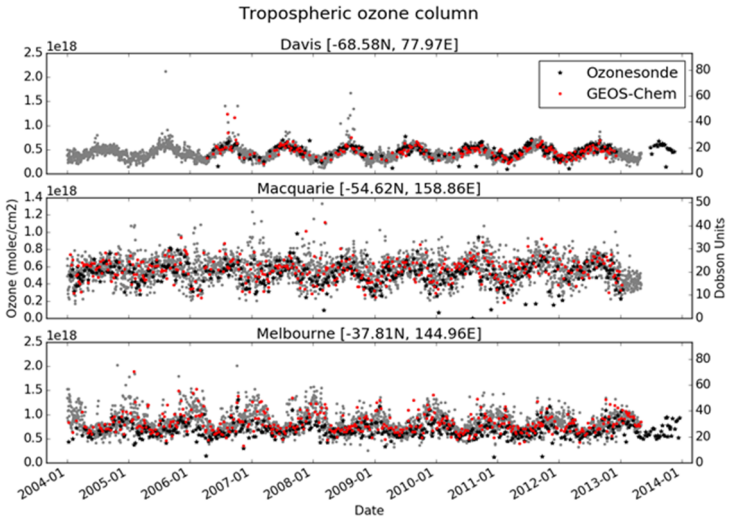


Figure: Tropospheric column from ozonesondes and simulated by GEOS-Chem.

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Revisiting deriving Total Column Ozone from solar spectral transmission in the Chappuis band (450-800 nm) from a 100 station-year record

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Abstract

The measurement of total column ozone (TCO) has typically been measured with specifically designed spectrophotometers (Dobson, since 1930), spectroradiometers (Brewer, since the late 1960s) and more recently by satellite-based remote sensing (1978+); all these methods use ozone extinction in the UV (300-330 m, Hartley-Huggins bands). During the 1970s-1990s deriving TCO from solar extinction measurements in the Chappuis band was tried with varying degrees of success, and largely abandoned as impractical given the uncertainties both in the knowledge of the ozone cross-sections and the available metrology knowledge on interference filter solar radiometry.

This paper reports the initial results of a re-examination of the use of narrowband filter solar radiometry in the Chappuis band to derive TCO using signals of direct solar transmission at three nominal wavelengths, 500, 610 and 778 nm. A new method is introduced that uses an iterative process for solving the Bouguer-Lambert-Beer (B-L-B) law of extinction with multiple specie components to derive morning and afternoon values of TCO when clear sun periods are available and utilizes the correlation in extinction across wavelengths to reduce uncertainties.

The results reported comprise a summary of 100 station-years of spectral measurements from the 10 nm full-width-half-maximum filter interference SPO2 radiometers used at Bureau solar and terrestrial irradiance monitoring sites since 1998, and a smaller time series using the high spectral resolution CCD array radiometers (ASR) during a period between 2013-2015.

To test the validity of the results comparison with satellite-derived TCO data from 2004 was used.

The initial analysis of the narrow band Chappuis results shows that using the B-L-B method the standard deviations of the difference between the coincident daily B-L-B and OMI-TOMS determination were ~12 DU for the majority of sites and the B-L-B method provided a good representation of the annual TCO cycle as all sites. The magnitude of the mean difference between the satellite and SPO2/ASR estimates was found to be dependent on the knowledge of the actual representative wavelengths and the latitude of the site, with tropical sites having an offset of -20 DU, and sites between 35 and 40°S having near zero offsets. The latitudinal offsets were decreased but not eliminated through application of a season latitudinal variation of the ozone extinction scale height when deriving the ozone air mass in combination with the latest laboratory-based ozone extinction cross sections.

Melbourne Total Ozone 1972-2016

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Abstract

The Bureau of Meteorology has made high quality measurements of total column ozone using the Dobson spectrophotometer in the Melbourne area since 1983, continuing a dataset originally started by CSIRO at Aspendale. In 1972, the instrument then in use, Dobson 105, was simultaneously compared with Dobson #83 from Boulder, USA which in later years went on to be designated the World Standard Dobson. This comparison has now been re-analysed and the resulting calibration applied to the early data, enabling the creation of a homogenous and consistently processed dataset from 1972 to the present, of immense value to understanding decadal ozone trends in the southern hemisphere.

An analysis of the newly reprocessed 1972-2016 time-series will be presented, concentrating on the influence on the long-term trend of stratospheric chlorine from Ozone Depleting Substances of anthropogenic origin, as well as natural drivers of variability such as the 11-year solar cycle and the Quasi-Biennial Oscillation.

Reanalysis of Australian ozone profiles from Dobson observations

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Abstract

Spectral observation of UV sunlight from the ground is the oldest methodology employed to determine the vertical distribution of ozone in our atmosphere. There has been significant recent effort to revisit and reanalyse historical Dobson Umkehr data, for example Miyagawa *et al*. [2009] for the Japanese sites and Krzyscin and Rajewska-Wiech [2009] for Belsk in Poland (1963-2007). Globally, *Bojkov et al.*, [2002] examined 15 long-term Umkehr sites, of ~80 total, including some of the Australian sites presented here. The Australian sites examined were incomplete, with early records not digitized, of particular significance are Brisbane and Perth data records, which extend from the 1960s to present. The profiles retrieved from these Umkehr data represent ozone products that pre-date the satellite era and provide significant climate records. The Australian sites span latitudes from the tropics to the Southern Ocean. Data from six Australian sites are presented: Darwin (12.4°S, 130.8°E; 1966-1974, 1990-2001, 2001-present), Brisbane (27.5°S, 153°E; 1967-present), Perth (31.9°S, 116°E; 1969-present), Melbourne (38°S, 145.1°E; 1955-2001), Hobart (42.9°S, 147.3°E; 1967-1973, 1974-1992), Macquarie Island (54.5°S, 158.9°E). We present the full instrumental records for the six Australian Umkehr sites (including several site and instrumental changes). Ozone profiles are retrieved using the updated methodology of Stone *et al*. [2015] which takes full advantage of improved information content achieved when the complete SZA range and all A, C and D wavelength pairs are used. The Dobson partial columns are compared with the Bodeker ozonesonde homogenized dataset, with generally very good agreement for the total column amounts, however the partial columns highlight differences.

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Comprehensive analysis of global surface ozone observations in the first Tropospheric Ozone Assessment Report

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Abstract

The first Tropospheric Ozone Assessment Report (TOAR) aims at assessing the current state and changes in tropospheric ozone and at providing easily accessible, comprehensive datasets for the evaluation of ozone impacts on human health, agriculture, and climate. TOAR is an international activity involving about 200 scientists globally and it is coordinated by an eleven-member steering board.

As one central component of TOAR, the world's largest collection of surface ozone measurements from the 1970s through 2014 has been ingested into a database at Forschungszentrum Jülich. The data and metadata have been harmonized and quality controlled, and augmented by additional metadata information from high resolution geospatial datasets providing information on population, emissions, land use, climatic zones, etc. Data from more than 9,500 stations around the world are now available for the first time in harmonized form to allow for consistent analyses of about 20 different ozone metrics. Many datasets have been shared with the international community for the first time, including for example data from 56 Australian air quality stations. Standardized aggregate data products with stringent data selection criteria are provided for the analysis of present-day ozone distributions and ozone trends. A web site and REST web service provide free and easy access to the TOAR database.

TOAR is scheduled to submit its report (as a special issue in Atmospheric Environment) in April 2017. This presentation will summarize the current state of analysis with a focus on the surface ozone evaluation.

The semi-annual cycle in ozone in the marine boundary layer

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Abstract

Recently a semi-annual cycle in ozone in the marine boundary layer has been identified as a widespread phenomenon that is of use in the objective comparison of models and observations [Parrish *et al*., 2016, Derwent *et al*., 2016]. Here the physical basis of this semi-annual cycle is explored in terms of the underlying physics and chemistry.

We suggest that this semi-annual cycle is a basic property of the Sun-Earth system. The presence and amplitude of this semi-annual cycle is determined by the spherical geometry of the Sun-Earth system combined with a stratosphere that is optically thick in the Huggins Band (300 – 340 nm). These regulate a semi-annual cycle in the actinic flux that determines the rate of ozone photolysis to form O(1D). In the remote boundary layer where there are low NO concentrations, (<~20 ppt) this photolysis leads to ozone loss. We utilize a simple model of ozone in the marine boundary layer to demonstrate how well this description fits observations at remote sites and we present an uncertainty analysis to determine the role of other meteorological and chemical processes in determining the amplitude and phase of this semi-annual ozone cycle.

Also the broader implications of the semi-annual cycle as a test of the 3-D modelling of ozone chemistry and dynamics in the troposphere will be presented.

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Improving dry deposition of ozone to seawater in a global composition model

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Abstract

Ozone destruction at the Earth's surface, described as dry deposition, is an important sink of tropospheric ozone, and according to the previous estimates about one third of it is to the oceans. The deposition rate is equal to the product of the near-surface ozone concentration and a deposition velocity. Ozone dry deposition velocity to seawater in different models is formulated with regards to how the dominant term of surface resistance is modelled. We examine three schemes for ozone dry deposition to seawater and test them in a global composition model, viz. ACCESS-UKCA, that incorporates meteorological nudging and monthly-varying reactive-gas emissions. The first scheme uses the common assumption that the water surface resistance is constant. The other two schemes, named the one-layer and a modified two-layer reactivity schemes, include the simultaneous influence on the water surface resistance of ozone solubility in water, waterside molecular diffusion and turbulent transfer, and a first-order chemical reaction of ozone with dissolved iodide. Unlike the one-layer scheme, the two-layer scheme can indirectly control the degree of interaction between chemical reaction and turbulent transfer through the specification of a surface reactive layer thickness. A comparison of the modelled deposition velocities from the three schemes with cruise based observations is presented. The two-layer scheme with a surface reactive layer thickness specification of 2.5 microns, which is approximately equal to the reacto-diffusive length scale of the ozone-iodide reaction, is able to simulate the field measurements most closely. The global oceanic deposition of ozone determined using this scheme is approximately 12% of the total global ozone deposition, which is much lower than the previous estimate of about 30%. The influence of the deposition parameterisation on the predicted ozone mixing ratios is also discussed.

More than 15 years long-term monitoring of hydroxyl radicals at the GAW station Hohenpeissenberg

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Abstract

Hydroxyl radicals (OH) are known to be the major daytime detergent of the atmosphere playing a key role in the initiation of oxidation processes and in tropospheric ozone and particle formation. Their abundance is governed by the presence of water vapour, ozone, sun light, as well as by biogenic and anthropogenic pollutants. Anthropogenic emissions, change in land use and climate change have being altering the atmospheric composition constantly with potential impact on the atmospheric self-cleaning capacity. Whilst several field campaigns have measured OH radicals for short term studies, hardly any continuous long term observations are available. So far, the long term behaviour of global OH concentrations has been solely estimated indirectly by the decay of certain tracers with well-known emissions, such as methylchloroform.

Here, we report about long term OH measurements, performed since 1998 on an almost continuous base at the global GAW station Hohenpeissenberg, South Germany. OH is being measured alongside with sulfuric acid, other trace gases and meteorological parameters routinely, using chemical ionization mass spectrometry (CIMS). Diurnal and seasonal cycles, as well as the time series are presented and the possible impact of change in the atmospheric composition over almost eighteen years is investigated.

Gunn Point: from Infant to Preppie

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Abstract

The tropics play a major role in global climate through both natural and anthropogenic processes, for which many potential feedbacks are poorly understood. The tropical regions globally are expected to be sensitive to future climate change due to vulnerabilities in the large carbon pools of tropical vegetation, influenced by biomass burning, deforestation and the draining of the tropical peatlands [Canadell *et al*., 2007]. However tropical regions are the most sparsely sampled in global atmospheric observing networks. To address this deficiency, in June 2010 the Australian Tropical Atmospheric Research Station (ATARS) was established near Darwin in the Northern Territory at the existing BoM radar facility at Gunn Point.

Initially established to measure greenhouse gases, the remit of the station expanded to the measurements of aerosols and reactive gases with the addition of a second demountable laboratory in 2013, which includes a sophisticated aerosol conditioning system. Operating in a remote site in the tropic has its challenges so that instrument malfunctions and environmental factors have interrupted the collection of datasets. However, as we gain experience, these interruptions have become reduced in length and significance.

In this presentation activities at Gunn Point, over the first 6 years of its existence (from infant to toddler to preppie) will be described; including evolution of the measurement program, collaborations, long term data sets produced and results from two intensive measurement campaigns.

We will argue that the impact of information produced at Gunn Point is just beginning to be realised. However, we have reached a decision point about the continued operation of the station and we will propose that observation and research activities at Gunn Point enter the envelope of the Cape Grim Science Program.

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Traceability of measurements within the Global Atmosphere Watch Programme: Results from the World Calibration Centre WCC-Empa

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Abstract

Empa operates the World Calibration Centre for Carbon Monoxide, Methane, Carbon Dioxide and Surface Ozone (WCC-Empa) since 1996 as a Swiss contribution to the Global Atmosphere Watch (GAW) programme and has conducted over 80 system- and performance audits over the past 20 years. In order to increase the number of nitrous oxide audits, WCC-Empa also cooperates with the World Calibration Centre for N_2O at KIT/IMK-IFU. The activities of WCC-Empa significantly contribute to sustain and improve the data quality required for climate and environmental research. The concept of the performance audits was recently expanded by the addition of parallel measurements with a travelling instrument using an entirely independent inlet system and calibration scheme [Zellweger *et al*., 2016).

The presentation will highlight results of CO, CH_4, CO_2, N_2O and surface ozone audits with relation to the measurement technique. In particular, results of CO and N_2O comparisons will be discussed. Audit results of these two parameters showed that the WMO/GAW compatibility goals are often not met, and further improvements both on analytical techniques and calibration standards are needed to solve this issue.

Further, we will show the advantages of the new performance audit approach using a travelling instrument. Results of parallel measurements at various stations using different analytical techniques are analysed, and aspects such as water vapour interference, calibration frequency, data coverage, and aggregation times will be addressed.

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The impact and importance of intercalibration and intercomparisons within the UK GAUGE greenhouse gas observational network

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Abstract

Motivated by the UK 2008 Climate Change Act, which requires the UK to decrease its greenhouse gas (GHG) emissions by 80% of 1990 levels by 2050, the Greenhouse gAs Uk and Global Emissions (GAUGE) project aims to better quantify UK GHG emissions. As part of this project a UK-focused GHG observational network has been established, drawing together new and existing GHG data streams from regional to global scales. These included high-density regional studies, tall-tower sites, moving platforms (ferry and aircraft) and satellite observations. Under the project these observations will be combined with modelling approaches to better quantify and characterise UK GHG emissions and place them within a global context.

This presentation will describe the network and the efforts made to ensure that common calibration scales were used, along with an assessment of the intercomparability of the stationary sites and moving platforms (6 near surface regional focused sites, 6 tall tower sites, ferry and aircraft measurements). This assessment was undertaken using both a cylinder intercomparison program (ICP) and a comparison between co-located flask and *in situ* measurements. The majority of the sites agreed within the WMO comparability guidelines, however, small biases were identified at some sites. These biases generally increased with concentration, with differences up to 0.3 ppm in CO_2, 3 ppb CH_4 and 0.3 ppt SF_6 observed between tall tower sites and mobile platforms, while larger biases were found at some of the regional study sites.

In order to assess the impact of biases of these types two experiments were conducted - one using pseudo observations (based on emissions estimates for key GHG sources) and another using actual observations. In both experiments the 'true' observations were adjusted by the biases identified within the ICP program. Regional UK emissions were then determined using two different inversion methods both based on the Met office NAME model. Using these emissions estimates we quantified the impact of systematic site biases on derived fluxes, assessing the relevance of the WMO comparability guidelines for our UK study and highlighting the importance of rigorous intercalibration and comparability of data streams for regional emissions estimation.

Using and improving the Australian Greenhouse Gas Observation Network

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Abstract

We present an overview of the Australian Greenhouse Gas Observation Network (AGGON, see Figure). AGGON is the CSIRO run network of continuous *in situ* measurements of CO_2 and CH_4 that provides data to the World Meteorological Organisation (WMO) Global Atmosphere Watch (GAW) program via the World Data Centre for Greenhouse Gases (WDCGG) as hourly means.

The calibration, quality control and data processing protocols used in AGGON will be described. We will present some representative data and consider issues such as the impact of moving from minutely to hourly data, the effect of the number of minutes contributing to the hourly mean and the usefulness of the minutely standard deviation information to better understand the dataset. From these examples, we hope to point to some 'smart' data selection tools for utilising the data that can help validate Australian terrestrial carbon cycle models.

Finally, we consider how we should grow AGGON to provide an improved dataset for carbon cycle model validation and to deliver low uncertainty top-down estimates of greenhouse gas emissions on a regional to continental scale.

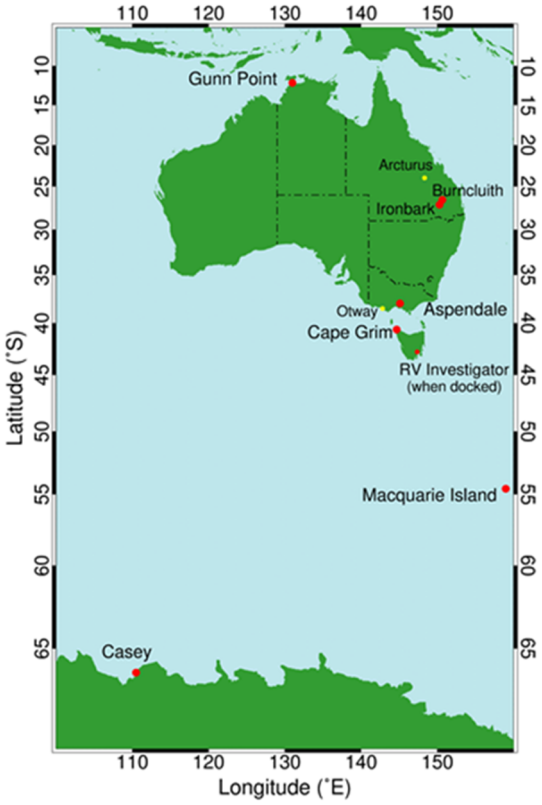


Figure: The Australian Greenhouse Gas Observation Network. Red dots indicate operational sites. Yellow dots indicate historical sites.

GEOS-Chem simulations of greenhouse gas measurements (CO_2, CH_4 and CO) from moving platforms in and around Australia

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Abstract

This study focuses on the analysis of *in situ* FTS measurements of the greenhouse gases carbon dioxide (CO_2) and methane (CH_4) along with carbon monoxide (CO) in the Australian region. The dataset includes *in situ* FTS measurements from ship cruises performed aboard the Australian research vessel RV Southern Surveyor operated by CSIRO/MNF (Commonwealth Scientific and Industrial Research Organisation/Marine National Facility) during several voyages in autumn, winter and spring in 2012 and 2013 in the South Pacific and Indian Ocean, and train-borne *in situ* FTS measurements taken on the Ghan railway that covers a north-south Australian continental transect, from Adelaide to Darwin and back for different time periods between 2008 and 2013. Since the measurement locations cover all latitudinal bands across Australia this dataset provides us the possibility to analyse the transport and latitudinal gradients of CO_2, CH_4 and CO in the Australian region. For the data interpretations a global 3D chemical transport model GEOS-Chem both v10-01 and v11-01, driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office is used. In this model tagged tracer simulations exist for CO_2, CH_4 and CO, which allows us to calculate the contributions of different regions and processes to the total amount of each gas and thereby quantify their relative contribution.

In this study we analyse and compare the model simulations to the previously mentioned measurements. Co-variations between the measured and modelled gas species will be used to understand the drivers of the differences between measured and modelled quantities. This will lead to improved estimates of the fluxes from processes in the Australian region.

Carbon isotope ratios suggest no additional methane from boreal wetlands during the rapid Greenland Interstadial 21.2

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Abstract

Samples from two Greenland ice cores (NEEM and NGRIP) have been measured for methane carbon isotope ratios (δ^{13}C-CH_4) to investigate the CH_4 mixing ratio anomaly during Greenland Interstadial (GI) 21.2 (85,000 years before present). This extraordinarily rapid event occurred within 150 years, comprising a CH_4 mixing ratio pulse of 150 ppb (~25%). Our new measurements disclose a concomitant shift in δ^{13}C-CH_4 of 1‰. Keeling plot analyses reveal the δ^{13}C of the additional CH_4 source constituting the CH_4 anomaly as −56.8 ± 2.8‰, which we confirm by means of a previously published box model. We propose tropical wetlands as the most probable additional CH_4 source during GI-21.2 and present independent evidence that suggests that tropical wetlands in South America and Asia have played a key role. We find no evidence that boreal CH_4 sources, such as permafrost degradation, contributed significantly to the atmospheric CH_4 increase, despite the pronounced warming in the Northern Hemisphere during GI-21.2.

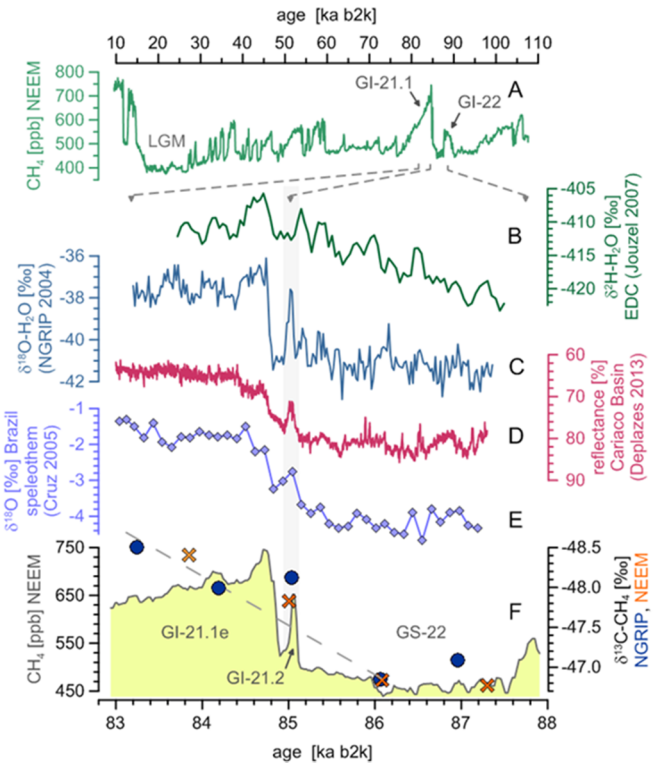


Figure: δ^{13}C-CH_4 in comparison to independent climate data. Panel A [CH_4] during last glacial [Chappellaz et al., 2013] on the top x axis. Panels B-F refer to the bottom x axis. The grey bar highlights the GI-21.2 event. (b) Antarctic and (c) Greenlandic water isotope ratios (Jouzel *et al.* [2007] and NGRIP [2004], respectively). (d) Reflectivity record of Cariaco Basin [Deplazes *et al.*, 2013] and (e) δ^{18}O from south Brazilian speleothems [Cruz et al., 2005]. (f) Left axis [CH_4] and right axis (inverted) δ^{13}C-CH_4 from NGRIP (circles) and NEEM (crosses). All data are shown on the GICC05_modelext timescale [Wolff *et al.*, 2010]. δ^{18}O Panel E is adjusted by +0.55ka to align GI-21.2, which is in line with Cruz *et al.* [2005].

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Study on variation of atmospheric CO_2 and source/sink characteristics based on stable carbon isotopes at SDZ in Beijing and LAN in Zhejiang

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Abstract

This study presents CO_2 concentrations of air flask samples obtained at regional background stations Shangdianzi (SDZ) and Lin'an (LAN) in China from January 2007 to December 2013, as well as its stable isotope ratios ($\delta^{13}C$) during 2009 and 2013 at SDZ and during 2011 and 2013 at LAN, respectively. Non-polluted, mixed-well and representative background air samples are selected to represent background characteristics of atmospheric CO_2 and $\delta^{13}C$ over Jing-Jin-Ji and Yangzte River Delta regions. The concentrations of atmospheric CO_2 observed at SDZ and LAN showed positive trend during the study period. Annual mean background CO_2 levels obtained at SDZ varied from 385.6 ppm in 2007 to 398.1 ppm in 2013, higher than global baseline levels, and the averaged growth rate is 2.0 ppm yr^{-1}. The average growth rate obtained at LAN is 2.7 ppm yr^{-1}, and the annual mean background CO_2 levels ranged from 388.3 ppm~405.8 ppm during 2007-2013, higher than SDZ, might owing to more energy consumption and carbon emissions observed at Yangtze River Delta regions than Jing-Jin-Ji areas. Atmospheric $\delta^{13}C$ (CO_2) levels obtained at SDZ and LAN present negative trends. The $\delta^{13}C$ (CO_2) values observed at SDZ decreased from -8.38‰ in 2009 to -8.52‰ in 2013, with a mean growth rate of -0.03‰ yr^{-1}. The annual mean values of $\delta^{13}C$ vary from -8.54‰ in 2011 to -8.75‰ in 2013 at LAN, with a growth rate of -0.080‰ yr^{-1}, lower than SDZ (-0.042‰ yr^{-1}) during the same periods. The absolute increase of CO_2 from 2007 to 2008 reached the lowest level during 2007 and 2013, and the lowest level of monthly CO_2 during July and September appeared in 2008, possibly due to relative less carbon emissions during 2008 Olympic Games period.

The analysed data of atmospheric CO_2, CO, $\delta^{13}C$ (CO_2) from flask samples obtained at WLG was compared with NOAA, and the probability distribution of data difference conformed to Gauss fitting curve. The probability is 82.2% for ΔCO_2 fall in the range of ±0.5 ppm, 78.7% for $\Delta\delta^{13}C$ fall in the range of ±0.1‰, and 93.4% for ΔCO fall in the range of ±5 ppb. Non-dispersive infrared (NDIR) and cavity ring-down spectroscopy (CRDS) CO_2 analyzers use $^{12}CO_2$ isotopologue absorption lines and are insensitive to all, or in part, the other CO_2-related isotopologues. This may produce biases in CO_2 mole fraction measurements of a sample if its carbon isotopic composition deviates from that of the standard gases being used. The CO_2 mole fractions error of natural atmospheric sample measured by LoFlo and PICARRO G1301 using industrial standards could reach 0.19 ppm and 0.2 ppm, respectively. According to the theoretical and experimental results, we concluded that the total CO_2 mole fractions of samples with depleted isotopic compositions can be corrected based on their $^{12}CO_2$ values calibrated by standard gases using LoFlo and PICARRO G1301, if we know the $\delta^{13}C$ and $\delta^{18}O$ values.

Cloud structure and phase at Cape Grim, Macquarie Island and over the Southern Ocean

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Abstract

Using depolarisation lidars located for seasonal campaigns at Cape Grim and Macquarie Island, we present the first (sub)-seasonal climatology of cloud structure and phases at these latitudes. The Bureau of Meteorology's UV Raman lidar with polarization capability was located at Cape Grim, Tasmania and collected data from July 203 – February 2014. The Australian Antarctic Division's depolarisation lidar was deployed to Macquarie Island in March 2016 and continues to collect data. We will demonstrate how a depolarisation lidar may determine cloud phase and discuss how cloud occurrence fraction varies with time and height. The presence of super-cooled liquid water clouds will be detailed and initial results on their occurrence and frequency of ice virga precipitating out of these clouds presented. We show how the surface-based lidar validates satellite-based observations of cloud and detail where and why differences occur. We will quantify the regular occurrence of very low level clouds and investigate the presence of multiple levels of super-cooled liquid water clouds at Macquarie Island. Lastly, we detail some cases of clouds observed by lidar on the CAPRICORN Southern Ocean cruise (autumn 2016).

Modelling and observations of organic carbon in the marine atmosphere

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Abstract

Marine aerosol is frequently characterized as being composed of sea-salt and DMS-derived sulphur. However, field measurements and laboratory studies in the last decade highlight a significant contribution to marine aerosol from organic carbon. Thus marine organisms may exert a significant influence on aerosol, cloud and radiative properties in the marine realm, potentially forming a feedback loop analogous to the CLAW hypothesis [Charlson et al., 1987]. Thus, emissions of both primary (emitted directly as particles) and secondary (organic carbon emitted in the gas-phase) marine organic carbon (MOC) need to be considered in aerosol-climate models. However, the emissions of MOC remain poorly constrained.

The complex evolution in the atmosphere of both primary and secondary aerosol sources requires a detailed microphysical aerosol model to accurately represent the relevant processes, and their role in climate. Here, the ACCESS-UKCA global composition-climate model [Woodhouse *et al*., 2015], which includes the aerosol microphysics module GLOMAP-mode, is used to simulate the emission and atmospheric processing of both primary and secondary MOC. Within ACCESS-UKCA, GLOMAP-mode is coupled to the cloud and radiation schemes, yielding the ability to assess the role of MOC in the climate system.

We use ACCESS-UKCA to quantify the contribution MOC makes to marine aerosol mass, number, cloud optical properties and short-wave radiation. Comparison with detailed observations derived from short-term campaigns (e.g. the SOAP cruise) and long-term stations (e.g. Cape Grim) allows us to constrain the uncertainty in the emission of both primary and secondary MOC. Including emissions of MOC may address some of the biases common to many climate models, in particular acting to reduce the Southern Ocean warm bias.

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Optimal estimation of sulfuryl fluoride emissions on regional and global scales using advanced 3D inverse modeling and AGAGE observations

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Abstract

Sulfuryl fluoride (SO_2F_2) is used increasingly as a fumigant to replace methyl bromide (CH_3Br), which was regulated under the Montreal Protocol (1986). Mühle e*t al*., [2009] showed that SO_2F_2 had been accumulating in the global atmosphere with a growth rate of 5±1% per year from 1978 to 2007. They also determined, using the 2D AGAGE box model, that SO_2F_2 has a total atmospheric lifetime of 36±11 years mainly driven by the oceanic uptake. In addition, the global warming potential (GWP) of SO_2F_2 has been estimated to be ≈4780 for a 100-year time horizon [Papadimitriou *et al*., 2008], which is similar to the CFC-11 (CCl_3F) GWP. Thus it is a potent greenhouse gas and its emissions are expected to continue to increase in the future. Here we report the first estimations of the SO_2F_2 emissions and its ocean sink from January 2006 to the end of 2015 on both the global scale using a 3D Eulerian chemical transport model (MOZART-4) solving a Main Chain Monte Carlo (MCMC) inversion, and on the regional scale using a 3D Lagrangian dispersion model (NAME) via the reversible-jump trans-dimensional MCMC approach [Lunt *et al*., 2016]. The mole fractions calculated on the global scale are used as boundary conditions for emission calculations over the NAME regions in North America, Europe, East Asia and Australia. For this 10-year inversion we use observations from the AGAGE (Advanced Global Atmospheric Gases Experiment) starting with six stations in 2006, which are La Jolla (California), Mace Head (Ireland), Cape Grim (Australia), Ragged Point (Barbados), Trinidad Head (California) and Cape Matatula (Samoa). We then add observations from Gosan (South Korea) in 2007, Jungfraujoch (Switzerland) in 2008, Shandiangzi (China) and Ny-Alesund (Norway) in 2010, and Monte Cimone (Italy) in 2011, reducing the uncertainty associated with the regions located close to these stations. Results are compared to (i) the total global SO_2F_2 emissions estimated by the 2D AGAGE box model [Mühle *et al*., in prep] and (ii) the global industrial estimates. This work provides a unique quantitative understanding of the SO_2F_2 industrial emissions and its oceanic and photochemical sinks over the last decade.

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Emissions of particulates from tropical peat burning in Malaysia

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Abstract

Tropical peat fires in Southeast Asia are a major contributor to greenhouse gases, aerosols, reactive gases and are responsible for poor regional air quality effecting hundreds of millions of people in the greater Asia region. Despite the severity of these impacts, very little is known about the contents of peatland fire emissions. Until recently, published research consisted of emission factors derived from a laboratory based study in which a single peat sample was burnt. In a more recent study by [Stockwell *et al.*, 2016], particulate matter emissions were measured by gravimetric means, not real time results, and these values are currently being processed by R. J. Yokelson and are not published yet.

During July and August 2016 a field study in in Peninsula Malaysia was undertaken in which MODIS, VIRS and in-situ smoke sightings were used to locate fire events throughout peatland in the Malaysian province of Selangor on the east coast.

In this presentation we will show emission factors derived from this study and discuss interesting data that shows how these emissions change as the fire ages. The emission factors developed through this study, when coupled with peat fuel consumption estimates, will improve global emission estimates from tropical peat fires.

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Long term trends in tropospheric composition from Australian ground-based remote sensing measurements

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Abstract

In 2015 the ground-based solar remote sensing Fourier transform measurement program celebrated its 20th year of continuous operation from Wollongong. The site has been operated throughout its lifetime as part of the world-wide Network for the Detection of Atmospheric Composition Change (NDACC, since 1995) and the Total Carbon Column Observing Network (TCCON, since 2008). The spectral record contains a wealth of information about the trace gases that absorb in the infrared region and the changing composition of the atmosphere. The spectra have been analysed in a methodical and consistent manner, using NDACC and TCCON recommended analysis procedures. In this presentation we discuss some of the tropospheric gases of interest to atmospheric chemistry and climate and their trends as recorded from this Southern Hemisphere site.

Long term trends in stratospheric composition from Australian ground-based remote sensing measurements

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Abstract

Remote sensing measurements in the mid infrared from a high resolution Fourier Transform Spectrometer (FTS) have been routinely collected from the campus of the University of Wollongong (UoW, 34.41°S, 150.88°E, alt 30 m) since 1995. This measurement programme at UoW is part of the world-wide Network for the Detection of Composition Change (NDACC), a global network of stations using a range of remote sensing and *in situ* measurement techniques. The mid-Infrared spectra recorded with the FTS, by their nature, capture the absorption information in the column from the ground to the top of the atmosphere. The spectra contain absorption features from over 40 gases, most of which have some importance to chemistry and transport in the stratosphere (O_3, HCl, HNO_3, HF), and troposphere (CO, HCN, NH_3, CH_4, CFCs, OCS …).

The physics of the spectral absorption is such that there is some vertical information inherent in the spectra, depending on where the particular species resides in the atmosphere and its concentration. The UoW dataset, like many within the NDACC, therefore contains a very valuable long term record of a range of processes throughout the atmosphere. This dataset has been recently analysed in a methodical and consistent manner, using NDACC recommended analysis procedures. In this presentation we focus on the stratospheric gases listed above and discuss their trends throughout the 20 year record. A parallel presentation [Clare Paton Walsh *et al*.] will focus on tropospheric gases from the NDACC programme.

Total Carbon Column Observing Network activities in Australia: moving to portable technologies to complement primary sites

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Abstract

The Total Carbon Column Observing Network (TCCON) was established in 2004 with the primary aim of producing high precision and accuracy measurements of column-averaged mole fractions of greenhouse gases. TCCON fulfils three scientific roles: satellite validation, for instruments such as the Greenhouse Gases Observing Satellite (GOSAT) and the Orbiting Carbon Observatory-2 (OCO-2); independent validation of model simulations driven by satellite or *in situ* measurements; and providing insight into the global carbon cycle. The core of these sites is a high resolution Bruker IFS125HR Fourier Transform Spectrometer, an instrument that is capable of high spectral resolution and excellent precision and consistency between sites; however, these instruments are both expensive and difficult to transport. In Australia there are two TCCON sites: Darwin was one of the first operational sites and has been running since 2005, while an additional TCCON site has been running at Wollongong since 2008. These are two of approximately 23 sites in a growing network around the world. TCCON is a GAW affiliated network.

TCCON sites have been extensively used to validate CO_2 and CH_4 measurements from GOSAT and OCO-2; however, no TCCON sites exist in regions with high surface albedo, such as deserts. In such regions, the reflected light is such that GOSAT measures in a medium-gain mode instead of the high-gain used elsewhere, and a bias has been observed between the retrievals using these different gains. At high southern latitudes, OCO-2 exhibits a bias in retrieved CO_2 over the oceans in the southern winter. The lack of TCCON validation sites in these locations means that these biases are not yet understood and Australia is well positioned to provide information on both problems: for example, the Australian desert interior means that a large fraction of GOSAT measurements over the Australian continent employ the medium gain mode.

Recently, several more portable alternative instruments to the standard TCCON configuration have become available. These instruments are suitable for deployment on a campaign basis and more suited to short-term scoping studies than a full TCCON station. The most widely used to date, the Bruker EM27/SUN, shows good short-to-medium term stability relative to TCCON, but is not yet tested on long-term time scales, nor well set up for remote, automated operation. At the Wollongong TCCON station, an EM27/SUN has been operational alongside the standard instrument over a period over several years, along with an in house developed instrument based on a Bruker IRCube.

In this presentation, we give an update on the TCCON with an Australian focus. Comparisons between TCCON and alternative portable instruments will be presented. We describe the development of an automated Bruker EM27/SUN suitable for remote deployment. Results from a campaign in September-October 2016 at Alice Springs with the EM27/SUN and a comparison to GOSAT CO2 column measurements will also be presented.

Southeast Asian & Australian TCCON operations-readying for Ibuki 2-validation (SATORI): The TCCON-Philippines Story

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Abstract

To meet the need for better quantification of greenhouse gases and monitoring of the carbon cycle, Japan launched the Greenhouse gases Observing SATellite (GOSAT or *Ibuki* in Japanese) in 2009. Its successor, GOSAT-2 (Ibuki 2), will continue this important work and is scheduled to be launched into orbit by the Japanese fiscal year of 2017 (FY2017, http://www.gosat-2.nies.go.jp/). GOSAT-2 will require validation of its measurements especially in tropical Southeast Asia.

Tropical Southeast Asia is a region with high scientific importance for the understanding of the transport of trace gases into the stratosphere, but due to several limitations, it is also a region with very few ground-based measurements. Measurements of greenhouse gases from satellites remain invalidated to a large extent in this region. This makes the understanding of key atmospheric processes difficult. Understanding these processes is also key to better weather predictions and disaster response, which are crucial in a region constantly battered by typhoons, flooding and other natural hazards that can be worsened by a changing climate.

This presentation discusses the behind the scenes work and challenges of establishing a new measurement station and traces the history that brought about a project collaboration among the University of Wollongong (Australia), the National Institute for Environmental Studies (NIES, Japan) and the Energy Development Corporation (EDC, Philippines).

Preliminary comparison of biomass burning inventories processed by GEOS-Chem with surface, total column and maybe satellite measurements in Australia

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Abstract

Australia contributes approximately 6% of global biomass burning CO_2 emissions, mostly from savanna type fires [Shi *et al*., 2015]. This estimate comes from biomass burning inventories that use emission factors derived from field campaigns performed outside Australia. The relevance of these emission factors to the Australian environment has not previously been evaluated and therefore needs to be tested.
Here we compare predictions from the chemical transport model GEOS-Chem and the global chemistry-climate model ACCESS-UKCA run using different biomass burning inventories to total column measurements of CO, C_2H_6 and HCHO, in order to identify the most representative inventory for Australian fire emissions.

The measurements come from the Network for Detection of Atmospheric Composition Change (NDACC) and Total Carbon Column Observing Network (TCCON) solar remote sensing Fourier transform spectrometers and satellite measurements from AIRS over Australia.

We evaluate three inventories: the Global Fire Emission Database version 4s – GFED4s [Giglio *et al*. 2013], the Fire Inventory from NCAR version 1.5 – FINN1.5 [Wiedinmyer *et al*., 2011], the Quick Fire Emission Database version 2 – QFED2 from NASA and the MACCity emission inventory (from the MACC/CityZEN EU projects).

From this evaluation we aim to give recommendations for the most appropriate inventory to use for different Australian environments. We also plan to examine any significant concentration variations arising from the differences between the two model setups.

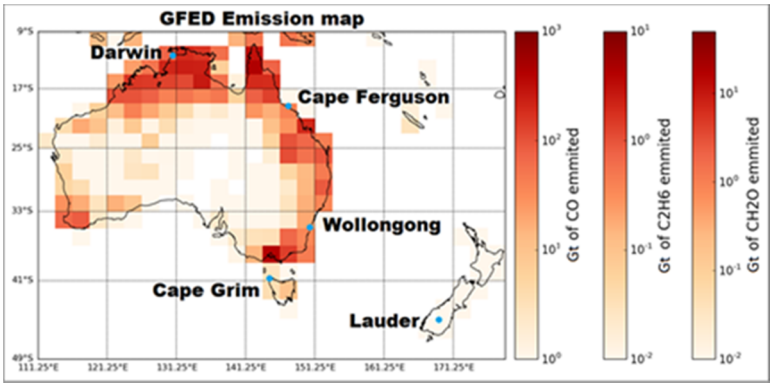


Figure: 2009 GFED4s biomass burning emission map over Australia (with GEOS-Chem resolution).

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Using box models to quantify zonal distributions and surface emissions of halocarbons in the background atmosphere

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Abstract

NOAA's Global Monitoring Division (GMD) began measurements of the major chlorofluorocarbons and nitrous oxide (three gases) in 1977 from flask samples collected at five remote sites around the world. Our program has expanded to over 40 gases at 15 sites, which includes six in situ instruments and 15 flask sites. The Montreal Protocol for Substances that Deplete the Ozone Layer and its subsequent amendments has helped to decrease the concentrations of many of the ozone depleting compounds in the atmosphere. Our goal is to provide zonal emission estimates for these trace gases from multi-box models and their estimated atmospheric lifetimes in this presentation and make the emission values available on our web site. We used our measurements to compare emissions estimated from the Harvard 5-box Model to the AGAGE 12-box Model. The emissions do not agree for some gases so our next step is to use our airborne measurements to calibrate the exchange times between the boxes for 5-box and 12-box models using sulfur hexafluoride, because its emissions are better understood.

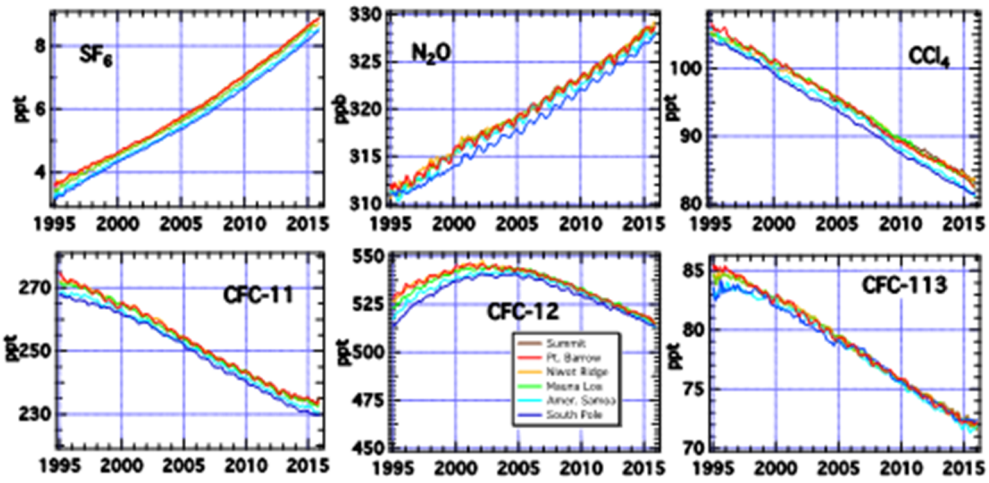


Figure: Monthly means of important trace gases from the GMD's halocarbon monitoring network (combined data from flasks and *in situ* GCs) versus time used to calculate emissions from two box models.

Does the Great Barrier Reef have an impact on local climate?

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Abstract

The debate as to whether some form of climatic bioregulation exists via the sulfur cycle has gone on since the late 1970s. The CLAW hypothesis suggests that algae, when stressed by above average sea surface temperatures, produces oceanic dimethyl sulfide (DMS), which is released into the atmosphere. DMS is then oxidised and may have some impact on aerosol formation and hence the formation of cloud condensation nuclei. This in turn can affect solar radiation at the surface, reducing the stress felt by the algae. Many studies have tried to either prove or disprove the hypothesis, using a range of methods, models, observations and temporal and spatial locations. The Great Barrier Reef provides a unique opportunity to study DMS/aerosol production, as recent studies have found the symbiotic organisms within the reef produce large amounts of DMS, that are also inherently stationary (unlike other large algal blooms). In this work, a field campaign over the Great Barrier Reef aims to further understand the role DMS plays in the local and broader climate, in particular from the perspective of aerosol formation.

Preliminary land and ship-based observations, taken simultaneously, are presented here. Measurements taken include vertical profiles of wind, temperature, humidity, pressure and particle size distribution via a ship based tethered helikite balloon, and a land based Mini Micro-pulse LiDAR, providing vertical profiles of aerosol light scattering and cloud phase and structure. These measurements have been taken as part of two large, collaborative field campaigns.

Ultimately these measurements will help validate a climate-chemistry model, through which we can begin to understand the effect of DMS on climate, in particular sea surface temperatures and rainfall. Initial work has been done using the United Kingdom Chemistry and Aerosol (UKCA) global climate model, to determine the sensitivity of the broad-scale climate to changes in DMS. Through this work, we can then ask the questions, how does the Great Barrier Reef impact local climate, and what will happen if it's gone?

Four years of gaseous elemental mercury measurements at Cape Grim station

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Abstract

The Minamata Convention on Mercury, signed by Australia in 2013, is aimed at developing global policy designed to reduce the environmental and human health impacts of this potent neurotoxin [Kessler, 2013]. As the atmosphere represents the dominant pathway of environmental mercury [Driscoll *et al.*, 2013], development and evaluation of such policy is reliant on long-term monitoring capabilities undertaken at effective spatial resolutions [Cinnirella *et al.*, 2014]. Recently, data collected at Cape Grim were used in conjunction with those from other international sites to show that atmospheric mercury concentrations in the Southern Hemisphere are up to 30% lower than previously believed – between 0.85 and 1.05 ng m^{-3} [Slemr *et al.*, 2015]. Here we present gaseous elemental mercury (GEM) from the beginning of the Cape Grim programme – one of the longest datasets of its kind in the Southern Hemisphere. Using this dataset we explore daily, seasonal and interannual trends, along with case studies highlighting aspects of the biogeochemical cycling of mercury such as atmospheric depletion events and biomass burning. Furthermore, air parcel back-trajectories obtained using HYSPLIT are used to investigate spatial characteristics of the sampling fetch in order to better understand ocean-atmosphere mercury exchange, which is still a large source of uncertainty in global mercury modelling [Selin, 2009].

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Measurements of aerosol-cloud properties in the Southern Ocean

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Abstract

Better characterisation of aerosol and cloud processes in the pristine environment of the Southern Ocean region is key to drastically reducing the uncertainty in our understanding of radiative forcing, and thus improve our ability to simulate the global climate. Large discrepancies currently exist between satellite observations and both state-of-the-art models and reanalysis products. These discrepancies present themselves in both cloud properties, where products underestimate cloud fraction by up to 20%, and radiative forcing, where observations of reflected shortwave radiation are 40-60 $W.m^{-2}$ larger that simulations. Aerosols can effect numerous cloud properties, including cloud lifetime, precipitation, cloud structure, and albedo, to name a few. These interactions are often most pronounced in pristine environments where aerosol concentrations are low, such as the Southern Ocean. Unfortunately, because of its remoteness, measurements in this region are scarce. The recent commissioning of Australia's blue-water research vessel, the RV Investigator, has provided a number of new opportunities due to its frequent passage in the region, combined with its strong atmospheric measurement capabilities. Two intensive measurement campaigns have already been undertaken where simultaneous co-located measurements of both aerosol and cloud properties were made. Early results show distinct periods when the cloud condensation nuclei (CCN) fraction increases just prior to the appearance of low-level clouds (less than 1 km). Interesting changes in cloud properties are also observed when perturbations occur in the relatively constant background CCN concentration. This presentation will outline initial results from these campaigns, with a focus on these relationships between cloud and aerosol properties.

Using atmospheric Radiocarbon to Estimate Fossil Fuel Sources: A Test Case with a Known Source

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Abstract

Atmospheric radiocarbon is a promising tracer for estimating the contribution of fossil fuel sources to the changes in atmospheric CO_2. Using atmospheric transport models and inverse techniques we can hope to estimate fossil fuel sources from radiocarbon measurements. In this study, we test this method using a known source. We use the Weather Research and Forecasting model (WRF-Chem) to investigate the $FFCO_2$ emission of a point source on 24th and 25th August 2013. The data consists of 20 samples of atmospheric CO_2 dissolved in weak NaOH solution. We use four-nested model domains with horizontal resolution of 9, 3, 1 and 0.33km, centered on Kogan Creek Power Station, which is a coal-fired power station located in the western Darling Downs in Queensland. We use the observed meteorology data from the Bureau of Meteorology to verify our model. The C-14 integrating air samples are used to examine the skill at modeling the small scale $FFCO_2$ and to test whether we can estimate the emissions.

The comparisons between simulated and observed values of wind speed and wind direction show that our model is reasonably reliable, with correlation coefficients of 0.87 and 0.6 respectively. We extract the data of modeled FFCO2 from a 3x3x2 Square of grid cells centered on the observation then calculate the mean and maximum value. The comparisons of 24 August and 25 August are investigated separately.

The model simulates well the overall structure of the results in both d03 and d04 on 25 August (correlation of 0.73 and 0.94) but not on 24 August (correlation of 0.1 and 0.07). The simulated values in both two days underestimates the magnitude of spatial variations. We also use the FLEXPART Lagrangian model to investigate other possible influences on the observations and see differences in the connection between the power station and observations on the two days. The result suggests either an underestimate of the emissions or that the model is diluting the influence of the power station.

Update from the Cape Grim VOC monitoring program

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Abstract

An *in situ* volatile organic compound (VOC) monitoring program was commissioned at Cape Grim in November 2014. The configuration of the Cape Grim VOC instrument is based on an online instrument which has been running successfully at the Cape Verde observatory for several years. VOC measurements are made every hour via a dual channel online gas chromatograph (GC) with flame ionization detectors (FID). Two columns and two FIDs allows quantification of both light hydrocarbons (C2-C8) and a range of oxygenated VOCs such as aldehydes, ketones and alcohols. The system has run almost continuously for 2 years.

We will provide an update about developments from the past year including linearity and breakthrough tests, installation of heated inlet for ozone removal, and calibration on the WMO GAW VOC scale. We present two years of data for some simple hydrocarbons including ethane, propane, butane isomers and benzene. We provide a comparison between concentration data obtained on this system with other Cape Grim hydrocarbon measurements, including insitu data from the AGAGE Medusa, as well as analysis of canister samples collected for the NOAA Halocarbon and other Atmospheric Trace Species (HATS) group and the Carbon Cycle Network. Surface hydrocarbon measurements from Cape Grim will also be compared to aircraft measurements over the Southern Ocean measured using NCAR's Trace Organic Gas Analyser (TOGA) during the Atmospheric Tomography (aTOM) mission.

We will discuss future plans for the system, including changes to the pre-concentrating trap to increase sample volume, and plans to quantify oxygenated VOCs.

Impact of background gas composition on CO_2 measurement by cavity ring-down spectroscopy

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Abstract

The effect of background composition on CO_2 measurement has been investigated using wavelength-scanned cavity ring-down spectrometry (WS-CRDS), taking a spectral line centered at the R(1) of the $(3\ 0^0\ 1)_{III} \leftarrow (0\ 0\ 0)$ band. For this purpose, eight cylinders with various compositions were gravimetrically and manometrically prepared with $1\sigma = 0.1\%$, and they were introduced to the WS-CRDS analyzer calibrated against standards consisting of ambient air composition. Against the assigned values, excess N_2 exhibited a negative CRDS response, whereas excess Ar showed a positive deviation. The total pressure broadening coefficient (TBPC) based on the composition of N_2, O_2, and Ar corrected the deviations to up to 0.15%. Furthermore, this correction linearly drifted the CRDS responses for a wide extent of TPBC ranging 0.065 to 0.081 $cm^{-1}\ atm^{-1}$. To obtain accurate measurements using intensity-based optical measurement techniques such as WS-CRDS, it is imperative to calibrate the instrument based on TBPC.

Gravimetric standard gas mixture of SF_6 at ambient level

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Abstract

Standard gas mixtures of SF_6 in synthetic air were gravimetrically developed as a suite consisting of 6 mixtures with fractions of SF_6 ranging from 5 to 15 pmol/mol. For precision in weighing the gas fills, an automatic weighing system coupled with a high sensitivity balance was used. Impurity profiles of the raw gases were determined by various analyzers. In particular, trace amounts of SF_6 in the matrix (down to amol/mol levels) were analysed. Gravimetric dilution of pure SF_6 by the purity-assessed N_2 was performed in 6 steps. In the 7^{th}, O_2 and Ar were added in order to mimic atmospheric composition. Gravimetric fractions of SF_6 in each step were estimated theoretically and validated experimentally. Ultimately, the SF_6 fraction uncertainty of the standard gas mixtures combined with the uncertainties of the gravimetric preparation and the verification measurements were found to be approximately 0.15 % at *k=2*.

Minimal infrastructure air quality sensors as both dense networks and an isolated deployment: Case studies from DISCOVER-AQ and in Danum Valley Borneo

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Abstract

Emerging minituarised environmental sensing technologies have the potential to allow detailed investigation of air quality species in the urban environment at previously unachievable fine spatio temporal scales. These new technologies also have the potential to provide data in logistically difficult or resource limited areas. Currently there are knowledge gaps surrounding how to most appropriately use these emerging systems and how to integrate data from dense small sensor networks with existing relatively sparse reference network (and reference site) data. Fundamentally there are also issues to be addressed on data validation as well as identifying levels of information content at various measurement scales (spatially and temporally) when using heterogeneous, mixed capability sensing networks.

Shown here are data from a number of high density sensor networks deployed during selected NASA DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) campaigns. Preliminary data from a small scale sensor deployment in Malaysian Borneo is also presented. Both these studies are part of ongoing research into appropriate scales of measurement in a range of environments as well as instrument and network validation studies.

For DISCOVER-AQ a number of high density, relatively low-cost, sensor networks were deployed within sub domains of the much larger campaign area. These were primarily aimed at investigating horizontal variability within these sub domains and how to best link these types of network outputs to other available data. There were two main deployments; 10 sensors for 3 months in Houston and 15 sensors for 5 weeks in Denver/Boulder (CO, NO, NO_2, O_3, temp and %RH at I min resolution). Vertical distribution was also investigated with sensors on the 300m Boulder Atmospheric Observatory tower.

Two sensors have been deployed to study use of relatively low cost sensors in logistically difficult environments by University Malaya. These have been in place on at the Danum Valley research centre in Malaysian Borneo (relatively undisturbed dipterocarp forest) for >6 months (CO, NO, NO_2, O_3, temp and %RH at I min resolution).

This work shows selected data and preliminary outcomes for these two programs and discusses future directions.

Preliminary surface ozone data from RV Investigator

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Abstract

Ozone is a key species in the troposphere, acting as the main precursor for the OH radical which determines the oxidation capacity of the troposphere [Seinfeld and Pandis, 2006], is the third most important greenhouse gas after carbon dioxide and methane, and contributes to the absorption of UV radiation. Measurement of surface ozone is necessary for, amongst other reasons, determining the changing composition of the atmosphere. However, coverage of surface ozone measurements in the Southern Hemisphere over the oceans has been sparse. Two UV photometric ozone analysers were installed on the newly commissioned RV Investigator in January 2015 as permanent fixtures. The data presented here is the surface ozone data collected from a number of voyages during 2015 and 2016. While the data is very preliminary in nature, in provides a first look at surface ozone data sampled from a range of Southern Hemisphere ocean environments.

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The potential of ^{14}CO in glacial ice as a tracer for past cosmic ray flux and atmospheric hydroxyl radical abundance

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Abstract

The amount of ^{14}C-containing carbon monoxide (^{14}CO) in glacial ice is determined by trapping of atmospheric ^{14}CO into air bubbles in the ice and in situ cosmogenic production of ^{14}CO in firn and relatively shallow ice. Earlier studies of ^{14}CO in ice cores showed large disagreements in the rates of *in situ* cosmogenic production as well as whether ^{14}CO produced in the firn layer is well retained or largely escapes to the atmosphere via the interconnected pore space. We have reviewed previously published work that included ^{14}CO measurements in ice and firn air, and have compared this with our more recent high-precision measurements on very large ice and firn samples. The available evidence suggests that very little *in situ* cosmogenic ^{14}CO is retained in the diffusive part of the firn (the upper ≈ 40 – 100m). *In situ* cosmogenic ^{14}CO production rates below the firn diffusive zone are non-negligible, with production due to deeper-penetrating muons. At sites with low snow accumulation rates, the in situ cosmogenic ^{14}CO component is expected to be larger than the trapped atmospheric component. This potentially allows to use ice core ^{14}CO measurements from such sites to improve our understanding of past cosmic ray flux variations. In contrast, at sites with very high accumulation rates, trapped atmospheric ^{14}CO is expected to be dominant over the *in situ* cosmogenic component. This potentially allows ^{14}CO records from such sites to be used for reconstructions of past atmospheric hydroxyl radical (OH) variations.

DE08 on Law Dome in Antarctica is a well-characterised site that is a perfect candidate for a paleo-atmospheric ^{14}CO reconstruction. DE08 has an unusually high accumulation rate of 120 cm ice equivalent per year, and such rapid burial of the firn and ice layers minimises the amount of accumulated *in situ* cosmogenic ^{14}CO. Measurements of [CO] in air extracted from DE08 firn and ice indicate that CO is well preserved. The high accumulation rate at DE08 also encloses air with recent ages, as well as with relatively narrow air-age distributions. This means that air contained in the deep firn and in the upper closed off ice has ages that are in the range of the atmospheric ^{14}CO record from New Zealand and Antarctica. Overlap between air ages at DE08 and the atmospheric ^{14}CO record allows for a direct determination of the *in situ* cosmogenic ^{14}CO component in this depth range, by subtracting the known atmospheric component from measured values. This will provide estimates of ^{14}CO *in situ* production rates and allow for an accurate correction for the in situ cosmogenic ^{14}CO to be applied at other depths, yielding the atmospheric ^{14}CO history.

Extended Path Open-Path Spectroscopy: Measuring Urban Air Pollution Gas Species at Extended Spatial Scales

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Abstract

Open path Fourier transform infrared (OP-FTIR) spectroscopy in the mid infrared has been used extensively to characterise gaseous emissions from local sources. In particular the technique has been employed to quantify emissions from agricultural sources including CH_4 from grazing ruminants, N_2O and NH_3 from soils and pastures and CO_2, CH_4, N_2O and NH_3 from intensive animal management systems such as cattle feedlots, chicken and pig production facilities and manure management systems.

The open path system detects gases in the open atmosphere between the spectrometer and a distant reflector. Typically the system is located downwind from the source with the reflector located between 50 and 150 m from the spectrometer.

As part of the Western Air-Shed and Particulate Study for Western Sydney (WASPSS) within the Clean Air and Urban Landscapes (CAUL) Hub, a system with extended pathlengths has been deployed to an urban environment to characterise targeted trace gases relevant to urban air quality, including CO, CO_2, CH_4 and NH_3. The measurement path has been extended to around 500 m to allow for integration of trace gas concentrations at scales comparable to the spatial resolution achieved by air quality models.

WASPSS has objectives to better characterise the sources and sinks of air pollutants in the Western Sydney basin. As part of this WASPSS aims to build an ensemble of air quality models to better estimate potential exposure of individuals to air pollutants, and to assess the impact of policy decisions on air quality. Validation of air quality models requires measurements on similar scales to the model spatial and temporal resolution.

The deployment of the extended path OP-FTIR system, alongside an extended path UV-Visible DOAS system in Western Sydney encompassing major rail freight line and road networks plus the Auburn CBD, will provide measurements of a suite of trace gas species important to air quality at scales comparable to air quality models. This paper reports on initial measurements of target gases from the OP-FTIR system and the implications of deploying this technology to characterise targeted gaseous species relevant to urban air quality.

Trends in chlorobenzenes in air at marine background, urban and biomass burning-impacted sites in Australia

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Abstract

Over the past few decades, there has been growing concern about adverse effects of chlorobenzenes on the environment, including evidence of their deposition in remote Arctic and Antarctic regions and uptake by the food web. This concern culminated in listing hexachlorobenzene (HCB) and pentachlorobenzene (PeCB) as persistent organic pollutants (POPs) under the Stockholm Convention in 2004 and 2009 respectively. Signatories to the convention agreed to eliminate use and production and reduce unintentional production. Potential primary emission sources include residues in chlorinated pesticides, by-products of chemical processes and combustion of fossil and biomass fuels. Secondary emissions from re-volatilisation of chlorobenzenes from soil are also expected to be a major source.

The Australian region is heavily influenced by biomass burning with an average area of 502,000 km^2 of 7,692,000 km^2 burnt per year [Giglio *et al*., 2013]. While the area burnt each year has significant variability (CV=0.46), there are strong seasonal and spatial trends to fires, with the majority (86%) of carbon emissions associated with savanna fires in northern Australia in the dry season (April to October). Seasonal activities involving biomass burning in southern Australia include wildfire and fuel reduction burns in the period of October to April and use of wood-heaters during winter months from May to August.

Previous measurements of HCB in air in Australia include annual average levels of HCB and other POPs at 15 sites for 2012. Wang *et al*. [2015] found HCB to be the most abundant organochlorine POP, with a median of 41 pg m^{-3}. The authors found that levels were higher than previously reported for background southern hemisphere levels and suggested the higher levels may be due to biomass burning emissions. No PeCB levels in air have been reported for Australia to date.

In this study, monthly measurements of ambient HCB and PeCB were measured at three Australian sites from 2011 to 2013. The Darwin site is impacted by dry-season savanna biomass burning, the Melbourne site is impacted by urban emissions, summer fires and winter wood-heater use and the Cape Grim site is representative of background marine air. The aim was to determine whether there were:

1. long-term trends at these sites due to decreased primary emissions; and
2. seasonal trends from combustion emissions or temperature-driven secondary emissions

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The impact of the 2016 Tasmanian fires on air quality

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Abstract

In 2016 large bushfires started in January across Tasmania and continued burning into February. Two distinct smoke plumes were observed at the Cape Grim Baseline Air Pollution Station (CGBAPS), when measured pollutant concentrations were significantly elevated above background concentrations. Satellite images confirm the significant smoke impacts on 25 January and 12 February.

The poster presents preliminary data of carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), black carbon (BC) and ozone (O_3) concentrations measured between 22 January and 16 February 2016.

Enhancement ratios (ER) were calculated using measured concentrations of pollutants at the CGBAPS during the smoke plume events. The ERs were calculated by taking the ratio of the enhancement of the species of interest above background to the enhancement of a stable representative plume tracer, in this case CO. Using the calculated ERs we could derive EFs. A comparison to literature data suggests that the smoke plume was a mixture of emissions from organic peatlands and forests.

Sensitivity of terrestrial biosphere CO_2 to temperature derived from pre-industrial CO_2, $\delta^{13}CO_2$ and COS

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Abstract

Models of future carbon cycle-climate changes predict a large range in atmospheric CO_2, mainly because of uncertainties in the response of the land carbon cycle to the future temperature increase. The Little Ice Age (LIA, 1500-1750 AD) CO_2 decrease [MacFarling Meure *et al*., 2006; Etheridge *et al*., 1996] is the most significant pre-industrial atmospheric change over the last millennia and has been used to derive the climate sensitivity of the global carbon cycle. While a recent study [Bauska *et al*., 2016] confirms previous evidence [Trudinger *et al*., 1999] that pre-industrial CO_2 variations were caused by changes in land carbon stores, there are open questions about the size of the atmospheric LIA CO_2 decrease reconstructed from ice cores, and about what caused the land to sequester CO_2. To quantify the size of the LIA CO_2 decrease, we have produced new CO_2 measurements from DML ice that support the DSS LIA CO_2decrease as a real atmospheric feature. To partition the contribution of ocean and land, we have measured the $\delta^{13}C$-CO_2, showing that the cause of the CO_2 drop was uptake by the terrestrial biosphere. To identify whether the land uptake was caused by temperature, or by a decline in farming due to pandemics, we have simulated the effect of a temperature perturbation on atmospheric carbonyl sulfide (COS). In agreement with the previously published positive COS anomaly, our results indicate that Global Primary Productivity (GPP) decreased during the LIA, ruling out the early anthropogenic land use change hypothesis as the dominant cause of increased terrestrial carbon storage. Using this information in a recent publication [Rubino *et al*., 2016] we derived the temperature sensitivity of pre-industrial CO_2 fluxes for the terrestrial biosphere (-10/-60 Pg of C K^{-1}), implying a positive climate feedback and providing a benchmark to reduce model uncertainties.

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KIT World Calibration Centres within the Global Atmosphere Watch (GAW) programme of the World Meteorological Organization (WMO)

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Abstract

In 1989 WMO established the Global Atmosphere Watch Network (GAW) to study the atmosphere and its chemistry. The GAW mission focuses on activities (1) to reduce environmental risks to society and meet the requirements of environmental conventions, (2) strengthen capabilities to predict climate, weather and air quality, and (3) contribute to scientific assessments in support of environmental policy. To accomplish the mission of GAW, high-quality measurements of relevant parameters are essential. Now GAW with its global, regional and contributing stations consists of more than 1000 sites in more than 100 countries. Components measured can only be conclusively analyzed from a global perspective if all measurements are of high quality and comparable over a long period. To achieve this, a Quality Assurance / Quality Control (QA / QC) system was set up. World Calibration Centres (WCCs) are one of the central facilities of the WMO-GAW programme ensuring best data quality in the observing network. The WCC-N_2O and WCC-VOC activities are supervised and funded by the Quality Assurance / Scientific Activity Centre (QA/SAC), Germany, operated through the German Umweltbundesamt.

WCC-N_2O

The WCC-N_2O keeps 8 laboratory standards calibrated by the Central Calibration Laboratory (CCL; last recalibration in October 2013), and 17 travelling standards (5 calibrated by the CCL in 2011) suited for performance audits and inter-comparisons. Audits were conducted on the basis of previously developed guidelines that had been approved by the Scientific Advisory Group for Greenhouse Gases (SAG-GG).

Audits comprised a general inspection of the individual GAW station and a data quality check, in particular based on a comparison experiment involving five travelling standards of the WCC-N_2O, containing different N_2O amount fractions. Important details in the context of audits were related to the quality of the analytical procedure, notably the characteristics of the detector performance, and compatibility of the N_2O measurements within the GAW network. In most cases, there are no obvious parameters of the instrumentation for major improvements in the near future. However, calibration of the detector over the entire amount fraction range as recommended by GAW (290 – 350 nmol/mol) is strongly encouraged. At present, achieving the N_2O DQOs (0.1 nmol/mol) remain a challenge, even with an optical detection system.

WCC-VOC

The QA/QC measures of the WCC-VOC are currently based on 10 gas standard with a mol fraction range close to ambient conditions (pmol/mol to nmol/mol). In 2011/12, a new set of ambient air standards and a new laboratory standard referenced to the VOC scale was established. Currently, the GAW network data compatibility for target Non-Methane-Hydrocarbons, a subset of VOCs targets of the GAW program, amounts to better than 80% with no bias within the current DQOs, except for ethyne and n-pentane.

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Aerosol measurements at Gunn Point: an update

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Abstract

The Australian Tropical Atmospheric Research Station (ATARS) is a CSIRO facility on the Bureau of Meteorology's Gunn Point Research Weather Radar site at Gunn Pt, NT. The ATARS has been in operation since 2010, during which time there have been measurements of meteorology, greenhouse gases, reactive gases, radon, and aerosols.

We will discuss recent measurements of aerosol scattering, absorption, and number concentration and provide a short update on the status of the supporting infrastructure at the ATARS.

Using skydiving and AirCore for convenient atmospheric profile sampling

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Abstract

The AirCore technique [Karion *et al*., 2010] has been used in 3 continents to collect atmospheric samples which retain a measurable concentration profile. These samples have typically been collected from high-altitude balloon platforms. In July 2016, University of Wollongong researchers collected a profile up to 4000 m by carrying an AirCore whilst skydiving. This work shows potential to provide an additional dimension in local air quality measurements.

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Aerosol Optical Depth at Cape Grim, Tasmania 1978 – 2016

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Abstract

The Southern Ocean is a significant fraction of the Earth's surface but challenging to assess by remote sensing due to cloudiness, especially for properties like Aerosol Optical Depth (AOD), where it is difficult to separate cloud from the record. Under these circumstances a well-characterised, ground-based AOD record is especially valuable. The first measurements of AOD were made at Cape Grim nearly 40 years ago using a handheld 2-channel sunphotometer (Figure 1). Since then, there have been numerous changes to the instruments and measurement protocols used. Currently there are two SPO2 sunphotometers measuring at 6 aerosol relevant wavelengths (1 wavelength for water vapour) with an automated sun tracker.

The measurement and calibration record has been revisited, collating the previous assessments of the manual measurements and for the automated measurement record (from 1986) harmonising the calibration protocols as far as possible and a summary of this process will be presented.

The AOD record at 500 nm (Figure 2) shows the clear impact of the volcanic eruptions of Mt. Pinatubo in 1991 and El Chichón in 1982. However there are significant inconsistencies in this record that will be discussed. The tropospheric aerosol sources are primarily biomass burning and oceanic, and this can be seen in the annual variations of total optical depth and the aerosol size distribution as inferred from the optical depth.



Figure 1: EKO handheld sunphotometer, the last handheld sunphotometer used at Cape Grim.

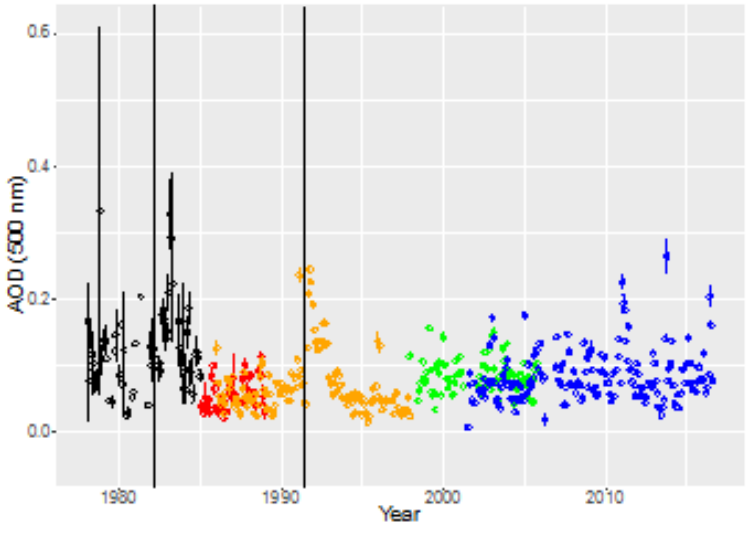


Figure 2: Monthly mean AOD at 500 nm since 1978. Colours denote model of sunphotometer, error bars are 95% confidence intervals of the mean. The two vertical lines indicate mark the two major volcanic eruptions, El Chichón (March 1982) and Mt. Pinatubo (June 1991).

First year of atmospheric reactive mercury measurements at Cape Grim and Macquarie University

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Abstract

Reactive mercury is an important component of the global mercury cycle, but there is little data on ambient concentrations in the Southern Hemisphere, and the most widely used monitoring technique has recently been shown to underestimate total reactive mercury. Here we show initial results from the first year of measurement at the Cape Grim Baseline Air Pollution Station and the Macquarie University meteorological station, using a recently developed cation exchange and nylon membrane filter collection system with better collection efficiencies than the standard method. The mean reactive mercury concentration at Cape Grim was 16 pg m^{-3} and ranged from 10 to 28 pg m^{-3}. Mean reactive mercury at Macquarie University was 20 pg m^{-3}, and ranged from 15 to 32 pg m^{-3}. The higher concentrations at the Macquarie campus are likely a result of greater exposure to local urban sources. There appears to be systematic variation in the minimum and maximum reactive mercury concentrations at Cape Grim, possibly due to seasonal factors, while the concentrations at Macquarie are more constant but punctuated with occasional spikes. The mercury speciation data showed a wide spectrum of mercury species in the urban environment compared to Cape Grim which showed the reactive mercury speciation (based on samples analysed to date) to be largely $HgSO_4$ and $Hg(NO_3)_2$.

Mercury emissions from Australian vegetation during prescribed burns

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Abstract

The occurrence of fires can substantially increase the emissions of mercury (Hg) into the atmosphere. The frequency of both prescribed and wildfires in Australia results in it being one of the largest contributor to Hg emissions from biomass burning, contributing 3% of total Hg global emissions of Hg from biomass burning [Fredli *et al*., 2009]. Little focus has been given to understanding Hg emissions and dispersion from Australian bushfires and no published work has been undertaken to measure real time Hg concentrations from fires, prescribed or otherwise. Hg emissions from vegetation was recorded during a joint experiment at the CSIRO Pyrotron facility in 2014 alongside CO, CO_2 and other commonly emitted gases to quantify emission factors (EF) and emission ratios (ER) from Australian vegetation during a prescribe burn. These EF and ER's were then used to determine the exposure of firefighters and populations to Hg. Two methods were employed during the experiment, the first a continuous Tekran intake place alongside the CO/CO_2 measurements. The second method employed the same grab sampling method where samples were collected in a 10 L tedlar Bags before being ran through a second Tekran unit. An average emission factor of 58.1 µg kg^{-1} continuous method (105.4 µg kg^{-1} bag method) was estimate for the Pyrotron experiment with an emission ratio of 26.25 Hg/CO_2 continuous (30.39 Hg/CO_2, Bag method).

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Emission factors of volatile organic compounds (VOCs) in savannah fires

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Abstract

Biomass burning emissions can have a large impact on atmospheric composition on local, regional and even global scales. In order to assess the impacts of biomass burning emissions on air quality and climate, models require input of emission factors (EFs) which are representative of local conditions. However, availability of EF for Australian fires are sparse, in particular for the wide range of volatile organic compound (VOCs) which are emitted from biomass burning. A study on EFs of VOCs has been carried out for the Savannah Fires in the Early Dry Season campaign (SAFIRED) in Darwin, Northern Territory of Australia in June 2014. The EFs in this study were calculated from VOC masses measured by PTRMS (Proton Transfer Reaction – Mass Spectrometry) which were identified using parallel measurements of VOCs by Thermal Desorption/GC/MS/FID and carbonyls via DNPH cartridge by HPLC. Previously published carbon monoxide (CO) EFs were also used in the calculation [Desservettaz *et al*., 2016]. The EFs for a wide range of VOCs including acetonitrile, methanol, acetaldehyde, etc. are reported and also compared to EF from elsewhere in Australia and oversees [Lawson *et al*., 2015]. This study provides a unique set of VOC EFs for Savannah Fires in Australia.

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Isoprene sensitivity to environmental and chemical factors

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Abstract

Isoprene is the major constituent of global biogenic volatile organic compounds, and is emitted in high concentrations in south east Australia. Isoprene contributes to ozone and secondary organic aerosol formation, particularly during summertime peak emission periods. Sydney is surrounded on three sides by dense eucalypt forest, a tree species with the highest emission factors for isoprene globally. The impact of isoprene emissions on the Sydney airshed is not fully known.

Emmerson *et al.* [2016] found that the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.1) [Guenther *et al*., 2012] predicted isoprene concentrations a factor of 6 higher than PTR-MS observations in Sydney suggested. However there are environmental factors such as water availability and uncertainty in land cover information which could supress these emissions of isoprene. The CSIRO-CTM also uses the carbon bond 5 chemistry scheme (CB05) which is over 10 years old, and does not represent the recent isoprene oxidation pathways which recycle more OH back to the atmosphere under low NOx conditions. These low NO_x conditions exist in south east Australian eucalypt forests.

These three uncertainties will be explored in this talk.

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Reversal of global atmospheric ethane and propane trends largely due to US oil and natural gas production

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Abstract

Ethane is the most abundant non-methane hydrocarbon (NMHC) in the background atmosphere at levels of ~0.4 – 2.5 nmol mol^{-1} (ppbv). Ethane peaked around 1970, followed by a downward trend for the next four decades. Here, we show that the near 40-year trend of declining global ethane halted between 2005-2010 in most of the Northern Hemisphere (NH), and that since it has reversed. These changes are occurring at a hemispheric scale, but are most evident in the eastern part of the North American continent. The largest increases of 5-9 % yr^{-1} in ethane, and of 8-22 % yr^{-1} for the shorter-lived propane are seen in the central and eastern U.S. and immediately downwind, identifying this region as the source of increased NMHC emissions. These NH increases starkly contrast the Southern Hemisphere data, where NMHC levels have been mostly stable. We calculate an approximate top down annual rate of increase in ethane emissions in NH of 0.42 Tg yr^{-1} (Tg = 106 metric tons), respectively a 2.1 Tg yr^{-1} flux increase between 2009.5 – 2014.5, with these emission increases primarily from North America, very likely from the growth of oil and natural gas development. Further, including other co-emitted oil and gas alkane hydrocarbons we estimate a total annual NMHC emission rate increase of 1.2 Tg yr^{-1} (5.9 Tg yr^{-1} total flux increase between 2009.5 – 2014.5). Model results suggest that this added flux of volatile organic compounds has the potential to increase annual mean surface ozone by several ppb and adversely affect air quality in oil and gas basins and downwind regions.

Impacts of C1-C3 alkyl nitrates on the NO_x budget in remote regions

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Abstract

Nitrogen oxides (NO + NO_2 = NO_x) are precursors to tropospheric ozone production, contribute to aerosol formation, and enhance nitrogen deposition to ecosystems. Due to the short NO_x lifetime, these impacts tend to be localised near major emissions, unless the NO_x is instead sequestered in long-lived reservoir species. With lifetimes of weeks to months, short chain (C1-C3) alkyl nitrates ($RONO_2$) are potentially important NO_x reservoirs. $RONO_2$ are formed during volatile organic compound oxidation in the presence of NO_x and removed via photolysis and OH oxidation. As a result, these species could serve to export NO_x from major source regions to the NO_x-limited remote marine troposphere. However, the global budget and impacts of C1-C3 $RONO_2$ remain poorly constrained. Here we will present a newly developed global simulation of C1-C3 $RONO_2$ in the GEOS-Chem global chemical transport model, evaluated using airborne observations from the 2013 NASA SEAC^{4}RS campaign in the Southeast US and the 2009-2011 HIPPO campaigns over the Pacific Ocean. Using the new simulation, we will provide a preliminary assessment of the impacts of $RONO_2$ formation on the NO_x budget.

Atmospheric composition in a region of intensive coal seam gas production

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Abstract

Coal seam gas (CSG) production has expanded rapidly in the Surat Basin Queensland, Australia in the past decade, driven by three major CSG to liquefied natural gas (LNG) export projects.

A new 2 year ambient air quality study has been instigated in the Surat Basin as part of GISERA (Gas Industry Social and Environmental Research Alliance), a partnership between CSIRO and industry. This is the first comprehensive air quality study to be undertaken in an unconventional gas region of Australia. The study involves monitoring NO_x, O_3, total VOCs, CO, CH_4, CO_2, PM2.5 and PM10 at three ambient air quality sites in the centre of the gas fields. In addition, NO_x, O_3 and CO measurements are made at two upwind and downwind sites, which were commissioned as part of the GISERA Methane Seeps project. To provide maximum transparency for concerned communities, ambient air quality data from the 5 sites is being live streamed every hour to the Queensland Government air quality site http://www.ehp.qld.gov.au/air/data/search.php. Additional measurements include fortnightly passive speciated VOC and H_2S measurements, and detailed characterisation of potential CSG related emission sources, including fugitive gas, gas fired compressors and engines, feed pond water and naturally occurring radioactive materials.

A CSIRO meteorological model coupled with a chemical transport model is being used to examine the distribution of primary pollutants (NO_x, CO) in the Surat Basin as well as secondary pollutants (O_3 and secondary PM). The modelling study requires building a detailed emissions inventory of all potential natural and anthropogenic emission sources in the Basin which is a significant task. CSG companies in the region are providing CSG-related emission estimates to the inventory. The study will explore the contribution from the CSG industry and other sources to observed levels of pollutants over one year at 1km, hourly resolution.

We provide an overview of the study including motivation for the work and preliminary ambient air monitoring data, including comparison with air quality standards and source identification of some observed enhancements. Future unconventional gas development in Australia will be discussed, with potential implications for air quality.

Observation of new particle formation in an urban marine coastal environment

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Abstract

Particle number size distribution data in the range of 14 nm to 660 nm were measured from 16 Jan to 18 Feb 2013 during Measurement of Urban, Marine and Biogenic Air (MUMBA) campaign in the Wollongong, Australia. The particle number size distribution was measured using scanning particle mobility sizer (SMPS). One of the goals of this research was to detect new particle formation (NPF) events and identify factors that promote the events. The overall mean (median) of 10-minute averaged concentration of the particle number over the entire size (N_T : 14 – 660 nm) is 5.1 x 10^3 (3.1 x 10^3 cm^{-3}). Overall, six NPF events were observed during the campaign. All NPF events occurred during the day time. The average particle growth rate of the events was 7 nm h^{-1} (ranged between 5 nm h^{-1} and 10 nm h^{-1}). Traffic emissions were the important contributor to the particles observed in the mornings linked to the increase in concentration of primary pollution plumes (e.g. NO and CO) and traces from motor vehicles (e.g. benzene and toluene) before the events. Secondary aerosols were formed in the midday presumably as a combination of photochemistry and meteorology. Oceanic spray also appears to be one of the factors that promote NPF events. Both fine and coarse marine aerosols identified by chloride and show a strong correlation to the NPF events.

Evaluation of regional air quality models over Sydney, Australia

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Abstract

Air quality models are valuable tools to investigate the complex and dynamic interactions between meteorology and chemistry leading to poor air quality episodes. As part of the Western Air-Shed and Particulate Study for Sydney project, we are evaluating several regional air quality models by comparing their output to observations. The models were run for three time periods that coincide with intensive measurement campaigns in the Greater Metropolitan Region of Sydney (February-March 2011, April-May 2012 and December 2012 to February 2013). The project leverages current regional modelling efforts from several groups, including the University of Melbourne, CSIRO, the NSW Office of Environment and Heritage and ANSTO and is open to further contributions, including from international research groups.

Here we describe the project, and present preliminary results for surface meteorological parameters, ozone, particulates, selected volatile organic species and radon. It is hoped the project will advance the reliability of modelling of current and future air quality over the Greater Sydney region to a point where it is possible to use the models to test different policy scenarios and therefore contribute to the design of a Clean Air Plan for Western Sydney.