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# Atmospheric Composition & Chemistry Observations & Modelling Conference

**incorporating the**

## Cape Grim Annual Science Meeting 2019

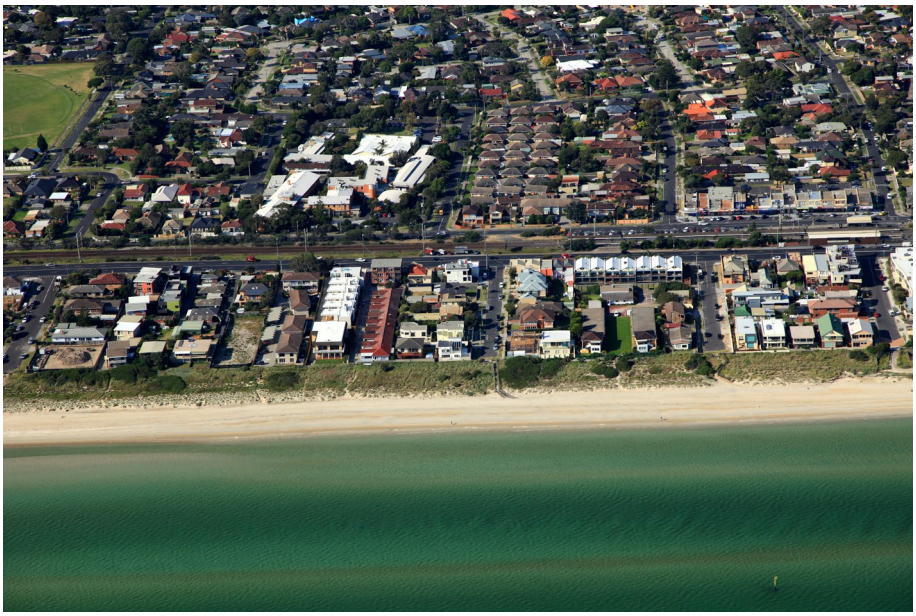
## Abstracts

**19-21 November 2019**

**hosted by**

**CSIRO Oceans and Atmosphere, Climate Science Centre**

**Priestley Lecture Theatre**
**Aspendale, Victoria, Australia**



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

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## Day 1: Tuesday, 19 November 2019

| | | |
|---|---|---|
| **09:00** | **Convenors** | **Welcome & general information on arrangements etc** |
| **Chair** | **Paul Krummel** | **Session 1 – Oral Presentations** |
| 09:10 | Neil Brough | Halley Research station in Antarctica and the influence of sea ice on the surrounding atmosphere |
| 09:25 | Keith Fifield | Bomb-pulse $^{36}Cl$ in the DE08 ice core from Law Dome |
| 09:40 | Cathy Trudinger | Rectifier effects in firn |
| 09:55 | David Etheridge | Quantifying atmospheric OH changes from 1870 AD to present using measurements of $^{14}CO$ in ice and firn from Law Dome, Antarctica |
| 10:10 | David Etheridge | Preindustrial $^{14}CH_4$ indicates that anthropogenic fossil $CH_4$ emissions are underestimated |
| 10:25 | Ingeborg Levin | Long-term changes of $\Delta^{14}CO_2$ in the global atmosphere |
| **10:40** | **Morning Tea** | **Canteen** |
| **Chair** | **Ann Stavert** | **Session 2 – Oral Presentations** |
| 11:10 | Alastair Williams | Cape Grim Radon Program Review: 2014-2019 |
| 11:40 | Alan Griffiths | Blueprint for a more compact high-sensitivity radon detector |
| 11:55 | Scott Chambers | Using $^{222}Rn$ as an indicator of diurnal - and synoptic-timescale changes in the atmospheric mixing state in urban air quality studies |
| 12:10 | Colin Allison | Assuring quality in the measurement and reporting of stable isotope measurements of atmospheric carbon dioxide |
| 12:25 | Andrew Crotwell | Improvements in NOAA analytical capabilities |
| 12:40 | | |
| **12:55** | **Lunch** | **Canteen** |
| **Chair** | **Matthew Woodhouse** | **Session 3 – Oral Presentations** |
| 14:00 | Ann Stavert | Regional trends and drivers of the global methane budget |
| 14:15 | Ray Langenfelds | The role of inter-hemispheric transport in Southern Hemisphere baseline $CO_2$ variation |
| 14:30 | Zoë Loh | Comparison of *in situ* measurements of $CO_2$ and $CH_4$ at Cape Grim Baseline Station |
| 14:45 | David Griffith | Spectronus at sea: greenhouse gas measurements on the *Aurora Australis* and *RV Investigator* |
| 15:00 | Tilo Ziehn | Australia's Earth System Model (ACCESS-ESM1.5) – Emissions-driven simulations for CMIP6 |
| **15:15** | **Afternoon Tea** | **Canteen** |
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| 16:00 | Alexander Geddes | Evaluating New Zealand's agricultural methane emissions using inverse modelling |
| 16:15 | Zoë Loh | Measuring Melbourne's greenhouse gas emissions |
| 16:30 | Xinyi (Lexie) Lu | Characterisation of $CH_4$ source signatures in Melbourne, Australia |
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| **17:00** | **End of Day 1** | |

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| | | |
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| **Chair** | **David Etheridge** | **Session 5 – Oral Presentations** |
| 09:00 | Jesse Greenslade | Top-down estimates of isoprene emissions in Australia inferred from OMI satellite data |
| 09:15 | Kathryn Emmerson | New measurements give insight into Australian isoprene emissions in 2050 |
| 09:30 | Clare Paton-Walsh | The COALA-JOEYS Campaign: Characterising Organics and Aerosol Loading in Australia- Joint Organic Emissions Year-round Study |
| 09:45 | Jhonathan Ramirez Gamboa | The COALA-JOEYS Campaign: Preliminary results from the biogenic ambient air sampling system |
| 10:00 | Hamish McDougall | The COALA-JOEYS Campaign and beyond: Thermal desorption tube measurements of volatile organic compounds |
| 10:15 | Sarah Lawson | Biogenic sulfur from the South West Pacific Ocean: estimating methanethiol fluxes and DMS measurement comparison |

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| **10:30** | **Morning Tea** | **Canteen** |
| **Chair** | **Clare Paton-Walsh** | **Session 6 –**<br>Parallel session with SOAR Workshop, Hobart (via weblink) – Chief's Conference Room |
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| 11:15 | Elise-Andrée Guérette | Exploring greenhouse gas seasonality in the top end: a collaboration between the Larrakia Rangers and CSIRO |
| 11:30 | Nicholas Deutscher | The Total Carbon Column Observing Network – update and future directions |
| 11:45 | Nicholas Jones | Novel ground-based remote sensing of greenhouse gases in the Arctic, tropics and subtropics |
| 12:00 | Travis Naylor | The GipNET measurement network – a potential measurement network for proposed carbon capture and storage |
| 12:15 | Jeremy Silver | Inverse modelling of $CO_2$ for offshore leak detection |
| **12:30** | **Lunch** | **Canteen** |
| **Chair** | **Paul Krummel** | **Session 7 – Shared session with SOAR Workshop, Hobart (via weblink)**<br>**Hobart (H); Aspendale (A)** |
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| 13:45 | Matthew Woodhouse (H) | Modelling marine organic contributions to aerosol |
| 14:00 | Imogen Wadlow (H) | Model-observation comparison of Southern Ocean aerosols in CMIP6 ACCESS |
| 14:15 | Ruhi Humphries (A) | Southern Ocean CCN - Temporal and spatial distributions |
| 14:30 | Robyn Schofield (A) | AIRBOX observations made on the *RSV Aurora Australis* over the 2018/2019 resupply season |
| 14:45 | Melita Keywood (A) | Decade-long aerosol scattering coefficient record from the global baseline atmospheric pollution station at Cape Grim, Australia |
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| Suzie Molloy | | Characteristics of the Baseline Cape Grim Ozone Record, 1982 - 2017 |
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| Ross Ursino | | Analysing trends in the climate-carbon cycle feedbacks and its drivers from observations |
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| 09:15 | Ian Galbally | Nitrogen oxides (NO, $NO_2$, $NO_y$) and nitrate in the air at Cape Grim |
| 09:30 | Nasimeh Shahrokhi | Applying the coupled WRF-CMAQ air quality simulation system for Tehran and comparison with surface and tropospheric column observations |
| 09:45 | Frances Phillips | Open-Path FTIR spectroscopy and measurement of trace gases important to urban air quality |
| 10:00 | Steven Utembe | A comparison of simulated air quality over Melbourne using EDGAR and EPA (Vic) emission inventories |
| 10:15 | Adhitya Sutresna | Characterisation of LA-ICP-MS and receptor modelling to measure, speciate and source apportion particulate matter |
| **10:30** | **Morning Tea** | **Canteen** |
| **Chair** | **Rachel Law** | **Session 10 – Oral Presentations** |
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| 11:15 | Stephen Wilson | Aerosol Optical Depth Measurements at dirty Cape Grim |
| 11:30 | Matthew Tully | Antarctic ozone increase measured by Davis ozonesondes |
| 11:45 | Paul Krummel | The 2019 Antarctic ozone hole |
| 12:00 | Paul Fraser | Global emissions of fluoroform (HFC-23) are increasing despite nations reporting reduced emissions |
| 12:15 | Paul Fraser | Gosan Station, Jeju Island, South Korea: a window into trace gas emissions from East China |
| **12:30** | **Lunch** | **Canteen – End of meeting, safe travels, we hope you enjoyed Aspendale** |
| **14:00** | **Ruhi Humphries** | **CGBAPS Voyage Workshop – Lecture Theatre** |

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# Halley Research station in Antarctica and the influence of sea ice on the surrounding atmosphere

N. Brough* and A. E. Jones

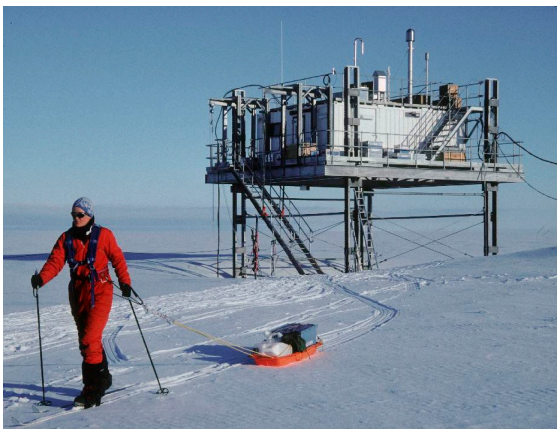
*British Antarctic Survey, Cambridge, UK, Antarctica*

* corresponding author: Neil Brough [nbro@bas.ac.uk]

## Abstract

In 2003, a new laboratory was commissioned at Halley research station in coastal Antarctica, dedicated to studies of the clean background atmosphere and air/ice exchange processes with the snow. With its position near the edge of the Weddell Sea, the laboratory is well placed to capture air masses with a variety of origins, including from the continent, over the sea ice zone and over the open ocean. Since its inception, the laboratory has been run in a dual mode, with specific measurement campaigns operating alongside long-term observations.

Here we introduce the laboratory and the instrument suite and present the type of measurements that we are now able to make. We present some observations of OH and ($HO_2$ + $RO_2$) carried out in Antarctica outside the summer season. Measurements were made over 23 days in spring at the coastal Antarctic station Halley. Air masses that had traversed the sea ice zone had both higher concentrations of OH and a larger OH:($HO_2$ + $RO_2$) ratio. We use steady state kinetic arguments and a 0-D box model to probe the chemical drivers. We find that differences in bromine chemistry, previously measured at Halley, are sufficient to account for the observed differences in OH concentration as well as the ratio. There is some evidence also that chlorine chemistry is influencing concentrations of $RO_2$.



## References

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# Bomb-pulse $^{36}$Cl in the DE08 ice core from Law Dome

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## Abstract

The radioactive isotope chlorine-36 ($T_{½}$ = 301,000 years) has been measured with annual resolution over the period 1945-1978 in the DE08 ice core from Law Dome. This covers the period of atmospheric nuclear weapons testing and its aftermath. The observed $^{36}$Cl was produced principally by thermonuclear (H-bomb) tests that were carried out on or over seawater at the islands of Enewetak and Bikini in the Marshall Islands in 1952-1958. Consequently, the injection of chlorine-36 into the stratosphere was rather well-defined in time. A strong peak in $^{36}$Cl that is a factor of 200 above natural levels is observed in 1958-1959. The subsequent decay of this signal over the next 20 years is driven by the rate of exchange of atmospheric gases between stratosphere and troposphere, which in principle can be deduced from the data. Further, similar measurements have been carried out by other groups in Greenland ice cores, and comparison between these Arctic and our Antarctic results contains information on the rate of exchange of stratospheric air between the northern and southern hemispheres. We will present the data and some preliminary conclusions.

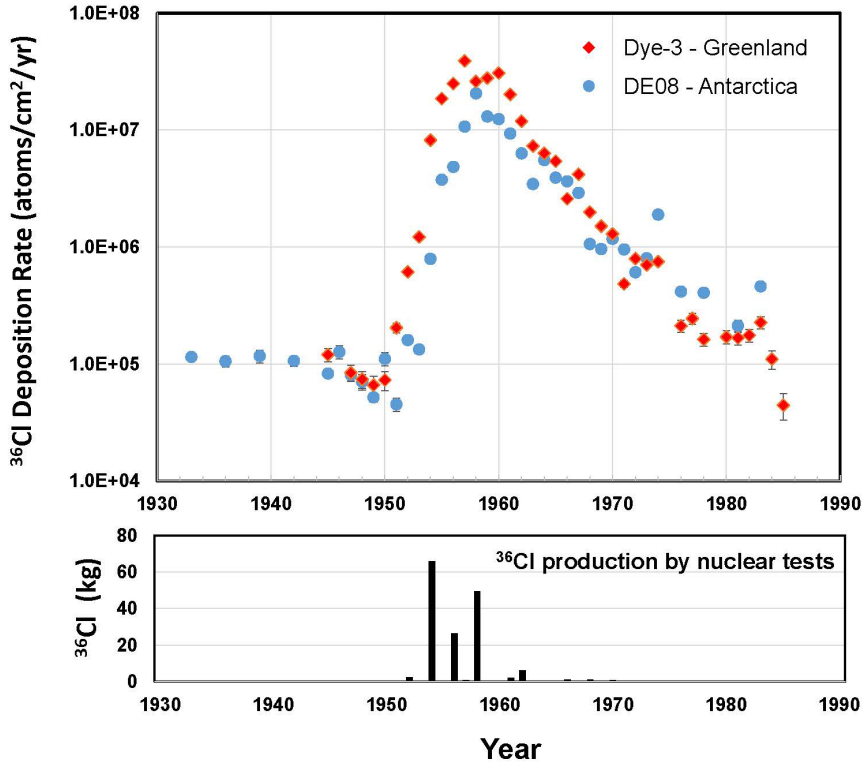



**Figure 1.** Deposition rates of $^{36}$Cl in Antarctica and Greenland are shown in the upper panel. The Antarctic results are from the present work, while the Greenland data are from Synal *et al.* The lower panel shows a preliminary production profile of $^{36}$Cl from nuclear weapons testing.

# Rectifier effects in firn

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## Abstract

Firn is the porous layer of compacted snow overlying an ice sheet. Air is contained in tiny channels that are open to the atmosphere, and this air can be extracted from the firn to provide an archive of old air. The channels eventually close to form discrete bubbles, and air from these bubbles (as well as firn air) can give information about past atmospheric composition. The composition of the air in firn and bubbles in ice is influenced by changes in the overlying atmosphere as well as by processes occurring in the firn.

Diffusive mixing is important throughout the firn, but convective mixing can dominate near the surface at some sites. Covariation of convective mixing in the upper firn and seasonality of the atmospheric abundance of some tracers has the potential to shift the mean level of a tracer in the firn relative to the atmospheric record, the so-called 'rectifier effect'. We have found evidence that rectifier effects have influenced carbon monoxide at the Antarctic DE08-OH site sampled in December 2018. We explore rectifier effects at DE08-OH and other firn sites for several trace gases with significant seasonality in the atmosphere. Better understanding of these effects will lead to improved reconstruction of atmospheric composition from firn and ice core measurements.

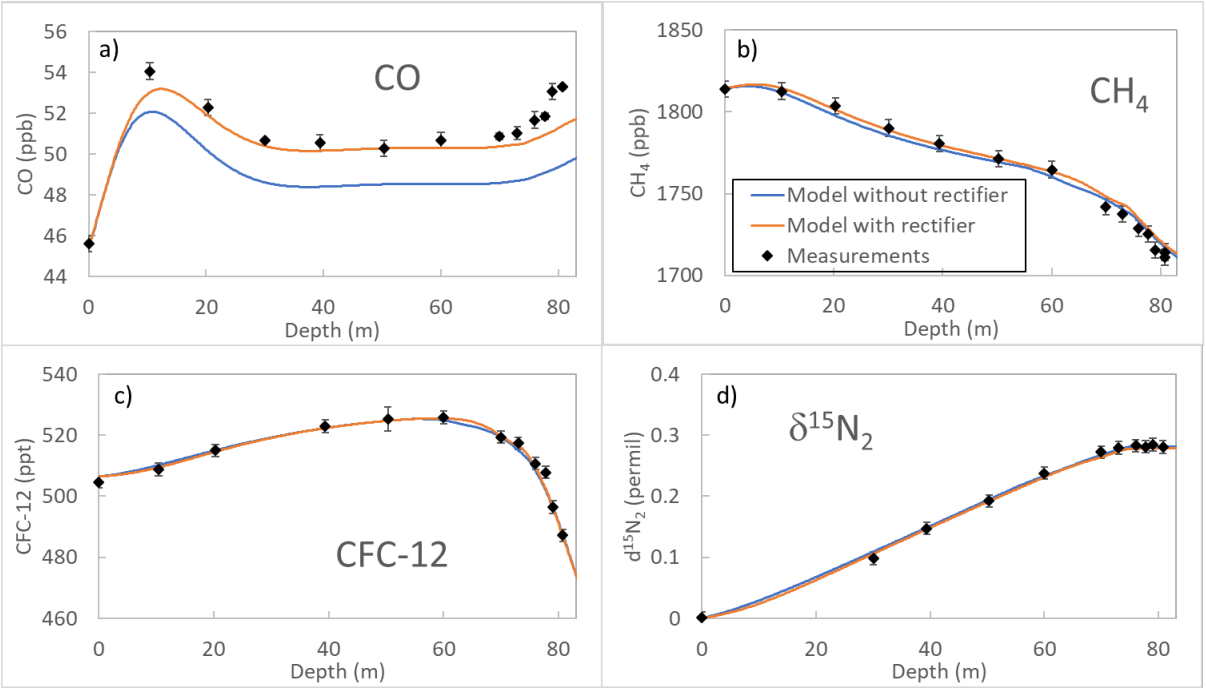



**Figure 1.** Modelled depths profiles of CO, $CH_4$, CFC-12 and $\delta^{15}N_2$ in firn air with and without rectifier effects, compared to observations at DE08-OH, Antarctica. Diffusivity parameters in the firn model were calibrated independently for the cases with and without rectifier effects, using 12 different tracers.

# Quantifying atmospheric OH changes from 1870 AD to present using measurements of $^{14}CO$ in ice and firn from Law Dome, Antarctica

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## Abstract

Hydroxyl, OH, is the main tropospheric oxidant and determines the lifetime of methane and most other trace gases in the atmosphere, thereby controlling the amount of greenhouse warming produced by these gases. Changes in OH concentration ([OH]) in response to large changes in reactive trace gas emissions (which may occur in the future) are uncertain. Measurements of $^{14}C$-containing carbon monoxide ($^{14}CO$) and other tracers such as methyl chloroform over the last ≈25 years have been successfully used to monitor changes in average [OH], but there are no observational constraints on [OH] further back in time. Reconstructions of $^{14}CO$ from ice cores could in principle provide such constraints but are complicated by in-situ production of $^{14}CO$ by cosmic rays directly in the ice. Recent work in Antarctica and Greenland shows that this in-situ component would be relatively small and can be accurately corrected for at sites with very high snow accumulation rates. From December 2018 to January 2019, a joint US and Australian team sampled and measured firn air and ice at Law Dome, Antarctica (site DE08-OH, 1.2 m a-1 ice-equivalent snow accumulation), to a maximum depth of 240 m. Trapped air was extracted from the ice using an on-site large-volume ice melting system. Preliminary comparisons of CH4 measured in the samples to existing ice core records and atmospheric measurements suggest ice core air sample ages spanning from the 1870s to the early 2000s. Firn-air samples from the snow surface to 81 m depth capture air from the early 2000s to present. Analyses of [CO] and halocarbons in the samples show a relatively low and stable procedural CO blank and demonstrate that the samples are unaffected by ambient air inclusion. $^{14}CO$ analyses in these firn and ice core air samples are in progress. This $^{14}CO$ history will be interpreted with the aid of the GEOS-Chem chemistry-transport model to place the first observational constraints on the variability of Southern Hemisphere [OH] since ≈1870 AD.

# Preindustrial $^{14}CH_4$ indicates that anthropogenic fossil $CH_4$ emissions are underestimated

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## Abstract

Atmospheric methane ($CH_4$) concentrations have more than doubled since the preindustrial era. $CH_4$ emissions from fossil fuel extraction and use are one of the largest anthropogenic sources, but the precise magnitude of this source is a subject of debate. Carbon-14 in $CH_4$ ($^{14}CH_4$) can distinguish between fossil ($^{14}C$-free) emissions and contemporaneous biogenic sources; however, poorly constrained direct $^{14}CH_4$ emissions from nuclear reactors complicate this interpretation after the mid-20$^{th}$ century. Further, it has been debated how total fossil $CH_4$ emissions are partitioned between anthropogenic and natural geological sources (such as seeps and mud volcanoes); emission inventories suggest the latter accounts for ~40-60 Tg $CH_4$/yr. Here we use preindustrial ice core $^{14}CH_4$ measurements to show that natural geological $CH_4$ emissions to the atmosphere were an order of magnitude smaller than currently used estimates. This result indicates that anthropogenic fossil emissions are underestimated by a corresponding amount (~25-40% of recent estimates). Our record highlights the human impact on the atmosphere and climate, provides a firm target for inventories of the global $CH_4$ budget, and will help to inform strategies for targeted emission reductions.

# Long-term changes of $\Delta^{14}CO_2$ in the global atmosphere

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## Abstract

Measurements of the distribution and long-term trends of radiocarbon in atmospheric carbon dioxide have provided invaluable insight into global and regional fluxes within the carbon cycle [e.g. Levin *et al*., 2003; Levin *et al*., 2010]. While $\Delta^{14}C$-$CO_2$ has been decreasing almost exponentially in the decades following the 1960s' atmospheric bomb testing, due to uptake of bomb-produced $^{14}CO_2$ by the ocean and the terrestrial biosphere, the currently observed mean $\Delta^{14}C$-$CO_2$ decrease rate of about 4-5‰ per year is almost solely caused by the input of fossil fuel $CO_2$ into the atmosphere. This $^{14}CO_2$-free anthropogenic $CO_2$ flux has increased globally by more than 50% compared to 1990 [Le Quéré *et al*., 2018], thus raising the atmospheric $CO_2$ burden by more than 4 PgC (1 PgC = $10^{15}$ gC) or >0.5% per year in the last decade (ESRL GMD, 2019). As the largest share of fossil fuel $CO_2$ is released in the Northern Hemisphere, the north-south $CO_2$ concentration difference also showed a steady increase. However, only since about 2005 we observe on average 5‰ higher $\Delta^{14}C$-$CO_2$ at the Antarctic coast compared to high latitudes of the Northern Hemisphere. This is due to still compensating $^{14}CO_2$ disequilibrium fluxes from biosphere and ocean, coupled with increasing $^{14}CO_2$ fossil fuel $CO_2$ emissions. Overlaying the long-term decreasing trend, we observe significant inter-annual variations, which must be attributed to natural signals, either varying disequilibrium fluxes with the southern (and global) ocean but also possibly due to natural $^{14}C$ production that is linked to solar variability (e.g. 11-year sunspot cycle). Our extended high-precision $\Delta^{14}C$-$CO_2$ observations from Alert, Jungfraujoch, Cape Grim and the German Antarctic Neumayer station are presented and discussed in view of the changes and variability of the atmospheric radiocarbon cycle in the last four decades.

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# Cape Grim Radon Program Review: 2014-2019

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## Abstract

At Cape Grim, changes in atmospheric radon concentrations are measured to high precision and temporal resolution using purpose-built high sensitivity instruments. Radon has been measured at the Cape Grim Baseline Air Pollution Station (CGBAPS) since 1980, as a collaboration between the Bureau of Meteorology (BoM) and the Australian Nuclear Science and Technology Organisation (ANSTO).

Radon provides an unambiguous indication of the degree of recent terrestrial influences on an air mass. It is an ideal tracer for a range of applications at baseline and remote stations such as Cape Grim. By observing the behaviour of radon as a tracer in the atmosphere, we provide knowledge and resources that aid in the monitoring and interpretation of changes in atmospheric composition, tracking of the movements of greenhouse gases and other trace species through the environment, and the prediction of future changes in regional and global patterns of pollution and climate.

The overarching aim of the Cape Grim Radon Program is to produce and apply high quality radon observations to advance our understanding of global atmospheric composition, pollution and climate. Specific goals of the Program are to:

- lead measurements of continuous, high temporal resolution (hourly or better), high sensitivity (≤10 mBq $m^{-3}$ lower limit of detection) time series of atmospheric radon concentrations;
- maintain and strengthen the world-leading expertise in measurement techniques of very low radon concentrations in marine air least perturbed by recent land contact;
- refine baseline and distant fetch analyses of greenhouse and other trace gases and aerosols, to exploit the CGBAPS' unique location for probing a large area of the Southern Ocean and mainland Australia south of 28°S;
- exploit the potential that radon offers for: (a) calibrating and/or constraining regional terrestrial emissions of greenhouse and other trace gases and aerosols, including carbon dioxide, methane, nitrous oxide, molecular hydrogen and black carbon; and (b) evaluation of transport and mixing schemes in weather, climate and air chemistry models on various scales;
- expand measuring programs of atmospheric radon at locations of national / international importance with strong links to the CGBAPS program; and
- further strengthen the CGBAPS role within the WMO-GAW community.

In this presentation, we will give an overview of the current Cape Grim Radon Program, focusing on research achievements and technical challenges since the previous review in 2013, and suggesting possible future directions.



Figure 1. Scott Chambers and Ot Sisoutham in front of the 5000L primary radon detector at CGBAPS, with the 1500L backup radon detector in the background (February 2015).

# Blueprint for a more compact high-sensitivity radon detector

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## Abstract

The concentration of radon–222 in clean marine air is very low indeed; the 100 mBq $m^{-3}$ threshold concentration, below which we say the air is free of recent terrestrial influence, equates to about 50 radon atoms per litre of air.

The dual-flow-loop two-filter radon detector, whose central design features have remained unchanged for more than two decades [Whittlestone and Zaharowski, 1998], does a remarkable job of measuring radon at extremely low concentrations. It is simple to operate, robust, and reliable; but not perfect. The current design calls for a large delay volume (1.5 $m^3$ is typical) meaning that the installation of these detectors must be planned carefully, and they can be difficult to retrofit to existing sites or mobile platforms. Often, due to their bulk, the detectors are installed outdoors thus increasing the maintenance burden and impacting on reliability.

To reduce the overall size of the detector, we are currently focusing on the so-called 'detector head', a combined filtration and alpha detection unit. This is because the sensitivity of the detector – a requirement dictated by the expected radon concentration – scales linearly with the product of the head efficiency and the size of the delay volume. A more efficient head, therefore, allows for a smaller delay volume. With the present design, though, head efficiency is fundamentally limited to 0.25 detected counts per alpha particle, and the detectors currently in use have an efficiency approaching this limit.

Taking advantage of wavelength shifting optical fibre and silicon photomultipliers, technologies which were not widely available in the 1990s, we are developing a new head design which uses a different approach for both filtration and detection. In principle, head efficiency in the new design can approach 100%. If achieved, the new head design could be used to improve the signal-to-noise ratio of radon measurements or used to reduce the size of the 5000 L detector presently at Cape Grim to a size comparable with a 19-inch instrument rack or domestic refrigerator.

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# Using $^{222}$Rn as an indicator of diurnal- and synoptic-timescale changes in the atmospheric mixing state in urban air quality studies

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## Abstract

Urban air quality is strongly influenced by the atmosphere's ability to disperse primary emissions and opportunities for secondary pollution formation. In northern mid- to high-latitude regions that experience enduring winter snow cover, regional subsidence associated with lingering anti-cyclones can lead to shallow 'persistent temperature inversions' (PTIs), which can result in life-threatening pollution episodes. While often associated with regions of complex topography [Baasandorj *et al*., 2017; Kikaj *et al*., 2019], PTI events can also influence the air quality of urban centres in flat, inland regions [Chambers and Podstawczyńska, 2019]. This presentation will introduce a recently-developed radon-based technique for identifying and characterising synoptic-timescale PTI events, which is proving to be a simple and economical alternative to contemporary meteorological approaches that require regular sonde profiles. Furthermore, some key assumptions for applying the radon-based technique to characterise diurnal-timescale changes in the atmospheric mixing state described by Chambers *et al*. [2019] are violated during PTI conditions. Here we demonstrate how successive application of radon-based techniques for classifying synoptic- and diurnal-timescale changes in the atmospheric mixing state can significantly improve our understanding of atmospheric controls on urban air quality in non-summer months across the full diurnal cycle.

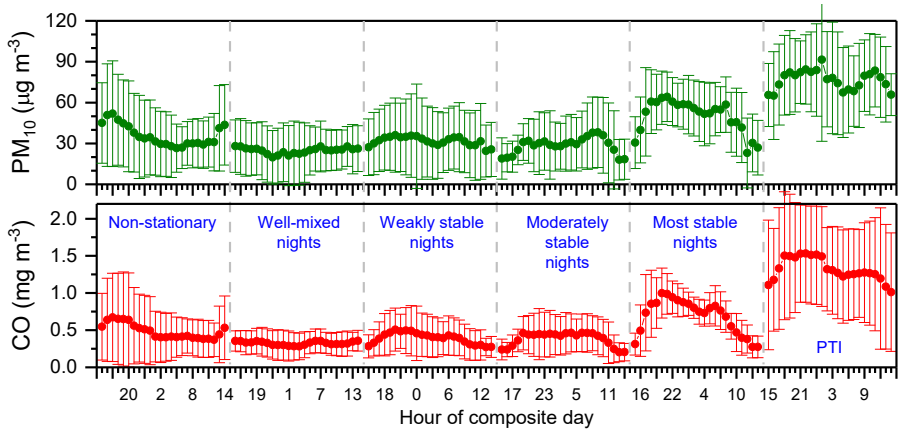



Figure 1. Mean-hourly CO and $PM_{10}$ in Ljubljana, Slovenia, as a function of radon-derived atmospheric mixing state.

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# Assuring quality in the measurement and reporting of stable isotope measurements of atmospheric carbon dioxide

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## Abstract

Of the forty-four laboratories that participated in the sixth WMO/IAEA Round Robin (RR6) only fifteen laboratories reported $\delta^{13}C$ (and only thirteen reported $\delta^{18}O$) measurements of the $CO_2$ [NOAA, 2019] and only two of those laboratories submit data from multiple sites to the WDCGG [GAW, 2019]. Differences between the fifteen laboratories that reported $\delta^{13}C$ of $CO_2$ data in RR6 significantly exceed the recommended targets for network compatibility (Table 1 in [WMO, 2018]). Is this 'disagreement' a limiting factor in the submission of data to the WDCGG by RR participants? If so, what could be done to improve this situation?

The WDCGG accepts $\delta^{13}C$ and $\delta^{18}O$ (of $CO_2$) data on two 'scales', PDB and VPDB-CO2, but the recommended scale is VPDB-CO2 and, as GGMT Meeting Reports have stated for several years "JRAS-06 is the recommended VPDB-CO2 scale realization for stable isotopes of $CO_2$" [WMO, 2018]. For several decades, both CSIRO and NOAA/INSTAAR, the laboratories that submit multiple data sets to the WDCGG, have maintained independent VPDB-CO2 calibration schemes; recently both have implemented the JRAS-06 realization of the VPDB-CO2 scale and so should be reporting data on the same scale.

The CSIRO implementation of the JRAS-06 realization of the VPDB-CO2 scale will be presented and compared with data from several laboratories in order to elaborate on some difficulties that need to be overcome in order to attain the recommended GGMT compatibility targets. Implications of the JRAS-06 scale realisation for the Cape Grim flask intercomparison with NOAA and for optical measurements of $CO_2$ stable isotopes will be discussed. Concluding remarks will emphasize the importance of a single calibration scale with robust uncertainty estimates in order to achieve data comparability and compatibility for stable isotope measurements of $CO_2$ in the upcoming seventh WMO/IAEA Round Robin (RR7).

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# Improvements in NOAA analytical capabilities

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## Abstract

The WMO GAW community has set network compatibility goals (a measure of bias between networks) to ensure data from various programs can be combined without biasing estimates of emissions. One of the ways in which the community ensures minimal bias is through the use of common calibration scales, maintained and disseminated by a single central calibration laboratory (CCL). NOAA serves as the CCL for carbon dioxide ($CO_2$), methane ($CH_4$), nitrous oxide ($N_2O$), sulfur hexafluoride ($SF_6$) and carbon monoxide (CO). The ability of NOAA to propagate scales to other institutions directly affects their ability to meet targeted WMO network compatibility goals. In the case of $N_2O$, and for $CO_2$ measurements from the Southern Hemisphere where gradients are small, the WMO network compatibility goals are extremely demanding and limitations in scale propagation have been a significant impediment to achieving them.

In 2016 the $CO_2$ calibration system at NOAA was updated to improve the ability of NOAA to propagate the scale. The system now uses laser spectroscopic techniques and properly accounts for isotopic differences between standards. The reproducibility for scale transfer is now ±0.01 (68% CI), a factor of three better than the historical NDIR based system. This may make it easier for labs measuring $CO_2$ in the SH to meet the WMO network compatibility goal (0.05 ppm).

In addition, the WMO GAW community stresses the need for on-going comparison activities to ensure biases between networks are continuously evaluated. NOAA contributes to this process through same-air and co-located comparisons of measurements at its network sites. The flask analysis system used to measure discrete air samples collected through the NOAA global network was updated in 2019. This system also relies on laser spectroscopic methods (for $CO_2$, $CH_4$, $N_2O$, and CO) along with gas chromatographic methods (for $SF_6$ and $H_2$). $N_2O$ measurements in particular have improved significantly. Agreement between pairs of flasks sampled in series is ~0.03 ppb, a factor of 10 better than before. The improved analytical methods along with an improved calibration strategy should greatly improve the consistency of the long-term NOAA records and improve their usefulness for comparison activities with other international networks.

# Regional trends and drivers of the global methane budget

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## Abstract

Methane ($CH_4$) is a key and concerning greenhouse gas with a global warming potential 28 times that of carbon dioxide ($CO_2$) over a 100-yr time horizon [Myhre *et al*., 2013]. While significant sinks of $CH_4$ exist, its atmospheric concentration continues to grow, largely due to anthropogenic emissions, albeit with some controversy as to the relative importance of different source types. This study focuses on temporal trends in the anthropogenic and natural sources of methane, decomposing them regionally into sector type – agriculture and waste, biomass burning and biofuels, fossil, wetlands and other natural sources (including freshwaters, geological and termites) – using both top-down (TD) and bottom-up (BU) methods. These two complementary methods utilise different sources of information, with BU methods using either an inventory or process model-based approach while TD methods are based on measurements of atmospheric $CH_4$ and atmospheric inversion systems (top-down methods - TD). As such, there is increased confidence in temporal trends when they are evident in both methods.

Regional trends in total methane emissions are very robust with all but one (Canada) of the 19 regions examined (18 land and one ocean) showing consistent temporal trends over 2000-2017. Of the regions with consistent trends, ten regions show an increase in total methane emissions, six no clear trend and only one region (Europe) a decrease. A sectorial decomposition of this region finds this decrease to be driven by a 25% and 17% decrease in fossil fuel and agriculture and waste emissions, respectively, again consistent between the TD and BU approaches. While the sectorial breakdown for the majority of regions with increasing total sources estimates are consistent between the TD and BU methods there are some differences both in trend and absolute magnitude that require further investigation.

Studies such as this which develop our understanding of trends in regional methane emissions are key for understanding the drivers of regional methane emissions and allow not only evaluation of current mitigation strategies and policies but for the identification of regional specific foci for future emissions reduction efforts.

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# The role of inter-hemispheric transport in Southern Hemisphere baseline $CO_2$ variation

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## Abstract

A precise three-decade composite record of monthly interhemispheric difference in baseline $CO_2$ reveals systematic variation around the trend due mainly to increasing Northern Hemisphere industrial emissions.

Mid-to-high northern latitude air-surface $CO_2$ exchange with terrestrial forests induce strong seasonality in the interhemispheric $CO_2$ difference However variations around the mean seasonal cycle show little correspondence with predicted climate induced air-surface flux anomalies obtained from Dynamic Vegetation Models.

Terrestrial and oceanic flux anomalies are demonstrated to be small by comparison with cross-equatorial upper troposphere $CO_2$ flux variations. Cross-equatorial fluxes are increasingly uni-directional as the $CO_2$ removal phase due to photosynthetic uptake by northern forests is offset by the increasing north difference due to industrial emissions, resulting in minimal partial pressure difference in this season. (Note: Exchanges with equatorial regions that contribute the semi-periodic ENSO variation in global $CO_2$ growth rates are not evident in the $CO_2$ inter-hemispheric difference).

Transport models of the global carbon budget have so far failed to capture the interhemispheric $CO_2$ transport variability, in particular that by eddy processes that peak around February-April; this is also when the $CO_2$ interhemispheric difference approaches a maximum. At this time, northern terrestrial $CO_2$ respiration adds to vertically transported industrial emissions from the SE Asian region, which is transported to the Southern Hemisphere when sufficient equatorial upper troposphere transient kinetic energy is present (mostly generated by intermittent westerly winds over the Himalayan region). Exchanged $CO_2$ carries a stable isotopic ratio characteristic of forest photosynthesis (~18 per mil discrimination against atmospheric $^{13}C$), similar to that from recently combusted fossil fuels.

Interhemispheric $CO_2$ in the May-Jul period partial pressure difference is similarly large. At this time there is descending air in the SE Asian region, with the consequence that emitted $CO_2$ has more opportunity and time for gross exchange and rapid isotopic equilibration with surface carbon reservoirs. In this the $CO_2$ eventually transferred to the Southern Hemisphere by mean transport (Hadley circulation) can have isotopic composition similar to Southern Hemisphere background air.

Evidence supporting the relevant magnitudes of air-surface and cross-equatorial $CO_2$ fluxes is presented, and the uniquely calibrated 3-decade Cape Grim baseline $\delta^{13}C$ flask record is examined for evidence of eddy incursions of recent heavily-labelled Northern emissions.

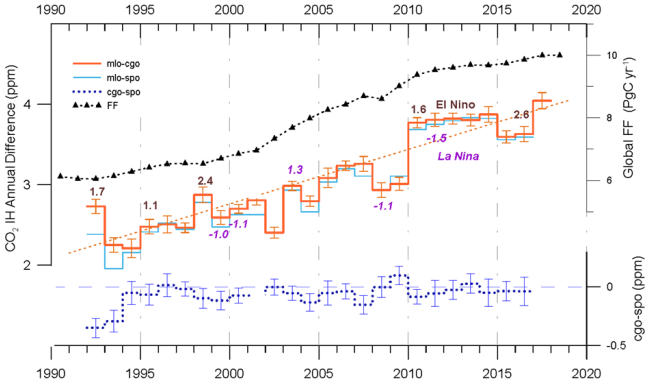



**Figure 1.** Annual changes in the baseline $CO_2$ difference between sites. Interhemispheric differences mlo-cgo (orange, with dashed linear regression) and mlo-spo (light blue) are plotted on the left axis. The peak magnitudes of strong El Niños (brown, ONI index > 1) and strong La Niñas (purple, ONI index < -1) are indicated. The cgo-spo annual differences are plotted on a doubled right-hand scale. Annual fossil fuel emissions from Frederiksen and Francey [2018], are shown on the top right axis.

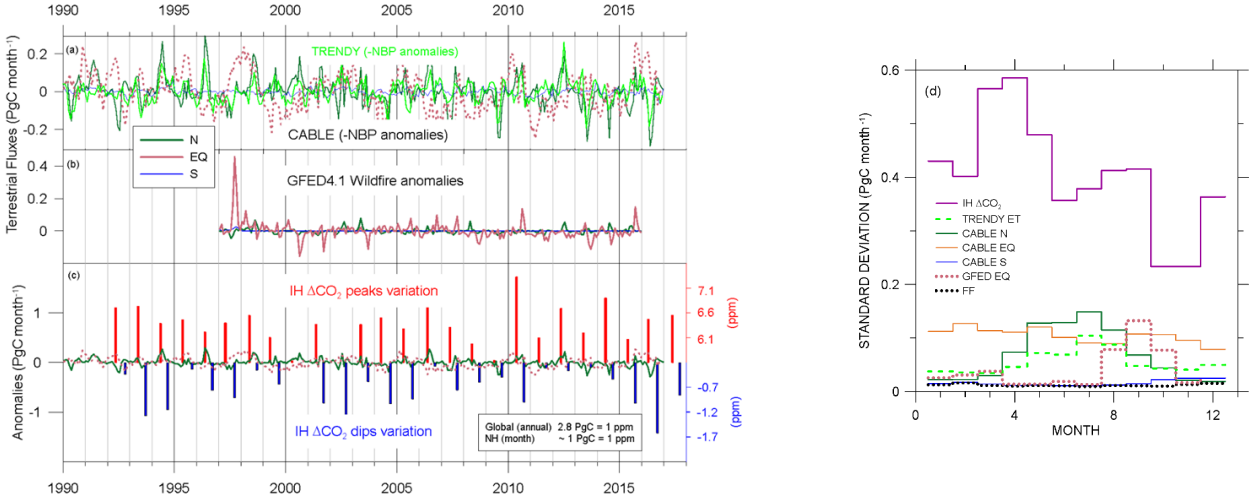



**Figure 2.** Comparison of the timing and amplitude of terrestrial emission anomalies (i.e. mean seasonality subtracted) with variations of the peaks and dips of the monthly mlo-cgo differences. (a) shows seasonal anomalies in CABLE emissions (dark green) and in 16-DGVM TRENDY extra-tropical emissions (light green) [Bastos *et al*., 2018] and (b) shows GFED4.1 wild fire seasonal anomalies, for NH (green), EQ (pink) and SH (blue, SH/EQ for GFED4.1). In (c) The largest anomalies (CABLE NH, CABLE EQ, and GFED4.1 EQ) on the left axis are compared to the ppm variation in peaks (red) and dips (blue) on separate right axes. The axes scaling equates 1 PgC with 1 ppm (see text). To highlight seasonal differences, Figure 2(d) shows the standard deviation in the seasonal anomalies for each month, including those in anthropogenic emissions [Francey *et al*., 2019].

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# Comparison of *in situ* measurements of $CO_2$ and $CH_4$ at Cape Grim Baseline Station

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## Abstract

*In situ* measurements of atmospheric $CO_2$ and $CH_4$ have been made at Cape Grim Baseline Air Pollution Station, in northwest Tasmania since 1976 and 1984, respectively. The instruments providing $CO_2$ measurements have been upgraded and replaced several times over the past 40 years, with a LoFlo Mk2 high precision non-dispersive infrared (NDIR) instrument functioning as our workhorse since 2004. Methane measurements have been made with a gas chromatography-flame ionisation detector method, a component of a gas chromatograph multi-detector (GC-MD) system (one ambient aliquot every 40 minutes) operated as part of the Advanced Global Atmospheric Gases Experiment (AGAGE) since 1993.

In mid-2012, a cavity ring-down spectrometer (CRDS) instrument (Picarro Inc., model G2301), measuring both $CO_2$ and $CH_4$ (as well as $H_2O$), was installed at Cape Grim, for trial ahead of retirement of the LoFlo instrument, and to significantly improve the temporal coverage of our $CH_4$ record. In January 2017, a second Picarro G2301 CRDS instrument was installed at Cape Grim to provide redundancy for these key measurements.

The technical details of gas handling and calibration are quite different for each type of measurement. For instance, the LoFlo instrument uses chemical drying (anhydrous magnesium perchlorate; $Mg(ClO_4)_2$) plus a Nafion membrane to measure dry air, whereas the Picarro instruments measure untreated ambient air (including its water content) and we make an empirical correction to obtain dry air mole fractions.

This presentation will provide a detailed comparison between pairs of *in situ* records for each trace gas, examining how effective our gas handling and calibration regimes are at attaining and sustaining the desired WMO compatibility goals of ±2 ppb $CH_4$ and ±0.05 ppm $CO_2$, at this Southern Hemisphere site.

# Spectronus at sea: greenhouse gas measurements on the *Aurora Australis* and RV *Investigator*

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## Abstract

The Spectronus FTIR analyser provides continuous, simultaneous measurements of $CO_2$, $CH_4$, CO, $N_2O$ and $\delta^{13}C$ in $CO_2$ in air with high precision and accuracy. An early University of Wollongong Spectronus took part in the first cold water trial of the *RV Investigator* in 2015, beside CSIRO analysers. The Airbox Spectronus operated on the *Aurora Australis* for four Antarctic supply voyages in 2018-2019 and is currently on board the *RV Investigator* in northern Australia until January 2020.

This talk will provide an overview of existing shipboard measurements, a preliminary look at the current voyage, comparisons to other instruments, impact of ship's exhaust, and lessons learnt for future deployments on ships.

# Australia's Earth System Model (ACCESS-ESM1.5) – Emissions driven simulations for CMIP6

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## Abstract

CSIRO and collaborators are preparing two submissions for CMIP6 using different versions of the Australian Community Climate and Earth System Simulator (ACCESS). Here we focus on one of those submissions, namely ACCESS-ESM1.5, which will be the first Australian submission to include an interactive carbon cycle. CMIP6 comprises core experiments that all models are expected to perform, and more than twenty optional model intercomparison projects (MIPs) that build on the core experiments and address different science questions. Here we explore emissions driven simulations covering the historical period (1850-2014) and future scenarios (2015-2100). We will discuss a selection of results, with the main focus on the carbon cycle and the interaction between atmosphere, land and ocean.

# CarbonWatchNZ: Regional to National Scale Inverse Modelling of New Zealand's Carbon Balance

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## Abstract

Atmospheric observations of $CO_2$ and other greenhouse gases have been widely used to constrain estimates of terrestrial and oceanic $CO_2$ fluxes through atmospheric inverse modelling. Yet, applying these methods at national scale to verify and improve the National Inventory Report (NIR) and support the Paris agreement remains at the frontier of $CO_2$ science.

A pilot study investigating New Zealand's national carbon budget for 2011-2013 found a significantly stronger (30-60 %) sink relative to the NIR [Steinkamp *et al*., 2017]. The results of the study pointed to a strong $CO_2$ uptake in Fiordland, a region covered by indigenous temperate rainforest in New Zealand's Southern Island. However, this pilot study covered a relatively short time period and included only two atmospheric observing stations, Baring Head (North Island, 41.41°S, 174.87°E) and Lauder (South Island, 38.33°S, 176.38°E).

We present the first results from the CarbonWatchNZ project, which will expand New Zealand's observing network to a total of eleven in situ greenhouse gas measurement sites and improve our atmospheric model resolution by roughly a factor of ten. We combine the measurements with models to develop a complete top-down picture of New Zealand's carbon balance, by studying the three landscapes that are most important to New Zealand's carbon emissions and uptake: forest, grassland and urban environments. In addition to quantifying New Zealand's carbon emissions on a national scale, the results of these studies will help identify the prevailing processes driving $CO_2$ changes in New Zealand to support climate mitigation.

In this initial year of the project, we have extended the $CO_2$ inversion described by Steinkamp *et al*. [2017] to the near-present (2018-2019), and included measurement from our newest observing site, Maunga Kakaramea (North Island, 45.034°S, 169.68°E). The inversion is based on back trajectory simulations from a Lagrangian model using the Numerical Atmospheric dispersion Modelling Environment (NAME) dispersion model driven by New Zealand Limited Area Model (NZLAM, 12 km) meteorology. To analyse the sensitivity and the robustness of the results we also performed simulations using higher resolution New Zealand Convective Scale Model (NZCSM, 1.5 km) meteorology.

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# Evaluating New Zealand's agricultural methane emissions using inverse modelling

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## Abstract

Agriculture accounts for nearly half of New Zealand's greenhouse gas emissions, primarily from enteric fermentation. Knowing this and with the recent announcement that the farming sector must make progress in reducing emissions at the farm scale before joining the emissions trading scheme by 2025, it is vitally important to be able to monitor emissions rather than relying solely on bottom-up estimates.

In this paper we present early results of a study into regional methane emissions using inverse modelling. Using in-situ measurements of methane at NIWA's world class observations sites, the NAME dispersion model [Jones *et al*., 2007] and a Bayesian regional inversion scheme [Steinkamp *et al*., 2017; Mikaloff Fletcher, 2004]. we have performed inversions for 2011-2013. While the results are promising, there are significant challenges and limitations of the method as it stands. Here we discuss these issues and how to address them, particularly focusing on the importance of transport with a comparison between the original numerical weather model; NZLAM [Davies *et al*., 2005], operating at a 12 km resolution, with the new NZCSM model which has a 1.5 km resolution.

These early results using NZCSM show marked improvement over NZLAM at modelling windspeed at the observations sites, the impact this has on the inversion is substantial. Far fewer erroneous results are found and for some regions, the degrees of freedom, a measure of the information content of the inversion, has doubled, approaching 8 per year out of a total of 12. While there is still a long way to go, these early results show that this is a promising method of quantifying regional emissions of methane at a monthly time scale.

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# Measuring Melbourne's greenhouse gas emissions

Z. M. Loh[1*], J. C. Turnbull[2], P. J. Rayner[3], W. Huo[3], Y. Villalobos Cortés[3], D. P. Thornton[1], B. Mitrevski[1], E.-A. Guérette[1], R. L. Langenfelds[1], D. A. Spencer[1], J. A. Collins[2] and P. B. Krummel[1]

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## Abstract

To meet the aspirations of the Paris Agreement, deep and rapid cuts in greenhouse gas (GHG) emissions are required. Since cities are both the epicentres of anthropogenic GHG emissions and often more progressive than states and nations in enacting policies to curb emissions, there is considerable value in developing atmospheric observational and modelling tools at city scales to inform smart mitigation options.

To complement *in situ* measurements of $CO_2$, $CH_4$, and a large range of synthetic GHGs at CSIRO's Aspendale site, we have been taking weekly flask samples at both Aspendale and the University of Melbourne's Parkville campus for radiocarbon ($^{14}CO_2$) analysis. The radiocarbon data are a direct tracer of fossil fuel derived $CO_2$ in the air sample and are thus a powerful constraint in determining the anthropogenic $CO_2$ emissions. In conjunction with radiocarbon measurements, we have also been making measurements of our suite of synthetic GHGs on flask samples from the University of Melbourne.

In this presentation, we will introduce this dataset and explore both what it already tells us about Greater Melbourne's GHG emissions and what additional observations would be needed to provide rigorous estimates of the city's GHG emissions based on atmospheric techniques.

# Characterisation of $CH_4$ source signatures in Melbourne, Australia

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## Abstract

Melbourne was founded in 1835 and has grown to become Australia's second most populous city with a population of almost 5 million people [ABS, 2019]. As Melbourne's population has expanded, land use and infrastructure has changed many times and this has resulted in a variety of potential methane ($CH_4$) sources, such as legacy and new landfills, leaks from the gas distribution and sewage networks, and houses with a range of heating systems including gas and wood fires. There are also large wetland areas and coastal ecosystems. Few of these potential sources of $CH_4$ have been well characterised. Here we present the results of 500 km of mobile methane surveying throughout the south-eastern suburbs of Melbourne, during day and night time, that mapped the location of major $CH_4$ sources in July 2019. Where major plumes could be attributed to a source based on proximity or smell (for example a domestic distribution gas leak or wood burning fire) the $\delta^{13}C_CH_4$ (isotopic signature) of that source was characterised.

The methane mole fraction ($[CH_4]$) was measured using a Los Gatos Research (LGR) greenhouse gas analyser that was placed within the car. The air intake was mounted on the roof of the car approximately 2.5 m above the ground, and the air was drawn into the analyser via a Teflon tube. The $CH_4$ data were geopositioned using a Hemisphere (model A21) GPS unit and all the results were converted to kml files for displaying in Google Earth. For each major plume 10 air samples were collected in SKC FlexFoil bags and later analysed for $[CH_4]$, $\delta^{13}C_CH_4$, $[CO_2]$ and $\delta^{13}C_CO_2$ using a Picarro G2201-i cavity ring-down spectrometer (CRDS) at the UNSW Sydney greenhouse gas laboratory. To determine the plume $CH_4$ source signature the ten sample $\{[CH_4], \delta^{13}C_CH_4\}$ sets were then analysed using Bayesian regression in Miller-Tans plot [Miller and Tans, 2003].

The measured mole fractions of methane ($[CH_4]$) ranged from 1.83 ppm to 147 ppm (associated with gas leak in a residential area). Our preliminary results indicate that there is a need to further investigate the difference between bottom-up inventory emissions estimates and top-down measurements from surveys like this one.

Our research demonstrates that mobile surveys are a highly efficient way to identify and characterise unknown $CH_4$ sources in cities. Our future work aims to combine the insights from the mobile methane survey results with the permanent *in situ* methane mole fraction record and yearlong isotopic methane observations recorded at CSIRO Oceans and Atmosphere, Aspendale, to quantify the rate and duration of emissions from various sources, and to determine which sources should be prioritised for mitigation actions.

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# Top-down estimates of isoprene emissions in Australia inferred from OMI satellite data

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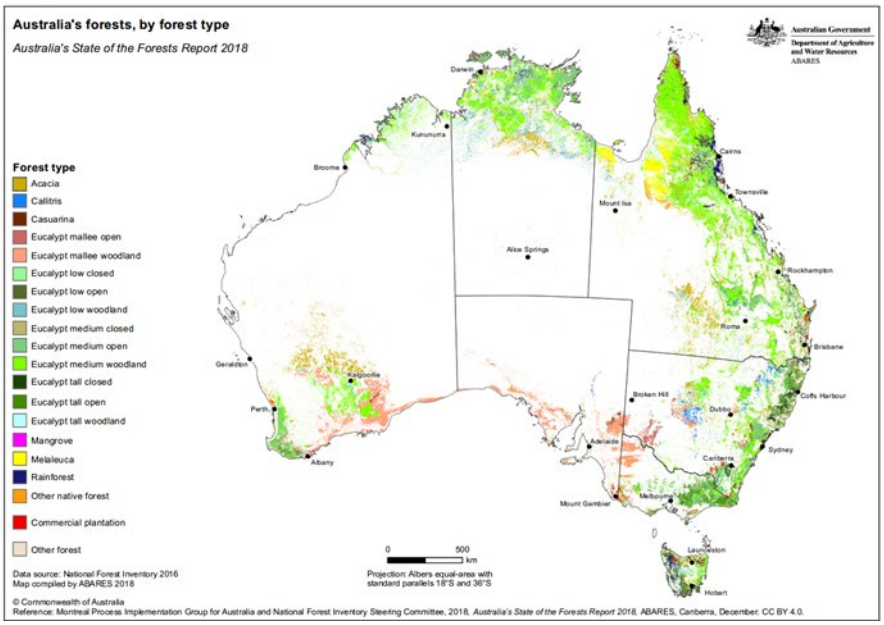
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## Abstract

Australia is a global hotspot for biogenic isoprene emission factors predicted by process-based models such as the Model of Emissions of Gases and Aerosols from Nature (MEGAN). It is also prone to increasingly frequent temperature extremes that can drive episodically high emissions. Estimates of biogenic isoprene emissions from Australia are poorly constrained, with the frequently used MEGAN model overestimating emissions by a factor of 4-6 in some areas. Evaluating MEGAN and other models in Australia is difficult due to sparse measurements of emissions and their ensuing chemical products.

In this talk I will present the results of a top-down emissions estimate performed using observations of formaldehyde (HCHO) from the OMI satellite instrument, combined with modelled isoprene to HCHO yields obtained from the GEOS-Chem chemical transport model. Where available results are evaluated using in situ observations from field campaigns conducted in southeast Australia. GEOS-Chem is also used to evaluate how updated emission estimates affect modelled ozone concentrations.

Results are summarised at a regional scale and show that the top-down isoprene emissions are lower than MEGAN estimates by half generally, but up to 80% lower during individual seasons/years. The overestimates are greatest along the eastern coast, including areas surrounding Australia's major population centres in Sydney, Melbourne, and Brisbane. Northern Australia shows different patterns of overestimation, with some caveats. The impacts of modelled overestimates of isoprene emissions translate to approximately 2-5% of the modelled ozone budget.



# New measurements give insight into Australian isoprene emissions in 2050

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## Abstract

Isoprene is the most abundant biogenic volatile organic compound emitted to the atmosphere. Isoprene is highly reactive, forming ozone and secondary organic aerosol which can cause air quality problems for cities surrounded by forest ecosystems. Previous attempts to reconcile model estimates with observations in Sydney have shown large model biases [Emmerson *et al*., 2016, 2018, 2019], which we seek to correct through the use of new chamber measurements on four eucalypt species common to NSW. The eucalypts have been grown under two different treatment regimes, the first to represent current day summer conditions and the second under conditions predicted for 2050 assuming the RCP 8.5 climate change scenario. This amounts to the trees experiencing a +3.5 °C average temperature. Isoprene emissions have been measured from both treatment regimens and compared with those predicted by the Model of Emissions of Gases and Aerosols from Nature [Guenther *et al*., 2012]. We have then adjusted the model to represent these new measurements, comparing the present day and future emissions of isoprene across Australia. The results represent the changes due to the eucalypt response to climate change alone (i.e. land use and meteorology have not changed).

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# The COALA-JOEYS Campaign: Characterising Organics And Aerosol Loading in Australia- Joint Organic Emissions Year-round Study

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## Abstract

The Charaterising Organics and Aerosol Loading in Australia (COALA) campaign, originally comprised three complementary components. It is now unclear whether or not the US component (COALA-BAIR) and the UK component (TALENT) will go ahead. Nevertheless, the Australian component (COALA-JOEYS) has been ongoing with measurements at the ANSTO, Lucas Heights campus that started in February 2019. An intensive phase of the campaign will go ahead this Austral summer at Cataract Scout Park near Sydney, with campaign dates of 16 January – 16 March 2020.

In addition to the AIRBOX instruments, there are plans to deploy a Tof-ACSM, SMPS, CCN, CN, PTRMS-TOF, SIFT-VOICE-200, aerosol filter sampling and VOC tube sampling. This presentation will outline the plans for this measurement campaign including some basic details of the instrumental techniques that will be deployed, the international groups who are planning to join and the overall aims of the campaign.

# The COALA-JOEYS Campaign: Preliminary results from the biogenic ambient air sampling system

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## Abstract

Biogenic Volatile Organic Compounds (BVOCs) are a group of carbon-based gases characterized by their high vapour pressure at ambient temperatures. Isoprene and monoterpenes together comprise most of the BVOC emissions and are the species that have been proven to be related to Secondary Organic Aerosol (SOA) and tropospheric ozone formation. The contribution of BVOC to tropospheric ozone and SOA is particularly important in low polluted areas where the BVOC emissions will drive the concentration of these pollutants. Eventhough several studies of biogenic VOC emissions have been made, most of them were in the northern hemisphere. The few studies made in the southern hemisphere are not enough to characterize and understand the effects of BVOC in local atmospheric chemistry. In an attempt to improve our knowledge of these compounds the Biogenic Ambient Air Sampling System was deployed at the ANSTO campus at Lucas Heights from February to June of 2019 as a preparation for the Characterising Organics and Aerosol Loading in Australia-Joint Organic Emissions Year-round Study (COALA-JOEYS) campaign, in order to characterize different species of BVOC in ambient air samples. The system uses a GC-MS, and the sampling method allows the capture and analysis of samples every hour. Many compounds were identified during the campaign but isoprene, BTEX and a group of monoterpenes were targeted during this period. The samples were taken through a quality check process prior to concentration analysis. This presentation will show the concentration time-series obtained from the samples and some preliminary results of daily cycles, ratios between gases and season cycles.

# The COALA-JOEYS Campaign and beyond: Thermal desorption tube measurements of volatile organic compounds

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## Abstract

Understanding of the contributions of Volatile Organic Compounds (VOCs) to ambient air quality is limited due to the complexity of their interactions in the atmosphere. Poor air quality is the fourth leading, and leading environmental, cause of premature death in humans worldwide [World Bank and IHME, 2016]. With this in mind, understanding and characterizing emissions from man-made sources in contrast with those of nature is essential in quantifying our impact on the environment. Measurements of VOCs have been conducted using thermal desorption tubes for selective capture of chemical species of interest. Well-validated procedures for this sampling technique have been described in the literature, but often see discrepancy between isolated field and laboratory studies. Thermal desorption tube manufacturers offer comprehensive guidelines for application of their instruments, though require further development to account for methodological parameters from experimental preparation up until output of results.

The characterisation of emissions from anthropogenic VOC sources using an optimised thermal desorption sampling protocol has been proposed. This study aims to understand/quantify discrepancies found between validated sampling methods using thermal desorption tubes in literature, while applying additional controls to produce valid and consistent results suitable for quantifying analytes at the ppb range. Refinement of this methodology will focus on areas explored briefly in the literature. This will include, but is not limited to, refrigerated sample preservation and storage, *in situ* drying, pre-sample tube conditioning and tube preparation, and active sample temperature and humidity controls.

Thermal desorption tube measurements will be used during the COALA-JOEYS campaign to complement continuous ambient VOC sampling. In preparation, an inter-comparison of thermal desorption tube measurements between several research groups will be performed, to compare analytical measurements of VOCs using GCMS across multiple analytical setups.

We will also discuss measurements made from a variety of potential VOC emission sites. Using the experimental parameters examined so far in this project, landfill sites and wastewater treatment ponds were sampled at the Gippsland Industrial Wastewater Treatment and Soil and Organic Recycling facilities. Results between sites are to be compared, with concentrations of benzene, toluene and xylene appearing above typical ambient levels at the industrial wastewater site but these compounds were not present at the soil recycling facility. Extension of this research would see comparisons of anthropogenic and biogenic VOCs in urban atmospheres, along with correlations of VOC concentrations to observed methane fluxes.

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# Biogenic sulfur from the South West Pacific Ocean: estimating methanethiol fluxes, and DMS measurement comparison

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## Abstract

Dimethylsulfide (DMS) is the most well-studied of the reduced sulfur gases originating from the ocean. In contrast, ocean-derived methanethiol (MeSH), or methyl mercaptan, is much less studied. Available measurements suggest MeSH has a global ocean source ~17% of the DMS source [Lee and Brimblecombe, 2016]. MeSH is short-lived and is oxidised by OH in a few hours, but the impact of MeSH and its oxidation productions on atmospheric chemistry and climate is not well understood.

Atmospheric MeSH and DMS were measured *in situ* using proton transfer reaction mass spectrometry (PTR-MS) during the Surface Ocean Aerosol Production (SOAP) voyage [Law *et al*., 2017]. MeSH levels were observed up to 65 ppt over the biologically productive subtropical front of the SW Pacific Ocean, the highest MeSH mixing ratios observed to date, and between 3-36% of parallel DMS mixing ratios [Lawson *et al*., 2019]. The nocturnal accumulation method [Marandino *et al*., 2007] was used to estimate the sea-air flux of MeSH; the resulting average flux (4.7 µmol $m^{-2}$ $day^{-1}$) was > 4 times higher than average MeSH fluxes from previous studies and is similar to maximum MeSH flux values reported in coastal/upwelling regions of the Atlantic [Kettle *et al*., 2001]. The average flux of MeSH compared to DMS (MeSH/(DMS+MeSH)) was higher in this study (20%) than other studies (4-16%) [Kettle *et al*., 2001; Leck and Rodhe, 1991; Kiene *et al*., 2017]; this suggests MeSH plays an important role in the transfer of sulfur to the atmosphere in this region.

Finally, an atmospheric DMS method measurement comparison was carried out at sea using three independently calibrated instruments – the PTR-MS (CSIRO), Gas Chromatograph with Sulfur Chemiluminescence Detector (GC-SCD; NIWA) and Atmospheric Pressure Ionisation-Chemical Ionisation Mass Spectrometer (mesoCIMS; UCI). Comparison of ambient levels of DMS in tedlar bags showed good agreement between PTR-MS and GC-SCD, with the mesoCIMS ~15-25% higher. In ambient air, DMS mixing ratios measured by mesoCIMS (inlet height 12 m) were ~25% higher than PTR-MS (inlet height 28 m). Smith *et al*., [2018] showed significant atmospheric vertical gradients of DMS were expected during the voyage, particularly during stable conditions with strong DMS fluxes. Correcting for the expected difference in DMS mixing ratios at the two intake heights improved the agreement between PTR-MS and mesoCIMS DMS measurements.

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# Recent long-term measurements of aerosol optical properties in the Northern Territory

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## Abstract

The Northern Territory Baseline Air Pollution Station (NTBAPS) is a CSIRO facility on the Bureau of Meteorology's Gunn Point Research Weather Radar site at Gunn Pt, NT. The NTBAPS has been in operation since 2010, during which time there have been measurements of meteorology, aerosols, greenhouse gases, reactive gases, radon, and total gaseous mercury.

Aerosol measurements at the NTBAPS are made with a three-wavelength integrating nephelometer and a multi angle absorption photometer. Continuous measurements throughout the northern Australian monsoon season are facilitated via a unique continuous automatic aerosol diffusion dryer custom built to overcome the challenge of accurate aerosol measurements in high humidity atmospheres.

Data from recent multi-year measurements of aerosol scattering coefficient and absorption coefficient will be presented. Diurnal cycles and seasonal trends in the aerosol optical properties of the region based will be examined. Impacts from biomass burning on aerosol optical properties and composition will be discussed.

# Exploring Greenhouse Gas Seasonality in the Top end: a collaboration between the Larrakia Rangers and CSIRO

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## Abstract

Tropical ecosystems store vast carbon pools, that are potentially highly vulnerable under the climate change scenarios likely to play out over the coming century. For example, methane production from wetlands is highly temperature sensitive, and prolonged dry spells may result in hotter and more extensive biomass burning during the dry season. Both mechanisms could displace long-stable land surface carbon pools into atmospheric greenhouse gases, acting as a positive feedback on the climate system. Despite this, there are few high-quality greenhouse gas observations in tropical regions.

Since 2018, CSIRO has renewed its atmospheric composition measurements at the Northern Territory Baseline Air Pollution Station (NT BAPS) at Gunn Point, through engagement with the Larrakia Rangers. The Rangers conduct monthly flask sampling for CSIRO's Global Flask Network and undertake routine site visits to support the *in situ* measurements of carbon dioxide, methane and carbon monoxide.

The collaboration with the rangers also includes a joint exploration of the observed seasonality in greenhouse gas mixing ratios. Drawing on work previously co-produced between CSIRO and Larrakia Elders, led by Lorraine Williams [Williams *et al*., 2012], the aim is to see how the observed seasonality corresponds to the Indigenous understanding of seasons on Larrakia country. This talk will give an update on the greenhouse gas program at NT BAPS and present some preliminary exploration of greenhouse gas seasonality in the Top End.

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# The Total Carbon Column Observing Network – update and future directions

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## Abstract

The Total Carbon Column Observing Network (TCCON) was established in 2004, and continues to expand, reaching 27 sites around the world, including two in Australia. TCCON provides high precision and accuracy ground-based total column measurements of $CO_2$, $CH_4$, $N_2O$, CO, $H_2O$ and HF. These measurements serve three purposes:

1. a *de* facto *gold-standard* for validation of satellite greenhouse gas measurements;
2. independent validation for model simulations, including inversions based on satellite or *in situ* data
3. improving our understanding of processes affecting the composition of the Earth's atmosphere.

As satellite-based instruments and retrievals improve, the demands on the TCCON as a validation resource increase, especially ensuring that inter-site biases are minimised. TCCON has made several recent developments to improve both the accuracy and the consistency between sites. These will be incorporated in a major upcoming new data release.

In this presentation we highlight recent developments and applications of TCCON and its data and discuss some future directions of the network, focussing on the Australian sites.

# Novel ground-based remote sensing of greenhouse gases in the Arctic, tropics and subtropics

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## Abstract

In a project funded by the European Space Agency (ESA) towards validation of the Sentinel-5P satellite, the University of Wollongong (UoW) is part of a team assessing the ability of several, low-cost, portable ground-based infrared spectrometers to measure total column amounts of the greenhouse gases $CO_2$ and $CH_4$. The aim of this campaign is to provide a suite of independent, fully characterised, and traceable ground-based total column measurements from a variety of instruments. The Centre for Atmospheric Chemistry at UoW has continued its involvement since the project began in Finland in 2017 with an extension to include Australia this year.

From March 2017 to the end of 2018, UoW participated in the FRM4GHG campaign at the Sodankylä (Finland) Total Column Carbon Observing Network (TCCON) site by deploying its Bruker IRCube FTIR. During 2019, as an extension to the Finish campaign, the IRCube was run alongside the TCCON instrument at UoW and then deployed to the tropical TCCON site in Darwin. The IRCube novelty concerns the use of a fibre-optic cable that feeds sunlight from an external tracker to the spectrometer, a method of optical feed not normally applied to this type of spectrometer. Using standard TCCON processing procedures, column averaged mole fractions of $CO_2$ and $CH_4$ were computed. These data have been compared with other collocated small portable instrument from our European participating partners. Following on from these 2017/2018 measurements, the extended campaign in Australia tested the portability of the IRCube to different locations to test its robustness and any potential biases that may be present in very different geolocations (Arctic, mid-latitude and tropical) and in the instrument performance itself. These datasets will provide important comparative validation with respect to the standard TCCON spectrometer for precision and stability, while giving insight into the use of small low-cost potable spectrometers as a supporting network for TCCON.

This presentation will give an overview of the FRM4GHG project and report results from 2017 and 2018, and report on preliminary data for the 2019 Wollongong and Darwin campaigns.

## Reference

FR4GHG, Fiducial Reference Measurements for Greenhouse Gases, http://frm4ghg.aeronomie.be/

# The GipNET measurement network – A potential measurement network for proposed carbon capture and storage

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## Abstract

Carbon capture and storage (CCS) is proposed as a solution to attempt to limit the quantity of carbon dioxide ($CO_2$) entering the atmosphere, especially from energy. The work here describes the GipNet atmospheric monitoring network established to assess a potential atmospheric assurance system for a proposed CCS location in the near-shore Gippsland region of Victoria, Australia. The network consisted of four measurement sites, and a regional scale model. In this talk we focus on the measurement network, while modelling work will be presented in a companion paper.

The near-shore environment presents challenges for atmospheric leak detection due to logistical difficulties in measuring near the injection site. One potential solution is to employ an open-path measurement technique to increase the potential of sampling an emitted plume. The project allowed development and testing of an open-path near infrared (OP-NIR) spectrometer.

We describe the setup of the instrument and its deployment during field trials. The spectrometer was tested over multiple pathlengths up to 1500m. At 1500m, the system achieves repeatabilities of better than 0.1% for $CO_2$ and $CH_4$. Comparison to in situ measurements under well-mixed conditions shows a bias of +2% relative to WMO scales. The instrument has now been deployed to Gippsland.

The GipNet measurement network also consisted of two Picarro G2301 analysers, located at Churchill and Yarram, and a Spectronus™ Trace Gas Analyser at Dutson Downs. We will also present data acquired by these instruments since their deployment to Gippsland.

# Inverse modelling of $CO_2$ for offshore leak detection

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## Abstract

Carbon capture and storage (CCS), or carbon sequestration, is one technology among many that have been proposed to mitigate emissions of greenhouse gases from electricity production. For CCS to be effective in its goal, the carbon dioxide ($CO_2$) that would be injected into the ground must not leak out, at least not at rates exceeding 1% per year. Thus, monitoring for leakage at CCS sites should be undertaken if this technology is to be adopted. This study considers the case of a proposed CCS site off the coast of Gippsland, in south-eastern Victoria. In this trial, the $CO_2$ would be sequestered in a disused gas well under the sea, roughly 10 km off the coast of the Australian mainland. Unlike a trial carbon sequestration injection that took place in the Otways (Victoria) that began in 2008, the offshore nature of the injection site makes nearby monitoring very challenging. However, measurements of the geological subsurface, the nearby sea-water and the air at on-shore sites is in place to first characterise the natural variability, and later to monitor for leakage.

In this work, we use atmospheric transport-dispersion modelling and inverse modelling techniques to assess whether the on-shore monitoring can potentially detect any major leakage of $CO_2$ to the air. Atmospheric dynamics are estimated via the Weather Research and Forecasting (WRF) model, and a tracer-only version of the Community Multi-scale Air Quality (CMAQ) model and its adjoint are used to simulate atmospheric transport. The $CO_2$ flux prior incorporates contributions from the terrestrial biosphere (using the Vegetation Photosynthesis Respiration Model, or VPRM), anthropogenic emissions (based on the Open-source Data Inventory for Anthropogenic $CO_2$, or ODIAC) and fire emissions (from the Global Fire Assimilation System, or GFAS). A newly-developed general-purpose inverse-modelling toolkit (py4dvar) was used for the flux inversions. The modelling results were either compared with or used (i.e., assimilated) $CO_2$ mixing ratios measured by a network of in-situ and open-path instruments that were run as part of this project.

Four groups of $CO_2$ simulations were undertaken: forward simulations, observing system simulation experiments, signal detection experiments, and assimilation of data collected within this project. The forward model matched the measured $CO_2$ reasonably well (in line with similar modelling studies). The observing system simulation experiments suggested that the flux inversion resulted in a moderate reduction in uncertainties at the injection site. The signal detection tests showed that the system was able to detect a likely leak, although with limited precision as to its magnitude. However, the inversions involving assimilation of the observations made in this project suggested that there would be a very high false-positive rate, and that further work may be required to solve for variables not constrained by these assimilations (in particular, boundary conditions).

This work suggests that even with state-of-the-art observations, atmospheric modelling and flux-inversion, the distance between the location of a potential leak and the nearest possible monitoring site is too large to be a reliable detection mechanism. More proximate monitoring, in particular with unmanned aerial vehicles (drones), may be more sensitive for the detection of leaks.

# Using hemispheric contrasts in satellite-derived cloud microphysical properties to constrain aerosol forcing

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## Abstract

Not available – Presentation via weblink from the SOAR workshop – Hobart, Tasmania

# Modelling marine organic contributions to aerosol

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## Abstract

Not available – Presentation via weblink from the SOAR workshop – Hobart, Tasmania

# Model-observation comparison of Southern Ocean aerosols in CMIP6 ACCESS

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## Abstract

Not available – Presentation via weblink from the SOAR workshop – Hobart, Tasmania

# Southern Ocean CCN – temporal and spatial distributions

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## Abstract

Being so far from anthropogenic activity, the Southern Ocean region is a unique environment that experiences the most pristine atmosphere in the world. This makes it a perfect testbed for understanding pre-industrial conditions that are so vital for reducing the uncertainties inherent in global climate and earth system models today [Carslaw *et al*., 2013]. Clouds are of particular interest in this region because of the large biases in shortwave radiation and sea surface temperatures that are present in most models. However, due to its remoteness and wild environment, the Southern Ocean is one of the most under-sampled regions of the Earth's atmosphere, making high quality observational data from the region invaluable.

The past few years have seen a relative abundance of atmospheric measurements in the region, with multiple ship-based campaigns aboard the *RV Investigator* and the *Aurora Australis*, an aircraft campaign, as well as a two-year intensive measurement campaign at Australia's sub-Antarctic research station at Macquarie Island (54.6°S, 158.9°E). Amongst the suite of instrumentation deployed are those for the measurements of condensation nuclei ($CN_{10}$) and cloud condensation nuclei (CCN) which are vital components in the formation of clouds and determining their properties. In this presentation, a number of these datasets will be explored in relation to the temporal and spatial variability.

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# AIRBOX observations made on the *RSV Aurora Australis* over the 2018/2019 resupply season

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## Abstract

We present an overview of the motivating science questions for atmospheric chemistry over the Southern Ocean and Antarctic sea-ice zones. The AIRBOX consortium conducted aerosol and atmospheric composition measurements made over the 2018/2019 resupply voyages on the *RSV Aurora Australis* from October 2018 to March 2019 representing four return voyages from Hobart to the Australian Antarctic bases. Mercury, radon, greenhouse gases, ozone, aerosol size, number, scattering, absorption, and composition properties were observed. Additionally, eddy covariance flux and sea-state conditions were monitored. We show initial results and interpretations from what promises to be an extremely valuable dataset in this sparsely monitored region of the Earth.

# Decade-long aerosol scattering coefficient record from the global baseline atmospheric pollution station at Cape Grim, Australia

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## Abstract

Marine aerosol is globally one of the most important natural aerosol systems, contributing to the Earth's radiative budget, biogeochemical cycling, impacts on ecosystems and regional air quality. In addition, the Southern Ocean aerosol is most likely the closest representation of natural aerosol on the globe due to relatively minimal impact of anthropogenic sources.

The contribution of atmospheric aerosol to global climate forcing is significant but highly uncertain (IPCC 2013). Both direct and indirect aerosol effects have been identified as significant, and globally, indirect effects through aerosol modification of cloud properties contribute around 60% of the aerosol effect. Aerosols interact directly with solar radiation by absorbing and scattering light. The amount of scattering and absorption is a function of particle size, composition, and shape, as well as external variables like relative humidity and wavelength of light.

The measurement of aerosol scattering coefficient has been made at Cape Grim since 2006. In this presentation we describe the measurement system used to collect this record, the quality assurance and control procedures and corrections used to ensure high quality data and an estimate of the uncertainty of the data. We present the time series and a brief analysis of the climatology of the aerosol scattering record both under baseline and non-baseline condition.

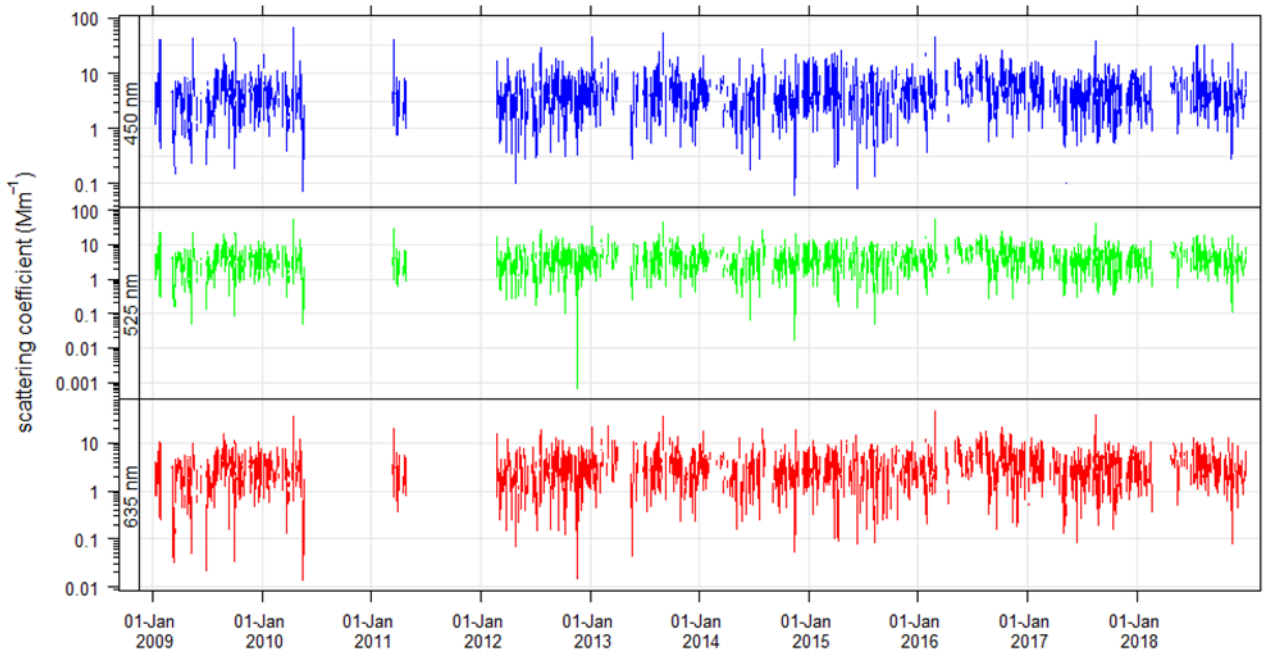



**Figure 1.** Time series of scattering coefficient at three wavelengths for baseline criteria of radon concentration less than 100 Bq m$^{-3}$ and wind direction between 190 and 280 degrees.

# Radon-222 measurements at the Gunn Point Northern Territory Baseline Air Pollution Station (BAPS)

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## Abstract

Global 'baseline' stations specialise in monitoring long-term regional or hemispheric-mean changes in anthropogenic emissions that have the potential to alter climate or adversely affect the environment. This ability is contingent upon reliably identifying well-mixed air masses that have been removed from localised pollution sources for a long time. Thanks largely to a unique collection of physical characteristics, radon provides a convenient, consistent, effective alternative to conventional meteorological approaches for baseline air mass identification [Chambers *et al*., 2016]. Furthermore, under 'non-baseline' conditions (particularly periods of extended terrestrial fetch), radon proves to be a valuable tool for classifying the near-surface atmospheric mixing state [e.g. Chambers *et al*., 2019; Chambers and Podstawczyńska, 2019]. This poster provides a brief introduction to the ongoing radon ($^{222}$Rn) measurement program at the Gunn Point Baseline Air Pollution Station site, 36 km northeast of Darwin. Radon measurements at this site are being conducted using a 700 L, dual-flow-loop, two-filter detector designed and built at ANSTO [e.g. Griffiths *et al*., 2016]. The seasonal radon cycle at Gunn Point is characterised by a wet-season (September – March) minimum, during which radon can be employed as a baseline selection tool, and a dry-season (April-August) maximum, during which radon observations can be used to elucidate surface-atmosphere exchange processes of key atmospheric constituents and pollutants [Howard *et al*., 2017].

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# ACCESS-CM2-Chem

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## Abstract

ACCESS-CM2-Chem is the latest iteration of chemistry-climate model for the Australian community. The model combines the Community Atmosphere–Biosphere Land Exchange (CABLE) model and the MetOffice's Unified Model (UM) atmosphere, which includes the United Kingdom Chemistry and Aerosol (UKCA) module. We run UKCA in its full 'StratTrop' configuration, comprising 81 chemically active species and 299 different reactions. Here we detail some recent developments to the chemistry scheme and the subsequent improvements to the simulation of ozone relative to the prior model generation (ACCESS-CCM). We also outline plans for model runs to be conducted in 2020 which will contribute to the Chemistry Climate Model Initiative (CCMI) phase 2 activities in support of the 2022 WMO Ozone Assessment.

# Towards tracking east Asian transport of pollution using the Burgos TCCON site and the GOSAT series satellites

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## Abstract

The Burgos TCCON station, which started operations in March 2017, has been validated using aircraft profiles within the EMeRGe mission, providing the last step towards a 'full TCCON' site status. In this new project, we plan to combine existing resources at the Burgos TCCON site in the Philippines and nearby sondes from Laoag airport with GOSAT and GOSAT-2 data to measure, monitor and understand the formation and transport of pollutants from East and Southeast Asia. As a first step, we aim to analyze measurements taken at the Burgos TCCON site and investigate the correlation of aerosols properties, water vapor and thin clouds on the column of trace gases measured from TCCON. We will combine this analysis with data from an aircraft and ship campaign (NASA-CAMP2Ex), undertaken in August to October 2019. As a second step, we plan to compare GOSAT-2 retrievals over Ocean of $XH_2O$, XCO, $XCO_2$, and $XCH_4$ with the ground-based measurements. In the third step, we will utilize a model to analyze the transport and address the question of how anthropogenic emissions from East Asian countries are formed and transported to the Tropical Western Pacific (TWP).

# ACCESS-Fire modelled weather case studies

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## Abstract

Fires are a damaging and wide-ranging seasonal occurrence for much of Australia, mitigated in part by national and state-based fire-fighting agencies and volunteers. A major factor in the effectiveness of their efforts is the fire danger index that attempts to predict where the worst fires may occur. One issue with the current system is that it does not consider atmospheric processes that are affected by local topography (such as down-slope wind flows) or wind and weather feedback (such as cumulonimbus driven wind gusts) [Peace, 2016].

This project has two intents: first that these processes can be adequately modelled by ACCESS when coupled to a fire model, and second to provide information on fire-weather feedbacks. Simulating how a fire changes local meteorological flows also enables enhanced forecasting of fire impacts and coverage [Toivanen, 2019].

My poster will detail progress so far in the analysis of ACCESS-Fire coupled outputs over two case studies (Bush fires at Waroona in January 2016, and Sir Ivan in February 2017), highlighting both meteorological features seen in the model and some model sensitivities.



**Source: R. Rampling (9 News Perth)**

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# Calibration of the Burgos TCCON site in the Philippines during EMeRGe

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## Abstract

The Total Carbon Column Observing Network (TCCON) is a network dedicated to the precise remote sensing measurement of the column-averaged dry-air mole fractions (DMF) of $CO_2$, CO, $CH_4$, $N_2O$ and $H_2O$. Column-averaged DMF of gases (denoted as XG for gas G, e.g. $XCO_2$) can be used in combination with in situ measurements to disentangle the effects of atmospheric mixing and surface exchange of atmospheric gases, providing a better understanding of the regional-scale fluxes. However, the accuracy of TCCON data products relies on the knowledge of spectroscopic parameters of the target and potential interfering trace gases and radiative transfer. These are improving but not necessarily known with sufficient accuracy. TCCON measurements of XG, such as $XCO_2$, must therefore be compared to in situ trace gas measurement scales. Here, we present validation results of the Burgos TCCON data using instrumentation aboard the HALO aircraft (www.halo.dlr.de) that measured profiles during its transfer flight in the scope of the EMeRGe mission in Asia (www.iup.uni-bremen.de/emerge). Measurements of $XCO_2$, $XCH_4$ and XCO with a portable spectrometer (Bruker EM27/SUN) were also made, coincident with TCCON and the HALO EMeRGe flight. This represents a unique demonstration opportunity and experiment.

# Investigation of drivers of variability in atmospheric carbon monoxide over Australia using column measurements

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## Abstract

There remains much uncertainty in estimating and characterizing the Southern Hemisphere Atmospheric composition compared to the better explored Northern Hemisphere. This poses significant challenges for analysing and modelling short-lived tropospheric trace gases, especially carbon monoxide (CO). Due to limited influence from anthropogenic emissions compared to Northern Hemisphere, sites studied in the southern hemisphere show that CO tends to be affected by a balance of long-range transport, secondary organic production, and removal via its primary sink, OH. This study will be particularly focussed on using a step-change in measurement density due to measurements from the newly launched Greenhouse Gases Observing Satellite-2 (GOSAT-2) combined with ground-based measurements from the Total Carbon Column Observing Network (TCCON) to investigate the variability of CO and the role of climate drivers in the Southern Hemisphere, especially the Australian region.

GOSAT-2 will measure column-averaged dry-air mole fractions of CO (XCO) with global coverage. To use these measurements to better understand of the seasonal cycle and variability of CO requires that the satellite measurements are validated relative to reliable, accurate ground truth. This study will provide comparison, validation and bias correction of GOSAT-2 XCO retrievals with respect to the ground-based TCCON site measurements, focussing on Darwin and Wollongong.

While the validated satellite retrievals of $X_{CO}$ along with the ground-based data will act as a foundation for this study, GOSAT-2 $X_{CO}$ retrievals are not yet available, so to date we focus on using the 12-year TCCON measurement record from the Wollongong site, and investigate $X_{CO}$ anomalies in this time series. We identify $X_{CO}$ anomalies from the Wollongong TCCON data record, and the results indicated significant enhancements of other related atmospheric constituents also measured by TCCON, including $CO_2$, and $CH_4$; these correlated enhancements are used to identify the influence from different possible sources. In addition, we use the combination of $X_{CO}$ anomalies with Aerosol Optical Depth (AOD) measurements from the site to provide a further method of identifying and characterizing the past anomalous CO occurrences.

Combining the TCCON ground-based data with the spatial coverage of GOSAT-2 will allow a detailed investigation on the seasonality, anomalies and temporal variability of CO with a particular emphasis of its possible sources from direct emissions and long-range transport in Australia. The influence of Climate drivers, El Nino Southern Oscillation (ENSO) and Indian Ocean Dipole (IOD) on the variability of CO and other trace gases in Australia will be investigated. We plan to test the conceptual understanding of the processes influencing CO in the southern hemisphere by comparing the measurements to model simulations using a chemical transport model.

# Satellite constraints on anthropogenic emissions of air pollutants in Australia

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## Abstract

Nitrogen dioxide ($NO_2$) in the troposphere has multiple ramifications for human health and the environment, especially through ozone and aerosol formation. The spatial and temporal understanding of nitrogen oxide ($NO_x$) emissions in Australia is limited. The use of satellite-derived $NO_2$ vertical column densities (VCDs) as a proxy for surface level $NO_x$ emissions has significantly contributed to improving the understanding of $NO_x$ worldwide. Satellite data features heavily in research of regions with the highest levels of $NO_x$ pollution including east China, northeast USA and Europe. Australia has been mentioned in global studies however, these have offered limited interpretation of Australian trends or development of a deeper understanding.

In this presentation, $NO_2$ in Australian urban areas is studied using a previously developed merged GOME-SCIAMACHY-GOME2ab dataset [Georgoulias *et al*., 2019]. Gridded satellite data ($0.25^o$ x $0.25^o$) and emissions inventories (CEDS, EDGARv4.3.2 and the 2008 OEH Air Emissions Inventory for the GMR in NSW) are collected and processed in python. CEDS and EDGAR are both bottom-up emissions inventories with global coverage. In the absence of a national or regional inventory for most of Australia, these are used to compare to the satellite dataset. The aim is to determine if major hotspots can be identified in satellite data and to assess the consistency between satellite and inventory derived $NO_x$ distributions. The greater temporal coverage of the satellite dataset is then used to assess interannual variability and trends in NOx emissions. Results to date have identified the major urban $NO_2$ hotspots and the spatial extent of elevated $NO_2$ at each. Comparison of the satellite and OEH inventory also shows good agreement in terms of elevated $NO_2$ VCDs and $NO_x$ emissions over a major power plant near Singleton, NSW. These initial results suggest there is more information about $NO_2$ emissions in Australia that can be drawn from the merged satellite dataset than has been published previously. Ongoing analysis aims to identify additional NO2 point sources and quantify the trends in $NO_2$ VCDs. These will be applied to inventories to derive model-ready emissions estimates of Australian NOx emissions.

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# Characteristics of the baseline Cape Grim ozone record, 1982 - 2017

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## Abstract

Ozone is a key component of the atmosphere, acting as a precursor in the formation of the hydroxyl radical, is a greenhouse gas, participates in many important atmospheric photo-oxidation reactions and is a secondary air pollutant. Given its highly reactive nature, there is considerable spatial and temporal variation in ozone concentrations across the globe. Monitoring of ozone concentrations across the Southern Hemisphere is very spare compared with the North, and the Southern Ocean region even more so. Continuous measurements of ozone mixing ratios in the marine boundary layer have been, and continue to be, collected at Cape Grim since 1982 through to the present. Under certain meteorological conditions, the air being sampled at Cape Grim is classed as baseline and is representative of air masses in the remote marine boundary layer in the Southern Ocean, free from the effects of local and regional sources and sinks of ozone.

In January 2019 the Cape Grim ozone measurement record from 1982 – 2017 (inclusive) was finalised and submitted to the World Data Centre for Reactive Gases (WDCRG). This included extensive quality assurance checks, corrections and incorporation of calibration data. The resulting data is fully traceable to BiPM standards and was subsequently also submitted to the TOAR database [Schultz, *et al*., 2017].

This 36-year record has since been filtered to produce a baseline flagged version using criteria defined in Molloy and Galbally [2014]. Radon concentrations are the primary component of the baseline identification criterion, supplemented by wind speed and direction when radon data is unavailable. An additional criterion is applied to the data set as a secondary filter based on the standard deviations and is intended to remove any additional non-baseline data points which may have been missed by the earlier filtering.

Statistics and characteristics, including diurnal and seasonal cycles, long-term trends and frequency distributions of the baseline data are compared with the complete, unfiltered data and the non-baseline data sets.

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# Ship-based measurements of BrO over the Southern Ocean

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## Abstract

The Southern Ocean is one of the most remote regions on Earth. However, previous studies have highlighted mercury contamination in the Southern Ocean and identified its biomagnification in marine food webs. Mercury is mainly emitted to the atmosphere as volatile gaseous elemental mercury ($Hg^0$). The deposition of mercury to ecosystems is largely controlled by the oxidation of $Hg^0$ to the water-soluble gaseous oxidised mercury ($Hg^{II}$). Research suggests that bromine is the dominant atmospheric oxidant for $Hg^0$. A lack of observational constraints on the distribution of bromine oxide (BrO) over the Southern Ocean is a major contributor to the uncertainty in mercury cycling in the region. Further uncertainty in this region can be attributed to the presence of sea ice that affects BrO concentrations and mercury exchanges between the ocean and atmosphere.

Bromine chemistry is especially active in the polar regions with both satellite and ship-based measurements observing high concentrations of BrO above the sea ice margins and the occurrence of bromine explosions shortly after polar sunrise. Connections have been made between elevated bromine concentrations and the subsequent depletion of both ozone and gaseous elemental mercury ($Hg^0$).

Presented here are Multi Axis-Differential Optical Absorption Spectroscopy (MAX-DOAS) observations of BrO carried out on board the Australian icebreaker Aurora Australis as part of the Chemical and Mesoscale Mechanisms of polar Cell Aerosol Nucleation (CAMMPCAN) campaign between October 2017 and March 2019. The campaign involved 6 research voyages across two seasons that traversed the Southern Ocean from Australia to three Antarctic locations: Davis, Casey and Mawson.

Our preliminary results indicate that that there is a lack of variability in BrO concentrations between Antarctic locations and across seasons. Baseline BrO concentrations were 4.99 pptv and 4.92 pptv for the 2017-18 and 2018-19 seasons respectively. The relationship between BrO and sea ice cover will also be discussed. Uncertainties remain in the distribution of BrO and Hg over the Southern Ocean and the ocean-atmosphere exchange of Hg, and these are the focus of ongoing and future work.

# Bridging the gap between atmospheric models and observations: A sensitivity analysis of methacrolein photochemistry

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## Abstract

Biogenic sources contribute to about 90% of the VOCs found in the atmosphere, with isoprene being the most abundant non-methane VOC [Berndt *et al*., 2019; Wennberg *et al*., 2018). The main carbonyl products from the gas-phase atmospheric oxidation of isoprene are methyl vinyl ketone and methacrolein [Liu *et al*., 2012]. Processes like photodegradation of the latter and its pathways have been relatively poorly studied [So *et al*., 2018]. For this, the UNSW Sydney Laser group of the School of Chemistry has been working on researching photolysis of methacrolein, finding new mechanisms and quantum yields [Jacob, 2019]. However, in order to test the relevance of such new mechanism and its impacts on other airborne species and atmospheric processes, an additional tool is required. Atmospheric chemical transport models, like GEOS-Chem, a widely used 3-D model with full chemistry and meteorology, are of use. Our project seeks to modify the current photolysis mechanism and quantum yields of methacrolein within GEOS-Chem, with the ones experimentally derived by Jacob [2019]. We test the impact of the newly found pathways on simulation results and concentrations of methacrolein, as well as products like $H_2$, $HO_2$, and $CH_2O$. This is achieved by performing sensitivity analyses using the original configuration of GEOS-Chem as the baseline, against model results after changing the quantum yields. The results we show here comprise the first stage of the project. Further tests will include wavelength dependent methacrolein quantum yields, as to date GEOS-Chem has fixed values across wavelength bins. By doing so, we want to close the known gap between the model and observations. Ultimately, we expect to provide a more refined tropospheric chemistry into the model by linking active modelling with experimental photochemistry data. Thus, having a two-way feedback, where the model helps drive experimental work, and the latter aids the model to be more precise.

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# Evaluating WRF-Chem against the 2016 R2R field campaign

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## Abstract

The Reef to Rainforest (R2R) field campaign, carried out over September and October of 2016, aimed to characterise marine aerosol over the Great Barrier Reef (GBR) and determine if a local source of aerosol, derived from dimethyl sulfide (DMS) produced by phytoplankton and coral reefs, is climatically relevant. This campaign involved concurrent observations taken aboard the RV Investigator circumnavigating the GBR and the land based atmospheric composition laboratory AIRBOX, located south of Cairns at Mission Beach. In this study, we use the observations taken during this extensive field campaign to evaluate the Weather Research Forecast – Chemistry (WRF-Chem) coupled regional model. The model outputs hourly timesteps and consists of three nested domains covering the Australian continent and Coral Sea (at 27 km resolution), the state of Queensland and GBR (9km) and an area at the southern end of the GBR at which the RVI was stationed for a period of time (3 km). To ensure consistency with observations, the model is nudged to the Bureau of Meteorology's Atmospheric high-resolution Regional Reanalysis for Australia. Initial results indicate that the model performs reasonably well against the ship meteorological fields (although this is unsurprising, given they are incorporated in the reanalysis product), and less well against the AIRBOX meteorological fields. After reducing the model's surface water DMS concentrations by more than half to match the average ship measurements, atmospheric DMS concentrations and sulfate aerosol mass are captured well by the model. This is likely due to strong performance of the wind fields in the model. Other aerosol fields ($NH_4$, $NO_3$, black carbon, organics) are generally underestimated, while chloride is overestimated. The number concentration of particles greater than 3nm in diameter ($N_3$) is overestimated, as is the cloud condensation nuclei (CCN) concentration. These overestimations were at first thought to be a result of the sea salt flux being too high in the model, which was subsequently halved. However, halving this flux made little difference to the $N_3$ and CCN suggesting that the overestimate is a result of the initial and boundary conditions for the region. In particular, we suggest that continental air masses are a key cause of higher model values of $N_3$ and CCN. WRF-Chem will later be used to further examine the question of whether DMS produced by GBR marine environment is climatically important, and if so, examine the impact of mass coral extinction for Queensland's climate.

# Analysing trends in the climate-carbon cycle feedbacks and its drivers from observations

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## Abstract

Emissions of carbon dioxide ($CO_2$) into the atmosphere are taken up by the biosphere and hydrosphere, regarded as carbon sinks or 'uptake'. Two main feedback processes within the carbon cycle control the rate of carbon uptake: 1. The increase in $CO_2$ concentrations enhances uptake, therefore the perturbed increase of concentrations is damped (β), and 2. The rise in the global mean surface temperature weakens uptake, therefore amplifying the perturbed increase in concentrations (γ). Quantifying β and γ on long-term timescales has been difficult to constrain, especially from $C^4$MIP and CMIP5 modelled results [Friedlingstein *et al*., 2006, Arora *et al*., 2013]. This project aims to analyse changes in the long-term trends of carbon uptake from the past and diagnose its response to the feedbacks β and γ. We will use observations from the Global Carbon Project (GCP, from 1959 to 2017) [Le Quéré *et al*., 2018], as well as six sets of atmospheric inversion model output (collectively ranging from 1976 to 2017) as observations. So far, linear regression analysis of GCP land and ocean uptake has shown that the gradient of consecutive 10-year trends from 1959-2017 is slowly decreasing. This indicates a deceleration of carbon uptake over the long-term. These results are currently being analysed for robustness. A deceleration of carbon uptake could be a response to a slowing of β and/or a strengthening of γ over the last six decades.

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# Was Australia a sink or source of $CO_2$ in 2015? Data assimilation using OCO-2 satellite data

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## Abstract

Estimates of the net $CO_2$ flux at a continental scale are essential to building up confidence in the global carbon budget. In this study, we present the assimilation of the satellite data from the Orbiting Carbon Observatory-2 (OCO-2) to estimate the Australia $CO_2$ surface fluxes for 2015. We used the Community Multiscale Air Quality (CMAQ) model and a four-dimensional variational scheme. Our preliminary results suggest that Australia was a slight carbon sink during 2015 of about -0.15 ± 0.11 PgC/y. To understand the seasonal cycle of our monthly $CO_2$ fluxes, we aggregated them into six regions over Australia using Land Cover Type Product the Moderate Resolution Imaging Spectroradiometer (MODIS). We also divided them into two areas (north and south). In southern Australia, the time series indicate the uptake of $CO_2$ occurs mainly in July over grasses and cereal, whereas in the northern, this uptake occurs from January - February. Analysis of the overall inversion performance shows that most of the biases on the model improve in both northern and southern Australia, except over land category, which relies upon a few OCO-2 measurements.

# Improving the lightning-generated $NO_x$ in ACCESS-UKCA, and its impact on tropospheric ozone

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## Abstract

Oxides of nitrogen ($NO_x \equiv NO + NO_2$) play an important role in tropospheric chemistry by influencing the production of ozone ($O_3$) and hydroxyl radical (OH), the main tropospheric oxidants. $O_3$ is a greenhouse gas and is most potent in the upper troposphere, whereas it is an air pollutant near the earth's surface. An important natural source of tropospheric $NO_x$ is lightning. Although lightning accounts for only about 10% (2–8 Tg N $yr^{-1}$) of the global $NO_x$ source, it is the most dominant source in the middle to upper troposphere [Schumann and Huntrieser, 2007]. Lightning $NO_x$ ($LNO_x$) also has a disproportionately large contribution to the tropospheric burdens of $O_3$ and OH. Using a climate-chemistry model, Grewe *et al*. [2007] found that $LNO_x$ is the dominant $NO_x$ source for tropospheric ozone (up to 40%) in the tropics and Southern Hemisphere. Of any major emission source, variability in global mean OH is most sensitive to $LNO_x$ because of the location and altitude of its emission.

We investigate the performance of the global climate-chemistry model ACCESS-UKCA in simulating $LNO_x$ and the impact of this source on the modelled tropospheric ozone. The UK chemistry and aerosol model [UKCA; Abraham *et al*., 2012] is the atmospheric composition component of the Australian ACCESS global climate model [ACCESS; Bi *et al*., 2013]. It includes both tropospheric and stratospheric chemistry and is used for computing the radiative feedbacks of ozone and methane within the coupled climate system and for other modelling purposes. Here we use ACCESS-UKCA (at UMvn8.4) in atmosphere-only mode with sea-surface temperatures prescribed [Woodhouse *et al*., 2015; Luhar *et al*., 2018]. The model simulations are conducted at 1.875° x 1.25° resolution, with 85 vertical levels extending to 85 km in height. The model uses monthly-varying anthropogenic and natural emissions, including aircraft and $LNO_x$ emissions and biogenic emissions of isoprene.

Lightning flash rate is a critical component in the calculation of $LNO_x$. ACCESS-UKCA uses the popular Price and Rind [1992] parameterisations for lightning flash rate, which are formulated in terms of convective cloud-top height over land and ocean. We test these parameterisations within ACCESS-UKCA using satellite data [Cecil *et al*., 2014] and examine their impact on tropospheric ozone and its budget. Alternative parameterisations are proposed based on lightning data, and their influence quantified. The modelled ozone is compared with observations from global *in situ* and ozone-sonde measurements. The impact of these model changes on OH may also be presented.

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# Nitrogen oxides (NO, $NO_2$, $NO_y$) and nitrate in the air at Cape Grim

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## Abstract

At Cape Grim, Australia's baseline atmospheric monitoring station and contributor to the World Meteorological Organisation Global Watch Program, we have conducted research developing reliable methods and quality control to measure nitrogen oxides and, more recently conducted monitoring of these nitrogen oxides in the background atmosphere of the Southern Hemisphere, where concentrations are amongst the lowest found worldwide. A technical manual outlining this work, the instrument characteristics and data processing has been completed [Galbally, 2019]. Similar work has been undertaken for nitrate in aerosol and rainwater [Cainey, 1998].

The observed average nitrogen oxide levels in baseline air at Cape Grim is 20 ppt $NO_x$. There is a diurnal variation with a daytime increase in $NO_x$ which is not currently well understood. Potential sources include 'renoxification' from photolysis of nitrate aerosol [Kasibhatla *et al*., 2018] and organic nitrates [Fisher *et al*., 2018]. The levels of nitrogen oxides are between 20 to 1000 times smaller in the background atmosphere at Cape Grim compared to the urban air-sheds in southern Australia and New Zealand. There is approximately double the level of nitrate in summer compared to winter in both PM2.5 and PM10 size ranges. This is presumably driven, at least in part, by the increased rate of nitrate aerosol production in summer due to higher levels of gaseous $NO_x$ and OH producing more gaseous $HNO_3$ and subsequently nitrate aerosol. One of the removal processes from the atmosphere of these nitrogen oxides and nitrate is via rainfall in wet deposition. Wet deposition of nitrate is highest in summer and decreases in winter. The lower nitrate deposition in winter is associated with a decrease in nitrate in rainwater coupled with a higher rainfall. This links back to lower nitrate aerosol in winter, and in turn to lower $HNO_3$ production in winter due to both lower $NO_x$ and OH.

The nitrogen oxide levels in baseline air at Cape Grim of 20 ppt $NO_x$ are sufficiently low that it is unlikely that they support any production of ozone in the background atmosphere. This is a key feature in understanding regional variations in the chemistry of the background atmosphere. In particular, the Southern Ocean region is very susceptible to $NO_x$ pollution because of the high photochemical efficiency of $NO_x$ at low levels.

For urban air-sheds in southern Australia and New Zealand, the air from remote marine atmosphere will be virtually free of nitrogen oxides when it enters the air-shed. In event of elevated nitrogen oxides being in the incoming air, this will most likely originate as transport from other urban areas or in the surrounding rural locations.

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# Applying the coupled WRF-CMAQ air quality simulation system for Tehran and comparison with surface and tropospheric column observations

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## Abstract

This study utilises the coupled WRF-CMAQ system as a forward air quality modelling process to estimate the concentration of atmospheric nitrogen dioxide ($NO_2$) for Tehran. A more accurate air quality modelling relies on decreasing uncertainties in the simulation system and, principally, emission inventories (EIs). Emission inventories contain spatiotemporal data about the emission sources and released ratio of pollutants. It is essential to improve EIs since it will improve air quality predictions and their impact.

This study uses some global datasets to prepare fine-resolution inventory data for an urban area. Emissions Database for Global Atmospheric Research (EDGAR), Gridded Population of the World, Night-time Lights Composite datasets, and Fossil Fuel Data Assimilation System are used to prepare hourly EIs at 9$Km^2$ resolution. The resultant high-resolution inventory is applied to run the forward WRF-CMAQ Modeling System to simulate the concentrations of air pollutants. The surface $NO_2$ measurements and TROPOspheric Monitoring Instrument (TROPOMI) $NO_2$ data product are used in this study to compare with the modelled concentration of $NO_2$. The comparison shows that the CMAQ forced with a priori EIs does not simulate the magnitude of surface and satellite data well; however, this system is able to capture some of the spatial variations of TROPOMI $NO_2$ tropospheric column. As a longer-term goal, an established inverse framework will be applied to improve the $NO_x$-EIs in this urban area.

# Open-Path FTIR spectroscopy and measurement of trace gases important to urban air quality

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## Abstract

Fine particulate matter, $PM_{2.5}$, is recognised as a major health risk in urban environments. Ammonia ($NH_3$) is the only alkaline species, In the urban atmosphere, available for the formation of inorganic $PM_{2.5}$ as $(NH_3)_2SO_4$ and $NH_3NO_3$. In the urban environment, the major source of $NH_3$ is vehicle exhaust. $NH_3$ can be difficult to measure, particularly with higher temporal resolution, and emissions to the urban atmosphere are not well defined. Wood smoke, from domestic wood burning heaters or forest fires, is a significant source of $PM_{2.5}$ in southern Australian cities. An open-path Fourier transform infrared (OP-FTIR) spectrometer was deployed in western Sydney, to measure infra-red active gases important to urban air quality, as part of the Clean Air and Urban Landscapes (CAUL) hub. The measurement path of the OP-FTIR spanned between the roofs of two buildings (path-length of ~400 m), and provided continuous measurements over 11 months, with high precision and temporal resolution. Identified gas species, relevant to vehicle emissions and wood smoke, included $NH_3$, carbon dioxide ($CO_2$) carbon monoxide (CO), methane ($CH_4$) methanol ($CH_3OH$), sulphur dioxide ($SO_2$), nitrogen dioxide ($NO_2$), formaldehyde ($CH_2O$), formic acid (HCOOH), acetylene ($C_2H_2$), ethylene ($C_2H_4$) and ethane ($C_2H_6$). The OP-FTIR system and analysis algorithms, developed at the University of Wollongong, are described together with the operational details, as deployed in western Sydney. $NH_3$ ranged between a background of ~1 ppbv to a ~20 ppbv and CO from a background of ~50 ppbv to a maximum 2000 ppbv and were substantially higher in winter (Figure 1). Vehicle $NH_3$ emissions were identified by the correlation with CO or $CO_2$, which corresponded well with peak traffic on nearby roads. Wood smoke events were identified by the correlation between sharp increases in CO and $PM_{2.5}$ during the winter evenings, or during smoke haze events from distant hazard reduction burns.

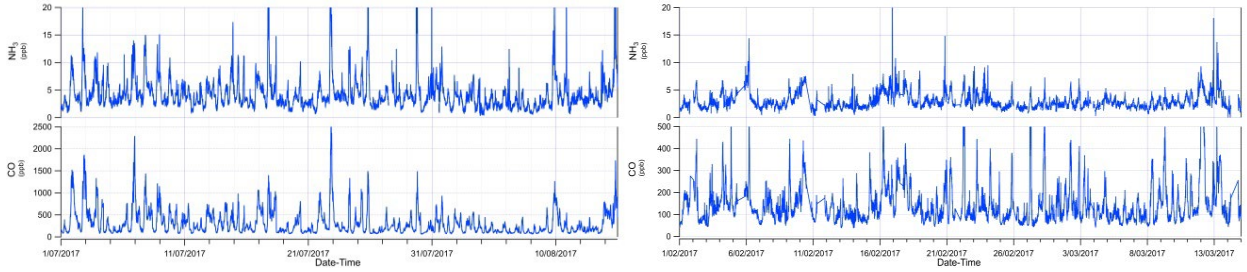



**Figure 1.** $NH_3$ and CO measured during summer (left panel) and winter (right panel).

# A comparison of simulated air quality over Melbourne using EDGAR and EPA (Vic) emission inventories

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## Abstract

It is often said that a model is as good as its inputs. The GIGO (Garbage In, Garbage Out) principle is certainly true when it comes to modelling air quality. Better and more accurate prediction of air quality depends, not only on accurate description of the chemistry and transport processes, but also on emission inventories that are accurate and up to date. In this study, the Weather Research and Forecasting Chemistry (WRF-Chem) model is used to simulate air quality over Melbourne using two emission inventories. The Emission Database for Global Atmospheric Research (EDGAR) inventory is a popular emission inventory used globally [Crippa *et al*., 2018]. However, it is available at a coarse resolution (typically about 10 km x 10 km). In this study, model runs using EDGAR are compared with those using the latest version of Vic EPA (2016) emission inventory (available on 1 km x 1 km resolution), and the results from the two runs are compared where possible with observed air quality.

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# Characterisation of LA-ICP-MS and receptor modelling to measure, speciate and source apportion particulate matter

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## Abstract

Particulate matter (PM) has been observed to produce negative health impacts in humans [WHO, 2016; OECD, 2014]. However, these health impacts can vary depending on several factors, including their sources [Jalava *et al*., 2007; Pun *et al*., 2014]. One approach to source apportioning PM is to determine its elemental composition, which can then be used as data inputs for receptor models that can characterise distinct source fingerprints. This project aims to identify a protocol for conducting elemental analysis of PM using Laser Ablation Inductively-Coupled Plasma Mass Spectrometry (LA-ICP-MS), which can serve as a high-throughput, low-cost alternative to analytical techniques employed for this purpose. Data from this analysis is then processed through a combination of the factor analysis receptor model Positive Matrix Factorisation (PMF), atmospheric measurements and back trajectories to estimate the contributions of different sources of PM.

$PM_{2.5}$ samples (as determined by Scanning Electron Microscope (SEM) analysis) were collected from Garden Island, Western Australia between April and May 2018. SEM was also used to identify spatial distributions of particles on sample papers, which was used to determine the ideal proportion of sample to be scanned to representatively detect the particles present. It was found that LA-ICP-MS analysis was able to reflect atmospheric conditions of the sampling periods. For example, samples that showed strong contributions from a sea spray source (i.e. those high in elements like Na, Cl and Br) corresponded with westerly winds from the Indian Ocean and low radon concentrations (as radon is a reliable tracer of terrestrial air). Conversely, sample that showed strong contributions from terrestrial sources (i.e. those high in elements like Al, Fe and Ti) aligned with winds from the mainland and high radon concentrations. A biomass burning source was also identified, with HYSPLIT-derived back trajectories showing that particles in these samples were transported by air that passed over regions where burning had occurred.

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# Analysis of solar radiation measurements in Australia

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## Abstract

The amount of solar radiation that reaches the Earth's surface is a critical measure of various aspects of climate, biological processes and renewable energy resources. Here, the historical solar radiation flux densities in Australia at 8 observatories within the mid-latitude zone (24-41oS) and 5 observatories at lower latitudes (12-23oS) are examined in terms of the total solar intensity (global) as well as the contribution of the direct (beam) and scattered (diffuse) components. Observational periods varied from nineteen years at Alice Springs to seven years at Mt Gambier.

The analysis focusses on seasonal and annual changes, including summer, winter and annual trends. Given the high degree of variability across time scales, trends at these observatories are poorly defined (generally statistically insignificant). Nevertheless, all mid latitude observatories recorded an increase in summer (January-March) global radiation through the observational period (approximately 1995-2015) equivalent to approximately 5% per decade and a decrease in the winter (July-September) at 6 of the 8 observatories, equivalent to 3% per decade. Annually the observations suggest an increase in annual global flux density at 6 of the 8 observatories, providing the first direct evidence of the global "brightening" phenomenon over Australia. Low latitude observatories generally have a less complete record, but those with longer and more complete records show a consistent positive trend in winter global radiation and no consistency in summer or annual trends. Changes in the relative contribution of the diffuse to global flux are variable between observatories with diffuse flux density decreasing during the winter at 6 of the 8 mid latitude observatories.

The potential significance of these patterns is discussed, with insights into the hypothesis that winter cloudiness may have decreased with the intensification of the semi-permanent high-pressure ridge over southern Australia potential impacts on photosynthetic activity.

# Aerosol Optical Depth Measurements at dirty Cape Grim

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## Abstract

Aerosol optical depth (AOD) measures the reduction in solar radiation at a particular wavelength due to aerosols. This is a key component of the radiation (energy) balance of the atmosphere and can be estimated by both satellite and ground based measurements. The southern hemisphere has typically much lower AOD than the northern hemisphere, and this poses some significant challenges; for satellite measurements the quantification of surface reflectivity becomes much more important and for ground-based measurements ensuring that the measured signal is not being reduced by other environmental factors, especially obstruction of solar radiation as It enters the sunphotometer, requires special attention. Both are challenged by clouds.

For measurements at a site like Cape Grim, contamination of the front window will reduce the observed signal, so that salt, water droplets, spider webs and bird excrement can all be mistaken for aerosols. An example of this impact can be seen in the weekly cycle of measured AOD (Figure 1), where the instrument is not cleaned on Saturday and Sunday, and there is a clear increase in observed AOD. Such an analysis is flawed, however, as cleaning is not carried out on all weekdays. We have therefore reassessed the AOD record since 1986 (the start of automated measurements) in an attempt to quantify the increase in AOD as a function of time since cleaning. This has involved digitising the logbook records of cleaning from 1986 – 1997. Secondly, we have considered whether this contamination has impacted significantly on the in-situ calibrations. Finally, we have also investigated the impact of using a shroud in front of the sunphotometer on the resultant AOD. The shroud is a solution being considered by the Bureau of Meteorology for their network of sunphotometers.

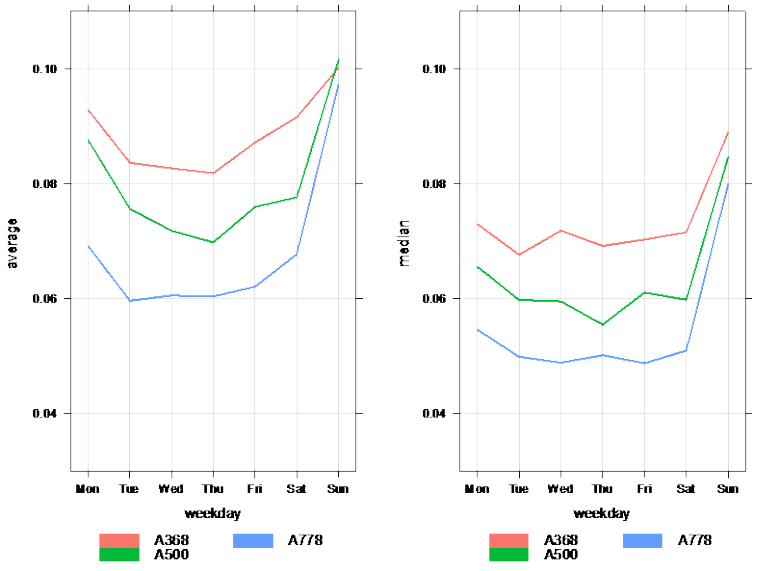



**Figure 1.** Observed Aerosol Optical Depth (AOD) at Cape Grim, 2001 – 2017 as a function of the day of the week. Data are shown for three wavelengths, 368, 500 and 778 nm. The left-hand panel shows the average for the period, with 95% confidence intervals for the average, and the right-hand panel shows the median values.

# Antarctic ozone increase measured by Davis ozonesondes

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## Abstract

We present long-term results from the ozonesonde program at Davis station in Antarctica (68.57°S 77.97°E) operated since 2003 in a partnership between the Australian Antarctic Division, the Bureau of Meteorology and the Chinese Academy of Meteorological Sciences. Ozonesondes provide a unique measurement of high-resolution vertical profiles of ozone from the surface to the mid-stratosphere, along with the co-located temperature profiles.

We present the annual cycle of ozone concentration at Davis over the period of record and calculate the mean ozone loss rate due to chemical depletion at each altitude in the depleted region over the course of winter and spring.

While the severity of Antarctic ozone loss in each year varies strongly with meteorological conditions in the Antarctic polar vortex, as seen for example in metrics of the Antarctic ozone hole, less variability is observed in late winter and early spring before ozone loss reaches its maximum for the year.

By regressing ozone loss against measured temperature, we find a significant increase of ozone is detectable in early spring over the period 2003-2018, with the clearest trend occurring around 70 hPa.

# The 2019 Antarctic ozone hole

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## Abstract

The 2019 ozone hole developed in earnest during the second week of August and initially the ozone hole area rose sharply following a familiar trajectory to past ozone holes, at least until late August. What occurred then in September was a sudden stratospheric warming event which saw record heat flux transported towards the pole with corresponding record warm temperatures in the Antarctic stratosphere. The impact on the development and overall size of the 2019 Antarctic ozone hole has been dramatic. For most of September and October, the daily area of the Antarctic ozone hole was approximately half to a third of what would be expected to be compared to recent ozone holes, with all other ozone hole metrics also being well below average. The rankings for the 2019 Antarctic ozone hole place it as one of the smallest on record, and similar in size to what was seen in the mid-1980s.

The development and recovery of the 2019 Antarctic ozone hole will be shown via a set of metrics based primarily on 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 2019 ozone hole metrics will be compared to the historical metrics.

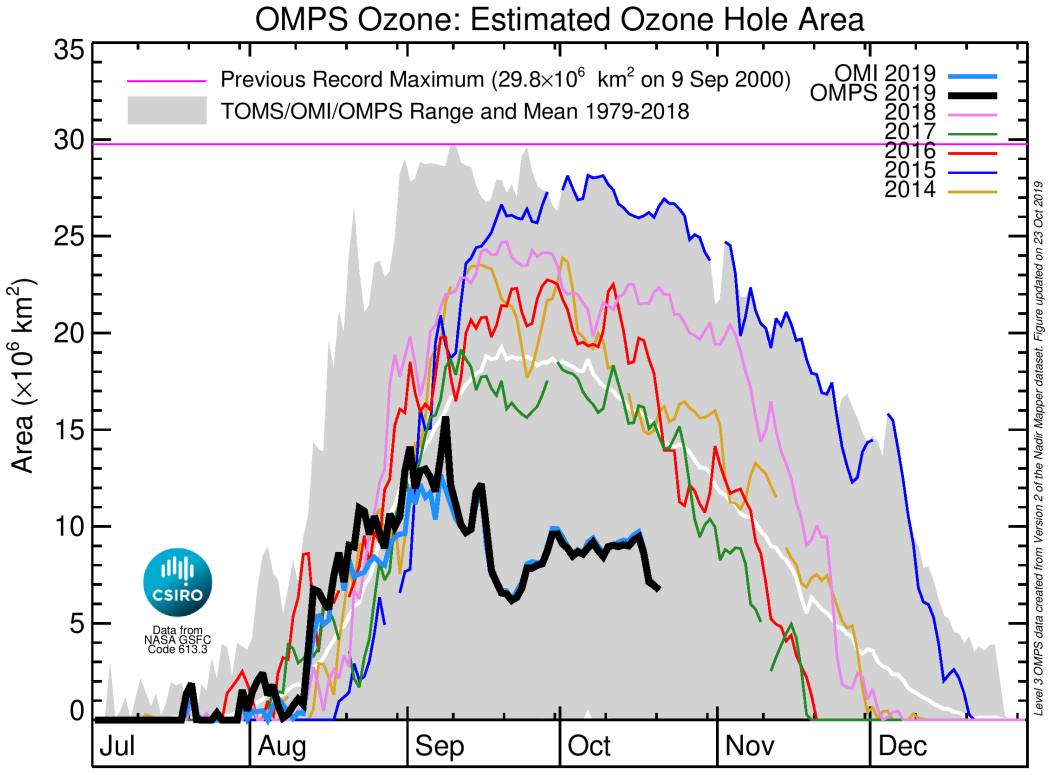



**Figure 1.** The Antarctic ozone hole area as of 20 October 2019, clearly showing the effects of the sudden stratospheric warming event that occurred in September.

# Global emissions of fluoroform (HFC-23) are increasing despite nations reporting reduced emissions

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## Abstract

The global emissions of hydrofluorocarbons (HFCs) are regulated under the Kyoto Protocol (1997) and the Kigali Amendment (2019) to the Montreal Protocol (1987). HFCs have large Global Warming Potentials (GWPs) and curtailing HFC emissions are attractive policy options for nations in meeting their greenhouse gas (GHG) emission reduction targets under the Kyoto Protocol and its Paris Amendments.

Fluoroform (HFC-23, $CHF_3$) is the most potent HFC in a global warming context on a mass-emitted basis with a 100-year integrated GWP of 12,700 [Montzka, Velders *et al*., 2018], whereby 1 tonne of HFC-23 emitted equates to 12,700 tonnes of carbon dioxide-$CO_2$-emitted. Most HFC-23 is vented to the atmosphere during the production of the hydrochlorofluorocarbon, HCFC-22 ($CHClF_2$), due to over-fluorination of chloroform ($CHCl_3$) in the HCFC production process. HCFC-22 is still a widely-produced and used refrigerant in the developing world, especially in China and India, where, unlike in developed countries, zero consumption of HCFCs is not required until 2030.

Atmospheric HFC-23 observations from the Advanced Global Atmospheric Gases Experiment (AGAGE) global network and from the Cape Grim air archive show that emissions grew rapidly from 1980 (4,200 tonnes) to 2006 (13,300 tonnes). They then declined in response to emission reductions in developed countries under the Kyoto Protocol and in developing countries under a UNFCCC (United Nations Framework Convention on Climate Change) Clean Development Mechanism (CDM), whereby developing countries received Montreal Protocol Multilateral Funds (MLF) during 2006-2014 to destroy the HFC-23 they produced, rather than releasing it to the atmosphere. The CDM was initially effective and by 2009 emissions had declined to 9,600 tonnes [Miller *et al*., 2010; Montzka, Velders *et al*., 2018].

However, the CDM was curtailed in the early 2010s, as it distorted the developing nations HCFC-production market, whereby it became profitable to over-produce HCFC-22 for the sole purpose of destroying the resultant HFC-23. In the post-CDM period, HFC-23 emissions have increased again to 15,900 tonnes in 2018, based on atmospheric observations, the highest level of emissions of this HFC ever recorded [Stanley *et al*., 2019]. This is despite significant reported abatement of emissions by China and India in the post-CDM period (2015-2018), such that so-called 'bottom-up' estimates of global HFC-23 emissions fell to 2,400 tonnes in 2017 compared to the 'top-down' estimate of 14,900 tonnes from atmospheric observations, a discrepancy equivalent to 160 M tonnes of $CO_2$ emitted, 30% of Australia's total GHG emissions. Either the reported abatements from China and India are in error or there is an unidentified major new source of HFC-23 emissions. The latter seems unlikely.

There is now no financial incentive for nations to overestimate abatements of HFC-23 emissions, as there was under the CDM, although they do help in meeting overall GHG emission reduction targets. The HFC-23 data show the ongoing need for atmospheric observations to derive top-down estimates of global and regional GHG emissions which can then be used to verify national and global GHG inventories reported to the UNFCCC.

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# Gosan Station, Jeju Island, South Korea: a window into trace gas emissions from East China

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## Abstract

China is now the largest emitter of greenhouse and ozone depleting gases globally and is in the process of developing a network of atmospheric observing stations to provide 'top-down' estimates of national emissions to compare to their 'bottom-up' inventories. For several reasons (short data records, measurement problems, political sensitivities) very little atmospheric data from within China on greenhouse and ozone depleting gases are available publicly.

The GAW-AGAGE baseline station at Gosan (33°N, 126°E) on Jeju Island, Republic of Korea (South Korea), is proving to be an ideal location for studying trace gas emissions from East China. Recent studies of chloroform ($CHCl_3$) and CFC-11 ($CCl_3F$) emissions from East China, based on Gosan data and 3-D inverse studies (Fang *et al.*, 2019; Rigby *et al.*, 2019), show the impact that these emissions are having on global trends in these trace gases and their potential impact on stratospheric ozone recovery.

The AGAGE program at Gosan measures an extensive suite of trace gases and preliminary insights into emissions from East China of a range of gases can be obtained by analysing the magnitude of pollution events arriving at Gosan from mainland China. In this presentation, updates and qualitative estimates of East China emissions of ozone depleting species - CFC-11, $CHCl_3$, methyl bromide ($CH_3Br$), dichloromethane ($CH_2Cl_2$) and carbon tetrachloride ($CCl_4$) - will be shown and their global significance discussed.

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