



COALA

Characterizing Organics and Aerosol Loading over Australia

White Paper for a tropospheric chemistry field campaign in Southeast Australia.

Steering Group:

COALA-JOEYS: Joint Organic Emissions Year-round Study

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COALA-BAIR: Biogenic Anthropogenic Interactions Research

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COALA-TALENT: Testing Analogues for the Low Emission of NO_x Transition

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Overview

COALA (Characterizing Organics and Aerosol Loading over Australia) is a broad project designed to study the emissions and atmospheric chemistry of Australian biogenic species in pristine conditions and as they interact with anthropogenic pollution. Recent technological developments now make it possible to study in unprecedented detail the reactive carbon budget for isoprene, monoterpenes and sesquiterpenes, including quantifying emissions, transformations and deposition. These new developments will allow us to address specific questions related to a major uncertainty associated with climate forcing: knowing how emissions were processed in a pristine environment representative of pre-industrial conditions. The isolated urban areas of Australia, located close to pristine forests, will also allow us to quantify the impact of isoprene, monoterpenes and sesquiterpenes on urban air quality, and to address questions associated with the emerging science of lower NO_x regimes in the mature industrialized world of the Northern Hemisphere.

COALA is an international collaboration bringing together resources and facilities from Australia (multiple funding sources), US (NSF), and the UK (NERC). COALA is the “umbrella” for three main projects: JOEYS (primarily Australian funding), BAIR (primarily US NSF funding), and TALENT (primarily UK NERC funding). Each project is designed as a standalone effort with specific addressable science questions but each will be greatly enhanced by the complementary measurements of the other projects. COALA’s main purpose is to ensure that the objectives of the individual projects are coordinated. An intensive COALA field phase will be held in January-February 2020 focused on Sydney and the surrounding areas in southeastern Australia.

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1 Introduction and Scientific Rationale

A quantitative determination of the changes in atmospheric chemistry and climate since pre-industrial times is urgently required in order to develop effective climate mitigation and adaptation strategies but this is complicated by the need to characterize natural emission sources, such as those from undisturbed terrestrial vegetation, wildfire, and oceans, and the identification of unique chemical processes without the influence from anthropogenic sources. Large areas of the southern hemisphere (SH) are near pristine and provide an opportunity to investigate the clean, natural atmosphere relatively free of anthropogenic influence, and thus to define the baselines from which human influences can be assessed. This opportunity has largely been lost in the northern hemisphere (NH), and the SH could serve as a proxy for potential future conditions in North America and Europe (under stricter pollution controls).

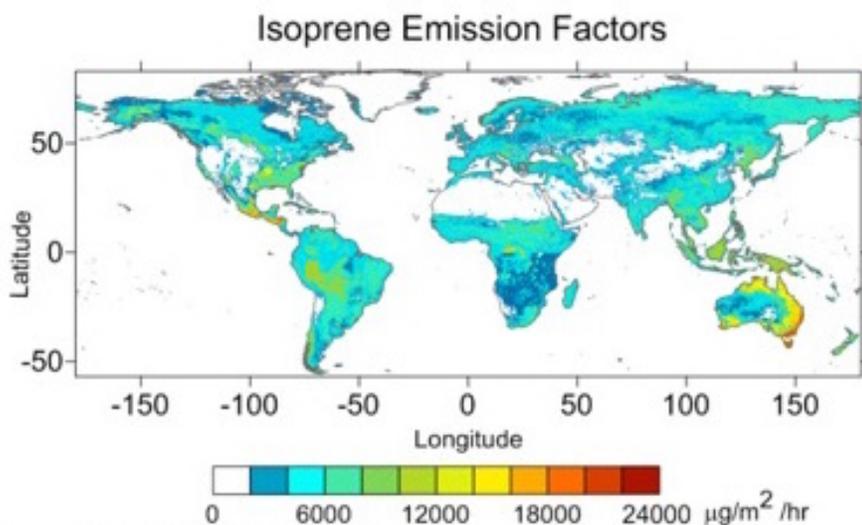


Figure 1. Isoprene emission factors from MEGAN (Guenther et al., 2012).

Southeast Australia presents a unique opportunity to evaluate these processes. Estimates of Biogenic Volatile Organic Compound (BVOC) emissions based on mechanistic models (Arneth et al., 2011; Guenther et al., 2012; Zeng et al., 2015) and satellite observations (Pfister et al., 2008) show Southeast Australian biogenic isoprene emissions are among the highest in the world (Figure 1), due largely to the prevalence of densely forested Eucalypt ecosystems. The

maximum emission factor in Southeast Australia is $24000 \mu\text{g}/\text{m}^2/\text{hr}$ which is more than double the global average for any Plant Functional Type ($11000 \mu\text{g}/\text{m}^2/\text{hr}$) from Guenther et al (2012). Eucalypts also emit large amounts of monoterpenes and sesquiterpenes (e.g., He et al. 2000), leading to a complex mix of ozone and organic aerosol precursors.

However, models of BVOC emissions in SE Australia vary widely across inventories, with differences of a factor of 2-3 for isoprene and a factor of 5-10 for monoterpenes (Zeng et al., 2015; Sindelarova et al., 2014; Emmerson et al., 2016). Field observations of ambient concentrations investigated by Emmerson et al. (2016) suggest that isoprene emissions in the inventories are overestimated while those of monoterpenes are underestimated. Furthermore, the model predictions of nearly uniform isoprene and monoterpene emissions across the region, as shown in Figure 1, are due to the simplistic approach for assigning emissions to plant functional types and we expect much higher diversity in reality with some landscapes dominated by monoterpenes and possibly even sesquiterpenes. Additionally, the chemistry of these emissions is challenging to simulate. For example, formaldehyde (a high-yield product of isoprene oxidation) in SE Australia is underestimated at Wollongong by $\sim 50\%$ irrespective of inventory choice (Zeng et al., 2015).

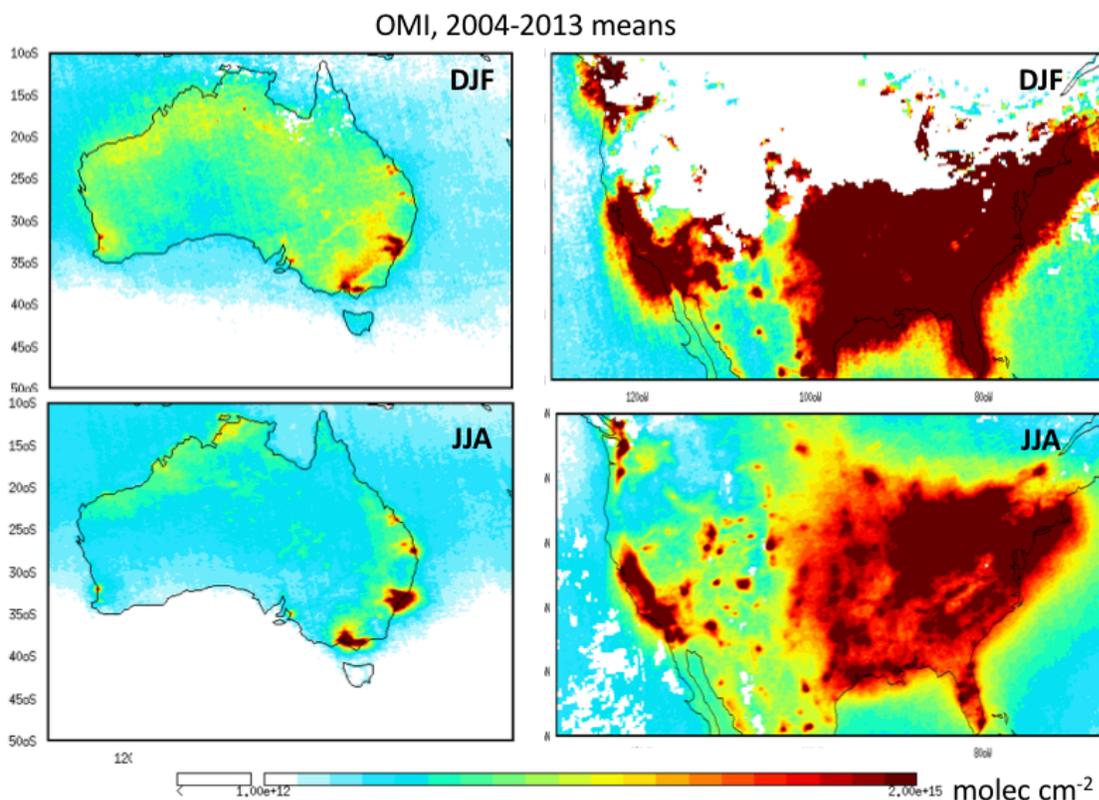


Figure 2. Tropospheric NO₂ column from OMI satellite for Australia (left) compared to the US (right), averaged over 10 years for December-February and June-August. Australian fire activity is low at this time of year (NW excepted), and NO₂ amounts highlight the urban influences around the SE coast (Sydney, Melbourne).

Anthropogenic NO_x emissions in SE Australia are extremely low and spatially isolated (Figure 2), with little anthropogenic influence outside the major urban centers of Sydney, Canberra, and Melbourne (National Pollutant Inventory: www.npi.gov.au). The population of Australia is densely located in urban hubs around the coast, with 90% of people living on just 0.22% of the land area (National Sustainability Council, 2013). The isolation of anthropogenic NO_x sources in SE Australia means that this region provides an ideal natural laboratory to study the impact of variable NO_x amounts on BVOC chemistry (from upwind of urban areas, through a gradient at the urban interface, to the downwind outflow).

Recent field campaigns investigated interactions between biogenic and anthropogenic emissions in the Southeast US (SOAS/SENEX/NOMADSS, SEAC⁴RS), where background NO_x levels are higher and much less spatially distinct (Fig. 2) and biogenic emissions are dominated by isoprene. The lower background concentrations in Australia will allow studies targeting the shift from moderate to low-NO_x conditions. This will provide a preview of the expected future trends and conditions in the U.S. and Europe, where NO_x levels have declined sharply in recent years and are expected to continue to decrease in response to strengthened air quality regulations (Duncan et al., 2016). Additional recent studies conducted in tropical forest regions have examined biogenic-anthropogenic emission and chemical interactions associated with an isolated urban plume in the Amazon forest (GOAmazon, Martin et al., 2016) and oil palm plantations in Borneo (OP3, Hewitt et al., 2010) which are both regions where biogenic emissions are dominated by isoprene. These studies have demonstrated the importance of isoprene emissions in determining anthropogenic impacts but have also shown that our current understanding is insufficient for quantifying the impacts and that there are substantial discrepancies between model simulations and observations. The COALA research program will synthesize knowledge from these recent studies and apply recent advances in observational techniques in order to achieve a comprehensive and systematic characterization of the reactive carbon budget including emissions, deposition and atmospheric transformations.

COALA will build upon recent advances in atmospheric chemistry, including field campaigns that have taken place around the globe in recent years, and will further characterize BVOC emissions, chemistry and implications for ozone and aerosols in an environment with vegetation types, oxidant levels, and climate conditions that are unlike what has previously been investigated. In addition to the moderate to low NO_x conditions in this region, the presence of some Eucalyptus forests with relatively high monoterpene emissions and others with high sesquiterpene emissions provides a potential opportunity to contrast regions dominated by isoprene with areas dominated by monoterpenes or even sesquiterpenes, unlike the previous isoprene-dominant studies (including SOAS/SENEX/NOMADSS, SEAC⁴RS, OP3, GOAmazon). The COALA project will address one of the central themes identified at the Atmospheric Chemistry Center for Observational Research and Data (ACCORD) Community Workshop (March 2015, <https://www2.acom.ucar.edu/accord/accord-2015-workshop>), which is to conduct a field campaign in a terpene-dominated landscape to close the reactive carbon budget.

The measurements and modeling associated with COALA will address the impacts of changing biogenic emissions due to weather extremes and climate factors. Sensitivity studies with regional and global coupled chemistry-climate models will explore the impacts of possible future climate conditions, such as rising temperature, increased frequency of drought and temperature extremes. Global model simulations will also assist in quantifying differences between Northern and Southern Hemispheres due to lower CO₂ and CH₄ in the Southern Hemisphere. Using models with specified dynamics allows for simulation of specific time periods of past observations to test and improve biogenic emissions algorithms, such as MEGAN, under stressed conditions.

The first COALA intensive experiment is planned for January-February 2020, coordinating flights of the U.K. FAAM BAe146 (TALENT) with NSF/NCAR tower-based facilities (BAIR) and ongoing Australian research that includes a pre-campaign (JOEYS) planned for preceding summers, and year-round measurements at one or more representative ground sites to capture seasonal and interannual variations. The science questions of the overall COALA project are discussed below, and the 2020 campaigns will begin to address these.

2 Key Science Questions for COALA

Q1. How have emissions changed since pre-industrial conditions?

Natural emissions, from undisturbed vegetation, wildland fires, and marine sources, controlled the composition of the atmosphere in pre-industrial times. These emissions have continued in the presence of changes in anthropogenic emissions in the past 150 years. To understand the changes in chemistry and climate since pre-industrial times, it is required to quantify both the natural emissions and the changing anthropogenic emissions of trace gases and particles. To do this, several specific issues need to be addressed:

Q1A. What are the gaseous and particulate emissions from natural sources in SE Australia, including biogenic, wildland fire, and marine emissions?

Southeast Australia has significant VOC and NO_x emissions from terrestrial vegetation (both undisturbed and disturbed) as well as from wildland fires and from the surrounding oceans. It is essential to quantify the emissions from these sources to predict the changes in composition since pre-industrial times and close the reactive carbon budget over this time. Recent model evaluations have shown significant disagreement between models and observations due, in part, to errors in emissions inventories. For example, Emmerson et al. (2016) show an over-estimate of isoprene emissions, but under-estimate of terpene emissions, in southeast Australia. Four different global models in a recent study also did a poor job of reproducing CO and formaldehyde observations in the SH, at least in part due to misrepresented emissions (Fisher et al., 2015; Zeng et al., 2015). Flux tower measurements in Eucalypt forest over extended periods will allow quantification of biogenic VOC emissions. Additional measurements from aircraft will improve estimates of canopy-scale emissions and wildfire (if present) emissions. There is significant uncertainty over DMS and VOC emissions from oceans,

and ship-based and airborne measurements over the sea will assist in quantifying those over the Southern Ocean.

Q1B. What are the magnitude and trends of anthropogenic emissions in the region?

The chemical composition of the atmosphere since pre-industrial times has changed due in part to the increases and evolution of anthropogenic emissions. Therefore, these emissions need to be elucidated in order to quantify atmospheric composition over the past 150 years. Surface measurements within the cities of southeast Australia (Sydney, Melbourne, Brisbane), and over power plants, fossil fuel extraction and refinery sites and other industrial sites, of source gases and primary aerosols (CH₄, CO, NO_x, SO₂, VOCs) will be used to verify present-day emissions inventories. Long-term observations from surface sites and satellites will be used to establish past trends.

Both ground-based and airborne measurements of NO_x, VOCs and aerosol concentrations and fluxes over SE Australia can quantify the emissions from biogenic and anthropogenic sources. VOC emission fluxes have been quantified from measurements on aircraft in past experiments (MIRAGE, NOMADSS, SEAC4RS, FRAPPÉ). Flight tracks of either racetracks, or straight legs over uniform landcover, at multiple altitudes within the boundary layer, have been used for flux determination, and would complement tower-based flux measurements in forests for biogenic emissions (Karl et al., 2009; Kaser et al., 2015). Quantification of VOC emissions over various landscapes and the ocean would be achieved through low altitude flights.

Q2. How are BVOC emissions chemically processed on local and regional scales?

A number of questions remain regarding the products and their rate of formation from biogenic VOCs in low NO_x environments, particularly from monoterpenes and sesquiterpenes. The production of ozone, organic nitrates and secondary organic aerosols have important impacts on downwind air quality as well as radiation budgets.

Q2A. How do low-NO_x environments affect the mechanism for BVOC oxidation in locations with similar concentrations of isoprene and terpenes, and how does this affect secondary organic aerosol formation?

The SE Australian region is characterized by low background NO_x concentrations, with existing sources limited to a few spatially distinct urban areas (Fig. 2), providing a clear NO_x gradient over which to investigate a number of related questions surrounding isoprene and terpene oxidation. Our current understanding of BVOC chemical processing (and subsequent ozone and aerosol formation) comes primarily from areas targeted by previous campaigns where isoprene dominates observed BVOC burdens (e.g. Southeast US, Amazon). In SE Australia, isoprene and monoterpenes are present at equal concentrations (Emmerson et al., 2016). The impacts of this much larger terpene-to-isoprene ratio on atmospheric oxidizing capacity, gas-phase chemical processing, and organic aerosol formation are not known.

The OH radical plays a critical role in this chemistry as the initiator of VOC oxidation during the day. However, there is a severe lack of OH measurements across the SH. Previous OH measurements in biogenic environments have highlighted major gaps in our understanding of OH formation and recycling during BVOC oxidation (e.g., Lelieveld et al., 2008; Whalley et al., 2011). Measurements of OH and reactivity across different NO_x levels in SE Australia will help further constrain OH chemistry in the presence of significant terpene concentrations.

SOA formation in balanced terpene-isoprene environments has not been observationally constrained. SOA yields from monoterpenes are larger than from isoprene. In high-isoprene environments, significant aerosol formation occurs via low-NO_x oxidation to produce isoprene epoxydiols. However, there is some evidence that isoprene dominance can suppress aerosol formation (Kanawade et al., 2011). In SE Australia, significant aerosol formation was observed at night at a site within a dense Eucalypt forest (Tumbarumba) (Suni et al., 2008), but without simultaneous gas-phase observations. Simultaneous observations of aerosol concentration, speciation, and gas-phase precursors are needed to better understand aerosol formation in terpene-isoprene balanced environments.

Q2B. How do organic nitrates form in high-BVOC, low-NO_x environments, and what are the implications for NO_x, ozone, and aerosols?

Organic nitrates exert a significant influence on the budgets of ozone (Fiore et al, 2005; Horowitz et al., 2007) and organic aerosols (Rollins et al., 2013; Lee et al., 2016), both within source regions and as a means for exporting NO_x (via PAN, for example). There is evidence that formation of organic nitrates can be the primary determinant of the NO_x lifetime in low-NO_x environments (Browne and Cohen, 2012; Romer et al., 2016). However, in most high-BVOC regions in the NH, anthropogenic NO_x sources are spatially distributed across a background of high BVOCs, and very little of the landscape has low enough NO_x emissions for organic nitrates to dominate NO_x loss (Fisher et al., 2016). Whether significant organic nitrate formation occurs in regions where NO_x sources are small and isolated remains uncertain, as do the implications for ozone and SOA. The SE Australian setting will allow us to better constrain pathways for organic nitrate formation and impacts in low-NO_x environments and at the urban-rural interface. It will also allow us to test whether NO_x from soil sources or urban outflow can lead to organic nitrate formation over forests, and whether this affects the lifetime of NO_x in these environments.

Q2C. To what extent does BVOC chemistry drive air quality in cities downwind of forests?

Dense anthropogenic NO_x sources are spatially isolated in Australia and surrounded by some of the world's most intense biogenic sources. The isolation of anthropogenic NO_x sources in SE Australia means that this region provides an ideal natural laboratory to study the impact of variable NO_x amounts on BVOC chemistry, from upwind of urban areas, through a gradient at the urban interface, to the downwind outflow. OH dominates oxidant chemistry during the daytime, and usually quenches most isoprene by sunset. Isoprene remaining after dark is typically removed by reaction with NO₃ radicals (Xie et al., 2013). However, when NO₃ radical formation is suppressed (as occurs in the presence of NO in urban environments), isoprene can persist at night and drive morning ozone formation (Millet et al., 2016). Through this

mechanism and through the formation and transport of BVOC-sourced organic aerosol, forests can contribute to poor air quality in cities downwind. Cities downwind of forests are prevalent across the tropics and the Southern Hemisphere, and a better understanding of these biogenic-anthropogenic interactions at the forest-urban interface is critical.

Q2D. Can isoprene emissions be inferred from satellite HCHO in low NO_x conditions?

Isoprene emissions are poorly constrained across much of the globe, particularly in the tropics and SH. In many regions where in situ observations are unavailable, isoprene emissions are inferred using satellite HCHO measurements (Kefauver et al., 2014, and references therein). However, low NO_x chemistry can hinder the interpretation of satellite measurements. While HCHO is produced from both NO and HO₂ oxidation, the HCHO production rate from NO oxidation is much larger. As a result, observed HCHO is more representative of local surface isoprene fluxes in regions where NO oxidation dominates. The slower HCHO production in low NO_x regions causes peak HCHO formation downwind of isoprene sources. This leads to “smearing” of the isoprene-HCHO relationship that can be a large source of error in inferred isoprene emissions (Palmer et al., 2003; Marais et al., 2012; Barkley et al., 2013). While this smearing has been quantified to some extent under moderately low NO_x conditions, it has not been observationally constrained at very low NO_x (Wolfe et al., 2015). Such conditions are representative of large swaths of the SH (Amazon, central Africa) that are typically difficult to reach and logistically challenging for making dedicated measurements. Direct comparison between in situ and satellite measurements are necessary to understand the reliability of the satellite data as a proxy for isoprene emissions in these regions (Zhu et al., 2016). Measurements of isoprene fluxes in SE Australia will provide tests of inverse modeling results from satellite formaldehyde retrievals.

Q2E. What conditions can we expect in future from declining NO_x emissions in the NH?

The much lower NO_x background in SE Australia relative to the NH means results from this study may provide a preview of the expected future conditions elsewhere. In the U.S. and Europe in particular, NO_x levels have declined sharply in recent years and are expected to continue decreasing in response to strengthened air quality regulations (Duncan et al., 2016). The implications for future organic aerosol burdens remain uncertain, with models showing conflicting results. Lower NO_x can reduce organic aerosol formation by shutting down organic nitrate mediated chemistry (Pye et al., 2015), but it can also enhance organic aerosol formation by shifting more isoprene oxidation to the low-NO_x pathways with higher SOA yields (Marais et al., 2016). The low NO_x backgrounds and strong gradients of SE Australia provide a natural laboratory to test these models and identify the dominant effects, improving our ability to predict future air quality in NH cities.

Q3. How will climate change and extreme weather (e.g., drought and temperature extremes) impact biogenic emissions?

Solar radiation, temperature and soil moisture conditions are important for plant growth, and non-optimal conditions such as drought may temporarily increase BVOC emissions but will ultimately inhibit emissions (Pegoraro et al 2007; Sharkey and Loreto, 1993). Australia has

experienced significant and long-lasting droughts, including the 2001-2009 Millennium Drought (Ummenhofer et al., 2009; van Dijk et al., 2013), with implications for both vegetation cover (especially leaf area index; Huang et al. 2014) and water stress, both of which impact BVOC emissions (Fortunati et al. 2008). Using a soil moisture activity factor to account for drought conditions, Sindelarova et al. (2014) found reductions in isoprene emissions of up to 50% globally, with even greater reductions in regions of Africa and Australia.

Southeast Australia is also a region with high sensitivity to ongoing global change. In the last decade, the region has experienced record-setting extreme high temperatures (Lewis and King, 2015), especially in summer (Lewis and Karoly, 2013) when BVOC emissions reach their annual peak. Isoprene emissions typically increase with temperature, but in at least some species of Eucalypts isoprene emissions flatten out around 35°C and begin to decline at temperatures above ~40°C (Guenther et al., 1991; Pacifico et al., 2009). Daily maximum temperatures in this range can occur in southeast Australia during summer. During the Measurement of Urban, Marine and Biogenic Air campaign (MUMBA) in the summer of 2012/2013, the air temperature surpassed 40°C on two days (Paton-Walsh et al., 2017). HCHO observations from Wollongong show increases in austral summer since 1997, especially during seasonal transitions (Nov, Feb), that may indicate biogenic emission responses to rising temperature and longer growing seasons (Lieschke, 2015).

Measurements of monoterpenes at Tumbarumba in November 2006 show the complex effects of plant stress on BVOC emissions. A normal profile shows the expected nighttime peak in monoterpene concentrations due to low mixing height and chemical loss rates. However, under stress conditions after a snowstorm the monoterpene emissions increased four-fold, and the diurnal profile looked similar to isoprene, with a midday peak, which suggests the emission of light dependent monoterpenes which are typically associated with stress induced emissions (Emmerson et al., 2016). Any extreme events that occur during the intensive field campaigns, or the year-round studies, will be analyzed as special case studies to understand the impact on biogenic emissions.

3 Experimental Design

The COALA observations include long-term measurements, including summers 2017 to 2019, of the JOEYS project and an intensive field campaign with the TALENT aircraft measurements and the BAIR tower-based measurements, planned for austral summer (January-February) 2020. BVOC emissions are expected to peak during summer when radiation and temperature are at their annual maxima (e.g., Fig. 5, below). The summer timing will also provide the possibility to capture Australian biomass burning plumes (Edwards et al., 2006). The possible sites of operation, as well as the Sydney area air quality monitoring stations, are marked on the map in Figure 3.

3.1 BAIR tower-based measurements

A comprehensive suite of emissions, micrometeorology and chemical concentration and flux measurements will be made at six heights, within and above the canopy, at both an open and a closed Eucalypt forest canopy. The measurements will fully characterize the leaf and canopy level emissions of isoprene, monoterpenes, sesquiterpenes and other biogenic VOC and the formation of oxidation products and secondary aerosol within and above the open and closed canopies. The dynamics of VOC/oxidant/reactive nitrogen interactions will be examined in the open and closed canopies over a wide range of NO_x levels. The Cataract Scout Park in the forest southwest of Sydney (northwest of Wollongong) has been identified as a likely location for the tower site. The deployment will enable the specific stand-alone BAIR research objectives and the measurements will also support the broader COALA objectives. A more detailed description of the BAIR scientific objectives and approach will be available from the COALA-BAIR science proposal.

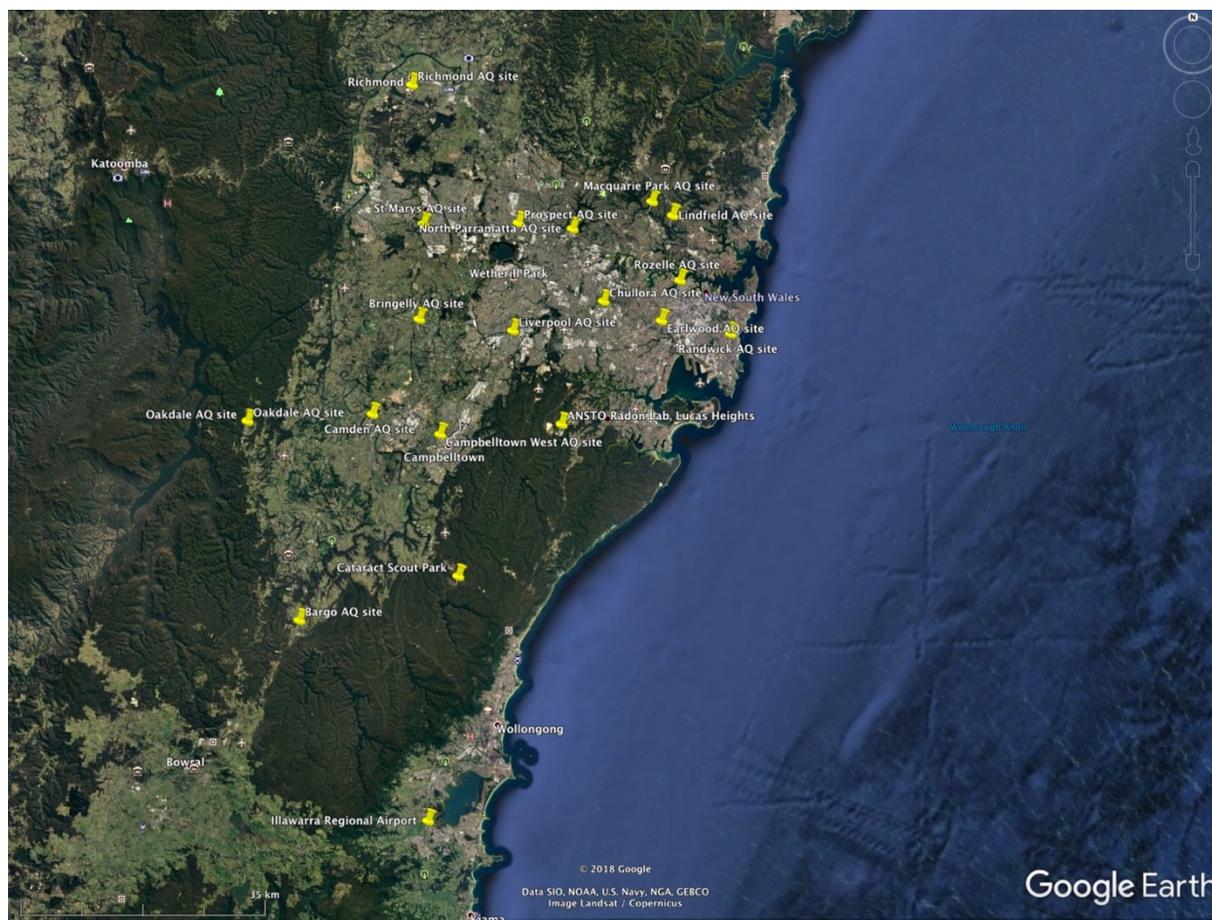


Figure 3. Map of COALA-BAIR tower site, airport and air quality stations.

3.2 TALENT aircraft-based measurements

Aircraft measurements will be a key component of COALA, providing regional context and linking the various ground-based measurements. The FAAM aircraft will be deployed in the

Sydney area in January-February 2020, allowing characterization of pristine, marine air over the ocean, while the impact of Sydney and its environs will be observed in inland flights that cross the Sydney urban area, the peri-urban fringe and unpopulated forests of the Blue Mountains. Lagrangian flights will be performed along wind-streams.

The BAE146 will be based at an airport within range of the Cataract Scout Park tower site, allowing sampling above the towers as well as the variety of Eucalypt forests in southeast Australia. A range of environments including desert, agricultural, forested, suburban, urban and ocean will be covered by flying east-west transects. A more detailed description of the TALENT aircraft scientific objectives and approach will be available from the COALA-TALENT science proposal.

3.3 JOEYS longer-term ground measurements

Ground-based measurements commenced during 2017 and will continue until the completion of the COALA-BAIR and COALA-TALENT intensive campaigns, in order to provide a longer temporal span of measurements in relevant environments. Whilst ground-based measurements will be a key component of the intensive campaign, the principle purpose of JOEYS is to provide a detailed characterisation of seasonal cycles of major biogenic VOCs and air quality in the relevant ecosystem. This will provide a contextual basis for interpreting the short temporal span of measurements over the greater spatial extent covered by the aircraft measurements.

Ground-based measurements funded through the Clean Air and Urban Landscapes hub (see www.nespurban.edu.au) will include:

- Measurements of BVOCs at a number of locations representative of different vegetation ecosystems around New South Wales over the summer of 2017-2018 using grab sampling techniques with absorbent tubes and subsequent analysis on a GC-MS system.
- Preliminary assessment of suitability of Cataract Scout Camp as main chemistry supersite (March 2018) using measurements conducted with the Atmospheric Integrated Research on Burdens and Oxidative capacity (AIR-BOX) facility and ancillary instrumentation. AIRBOX is a collaboratively owned Australian mobile atmospheric laboratory. Further details can be found at: smah.uow.edu.au/cac/index.html.
- Year-round measurements of volatile organic compounds using the Biogenic Ambient Atmospheric Sampling System (BAASS) installed at ANSTO (Australian Nuclear Science and Technology Organisation) in Lucas Heights. These will be accompanied by ancillary measurements including CO, CO₂, NO_x and O₃ and campaign measurements using an aerosol mass spectrometer (to be provided by CSIRO in the summer of 2018-2019).

JOEYS will seek further funding through Australian funding agencies to extend the Australian contribution to COALA with further measurements, modelling and scientific analysis during the COALA intensive campaign period.

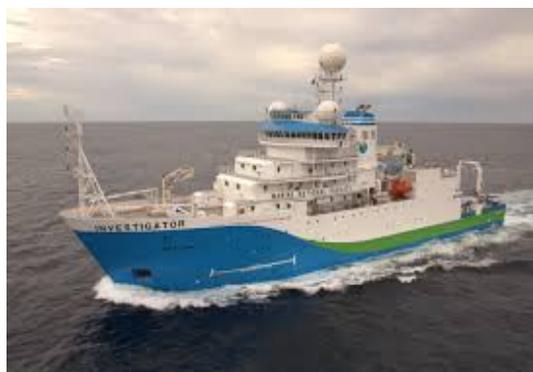
Ongoing air quality monitoring

In addition to the measurements described above, there are 42 air quality monitoring stations within New South Wales, making measurements of PM₁₀, PM_{2.5}, O₃, NO_x, CO and SO₂, along with meteorological parameters (wind speed, wind direction, temperature, pressure and relative humidity).

R.V. Investigator Campaign to Sample Clean Marine Air at Southern Hemisphere Mid-latitudes

Australia's new marine national facility, the RV Investigator, would have the capability to characterise the clean marine air background concentrations and latitudinal gradients of biogenic VOCs, O₃ and NO_x at SH mid-latitudes. The R.V. Investigator has a bespoke stainless steel air sampling tube through which air samples can be drawn from 6m up the foremast into the two dedicated laboratories for atmospheric measurements.

An aerosol laboratory is situated at the bow directly below the foremast and contains an absorption photometer, an atmospheric nephelometer, and a scanning mobility particle sizer as well as a radon detector. Sample air also passes through the aerosol laboratory and onto the larger air chemistry laboratory, which houses O₃ and NO_x monitors as well as cavity ring-down spectrometers for CO₂, CH₄, CO and N₂O. There is capacity for guest instruments aboard the R.V. Investigator, either within the air chemistry laboratory or inside a shipping container, which may be located on one of two purpose designed footings on the foredeck. The timing of this campaign is subject to availability of ship-time.



3.4 Satellites

Satellite observations of tropospheric constituents provide long-term and regional-to-global context for the COALA campaign. The long-time records from instruments such as MOPITT and OMI provide a global climatology of CO, NO₂, CH₂O, O₃, AOD, etc., allowing quantification of the seasonal cycle and variability. The newly launched polar-orbiting Sentinel-5P/TROPOMI instrument provides observations of numerous species at high horizontal resolution each day, and the geostationary satellite Himawari provides high spatial resolution AOD and visible imagery. In turn, COALA ground-based and aircraft observations will contribute to the validation of the satellite retrievals.

Satellite retrievals of CO, from MOPITT on the Terra satellite (see Figure 4) and IASI on MetOpA and MetOpB, will be particularly valuable during COALA to track the influence of wildfire emissions over Australia. Aerosol Optical Depths (AOD) from MODIS on Terra and Aqua will also show the location of fire plumes and pollution outflow. Satellite retrievals of tropospheric NO₂ from TROPOMI (similar to OMI (Figure 2) and GOME-2) highlight source regions such as

urban centers and will be useful for determining where to fly to best intercept the rural-urban gradient.

Satellite retrievals are now possible for several BVOCs and oxidation products, including methanol, glyoxal, formic acid (IASI), and formaldehyde (TROPOMI, OMI, GOME-2). These will facilitate identification of BVOC emissions and their temporal variability (see Figure 5). Formaldehyde in particular has been used previously to quantify the emission of BVOC precursors such as isoprene. However, retrieving CH_2O amounts from satellites is quite challenging and requires extensive averaging to establish a clear signal. There are known biases in current CH_2O retrievals (Zhu et al., 2016), and the isoprene- CH_2O relationship has not been quantified in a high-terpene environment. The coincident ground-based, airborne, and satellite measurements of BVOCs and CH_2O will help to better quantify satellite biases and improve understanding of the links between CH_2O and BVOC fluxes.

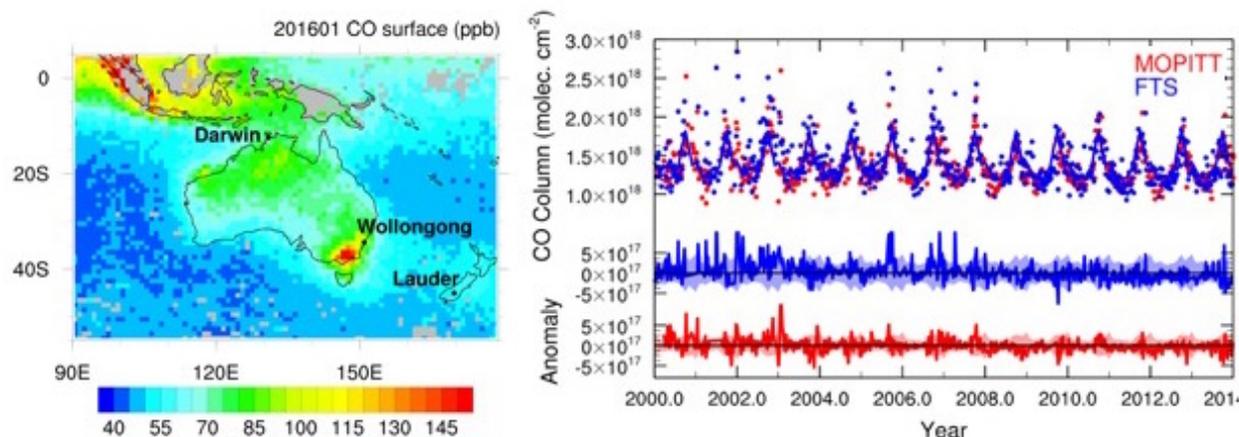


Figure 4. Left: MOPITT CO surface retrieval for Jan 2016, with location of FTS sites. Right: Wollongong FTS column CO time series with MOPITT CO, and weekly anomalies.

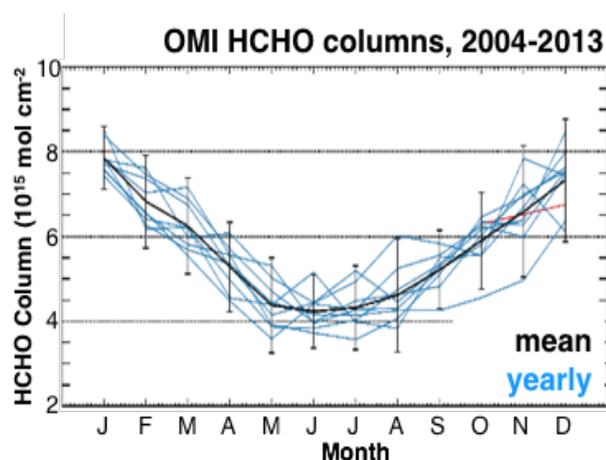


Figure 5. Monthly mean tropospheric HCHO columns from the OMI satellite, averaged over Southeast Australia, with individual years in blue and multi-year means in black (from Jesse Greenslade, University of Wollongong).

4 Integration with models

Chemistry models across scales from box models to regional and global 3D chemistry-climate models will be used to design and interpret the COALA experiment and results.

Chemical forecasts from global and regional models will be used for aircraft flight planning and to direct particular sampling at the tower site. Near-real-time simulations will also be compared to early observations during the campaign to further guide flight planning by identifying any major gaps between models and observations that may warrant investigation in future flights.

After the campaign, the aircraft and ground-based measurements will be used to evaluate the biogenic emissions models (e.g., MEGAN), the anthropogenic and wildfire emissions inventories, and the representation of chemical processing in the models. Improvements will be made to the models, as needed, to better represent the observations. The models will also be used to provide context for the observations, such as identifying when wildfire emissions may have impacted the observations.

Coupled chemistry-climate models will be tested against the COALA observations to evaluate the models' ability to reproduce an atmosphere strongly influenced by biogenic emissions, but weakly by anthropogenic sources.

5 International collaboration

COALA will bring all the benefits of a major international collaborative campaign, with involvement of research groups from at least 3 continents (America, Europe and Australia). The Steering Committee has representatives from Australia, US, and UK that include both researchers experienced in running aircraft and tower-based campaigns and local representatives, who can organize logistics on the ground.

The use of AIRBOX within JOEYS will involve a large consortium of Australian universities and research institutes, including the University of Melbourne, Queensland University of Technology, Macquarie University, the University of Wollongong, the University of Tasmania, Monash University, the Australian Antarctic Division, the Commonwealth Scientific and Industrial Research Organisation and Australian Nuclear Science and Technology Organisation.

A significant part of the collaboration will occur once the campaign data has been collected, with the many groups sharing data, coordinating modelling studies and working together to best understand, analyse and publish the findings from the campaign. This will ensure that the results are disseminated to as wide an audience as possible.

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