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COMMENTARY

Key challenges for tropospheric chemistry in the Southern Hemisphere

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This commentary paper from the recently formed International Global Atmospheric Chemistry (IGAC) Southern Hemisphere Working Group outlines key issues in atmospheric composition research that particularly impact the Southern Hemisphere. In this article, we present a broad overview of many of the challenges for understanding atmospheric chemistry in the Southern Hemisphere, before focusing in on the most significant factors that differentiate it from the Northern Hemisphere. We present sections on the importance of biogenic emissions and fires in the Southern Hemisphere, showing that these emissions often dominate over anthropogenic emissions in many regions. We then describe how these and other factors influence air quality in different parts of the Southern Hemisphere. Finally, we describe the key role of the Southern Ocean in influencing atmospheric chemistry and conclude with a description of the aims and scope of the newly formed IGAC Southern Hemisphere Working Group.

Keywords: Biogenics, Fires, Southern Ocean

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1. Introduction — Overview of issues for tropospheric chemistry in the Southern Hemisphere

Despite significant differences in atmospheric composition between the Northern and Southern Hemispheres, atmospheric chemistry in the Southern Hemisphere troposphere has received relatively little scientific attention. The Southern Hemisphere surface is dominated by oceans (approximately 80% of the surface area) and contains less than half the landmass of the Northern Hemisphere, so deposition processes to the oceans (e.g., for ozone) may play a more significant role than in the north. Additionally, anthropogenic activities influence a smaller proportion of the landmass in the Southern Hemisphere than in the Northern Hemisphere (i.e., less urban and agricultural development and more natural landscapes). These basic geographic aspects have a strong influence on the composition of the Southern Hemisphere troposphere, leading to a dominance of emissions from biogenic and fire sources on land alongside oceanic influences, especially from the Southern Ocean. Thus, the cleanest atmospheric conditions on the planet are to be found in the Southern Hemisphere, which may more closely resemble preindustrial conditions. In remote locations, the background atmospheric chemistry may be quite different from what is observed in the Northern Hemisphere, with low concentrations of nitrogen oxides making peroxyl radical chemistry of greater importance. Much of the Southern Hemisphere atmosphere is vastly undersampled, particularly around the Southern Ocean, Antarctica, and sub-Saharan Africa.

Historically, the atmospheric chemistry issue that has received most attention in the Southern Hemisphere is the ozone hole that is observed over Antarctica in austral spring (Farman et al., 1985). Total column ozone observations have played a pivotal role in understanding the development of the ozone hole and remain important for monitoring the expected ozone recovery in coming years (Solomon et al., 2016); however, aging infrastructure in some stations negatively impacts this important research area. In addition to total column measurements, balloon-borne ozonesondes are launched from several stations in the Southern Hemisphere (Thompson et al., 2003). Surface ozone photochemistry is substantially influenced by the fact that there is 41% more surface ultraviolet radiation in summer in the Southern Hemisphere than in the Northern Hemisphere due to the Earth's orbit being closer to the sun in the austral summer (McKenzie et al., 2006). Surface ozone measurements are mostly conducted in urban areas for air quality monitoring, although stations that are part of the World Meteorological Organisation's Global Atmosphere Watch (GAW) monitor ozone over longer timescales in marine environments (Cape Point, Cape Grim, Amsterdam Island and Baring Head) and other remote locations (Lauder and Arrival Heights; Cooper et al., 2020). These observations indicate that ozone chemistry differs between urban and marine environments and between hemispheres in the marine boundary layer in terms of seasonality and annual variation (Derwent et al., 2016). More observations are required to identify the underlying factors for the observed ozone dynamics.

The advancement of atmospheric chemistry in the Southern Hemisphere is hindered by a sparsity of observation sites (see **Figure 1**). This sparsity is driven by logistical issues, with the Southern Hemisphere containing only approximately a third of the global land area and a 10th of the global population. As an example of the paucity of surface measurements, only 3 of the 15 Advanced Global Atmospheric Gases Experiment (AGAGE) stations are located in the Southern Hemisphere (Mt. Mugogo, Cape Matatula, and Cape Grim), along with 9 of the 30 GAW global stations (Samoa, Ushuaia, La Réunion, Cape Grim, Lauder, Cape Point, Neumayer, Halley, and South Pole) and only 17% of the more than 400 global and regional GAW stations.

The current generation of satellite-based sensors provides improved observation density and better coverage for the Southern Hemisphere than can be achieved from ground-based instruments alone. Nevertheless, the satellite sensors require ground-based remote sensing instruments to validate their products within the Southern Hemisphere due to differences in viewing geometry and albedo that can impact retrieval accuracy (Velazco et al., 2019). As for the in situ networks, the number of ground-based remote sensing facilities in the Southern Hemisphere is limited. For example, the Total Carbon Column Observing Network (TCCON) (Wunch et al., 2011), which is used for validating satellite greenhouse gas measurements, has only 5 of the 23 stations in the Southern Hemisphere: Darwin, Wollongong, Lauder, Reunion, and Ascension Island. This means that there are no permanent TCCON sites in South America, mainland Africa, or Antarctica, hampering validation over much of the Southern Hemisphere landmass. Similarly, of the 64 current Baseline Surface Radiation Network sites, only 13 are in the Southern Hemisphere (Alice Springs, Brasilia, Cocos Island, Darwin, Florianopolis, Gobabeb (Namibia), Neumayer, Lauder, Momote, Nauru, Rolim de Moura, Sao Martinho da Serra, and Syowa), along with only 18% of the AErosol RObotic NETwork (AERONET) sites used to validate aerosol optical depth retrievals from satellite measurements. The situation is exacerbated by the fact that the next generation of geostationary satellites has been designed to provide detailed coverage of the Northern Hemisphere only, which will again disadvantage our understanding of Southern Hemisphere tropospheric chemistry.

Scientists studying the Southern Hemisphere atmosphere face a number of common challenges but have historically lacked a vehicle for sharing ideas and opportunities to collaborate. Recently, the International Global Atmospheric Chemistry (IGAC) project has established a Southern Hemisphere Working Group in order to meet this need. This article has been written by members of this group with the aim of presenting the current knowledge and challenges facing atmospheric chemistry in the Southern Hemisphere. We first discuss the importance of both biogenic volatile organic compounds (BVOCs) and fires in the Southern Hemisphere and then the common factors

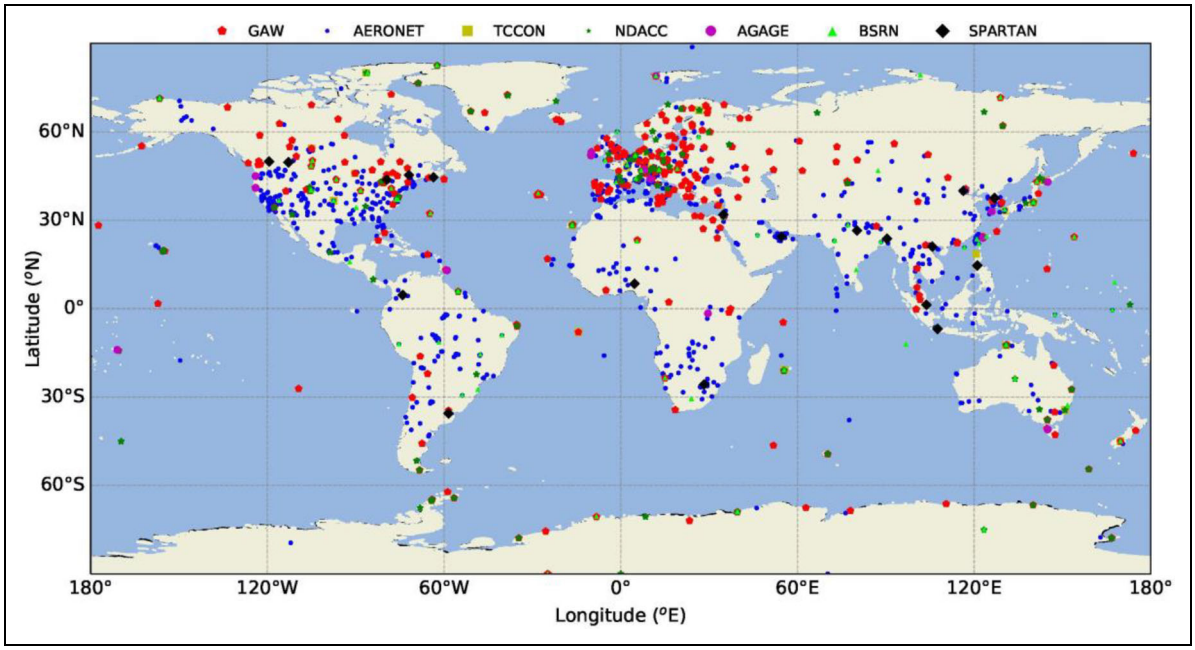

Figure 1. Global measurement networks. The percentage of sites located in the Southern Hemisphere for each network is Global Atmosphere Watch—17%, AErosol RObotic NETwork—18%, Total Carbon Column Observing Network—22%, Network for the Detection of Atmospheric Composition Change—32%, Advanced Global Atmospheric Gases Experiment—20%, Baseline Surface Radiation Network—27%, and Surface Particulate Matter Network—16%. DOI: https://doi.org/10.1525/elementa.2021.00050.f1

impacting air quality in Southern Hemisphere cities and regions. Finally, we conclude with a discussion of the Southern Ocean and associated atmospheric chemistry and outline the aims of the IGAC Southern Hemisphere Working Group. This article is not intended to be a comprehensive review of Southern Hemisphere research in tropospheric chemistry, as it has slightly more focus on aerosols and their precursors than ozone and its precursors; nor does it cover the complex environment of Antarctica, which is more the remit of IGAC's Cryosphere and ATmospheric Chemistry initiative project (Thomas et al., 2019). Instead, we focus on the research unique to the Southern Hemisphere that is needed to address environmental and societal issues that are influenced by tropospheric chemistry.

2. The importance of BVOCs in the Southern Hemisphere

The Southern Hemisphere is rich in vegetation, from the dense rainforests of the Amazon and central Africa to the eucalypt-dominated southeast coast of Australia. Vegetation emits BVOCs as a protection mechanism against changes in light, temperature, carbon dioxide (CO_2), drought, and herbivory; hence in a warming climate, the BVOC flux will change. BVOCs react chemically in the atmosphere, forming ozone, and secondary organic aerosol (SOA; Hoyle et al., 2011). It is therefore important to understand the biogenic contribution and the interactions with anthropogenic emissions. In this section, we describe findings from biogenic field and modeling studies that have focused explicitly on the Southern Hemisphere.

Various models have been developed to predict emissions of BVOCs. One of the most widely used Model of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2012) predicts global mean annual BVOC emissions of 760 Tg C yr^{-1} (1980–2010), with isoprene as the major contributor (Sindelarova et al., 2014). Most of the isoprene is emitted by broadleaf trees and shrubs, which dominate in the tropics (**Figure 2**). The equator falls in the middle of this high-emission region, but even so, the geographically defined Southern Hemisphere emits 10% more isoprene than the Northern Hemisphere.

2.1. Biogenic emissions and chemistry in South America

In South America, the Amazon Basin has been the focus of several local and international studies due to its large geographical extent and potential influence on global climate. The number of studies for the region is increasing, and therefore, the availability of data on VOC emissions is also increasing (see **Table 1**). The tropical South American region has become a good case study for testing algorithms to constrain isoprene emissions from satellite data (e.g., Barkley et al., 2013).

Isoprene and monoterpene emissions in the Amazon have been observed to have a light dependence (Rinne et al., 2002; Richter et al., 2016). However, the extent of light dependence of BVOC emissions in other Southern Hemisphere environments is still uncertain. At the Amazon Tall Tower Observatory, the diurnal cycle in monoterpene mole fraction varied by chemical species, with α-pinene more prevalent in the day and limonene more prevalent at night (Yáñez-Serrano et al., 2018), suggesting a light

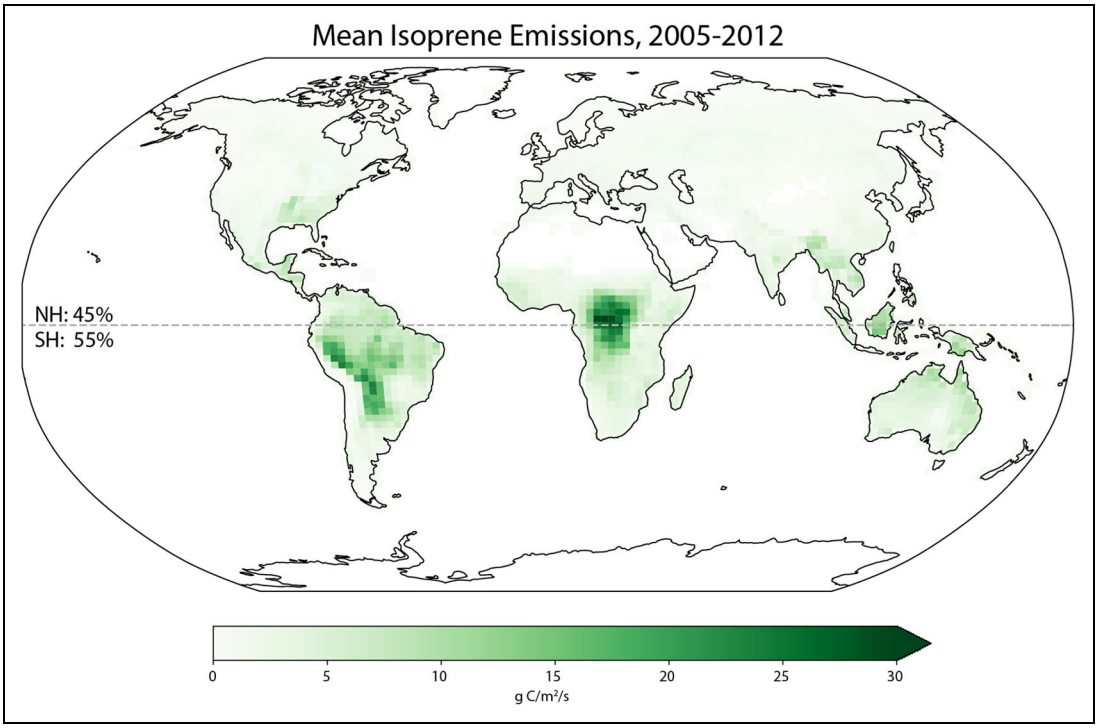

Figure 2. Annual mean isoprene emissions averaged over 2005–2012 predicted by the Model of Gases and Aerosols from Nature model as implemented in the Goddard Earth Observing System chemistry model (GEOS-Chem), along with the percentage of the total that is emitted in each hemisphere. DOI: https://doi.org/10.1525/elementa.2021.00050.f2

dependency of emission fluxes. Monoterpene emissions by plants may also vary by plant type and geographical region, according, for example, to Bracho-Nunez et al. (2013) who identified fewer monoterpene-emitting plants in the Amazon compared to the Mediterranean.

Aircraft measurements over the Central Amazon measured maximum isoprene and α-pinene mole fractions of 6.6 and 0.6 ppb, respectively, over the forest surface during the Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA)-Cooperative LBA Airborne Regional Experiment (LBA-CLAIRE-2001) field campaign (Kuhn et al., 2007). A single column model for the same period overpredicted isoprene by a factor of 2, suggesting a missing source of hydroxyl radicals (Kuhn et al., 2007). Aircraft measurements during the GOAMAZON campaign also showed that rapid oxidation of isoprene occurs when biogenic plumes from the Amazon interact with urban air masses of Manaus (Martin et al., 2017). Urban atmospheric chemistry accelerates oxidant cycling, producing ozone and increasing concentrations of isoprene oxidation products (da Silva et al., 2018).

Biogenic sources can contribute 70%–90% of all particle formation in the Amazon Basin (Graham et al., 2003), which can act as cloud condensation nuclei (CCN; Krejci et al., 2005; Ahlm et al., 2009). Fine and ultrafine particles were measured in Brazil showing strong biogenic influences, particularly isoprene-derived organosulfates in the presence of biomass burning and other anthropogenic emissions (Pauliquevis et al., 2012; Kourtchev et al., 2016; Wu et al., 2019). Interactions with anthropogenic NO_x from Manaus contributed to a 60%–200% enhancement of biogenic SOA in the Amazon via increased production of oxidants (Shrivastava et al., 2019). In Bolivia, new particle formation at high altitude can be influenced by the transport of organic compounds from the Amazon rainforest (Rose et al., 2015). Chamber measurements have also shown up to 10 μg m^{-3} of biogenic SOA formation in tropical ambient air (Palm et al., 2018). Biogenic measurements are not homogeneous across South America. Available data are more abundant from Amazonian and Andean regions, with only a few measurements in urban areas. Due to the broad meteorological conditions across the entire continent, more ground measurements would be required to improve model estimates and generate more accurate results regarding the atmospheric biogenic emissions.

2.2. Biogenic emissions and chemistry in Africa

Africa is a large source of BVOCs. While biomass burning emissions are often the focus of African research, there are strong indications that emissions of BVOCs are just as important. For example, Meyer-Arnek et al. (2005) found that when simulating formaldehyde concentrations over Africa for September 1997, there were similar amounts of formaldehyde generated from biogenic emissions (0.033 Tg) as from Africa-wide biomass burning (0.028 Tg). Aghedo et al. (2007) found in a simulation that biogenic emissions of VOCs, carbon monoxide (CO), and nitrogen oxides (NO_x) impact the global total ozone burden more than biomass burning, lightning, and anthropogenic emissions.

In general, biomass burning emissions are expected to have a greater impact on total emissions and ozone over

Table 1. Biogenic volatile organic compound emissions and mole fraction measurements in South America. DOI: https://doi.org/10.1525/elementa.2021.00050.t1

Region	Emission or Ambient Mole Fraction	Reference
Tropical South America	Isoprene (Global Ozone Monitoring Experiment):	Barkley et al. (2008)
	Annual mean: 15.1 (range = 8.2–23.0) Tg C yr^{-1}	
	Isoprene (Model of Gases and Aerosols from Nature):	
	Annual mean: 23.7 (range = 15.7–38.8) Tg C yr^{-1}	
Tropical South America	Isoprene: 144 Tg C yr^{-1}	Sindelarova et al. (2014)
Amazon	Isoprene: 2–10 ppb (dry season) and 1–6 ppb (wet season)	Wei et al. (2018)
	Emission rate: up to 4 mg m^{-2} h^{-1} (wet season) and up to 11 mg m^{-2} h^{-1} (dry season)	
Argentina (Northeastern Eucalyptus forest)	Isoprene: Daytime maximum 7.9 mg m^{-2} h^{-1}	Richter et al. (2016)
Argentina (Patagonia)	Monoterpenes: 1.3 ± 0.15 to 3.9 ± 0.64 mg m^{-2} h^{-1} (summer)	Centritto et al. (2008)
Brazil (Central Amazon near Manaus)	Isoprene: Mean of 12.1 ± 4 mg m^{-2} h^{-1} (dry season)	Karl et al. (2007)
Brazil (Rondonia)	Isoprene: 2.2–6.7 ppb (wet season) 9.8 mg m^{-2} h^{-1}	Greenberg et al. (2004)
Brazil (Central Amazon)	Isoprene: mean of 1.37 ± 0.7 mg m^{-2} h^{-1} (dry season)	Alves et al. (2016)
Brazil (Central Amazon)	Monoterpenes: Mean of 0.96 ± 0.27 ppb (dry season)	Yanez-Serrano et al. (2018)
Brazil (Central Amazon—GoAmazon)	Total sesquiterpenes + diterpenes: Mean of 41.8 ng m^{-3} (wet season) and 38.8 ng m^{-3} (dry season)	Yee et al. (2018)
Brazil (Central Amazon—GoAmazon)	Isoprene: Mean of 0.82 ppb (wet season) and 2.37 ppb (dry season)	Shilling et al. (2018)
Brazil (São Paulo)	Isoprene: Mean of 0.08 ppb (summer) and 0.06 ppb (winter)	Alvim et al. (2018)
Brazil (Rio de Janeiro during 2014 soccer World Cup)	Isoprene: 0.14–0.84 ppb (winter)	Souza et al. (2016)
Colombia (urban area—Bogotá)	Isoprene: 0.2–4.1 ppb (summer)	Franco et al. (2015)
Ecuador	Modeled annual isoprene: 1.1 Tg C yr^{-1}, 0.028–0.086 Tg C $month^{-1}$ (dry season), 0.092–0.118 Tg C $month^{-1}$ (wet season)	Viteri et al. (2017)
	Modeled annual monoterpenes: 3.8 Tg C yr^{-1}, 0.030–0.034 Tg C $month^{-1}$ (dry season), 0.028–0.033 Tg C $month^{-1}$ (wet season)	
Peru (Amazon)	Isoprene: 3.0–8.2 mg m^{-2} h^{-1} (dry season)	Helmig et al. (1998)

Note that the term "ppb" refers to the mole fraction nmol/mol.

Africa during the biomass burning season, but not at other times of the year. In tropical Africa, there is little seasonal variability in BVOC emissions, and thus biogenic emissions are important throughout the year. The southern savannahs, however, show a BVOC minimum in winter (Otter et al., 2003; Marais et al., 2014). Woodlands are the largest annual source of BVOCs in Africa south of the equator (Otter et al., 2003). Rainforests cover a relatively small part of Southern Hemisphere Africa (3.5%) but have a high emission rate with little seasonal variation (Otter et al., 2003).

There are still large uncertainties in BVOC emissions due to a lack of measurements and the need to characterize emissions in the context of the land surface and vegetation types. In the early 1990s, the Southern African Fire-Atmospheric Research Initiative (SAFARI) campaign (Lindesay et al., 1996) provided BVOC emission estimates for Africa (despite the campaign's focus on biomass burning). However, limited research has occurred since then in Southern Hemisphere Africa, and large gaps remain in understanding southern African BVOC emissions (Sowden et al., 2007).

Estimates of isoprene emissions for the entire African continent range from 60 to 150 Tg yr^{-1} (see **Table 2**). It is challenging to evaluate how much isoprene relates only to the Southern Hemisphere portion of Africa. There are emissions from western Africa in the Northern Hemisphere; however, a large fraction of the forests in equatorial and southern Africa is located within the Southern Hemisphere (**Figure 2**). Marais et al. (2012) found African

Table 2. BVOC emissions in Africa (from studies that include or focus on Southern Hemisphere Africa). DOI: https://doi.org/10.1525/elementa.2021.00050.t2

Region	Emission	Reference
Africa	Isoprene: 60 Tg C yr^{-1} (OMI)–77 Tg C yr^{-1} (MEGAN)	Marais et al. (2012)
Africa	Isoprene: 130 Tg C yr^{-1} (approximately 25% of global emissions)	Sindelarova et al. (2014)
Africa	Isoprene: 77 Tg C yr^{-1} (OMI)–104 Tg C yr^{-1} (MEGAN v2.1)	Marais et al. (2014)
Africa south of equator	BVOCs from savannahs: 18–74 Tg C yr^{-1}	Otter et al. (2002)
Africa south of equator	BVOCs: 80 Tg C yr^{-1} (70% isoprene, 22% oxygenated VOCs, 6% stored monoterpenes, and 3% light-dependent monoterpenes)	Otter et al. (2003)
Africa south of equator	Isoprene: 59 Tg C yr^{-1} Monoterpenes: 11.8 Tg C yr^{-1} Other VOC: 23.4 Tg C yr^{-1}	Guenther (1995; as reported in Otter et al., 2003)

BVOC = biogenic volatile organic compound; MEGAN = Model of Gases and Aerosols from Nature; OMI = Ozone Monitoring Instument.

isoprene emissions derived using the Ozone Monitoring Instument (OMI) satellite formaldehyde observations were 22% lower than those predicted by the MEGAN model, with significant spatial variability in these differences. The maximum isoprene emissions in MEGAN are located further north than indicated by the OMI formaldehyde. The OMI-derived estimates were 43% lower than MEGAN in central African rainforests and 21% higher in the southern deciduous broadleaf forests.

There have been very few BVOC flux measurements in Africa. For example, in Marais et al. (2014), there were flux measurements from campaigns available for only 10 sites across all of sub-Saharan Africa for comparison to the satellite-derived results (see **Table 2**). The small number of locations with data highlights the dearth of information on BVOC fluxes. In addition, these data are limited in their spatial and temporal coverage. The campaigns did not incorporate all seasons and took place at different times of year (and in different years), thereby introducing an extra degree of variability in these observations.

BVOC measurements were conducted over several years at Welgegund, a regional background site situated in grazed savannah–grassland–agricultural landscape in South Africa (Jaars et al., 2016). BVOCs from these grassland savannah measurements were substantially lower compared with measurements from landscapes with more woody vegetation. Annual median mole fractions of isoprene, 2-methyl-3-butene-2-ol (MBO), monoterpenes and sesquiterpenes were 14, 7, 120, and 8 ppt, respectively, from February 2011 to February 2012, and 14, 4, 83, and 4 ppt, respectively, between December 2013 and February 2015 (where the term "ppt" refers to the mole fraction pmol/mol). The total monoterpene concentrations were an order of magnitude higher than the concentrations of other BVOC species, with α-pinene being the most abundant. The highest BVOC concentrations occurred during the wet season, coinciding with increased soil moisture levels and temperatures. Soil moisture had the most significant impact on atmospheric levels of MBO, monoterpenes, and sesquiterpenes, while temperature had the greatest influence on isoprene levels.

The urban fringes, where biogenic and anthropogenic emissions mix, promote atmospheric chemistry not seen in other biogenic regions. In a regional site in South Africa, BVOCs were shown to drive the seasonal cycle of the growth rate of new particle formation (Vakkari et al., 2011) while sulfuric acid was found to drive the occurrence of new particle formation, implying that aerosol formation and growth processes are independent of one another. Ambient measurements from this permanent site later identified that the VOCs measured had both biogenic and anthropogenic origins (Jaars et al., 2018). In the Cross Border Air Pollution Impact Assessment project in southern Africa, Zunckel et al. (2006) found biogenic and anthropogenic VOCs are both important for accurate modeling of ozone concentrations.

2.3. Biogenic emissions and chemistry in Australasia

There are few field campaigns focused on BVOC measurements in Australia and none reported in New Zealand. Aircraft measurements near Darwin in tropical northern Australia yielded very low isoprene, while α-pinene was below the limit of detection (Allen et al., 2008). Low isoprene mole fractions were also measured at the Cape Grim Baseline Air Pollution Station on the northwestern tip of Tasmania, with 4 ppt under baseline conditions (Lewis et al., 2001) and peaks up to 15 ppt (Lawson et al., 2007). Higher amounts of isoprene and monoterpenes have been measured in greater metropolitan Sydney, peaking at up to 7 ppb in some locations (Paton-Walsh et al., 2017; Paton-Walsh et al., 2018; Guérette et al., 2019; Keywood et al., 2019). Sydney is surrounded by eucalypt forests and is dominated by BVOC-influenced airmasses in summer (Utembe et al., 2018). Measurements within those eucalypt forests revealed summer BVOC concentrations of up to 8 ppb (dominated by isoprene) with a winter minimum of 0.1 ppb (Ramirez-Gamboa et al., 2021). Among the

monoterpene species, eucalyptol and p-cymene were dominant (75% and 20%, respectively), whilst α-pinene made only a minor contribution at 5% (Ramirez-Gamboa et al., 2021).

Eucalyptus species are a dominant feature of the Australian vegetated landscape. Most emission experiments using *Eucalyptus* species have been conducted on individual laboratory specimens in the Northern Hemisphere and not on the mix of vegetation found in real-world Australian environments. One Australian study by He et al. (2000) using 15 potted *Eucalyptus* species inside a chamber showed a clear temperature dependence for emission of monoterpenes but no light dependence. The lack of light dependence for monoterpene emissions could be a unique trait in Australian trees (i.e., compared to South America where light dependency has been observed, see Section 2.1). Modeling produced more accurate simulations of monoterpene concentrations in southeast Australia when the light dependence of their emission was removed (Emmerson et al., 2018).

In another trait unique to Australia, the ratio of the total amount of carbon in isoprene to that in monoterpenes in Australian atmospheric measurements is in balance (neither dominates; Emmerson et al., 2016). The particle formation potential in this carbon-balanced environment has not been observationally constrained, but Suni et al. (2008) observed significant nighttime SOA formation when air masses originated from eucalypt-rich regions, again suggesting a light-independent emission source.

Sørensen et al. (2020) identified over 100 BVOC compounds from 9 species of mature *Eucalyptus* trees in southeast Australia. They identified different monoterpene compounds stored in the leaf reservoirs to those that were emitted to the atmosphere by the same leaves and that these compounds differed among the *Eucalyptus* species. This finding suggests that each *Eucalyptus* species regulates its own formation, storage, and emission of BVOCs. Similarly, Maleknia et al. (2009) demonstrated that the suite of monoterpene compounds emitted from *Eucalyptus grandis* leaves changes as they undergo pyrolysis and that storm damage to trees increased monoterpene emission 4-fold.

Modeling studies have shown large differences between biogenic emission inventories for the broader Australia/New Zealand region (Zeng et al., 2015; Bauwens et al., 2016) and large discrepancies between modeled BVOCs and atmospheric observations (Emmerson et al., 2016). Inclusion of soil moisture dependency in a model suppressed BVOC emissions by up to 70%, improving the model comparison to observations (Emmerson et al., 2019). These results suggest Australia's drought conditions have an important influence on local atmospheric composition.

Climatic conditions beyond drought also have the potential to influence BVOC emissions and impacts in Australia. Measurements from mature *Eucalyptus* trees grown in a 550 ppm CO_2 atmosphere showed a 15% decrease in isoprene and a 62% increase in monoterpene concentrations compared to trees grown in current CO_2 conditions, indicating how emissions might change by 2050 under climate change (Jiang et al., 2020a). Combining higher CO_2 with higher projected temperatures representative of a 2050 climate could increase summertime isoprene by 2 ppb in Australia from a daytime average initial value of 1.2 ppb, causing localized average ozone to rise by 4–8 ppb (Emmerson et al., 2020).

2.4. Future research priorities for our understanding of Southern Hemisphere biogenic emissions

The Southern Hemisphere emits the majority of global BVOCs, yet there remain insufficient measurements to accurately quantify their total contribution. Recently, a technique to directly infer isoprene emissions from satellite data was demonstrated in the Amazon (Fu et al., 2019) and then extended globally (Wells et al., 2020). Broadly, the technique uses the differences in the brightness temperature between the peak of the isoprene infrared absorption band (approximately 893 cm^{-1}) and nearby off-peak bands (approximately 894–895 cm^{-1}). The satellite results suggested missing sources of isoprene in existing inventories for southern Africa and that isoprene seasonal cycles are not well represented in models. For example, maximum measured isoprene occurred several months later than expected in the Amazon, and the measured summertime isoprene peak in Australia persisted well into the autumn rather than declining as expected. Further analysis of these new top-down constraints on isoprene is a priority as it will improve our understanding of the spatial and temporal fluctuations in Southern Hemisphere isoprene emissions. In addition, other BVOC emissions such as monoterpenes will need to be parameterized based on limited in situ measurements. Hence, further in situ measurements to characterize the emissions of BVOCs from Southern Hemisphere ecosystems are a priority for future research. Measurements are needed that can characterize emissions in terms of land surface and vegetation types and include seasonal variation and/or the light and temperature dependence of the emissions.

3. The importance of fire in the Southern Hemisphere

Fires are a major source of trace gases and aerosols to the atmosphere and are part of the biome in many regions of the Southern Hemisphere. These biomes include tropical and temperate forests as well as different types of savannah, shrublands, and grasslands. Fires in the savannah dominate in terms of total area burned in Africa, South America, and Australia (e.g., Giglio et al. [2013] estimated 90%, 82%, and 37%, respectively).

In South America, a large fraction of fires result from human activity. Most of the fires occur between July and October in Central South America on a very regular annual basis due to slash-and-burn practices of primary and secondary Amazonian rainforest as well as pasture maintenance activities (Artaxo et al., 2002; Soares-Filho et al., 2006; Alves et al., 2009; Bowman et al., 2009; Walker et al., 2009; Martin et al., 2010; Andreae et al., 2012). Other hot spots for fire activity in South America are the Amazon-adjacent Cerrado biome as well as sugar cane

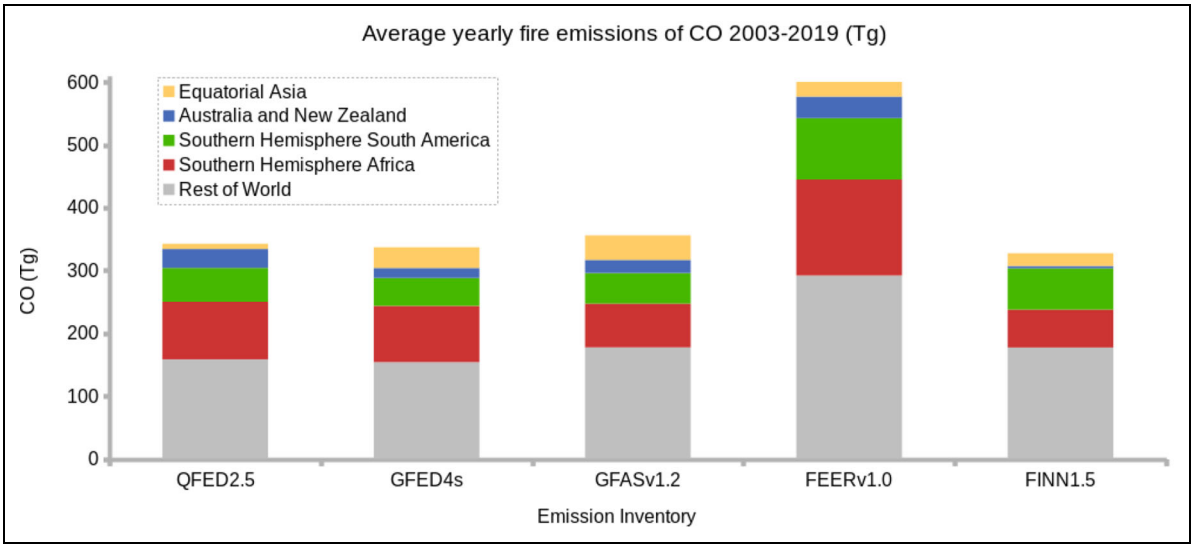

Figure 3. Average yearly fire emissions of carbon monoxide between 2003 and 2019 from the 5 commonly used global emission inventories, separated into Southern Hemisphere regions (plus Equatorial Asia) and the rest of the world, with regional boundaries as defined by Giglio et al. (2010). Numbers and letters after the base inventory names denote versions. DOI: https://doi.org/10.1525/elementa.2021.00050.f3

plantations in the southeast, south, and coastal northeast. Libonati et al. (2015) showed that typically 30%–50% of the annually burned area within the Amazon lies within the Cerrado biome.

In Southern Africa, around 70% of the ecosystems are fire-adapted (Lehmann et al., 2011), and two seasonal patterns of fire exist: one during the dry summer months in the winter–rainfall areas of South Africa (i.e., Western Cape) and one during the dry winter months elsewhere. For the latter, fires begin earlier in the northwest of the region and progress to the southeast of the region (Archibald et al., 2010). Central Africa has the highest fire frequency and the most extensive burned area in the world, with African savannah fires comprising the largest proportion (71%) of areas burned globally (Jiang et al., 2020b). Human influences are minor in the rainforest but are a major contributor to fires in the savannahs and grasslands (Zhou et al., 2014).

Much of Australia's vegetation is prone to burning due to its highly flammable foliage, litter and oils, and fire is an important part of the regeneration of many species and ecological communities (Metcalfe and Bui, 2016). Different fire regimes exist in northern and southern Australia (Russell-Smith et al., 2007). In northern Australia, where a wet and a dry season exist, the savannah landscape experiences fire in the dry season (April to October), while in southern Australia, the temperate forests predominately burn in the summer months (October to March). Anthropogenic fire activities include traditional land management (Altman, 2009; Price, 2015; Ansell et al., 2020), prescribed burning to reduce fuel loads, and agricultural burning.

3.1. Estimating fire emissions

Although the Southern Hemisphere contains less than half the landmass of the Northern Hemisphere, Southern Hemisphere fires are a major source of atmospheric pollution and produce about half of all global fire emissions (**Figure 3**). Two methods for estimating fire emissions are often used in computational modeling. The first method combines burned area estimates with knowledge of vegetation type, biomass loading, and burning completeness to calculate the amount of carbon burnt during a fire (Voulgarakis and Field, 2015). The second method uses fire intensity quantified as fire radiative power, combined with biome scaling factors, to calculate carbon burnt (Voulgarakis and Field, 2015). Once the fuel consumption is known, emission factors collated from field campaigns and laboratory studies are used to calculate emissions of trace gases and particles released into the atmosphere (Akagi et al., 2011). Global emission inventories use satellite observations of fire and vegetation parameters, and emission factors are aggregated into 4–6 vegetation types. The five commonly used global emission inventories are the two burned area-based inventories: Global Fire Emissions Database (GFED; Van Der Werf et al., 2017) and Fire INventory from National Center for Atmospheric Research (Wiedinmyer et al., 2011); and the 3 fire radiative power-based inventories: Quick Fire Emission Dataset (QFED; Darmenov and da Silva, 2015), Global Fire Assimilation System (GFAS; Kaiser et al., 2012), and Fire Energetics and Emissions Research (Ichoku and Ellison, 2014).

Carbon from fires is mainly released as CO_2 and CO (95%; Ward and Hao, 1991), and CO is a useful tracer to summarize yearly and regional differences in fire emissions that can generally be related to the variability of wildfire. The 5 commonly used inventories generally agree that the Southern Hemisphere produces about 50% of the global annual fire emissions of CO, ranging from 150 to 290 Tg yr^{-1} of CO (see **Figure 3**). Southern Africa provides the largest contribution to Southern Hemisphere fire emissions at 60–153 Tg yr^{-1}, followed by South America

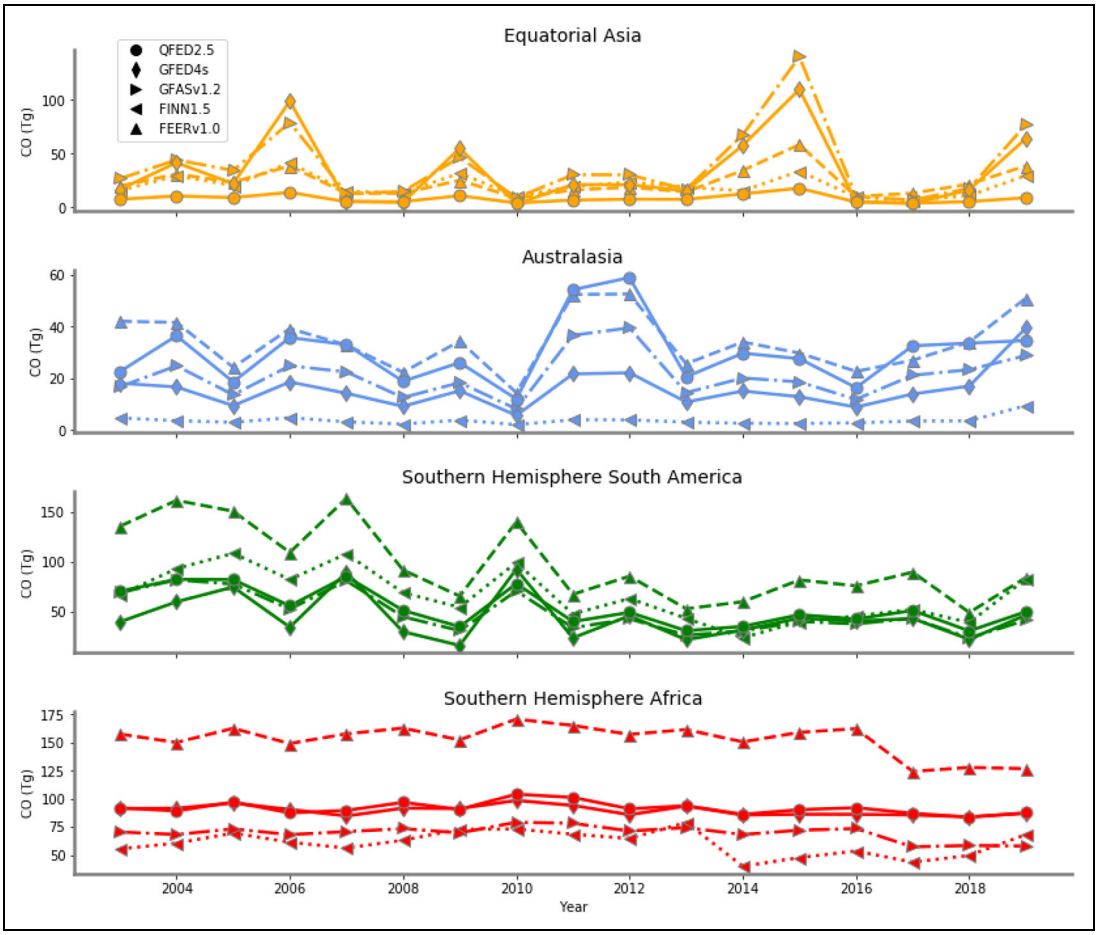

Figure 4. Yearly fire emissions of carbon monoxide between 2003 and 2019 from the 5 main global emission inventories in Southern Hemisphere regions (plus Equatorial Asia). Note the different scales. DOI: https://doi.org/10.1525/elementa.2021.00050.f4

at 44–98 Tg yr^{-1} (**Figure 3**). Australian and Equatorial Asian contributions differ in relative importance depending on the inventory. For example, QFED suggests that fire emissions of CO from Australia are on average higher than from Equatorial Asia (30 vs. 8 Tg yr^{-1}, respectively), while GFED suggests the opposite (16 vs. 33 Tg yr^{-1}, respectively; **Figure 3**). Inventories also generally agree on the interannual variability and temporal tendency in each region (see **Figure 4**).

Globally, fires decreased between 1998 and 2015 (Andela et al., 2017), with the strongest trends in tropical regions, driving a decrease in emissions. The large interannual variability of fire emissions often prevents the analysis of statistically significant trends over fire regions given the short time series of emissions developed from satellite observations. This large regional variability likely contributes to the lack of easily discernible downward tendencies in **Figure 4**. South America experienced a downward trend in fire emissions until around 2013 when the trend flattened. From 2018, there has been a significant increase in burnt area in the Amazonian region which will also be reflected in the emissions (Feng et al., 2021). Australia may also have experienced a downward trend in the emissions until around 2010, potentially followed by an upward trend since 2016.

The different inventories produce a large spread in CO emission estimates within each region, with no consistent bias between the inventories across regions (see **Figure 4**). For example, QFED generally produces some of the highest emissions in Australasia and the lowest emissions in Equatorial Asia. It is reasonable to assume the inconsistent spread will be similar for other species because emissions of other trace gases and aerosols are typically calculated relative to CO. The aerosol species (organic carbon and black carbon) were found to have similarly inconsistent variability between regions (Pan et al., 2020; Liu et al., 2021). Pan et al. (2020) compared emissions used in a global model with satellite and ground-based aerosol optical depth measurements in April and September 2008 and generally found underestimates for all inventories in the Southern Hemisphere.

Accurate fire emission estimates are important in the Southern Hemisphere due to their major contribution to atmospheric composition. Inventory differences imply that each region should be analyzed separately to identify the most accurate fire emissions. Comparisons need to be extended to longer time series and to emitted species besides aerosols. Future work will need to relate the accuracy of emissions to emission factors, the underlying vegetation profile, fire detection, and carbon burnt to

improve inventories. For example, Liu et al. (2020) analyzed inventory estimates for Equatorial Asia and deduced that GFAS was the best inventory because it used a cloud-clearing technique and could account for small, fragmented fires. Other improvements needed for more accurate fire emissions estimates include adding diurnal cycles and plume rise.

3.2. Impacts of fire emissions on atmospheric chemistry

Biomass burning smoke contains a complex and dynamic mixture of particles and gases that undergo chemical transformation and transportation in the atmosphere. Changes in photochemical activity that occur in smoke plumes can change the composition and microphysical properties of aerosol, potentially leading to changes in the optical and hydroscopic (and thus radiative) properties of the aerosol. It is important to simulate these processes accurately in climate models and chemical transport models.

Much of our current understanding of the importance of biomass burning emissions for global atmospheric chemistry stems from a series of international and interdisciplinary research campaigns on biomass burning in tropical, subtropical, and boreal biomes conducted during the 1990s as part of the International Geosphere-Biosphere Program IGAC Program (Keywood et al., 2013). Many of these experiments were focused on southern Africa (e.g., the Southern Africa Fire-Atmosphere Research Initiative [SAFARI]; Lindesay et al., 1996; Swap et al., 2003). During the 2000s, this large-scale international collaborative effort was replaced by numerous small-scale projects and campaigns (e.g., Wooster et al., 2011). The global nature of fire and the persistence of scientific questions and uncertainties on emissions and radiative effects all highlighted the need for a move back to coordinated international efforts including large-scale facilities (Kaiser and Keywood, 2015). Recently, in a follow up to the airborne measurements of SAFARI, there have been several large-scale field campaigns that have included a focus on the chemical and radiative properties of biomass burning smoke from central Africa and its transport over the Southeast Atlantic (Zuidema et al., 2016; Formenti et al., 2019; Haywood et al., 2021; Redemann et al., 2021). These campaigns used aircraft and ground-based observations to achieve a large-scale picture of the biomass burning aerosols, including aging, and the process-level interactions of the aerosols with radiation and low-level clouds. The campaigns provided new model-measurement comparison exercises (including remote sensing; e.g., Pistone et al., 2019; Doherty et al., 2021) and the momentum for the development of novel satellite products and new regional climate simulations (Mallet et al., 2019; Shinozuka et al., 2020; Chauvigné et al., 2021; Peers et al., 2021).

The presence of reactive species in fire emissions results in a significant influence of smoke on atmospheric chemistry. In fresh plumes, NO_x species titrate ozone leading to low ozone concentrations (Crutzen and Andreae, 1990); however, in aged plumes, high concentrations of secondary pollutants including ozone are observed. These relationships have been observed in Africa; for example, Capes et al. (2008) observed ozone formation in aged biomass burning plumes over West Africa. In southern Africa, Vakkari et al. (2014) observed a large increase in the aerosol scattering coefficient in aged plumes and an increase in the oxidation of organic aerosol in daytime plumes relative to fresh nighttime plumes. However, while ozone formation was only observed in 13% of the daytime plumes in Vakkari et al. (2014), changes in the size distribution, chemical composition, single-scattering albedo, and hygroscopicity of biomass burning particles in the daytime plumes were driven by atmospheric oxidation and subsequent secondary aerosol formation. Vakkari et al. (2018) also found that, on average, the mass of submicron aerosols more than doubles during 3 h of daytime aging. Recently, Wu et al. (2020) showed that biomass burning aerosol from the west coast of southern Africa, transported for 7 days into the Atlantic Ocean, consisted of highly oxygenated organic compounds but was also rich in black carbon, resulting in a low single scattering albedo relative to aged biomass burning aerosol observed elsewhere.

Central South American fires emit smoke plumes that cover hundreds of thousands of hectares every year over central Brazil and western Bolivia and Peru (Freitas et al., 2005). The smoke impacts the radiation budget and cloud formation in the region and leads to deterioration of the air quality in many inland cities and smaller villages, affecting not only human health (Schwartz et al., 2002; Barregard et al., 2006; Ignotti et al., 2007) but also visibility and closure of inland airports for many weeks (Andreae et al., 2001; Andreae et al., 2004; Barregard et al., 2006; Hoelzemann et al., 2009; Longo et al., 2009). The smoke plumes are regularly transported over the continent, first mostly to the south and then over southern and southeastern cities and megacities including São Paulo, Rio de Janeiro, Montevideo, and Buenos Aires. From there, the pollutants often travel over the South Atlantic and contribute, together with Southern Hemispheric African smoke emissions, to the formation of tropospheric ozone over the region, with elevated ozone concentrations typically observed every September (Andreae and Merlet, 2001; Artaxo et al., 2002; Freitas et al., 2005; Freitas et al., 2007; Hoelzemann et al., 2009).

A number of observational biomass burning studies conducted in the Amazon have reported a consistent aerosol size distributions for biomass burning aerosols, with modal diameter of around 100 nm (Andreae et al., 2004; Rissler et al., 2004; Rissler et al., 2006; Artaxo et al., 2013; Brito et al., 2014). Hygroscopicity measurements showed that the biomass burning particles were externally mixed (Rissler et al., 2004, 2006) with very low hygroscopic and moderately low hygroscopic populations. Despite this low hygroscopicity, the biomass burning aerosol acted efficiently as CCN under conditions of high supersaturation relative to water vapour (Sánchez Gácita et al., 2017). During the CLAIRE-2001 experiment, fresh biomass burning aerosol showed greater hygroscopicity than aged biomass burning aerosol, which may have resulted from efficient

high-temperature combustion that reduced the fractions of organic material and soot in the aerosol (Rissler et al., 2004).

During the South American Biomass Burning Analysis, the transformation and aging of biomass burning aerosol over the Amazon was characterized. Aged aerosol showed an increase in the presence of oxygenated organic aerosol in both ground and aircraft observations (Brito et al., 2014; Morgan et al., 2020b) but not in aerosol mass, contrasting with the observations from southern Africa reported by Vakkari et al (2018).

In Australia, up to a third of the tropical savannah region in the north burns each year, while the densely forested regions to the south experience less frequent but more intense fire activity (Russell-Smith et al., 2007). One of the most comprehensive investigations into the chemical composition and transformations of smoke in the tropical north of Australia was the Savannah Fires in the Early Dry Season campaign that took place at the Northern Territory Baseline Air Pollution Station at Gunn Point in June 2014 (Mallet et al., 2017). Over the course of the monthlong campaign, biomass burning signals were prevalent and emissions from several large single burning events were observed at the station (Desservettaz et al., 2017). The composition of the smoke aerosol was dominated by organic compounds with varying degrees of oxidation (Milic et al., 2017), with a clear increase in highly oxygenated organic compounds matched by a decrease in biomass burning organic signature. Plume emissions were photochemically active, resulting in the production of tropospheric ozone, with smoke from fires closer to the station showing lower ozone enrichment compared to smoke from fires at greater distances from the station (Mallet et al., 2017). Oxygenated biomass burning aerosol made up 25% of the regional background aerosol composition (Milic et al., 2017).

In southern Australia, Keywood et al. (2015) observed high concentrations of SOA compounds in aged smoke plumes from fires impacting Melbourne, particularly from the oxidation of cineole. This study also investigated nighttime plume evolution and found the resulting dominance of sulfates in SOA species. In Tasmania, smoke plumes impacting the Cape Grim Air Pollution Monitoring Station in February 2006 coincided with the Precursors to Particles Campaign (Cainey et al., 2007). Lawson et al. (2015) reported on the evolution of the composition of the smoke plume over several hours, showing decreases in particle number concentrations and increases in ozone concentration, particle diameter, and CCN.

Following anomalously large fires in the Amazon in 2019 (Silveira et al., 2020), the 2019/2020 Australian fire season was unprecedented in its extent and intensity, with an estimated 5.8 million hectares burned (Boer et al., 2020; Collins et al., 2021). The fires burned through areas that consisted mainly of temperate broadleaf forest and included the Gospers Mountain fire, which is the largest forest fire ever recorded in Australia (Boer et al., 2020). The severity and extent of the fires have been shown to be the result of climate drivers rather than forest management (Bowman et al., 2021; Jan Van Oldenborgh et al., 2021), and the impacts on atmospheric composition and chemistry provide a mechanism for future fire-driven climate-carbon feedbacks (van der Velde et al., 2021). The 2019/2020 Australian fires resulted in a vast injection of CO_2 into the atmosphere (Byrne et al., 2021; van der Velde et al., 2021) and record-breaking aerosol optical depth levels in the Southern Hemisphere, with smoke injection into the stratosphere comparable to a volcanic eruption that impacted dynamical circulation and radiative balance (Khaykin et al., 2020; Fasullo et al., 2021; Hirsch and Koren, 2021). The fires had significant impacts on tropospheric composition including elevated CO and ozone over the Oceania and southern Pacific regions (Bègue et al., 2021; John et al., 2021) as well as deposition into the Southern Ocean resulting in widespread phytoplankton blooms (Tang et al., 2021). Smoke from the fires caused some of the worst air pollution events on record in Australia, with large populations exposed to hazardous air quality over extended periods of time (Nguyen et al., 2021). Johnston et al. (2021) estimated that about 80% of the Australian population was affected by bushfire smoke at some point during this period. Exposure to fine particulate matter (PM) resulted in 417 excess deaths across Australia (Borchers-Arriagada et al., 2020), and the economic cost of this event was estimated to be almost $2 billion Australian dollars (Johnston et al., 2021). As well as fine PM, a number of toxic gases (including formaldehyde, hydrogen cyanide, and acrolein) were emitted by these bushfires (Lawson et al., 2015; MacSween et al., 2020; Mouat et al., 2021) and will contribute to the ongoing health burden of exposure to smoke from these fires.

3.3. Fire and smoke management

Human activity has influenced fire behavior for thousands of years. For example, cultural burning has influenced how Australian Indigenous peoples live on, with and through their land for millennia, with Aboriginal Australians skillfully using fire to adaptively manage their local environments (Preece, 2013). The 1990s saw a return to traditional Aboriginal fire management practices in northern Australia, which aimed to reduce fire intensity and total emissions by increasing the amount of burning in the early part of the dry season when the fuel moisture is higher (Russell-Smith et al., 2013; Ansell et al., 2020). Liu et al. (2021) recently reported a shift in fire activity to earlier in the year (detected by satellite-based thermal anomalies) caused by these interventions, implying a successful overall emissions reduction may have been achieved.

In addition to fire ignition and suppression, human activities influence the prevalence and nature of fires through agriculture and other land use changes (Pechony and Shindell, 2010). In recent years, a significant reduction in global burnt area (approximately 25% from 1998 to 2015) has been attributed to increasing urbanization and agricultural activity (Andela et al., 2017), but how this influence may change in coming years is uncertain. For example, a large proportion of South America's total fire

emissions stem from the Brazilian "arc of deforestation" (Aldrich et al., 2012). During the early 2000s, a reduction of these fire activities could have been related to law enforcement and politics directed toward protection of the Amazonian rainforest (Almeida et al., 2016). However, since 2013, this trend has been reversed, and in 2019 and 2020, the Amazon suffered from severe deforestation and corresponding fire activity, with the 2020 deforestation rate of the Brazilian Amazon being the largest of the decade (Silva Junior et al., 2021).

Prescribed burning (also known as hazard reduction burning) is a common method used to reduce fuel loads and thus reduce the risk of uncontrollable fires during the fire season. The effectiveness of prescribed burning is debated (e.g., Fernandes and Botelho, 2003; Bradstock et al., 2012; Price et al., 2012). Penman and Cirulis (2020) modeled the cost-effectiveness of prescribed burning in different Australian landscapes and found it varied widely with spatial configuration of assets (buildings and infrastructure) and natural vegetation. Their work showed that prescribed burning was most cost-effective in regions with continuous urban–vegetation interfaces. However, their work did not account for the impact of smoke on human populations (and the associated health costs), and at the urban–vegetation interface where asset protection-prescribed burning is most cost effective, smoke exposure is likely to be significant.

Exposure to smoke from prescribed burning can be reduced using smoke forecasting methodologies, particularly if used as part of a fire land management program. Smoke forecasting requires information on fuels, fire behavior, and emissions, combined with a meteorological model, smoke plume transport and chemical transformation model, and a real-time validation system. Many of these components are regionally specific, including emission factors (Guérette et al., 2018) under different burning conditions (Reisen et al., 2018) and fuel load maps (Volkova et al., 2018). Region-specific research is therefore required to generate accurate inputs to reliable smoke forecasting systems such as the smoke emissions and transport modeling system developed in Australia by Cope et al. (2019). This is a 3-tiered approach to forecasting smoke emissions from prescribed burning activities, with the first tier forecasting general and fire weather indices, the second tier forecasting regional air pollution out to 3 days, and the third tier producing high-resolution fuel reduction smoke forecasts out to 24 h, providing information to fire managers about the smoke exposure risk associated with planned burns. Factoring in the impacts of smoke pollution on the cost-benefit analysis of conducting prescribed burning is important for improved health outcomes in future, since fires play a key role in air quality in much of the Southern Hemisphere.

3.4. Future research priorities for our understanding of the impacts of Southern Hemisphere fires

One clear priority is to characterize the emissions from fires in undersampled ecosystems such as sub-Saharan Africa. Improvements in emissions inventories will require improved knowledge of the vegetation across each region, the appropriate emission factors for the vegetation type/ecosystem, and the total fuel carbon burned, which requires good fire detection and assessment of the burning efficiency.

The increasing impact of fires in the Southern Hemisphere makes it important to understand the evolution and chemical transformation of biomass burning plumes. There is a clear need for further investigations that include comprehensive chemical and physical assessments. Both observational and modeling activities will be needed in order to understand the effect of biomass burning emissions on air quality, radiative budgets, and transport of pollutants and nutrients at the large scale, all shaping future climate conditions in the Southern Hemisphere.

4. Air quality issues in the Southern Hemisphere

Despite the smaller population, air pollution is a major environmental issue in the Southern Hemisphere, where many countries suffer from a lack of regulation regarding air quality. Reliable and representative monitoring of air quality has been identified as one of the determinants for comprehensive air quality management and is needed for strengthened capabilities to improve air quality (e.g., Franco et al., 2019). The scarcity of air quality monitoring data in many regions in the Southern Hemisphere and information on the quality of these data prevent the understanding of the underlying drivers of air quality and thus the development of targeted policies to improve it. Additionally, knowledge of emission sources is also key for the development of policies aimed at reducing concentration of air pollutants. In many countries, the lack of national emission inventories integrating local information prevents the design of adequate mitigation strategies to improve air quality.

Rapid population growth and urbanization is causing worsening air quality across much of the Southern Hemisphere with significant health impacts. World Health Organization (WHO) recommended limits for ozone and PM with aerodynamic diameters of $\leq$10 μm and $\leq$2.5 μm (PM_{10} and $PM_{2.5}$, respectively) are exceeded in many urban and semiurban areas due to a combination of the formation of secondary pollutants from biogenic and anthropogenic precursors and direct emissions from biomass burning, industry, residential wood burning, and traffic from growing vehicle fleets and inadequate vehicle maintenance (e.g., Amegah and Agyei-Mensah, 2017; Paton-Walsh et al., 2019; Peláez et al., 2020).

4.1. Air quality in South America

South America is a highly urbanized continent (>80%) with approximately 412 million inhabitants (United Nations, 2018). Ambient $PM_{2.5}$ is estimated to have caused 112,000 premature deaths in South America in 2016 (WHO, 2020). Furthermore, 17% of the population in South America is exposed to annual mean $PM_{2.5}$ concentrations exceeding WHO guidelines (10 μg m^{-3}), while for PM_{10}, this percentage increases to 30% of the population. These statistics are however limited by the coverage of the

measuring network: Only 145 cities in South America have PM_{10} (and just 56 have $PM_{2.5}$) measurements, of which at least 84% present annual concentrations exceeding WHO (2018) guidelines.

Peláez et al. (2020) showed that the main air quality issues in South America are related to both PM_{10} and $PM_{2.5}$. The 11 metropolitan areas of South America considered in their study exceeded the WHO guidelines for annual concentration of PM_{10} and $PM_{2.5}$ between 2010 and 2017, while most of these cities complied with WHO guidelines for annual concentrations of nitrogen dioxide (NO_2) and sulfur dioxide (SO_2). In terms of daily concentration, all cities except one exceeded the WHO guideline for daily concentration for $PM_{2.5}$ (20 $\mu m\ m^{-3}$), whereas the 8-h running average ozone concentration was generally below WHO guidelines.

Multiple studies have highlighted the scarcity of air quality monitoring in South America and the deficient accessibility of air quality data and absence of information on the quality of these data and control protocols (e.g., Andrade-Flores et al., 2016; Riojas-Rodríguez et al., 2016; Peláez et al., 2020). Further, except in Argentina and Chile, no locally derived national emission inventories of criteria pollutants are available in South America, although local emission inventories exist for growing urban areas and/or megacities for air quality and public health studies (Huneeus et al., 2020). Therefore, studies exploring air quality at a national or regional scale in South America (or for cities without local emission inventories) rely on downscaled global emission inventories. Despite comparable total emissions between local and global emission inventories, there are large discrepancies in sectoral distribution of pollutants, and Huneeus et al. (2020) therefore advise against the use of global emission inventories for urban air quality modeling in South America. Accurate distribution of emissions into the different sectors is key when examining and designing mitigation strategies to improve air quality.

4.2. Air quality in Africa

South Africa has the largest industrialized economy in Africa, with significant mining and metallurgical activities, coal-fired power generation, and a large petrochemical industry. Most industries in South Africa do not apply scrubbing techniques to remove SO_2 and NO_x from emissions, while the vehicular fleet in South Africa is relatively old (Collett et al., 2010; Lourens et al., 2011). It has been estimated that if $PM_{2.5}$ concentrations across South Africa were to be reduced to meet the WHO annual average standard for $PM_{2.5}$, up to 28,000 (95th percentile CI [15,000, 52,000]) premature mortalities could be avoided, which would have an estimated monetary value of $29.1 billion (approximately 4.5% of South Africa's gross domestic product; Altieri and Keen, 2019). In addition, several atmospheric studies conducted in southern Africa over recent years have indicated the significant influence of household combustion for space heating and cooking (e.g., Balmer, 2007; Ross et al., 2007; Nkosi et al., 2018; Kapwata et al., 2019), as well as regional impacts of open biomass burning on high levels of atmospheric pollutants in the region (Hersey et al., 2015; Muyemeki et al., 2021). South Africa is well known for its NO_2 hot spot revealed by satellite retrievals over the Mpumalanga Highveld, a region where 11 coal-fired power stations are located (Lourens et al., 2012; Duncan et al., 2016). In addition, South Africa is regarded as a globally significant source of anthropogenic sulfur emissions (Stern, 2006; Fioletov et al., 2020). The northeastern interior of South Africa, which includes the Mpumalanga Highveld and the Johannesburg-Pretoria megacity, is the largest industrial area in South Africa, with substantial emissions from several industries, household combustion, and vehicles (Lourens et al., 2012; Laban et al., 2018). Consequently, ambient concentrations of PM and ozone in this region are regularly out of compliance with National Ambient Air Quality Standards (Venter et al., 2012; Hersey et al., 2015; Feig et al., 2019; Govender and Sivakumar, 2019). Atmospheric pollution in South Africa is also compounded by high population growth, which drives economic and industrial growth, leading to an ever-increasing energy demand and associated pollutant emissions (Tiitta et al., 2014).

Elsewhere in sub-Saharan Africa, air pollution from various sources including industrial emissions, wood burning, and transport emissions are increasing in both rural and urban environments (Katoto et al., 2019). The situation is exacerbated by the high densities of older vehicles, inadequate vehicle maintenance, poor traffic management and road conditions, and inadequate mass transport systems (Liousse et al., 2014). The WHO (2016) reported that air pollution in African homes contributed to almost 600,000 deaths in 2012. It is estimated that over the next 40 years, air pollution (Lacey et al., 2017) and climate change (Silva et al., 2017) in Africa will further increase mortality, overtaking unsafe water, and poor sanitation as the primary drivers of mortality (Organization for Economic Cooperation and Development, 2012).

Across Africa, urban air pollution observations remain very sparse. Much of the available data are satellite estimates that reveal that levels of airborne PM remain above the global median of the WHO (2016) ambient air quality guidelines. The main problems are the severe lack of local air pollution knowledge and inadequate funding to install air-monitoring stations. One proposed option is the use of low-cost sensors, which have been used in conjunction with satellite measurements of aerosol optical depth (Malings et al., 2020). The ever-growing stock of unconventional data from low-cost sensors represents a rich source of additional data to characterize air quality; however, their merit still needs to be further explored. They do not yet represent a replacement of reference monitoring observations but can be used successfully in combination with reliable and representative measurements from air quality networks used in mandatory monitoring (Peltier et al., 2020). **Figure 5** presents the locations of existing air quality monitoring stations superimposed on a map of global concentrations of $PM_{2.5}$ modeled by the WHO. It is very clear that the worst air pollution issues coincide with the areas with the fewest measurements, and in the Southern Hemisphere, these are primarily in Africa.

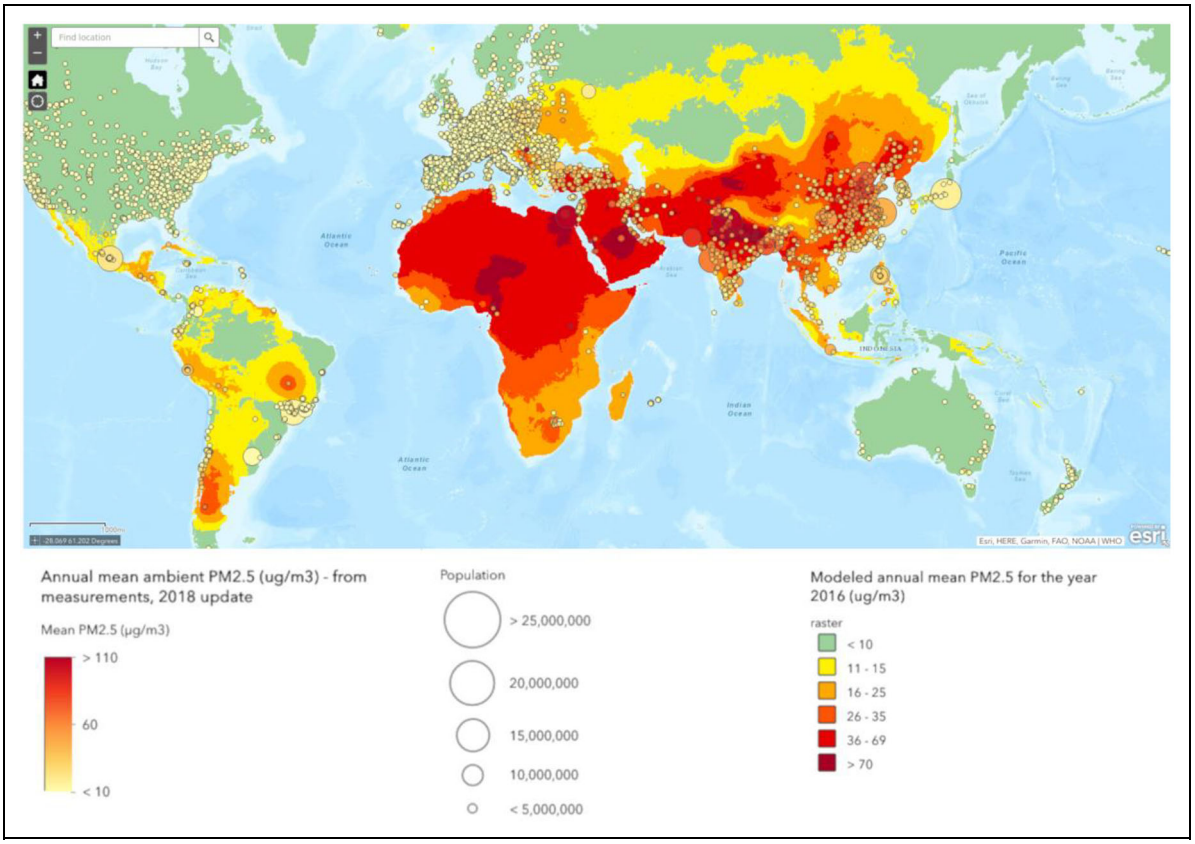

Figure 5. Global particulate air pollution (from worst = red to best = green) and national air monitoring network locations (circles), reproduced from World Health Organization (2016). Licence: CC BY-NC-SA 3.0 IGO. DOI: https://doi.org/10.1525/elementa.2021.00050.f5

Laws to reduce the exposure to air pollution from wood burning and vehicular emissions are being developed for some cities in sub-Saharan Africa. However, controlling those emissions alone will not achieve effective improvement of air quality, despite the urgent need underlined by strong evidence of impact on health including infant mortality (Heft-Neal et al., 2018). Integrated and inclusive control of biomass burning, electricity generation, industrial emission, and traffic-related pollutants via a robust regulatory framework based on rigorous scientific evidence is needed (Katoto et al., 2019; Marais et al., 2019). This is not currently possible due to lack of knowledge of the sources, causes, and impacts of African air pollution and inadequate funding for air quality monitoring stations in many parts of Africa. Further collaborative efforts are needed to assist scientists conducting research on urban air quality in Africa.

4.3. Air quality in Australasia

The literature about air quality in Australasia is mainly restricted to Australia and New Zealand with only a handful of publications in other Australasian countries like Fiji and Papua New Guinea. This is due mainly to a lack of resources and the perception that the South Pacific is a "pristine" area. However, the use of solid fuels for cooking and heating is widespread in the South Pacific region and has been shown to have significant health effects, with deaths attributed to air pollution being more prevalent than those due to traffic accidents (Isley and Taylor, 2018; Isley et al., 2018; Yeimo et al., 2018).

In Australia, significant air pollution is generally associated with episodic events such as wildfires, prescribed burning, or dust storms (e.g., Merrifield et al., 2013; Rea et al., 2016). Regardless of their anthropogenic or natural origin, these events can drive breaches of the air quality standards of PM_{10} and $PM_{2.5}$ across large regions, leading to significant health impacts. For example, Horsley et al. (2019) concluded that between 2001 and 2013, air quality in Sydney was impacted by smoke on 184 days, resulting in an estimated 197 premature deaths and 436 cardiovascular hospitalisations. Borchers-Arriagada et al. (2020) showed that the catastrophic 2019/2020 fire season in southeastern Australia resulted in more than 400 excess deaths and thousands of hospitalizations for cardiovascular and respiratory problems, including asthma. For the same period, Ryan et al. (2021) accounted for meteorology and found 250 excess deaths and 3,490 hospitalizations due to excess $PM_{2.5}$ and ozone exposures in New South Wales and Victoria from the mega-fires, over and above those cause by previous summertime air quality exceedances. For context, the 2019/2020 fire season was attributed to have caused more negative health outcomes for Sydney than the previous 12 years combined (Johnston et al., 2021). A limited number of studies have estimated the health impacts of exposure to smoke from prescribed (hazard reduction) burns specifically. Estimates based on

observations (Broome et al., 2016) and modeling (Nguyen et al., 2020) suggested around 14 premature deaths could be attributed to smoke from hazard reduction burning on 6 smoky days in Sydney (May 6–9, 18, and 22–23, 2016). There is still a need to understand the relative impacts of exposure to smoke from prescribed burning versus bushfires for proper cost-benefit analysis of prescribed burning (Morgan et al., 2020a).

Another air quality issue in Australia is ozone, which can exceed air quality standards in Australia under some circumstances and is associated with 0.8% of excess deaths per year (Broome et al., 2015). In a recent modeling study, Utembe et al. (2018) showed an association between increased temperature and ozone concentrations. When BVOCs were removed from their model, ozone exceedances on very hot days disappeared, demonstrating the dominant role of BVOC emissions from *Eucalyptus* trees (versus anthropogenic VOCs) in ozone formation in Sydney. In Melbourne, Ryan et al. (2020) confirmed that ozone is controlled by NO_x levels in all seasons, and therefore policy actions to limit NO_x will result in reduced ozone. This is of particular importance for mitigating future summertime ozone exceedances, as more extreme summer temperatures are expected due to climate change.

In New Zealand, outside of the main population centers of Auckland and Wellington where traffic is a significant source of pollution (Davy et al., 2007), air quality issues are primarily related to the extensive use of wood for heating (Tunno et al., 2019). During winter when the need for domestic heating is highest, wood smoke emissions coupled with poor dispersion conditions can lead to peak PM_{10} and $PM_{2.5}$ concentrations comparable to those observed in large cities in mainland China and Southeast Asia, with significant health effects (Barnett et al., 2006; Kingham et al., 2007). Domestic wood heating has also been shown to be a significant source of $PM_{2.5}$ in Sydney, with cumulative pollution impacts similar to those from prescribed burning (Desservettaz et al., 2019).

4.4. Future research priorities for our understanding of air quality in the Southern Hemisphere

Many commonalities exist in the barriers to improved air quality in Southern Hemisphere cities, including a lack of observations and regionally specific emissions information. Improved air quality modeling is required, not only as a mechanism to warn vulnerable people of pollution events but also to prioritize policy options via simulation of future mitigation scenarios. While modeling systems have been widely used in parts of the Southern Hemisphere to predict air quality (e.g., Saide et al., 2011; Andrade et al., 2015; Zhang et al., 2019) and/or study the factors modulating it (e.g., Paton-Walsh et al., 2018; Chang et al., 2019), their accuracy is hampered by a lack of observations to better constrain both emissions and meteorology (Monk et al., 2019; Zhang et al., 2019; Guérette et al., 2020). Thus, closer collaborations between researchers addressing the challenges, including reference-grade atmospheric composition measurements and improved emissions inventories that incorporate local knowledge, are likely to yield better outcomes for clean air in future. Research collaborations that combine state-of-the-science air quality modeling, assimilation of satellite data for improved meteorology, and local expertise should therefore be prioritized.

5. The important role of the Southern Ocean for atmospheric chemistry

The Southern Ocean is a globally important region for climate processes, yet much is still unknown about the atmospheric chemistry processes occurring there. It is an important global sink of CO_2 (Stephens et al., 2018) and is composed of diverse physical and biogeochemical zones. A latitudinal transect of the Southern Ocean from north to south crosses regions of low wind speeds, increasing to peak wind speed between 40°S–50°S ("the roaring 40s") and reducing again further south toward the pole. Sea surface temperature varies from 20°C to –2°C along such a transect, and biological diversity varies considerably, with iron-limited productivity in the open Antarctic zone (dominated by diatom phytoplankton communities) increasing to maximum productivity in the sub-Antarctic zone where nondiatom phytoplankton predominate (Henley et al., 2020).

Significant uncertainties remain on the feedbacks and links between surface ocean biogeochemistry, emissions of aerosol precursor gases, aerosol formation and composition, CCN, and transport and deposition. All of these factors influence clouds over the Southern Ocean, a region that plays a disproportionately large role in global climate forcing (McFarquhar et al., 2021). The wild weather and remoteness from major anthropogenic activity make the Southern Ocean one of the most pristine regions on the planet and one of the best proxies for the preindustrial baseline used to calculate the magnitude of the anthropogenic impact on Earth's climate (Carslaw et al., 2013; Hamilton et al., 2014; Schmale et al., 2019). Current climate models exhibit significant biases and uncertainties in the simulation of clouds, aerosols, and air–sea exchange in the Southern Ocean region, leading to overly strong absorption of solar radiation by the Southern Ocean atmosphere (Flato et al., 2014; see **Figure 6**). Reducing the uncertainty in our understanding of atmospheric chemistry and microphysical processes in the pristine Southern Ocean environment is key to improving simulations of a future changing climate. In fact, a recent study confirmed the overproportional efficacy of Southern Ocean aerosol measurements for reducing uncertainty of aerosol forcing in climate models (Regayre et al., 2020).

The large biases and uncertainties in the Southern Ocean region can largely be traced to the dearth of measurements in the region, which in turn drives poor physical understanding of the underlying processes in the pristine natural environment. For example, until recently, the majority of in situ cloud-aerosol observational data was derived from two large field campaigns, the Southern Ocean Cloud Experiment and the First Aerosol Characterization Experiment (both in the early 1990s), and a long time series of observations at Cape Grim, Australia. There

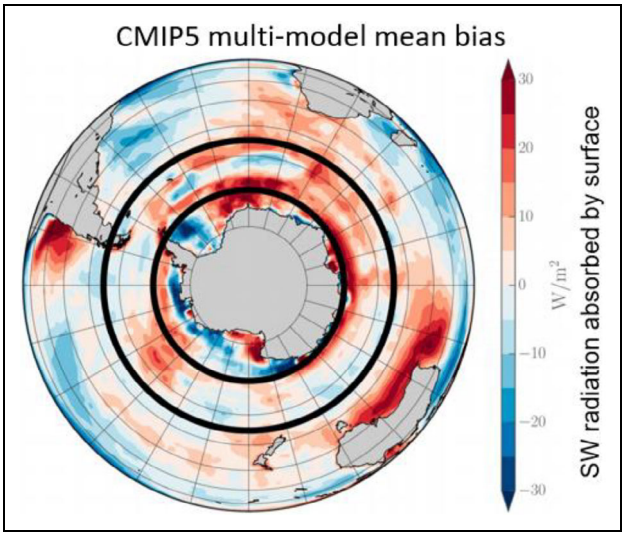

Figure 6. Multimodel mean shortwave absorption bias relative to austral summer satellite observations (Mason et al., 2014). The black lines indicate the zone with the strongest bias (50°S–65°S). Positive biases in this region suggest that clouds in the models do not reflect enough incoming solar radiation in the region. DOI: https://doi.org/10.1525/elementa.2021.00050.f6

are also records of aerosol optical depth at Cape Grim (Wilson and Forgan, 2002) and at Cape Point, South Africa (Nyeki et al., 2015). There is an annual cycle in observed aerosol optical depth at Cape Grim, driven by both biogenic activity and long-range transport (Mitchell et al., 2017); however, the magnitude and location dependence is poorly characterized. These data sets only measured the northern region of the Southern Ocean, which has recently been shown to be distinct from the mid- and high-latitude regions (Humphries et al., 2021). There are also a number of portable sun photometers measurements that have been made within the AERONET Maritime Aerosol Network (Smirnov et al., 2009; Smirnov et al., 2011) and used in validation and model development of important aerosol components such as oceanic sea salt (Bian et al., 2019).

The need for more measurements in the Southern Ocean atmosphere has more recently been recognized, resulting in a multitude of measurements in the last few years (see Schmale et al., 2019, for an overview), with at least 15 campaigns in the region since the turn of the century as well as the establishment in 2015 of the world's first mobile GAW station aboard the RV Investigator. The vessel frequents the Southern Ocean waters surrounding Australia. Satellite data have also provided vital information for this infrequently sampled region; however, the persistent cloudiness (low-level clouds typically present 90% of the time) and limited number of ground validation sites have limited the usefulness of these data. The recent flurry of campaigns and shipborne measurements has been critical in accelerating the science in this region and has highlighted that many questions still remain.

5.1. Aerosol sources, processes, and their relevance for CCN and ice nucleating particles (INPs)

Understanding aerosol–cloud interactions over the Southern Ocean is critical for the two reasons discussed above: poor representation in models resulting in cloud lifetime underestimation and large radiation biases (Flato et al., 2014) and preindustrial-like conditions that help constrain uncertainty in anthropogenic radiative forcing (Regayre et al., 2020). The observations from the last few decades, together with modeling studies, have helped to create a relatively clear understanding of the main processes, including emissions of aerosols and their precursors, atmospheric processing, activation as CCN and INPs, and aerosol removal processes.

A major area of research has been identifying the sources of aerosols over the Southern Ocean. Wind and sea state driven production of primary aerosol particles composed of sea salt and organic material are major sources of aerosols (e.g., Murphy et al., 1998; Fossum et al., 2018; Schmale et al., 2019). There is also a secondary source from marine microbial emissions (e.g., dimethyl sulfide [DMS] from phytoplankton), which are converted to particulate nonsea salt sulfate (nss-SO_4^{2-}) and methanesulfonic acid (MSA), either through condensation from the gas phase or heterogeneous chemistry (cloud processing; Hoffmann et al., 2016). Other mainly locally important sources are ammonia emissions from sea bird and mammal colonies (Schmale et al., 2013) and local volcanic emissions (Schmidt et al., 2012). Minor sources are from long-range transport of dust (Gassó and Stein, 2007), low-latitude volcanic eruptions (Solomon et al., 2016), forest fires (Fiebig et al., 2009), and continental anthropogenic emission outflow (Barbante et al., 1998). The minimal shipping activities in the Southern Ocean mean local anthropogenic emissions are only a small contribution.

A key research question has been whether primary sea spray or secondary marine emissions contribute more significantly to CCN. The answer differs by season and latitude. In winter, sea spray is clearly the more dominant source of CCN due to the absence of marine microbial processes (Vallina et al., 2006). In summer, marine secondary particles (nss-SO_4^{2-} and MSA) become more important at higher latitudes due to the highly biologically active waters near the Antarctic coast and around local hot spots (Gras and Keywood, 2017; Schmale et al., 2019; Yan et al., 2020; Sanchez et al., 2021).

A typical Southern Ocean size distribution features a prominent Aitken mode, followed by a Hoppel minimum and an accumulation mode and a coarse mode. Occasionally, a nucleation mode is also observed. In **Figure 7**, we highlight the submicron size distributions and separate typical shapes by *k*-means clustering. The Hoppel minium indicates particle processing by clouds, that is particles activated as cloud droplets grow further in size through aqueous phase processes, thereby creating the "gap" between the nonactivated and activated accumulation mode particles (Hoppel et al., 1986). This distribution has led to questions around the source of the sometimes-dominant Aitken mode, the relative importance of aqueous phase chemistry versus gas-phase

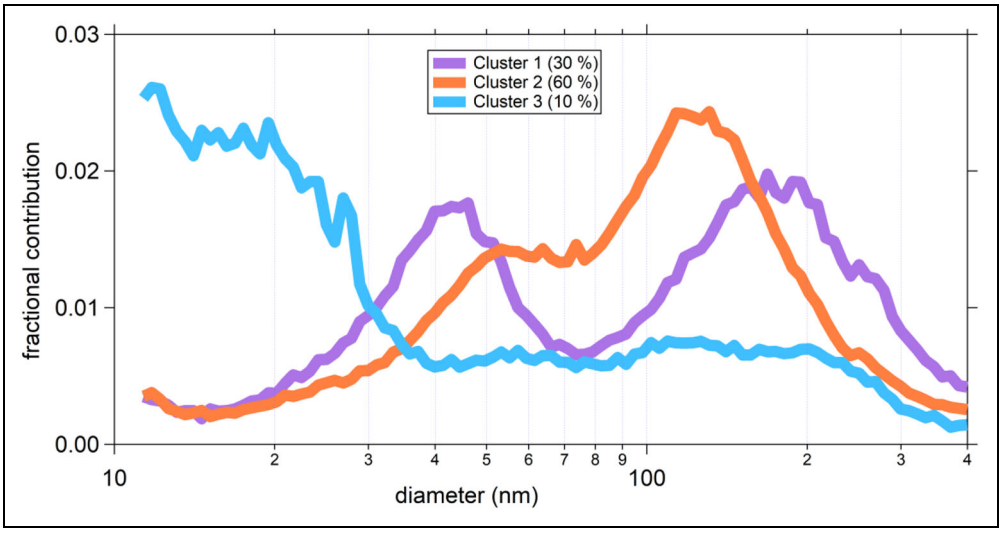

Figure 7. Typical aerosol size distributions over the Southern Ocean between 40°S and 72°S from the Antarctic Circumnavigation Expedition. Here, a *k*-means cluster algorithm was applied to the size distributions measured by a scanning particle mobility sizer to highlight the three dominant distribution types: cluster 1 (purple) shows a pronounced Hoppel minimum and occurs 30% of the time, cluster 2 (orange) shows Aitken mode particles growing into the accumulation mode and occurs 60% of the time, and cluster 3 (blue) represents nucleation mode particles, which occur 10% of the time, in absence of larger sized particles (i.e., larger than nucleation mode). The figure is adapted from Schmale *et al.* (2019). DOI: https://doi.org/10.1525/elementa.2021.00050.f7

condensation, and whether cloud droplets fed from CCN originate in the free troposphere or the boundary layer. While the coarse mode is mainly a result of sea spray emissions, the accumulation mode is a mix of sea spray primary particles and cloud-processed Aitken mode particles. This is evident from the persistent Hoppel minimum. The Aitken mode aerosol is typically formed from gas-to-particle conversion and subsequent growth. However, while there are several reports of new particle formation around the Antarctic coast (Covert et al., 1992; Koponen et al., 2003; Heintzenberg et al., 2004; Kyrö et al., 2013; Humphries et al., 2015; Weller et al., 2015; Jokinen et al., 2018; Kerminen et al., 2018; Kim et al., 2019), new particle formation in the marine boundary layer away from the Antarctic coast is not a strong source of particles (Baccarini et al., 2021).

After first indications by Clarke et al. (1998) that new particle formation happens in the outflow of clouds in the free troposphere, recent aircraft campaigns have shed more light on the mechanism. Marine trace gases are thought to be mixed into the free troposphere, undergo new particle formation there and then be down-mixed again into the marine boundary layer (Humphries et al., 2016) where they can grow further from marine trace gas emissions (McCoy et al., 2021). The question remains as to whether this growth process happens through condensation of gases or through cloud processing. While direct observations are scarce, the few existing observations and modeling studies provide strong evidence that cloud processing is the more important pathway to grow Aitken mode particles (Revell et al., 2019; Schmale et al., 2019; Alroe et al., 2020). The emerging evidence that down-mixing of free tropospheric air masses to the marine boundary layer is critical to grow small particles into sizes which act as CCN also helps to answer the question of whether clouds are nourished mainly by particles from above or below. Satellite-based observations suggest that the below-cloud particles are key to forming droplets (Efraim et al., 2020). This is consistent with down-mixed and growing particles (McCoy et al., 2021).

Finally, and equally important, is research regarding the abundance and sources of INP. The first measurements of INP in the Southern Ocean atmosphere were made in the 1960s (Bigg and Hopwood, 1963). Bigg (1973) found INP concentrations of roughly 10–100 m^{-3}, while more recent measurements found concentrations were 1–2 orders of magnitude lower (McCluskey et al., 2018; Schmale et al., 2019). Compared to elsewhere in the global ocean, INP concentrations are the lowest over the Southern Ocean (Welti et al., 2020). McCluskey et al. (2018) found contributions to INP from heat-labile organic substances, indicating a biological source. Generally, INP concentrations appear higher near land compared to over open ocean (Landwehr et al., 2021). The importance of accurate INP concentration for simulating Southern Ocean mixed phase clouds has been demonstrated in a modeling study (Vergara-Temprado et al., 2018), where overly high concentrations led to overly strong precipitation and overly short cloud lifetime. High INP concentrations are a common feature in the global models that underestimate cloud lifetime (Vergara-Temprado et al., 2018).

Open questions remain concerning the detailed process understanding of aqueous phase chemistry and air mass exchange across the top of the marine boundary layer for particle growth and CCN formation. Here, direct measurements are needed as well as more sophisticated DMS and MSA heterogeneous chemistry schemes in models. The discrepancies in INP concentrations from recent

measurements and those made several decades ago also raise questions as to the actual abundance of INP, while source identification remains elusive. Further questions are related to sea spray, as models tend to over predict sea spray concentrations (Revell et al., 2019; Alroe et al., 2020; Hartery et al., 2020). While more effort has been made to improve the sea spray emission source function, the importance of air mass exchange and deposition processes have received less attention (Landwehr et al., 2021). A major outstanding question is the link between aerosols and clouds. Recent measurements in the high latitudes show coincident increases in cloud droplet concentrations, decreases in droplet effective radius, and increases in CCN (Mace et al., 2021). As the climate changes, we have already witnessed the invigoration of the westerly wind belt (Schneider et al., 2015), and it is conceivable that microbial activity and hence emissions will change in response. Both factors will play a role for the Southern Ocean aerosol population and its impacts on clouds.

5.2. Ocean emissions of secondary aerosol precursors and reactive gases

In regions truly remote from anthropogenic sources, such as the open Southern Ocean, the sources of precursors to secondary aerosol formation are limited to long-range transport and local emissions from the surface ocean. As compared to other oceanic regions, the Southern Ocean has relatively high surface concentrations of ammonium (up to 2 μM; Gruber, 2008; Henley et al., 2020), DMS (Koga et al., 2014), and nitrite (which leads to supersaturation of alkyl nitrates (Fisher et al., 2018). High surface concentrations combined with high wind speeds across the Southern Ocean contribute to the potential for large sea–air fluxes. Interestingly, despite high concentrations of ammonium, global models suggest that the high latitudes are a sink for ammonia due to low surface temperatures (Paulot et al., 2015). Recent observations from the Atlantic sector of the Southern Ocean suggest that the Southern Ocean is indeed a net sink of ammonia in summer, but potentially a net source in winter as surface ocean ammonium concentrations increase 5-fold from summer to winter (Altieri et al., 2021). Despite this, its remote nature and distance from human activities renders the Southern Ocean the only region where ocean emissions of ammonia are expected to dominate over large anthropogenic sources (Paulot et al., 2015).

Annual DMS emissions from the Southern Ocean, a long recognized contributor to CCN (Charlson et al., 1987), account for 60% of global annual DMS emissions (Lana et al., 2011). Ammonia partitions rapidly between the gas and highly acidic submicron aerosol phases by neutralization reactions with sulfuric acid (the oxidation product of DMS), forming fine mode ammonium sulfate and ammonium bisulfate aerosols (Pye et al., 2020). Ammonia and DMS emissions therefore exert control on the size, chemical composition, and neutralization extent of aerosols in this region, all of which affect aerosol properties such as scattering efficiency (Martin et al., 2004), hygroscopicity (Petters and Kreidenweis, 2007), and ice/cloud nucleating ability (Abbatt et al., 2006). The Southern Ocean (along with the tropical Pacific) is the most important region for ocean emissions of alkyl nitrates ($RONO_2$, where R is an organic group; Fisher et al. 2018). This has implications for atmospheric chemistry, as $RONO_2$ can be photolyzed to produce NO_x. Indeed, methyl nitrate is modeled to be the dominant form of reactive nitrogen (NO_y) in the Southern Ocean marine boundary layer (Fisher et al., 2018).

5.3. Future research priorities for our understanding of the atmospheric chemistry of the Southern Ocean

Measurement campaigns are needed to address the key remaining uncertainties of particular importance for the atmospheric chemistry of the Southern Ocean, including:

1. the magnitude, direction, and controls on the air–sea flux of ammonia (Paulot et al., 2015);
2. the relative importance of DMS (vs. organic aerosols) for CCN formation (Quinn and Bates, 2011); and
3. the continued inability of global models to simulate atmospheric NO_x in the region (Fisher et al., 2018).

The lack of measurements in this remote region, and particularly measurements from interdisciplinary research expeditions that combine oceanography, biogeochemistry, atmospheric chemistry, and microphysics, greatly limits our ability to establish a comprehensive mechanistic understanding for this highly complex region and the interactive ocean-trace-gas-aerosol-cloud-climate system. Strong collaborations across both national and disciplinary borders will be needed to address these research priorities in future.

6. Conclusion — A role for the IGAC Southern Hemisphere Working Group

Scientists studying atmospheric composition and chemistry in the Southern Hemisphere face many common challenges, including the dominance in the reactive chemistry of the atmosphere of emissions from vegetation fires and poorly characterized biogenic emissions. Despite lower anthropogenic emissions on the hemispheric scale, many urban and semi-urban regions in the Southern Hemisphere suffer from very poor air quality. Lax regulations and inadequate funding for air quality monitoring are common issues across many countries. Sparsity of observations is a pervasive problem and is especially acute in Africa and in the Southern Ocean, where atmospheric scientists are striving to understand the mechanisms that drive aerosol and cloud formation and the globally significant chemistry-climate feedbacks in this region. In all these areas, there is a need for better collaboration in future years.

The IGAC Southern Hemisphere Working Group has recently been established to provide a forum for collaboration amongst atmospheric chemists working in and

studying the composition of the Southern Hemisphere atmosphere.

The IGAC Southern Hemisphere Working Group aims to:

- identify key challenges unique to the Southern Hemisphere atmosphere,
- understand commonalities in challenges to improve air quality in Southern Hemisphere cities and regional areas,
- facilitate closer collaborations,
- share resources and design future collaborative measurement campaigns, and
- highlight Southern Hemisphere research (which is sometimes considered to be of marginal interest to scientists in the Northern Hemisphere) and its global importance.

Through our early collaborations, we have identified the following priorities for future research:

1. Additional measurements of BVOC emissions in poorly sampled ecosystems, and in particular measurements that include an assessment of the light and temperature dependence of the emissions.
2. Further research into understanding the chemical processing of BVOCs that results in the formation of SOAs and ozone.
3. Improvements in fire emission inventories for poorly characterized parts of the Southern Hemisphere including better vegetation mapping, appropriate emission factors, and improved fire detection and assessments of total fuel consumed.
4. Combined observational and modeling studies into the chemical and physical evolution of smoke plumes from Southern Hemisphere fires in order to understand the impacts on radiative budgets, air quality, and the large-scale transport of pollutants and nutrients.
5. Additional monitoring of air pollutants and work on improved emissions inventories in many regions in the Southern Hemisphere, especially in Africa.
6. VOC speciation in emissions inventories to better model the formation of ozone and SOAs in urban areas.
7. Research collaborations between atmospheric scientists with localized knowledge of emissions and pollution issues and air quality modelers using modeling systems with capabilities for satellite assimilation of meteorological and atmospheric composition data.
8. Campaigns that can help address the lack of measurements in the remote Southern Ocean with particular focus on studies that can help determine the sources and sinks of NO_x, the air–sea flux of ammonia, and the relative importance of organic aerosols and DMS in the formation of CCN.

Strong international collaborations will be needed to address these research priorities, and the IGAC Southern Hemisphere Working Group aims to help facilitate such collaborations in the future. The IGAC Southern Hemisphere Working Group welcomes new members and is open to all scientists, researchers, air quality professionals, and others interested in collaborating to better understand atmospheric composition in the Southern Hemisphere. For more information, see https://igacproject.org/working-groups/SH-working-group.

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Competing interests

The authors have declared that no competing interests exist.

Author contributions

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