The sensitivity of Southern Ocean aerosol concentrations to sea spray and DMS emissions in the HadGEM3-GA7.1 chemistry-climate model

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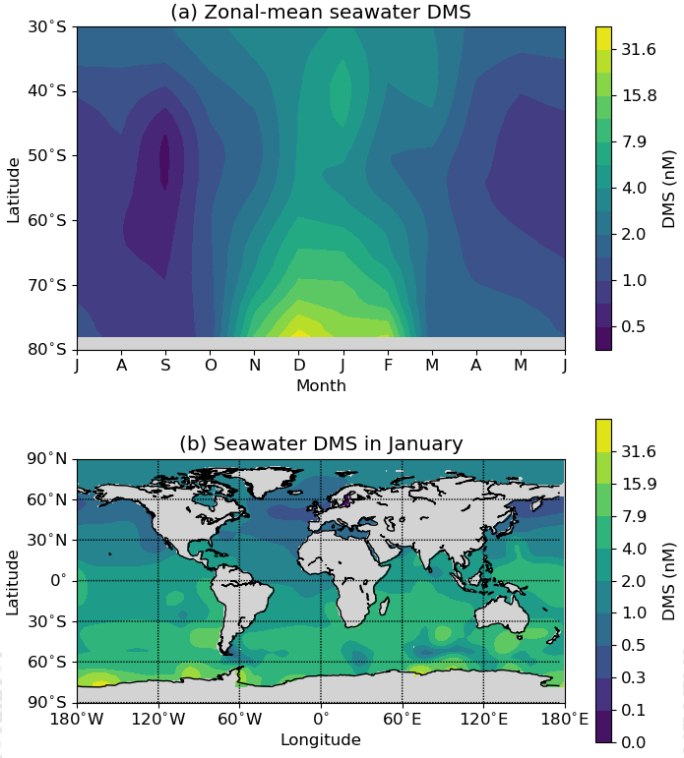
Model simulations

- Simulations performed for the recent past (1989-2008) using the New Zealand Earth System Model AKA **HadGEM3-GA7.1** (Hadley Centre Global Environmental Model v3, Global Atmosphere 7.1).
  - 85 levels between the surface and 85 km; $1.875^\circ \times 1.25^\circ$ horizontal resolution
  - Sea spray aerosol parameterised via Gong (2003) i.e. dependent on the 10 m wind speed $u_{10}^{3.41}$
  - Seawater DMS climatology of Lana *et al.* (2011)

Seawater DMS maximises at southern high latitudes during austral summer



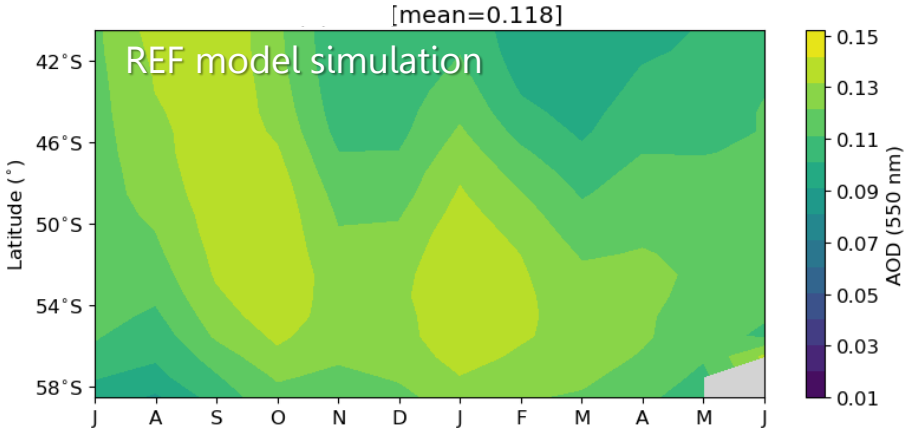
Gong (2003), Glob. Biogeochem. Cycles
Lana et al. (2011), Glob. Biogeochem. Cycles

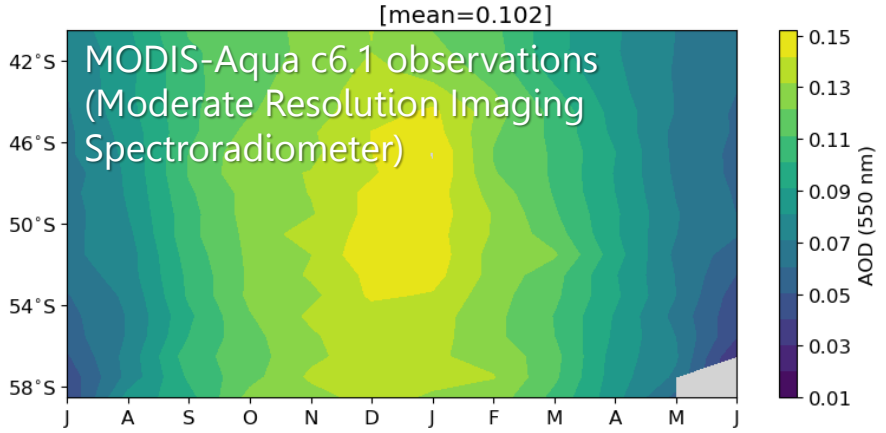
Marine aerosols in the model

- Aerosol emission, evolution and deposition are simulated with the Global Model of Aerosol Processes (GLOMAP-mode)

- Aerosols are represented in four log-normal size modes:
  - Nucleation (< 10 nm)
  - Aitken (10 – 100 nm)
  - Accumulation (100 – 1000 nm)
  - Coarse (> 1000 nm)

Seasonal biases in total AOD





Sea spray aerosol dominates total AOD during JJA

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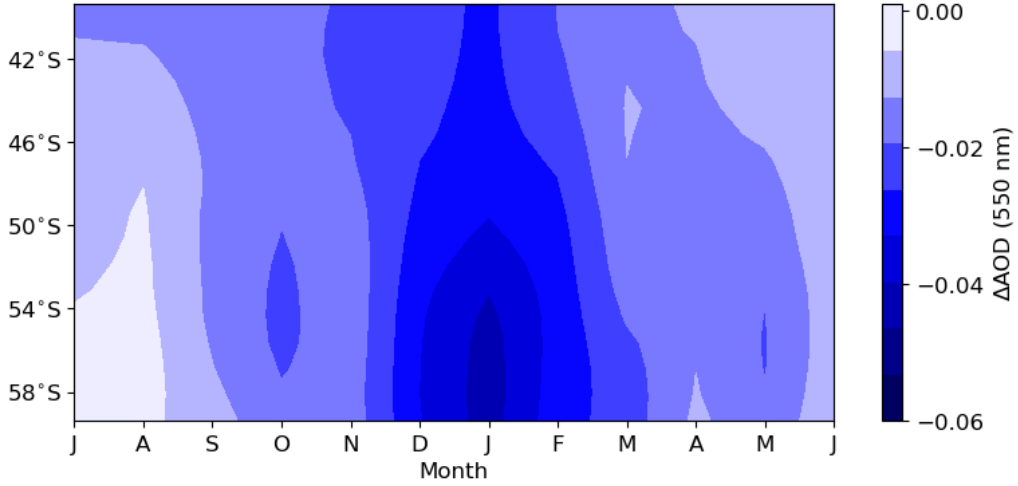
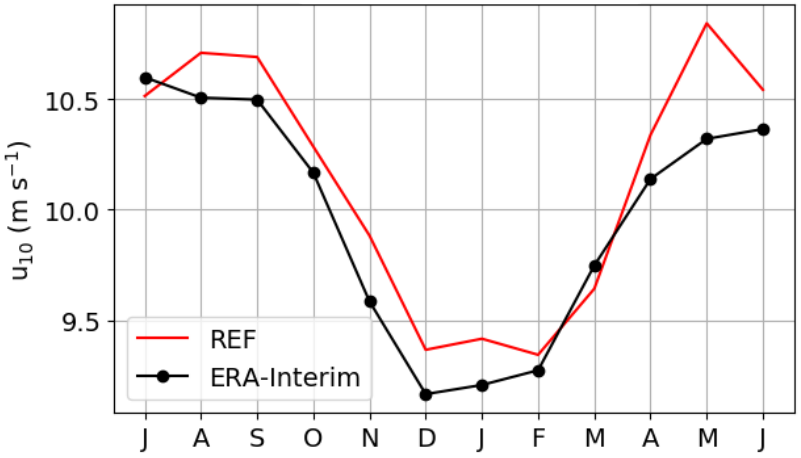
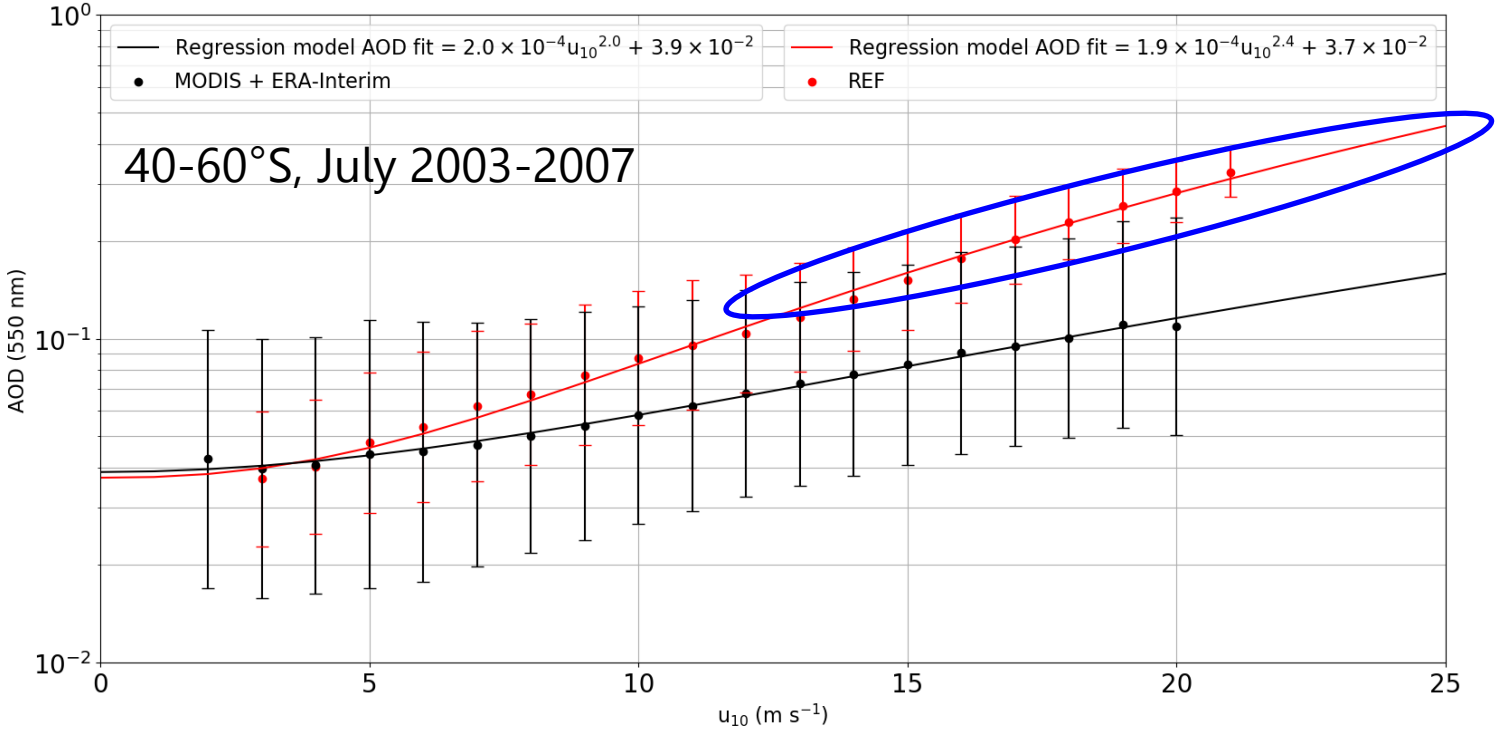
ΔAOD: 0*DMS simulation minus REF



10 m climatological monthly-mean wind speed, 40-60°S, 2003-2007



Simulated sea spray aerosol is overestimated at high wind speeds



The Gong (2003) source function (with $u_{10}^{3.41}$) causes too much sea spray aerosol to be produced over the Southern Ocean

We tested a new sea spray aerosol source function in HadGEM3-GA7.1

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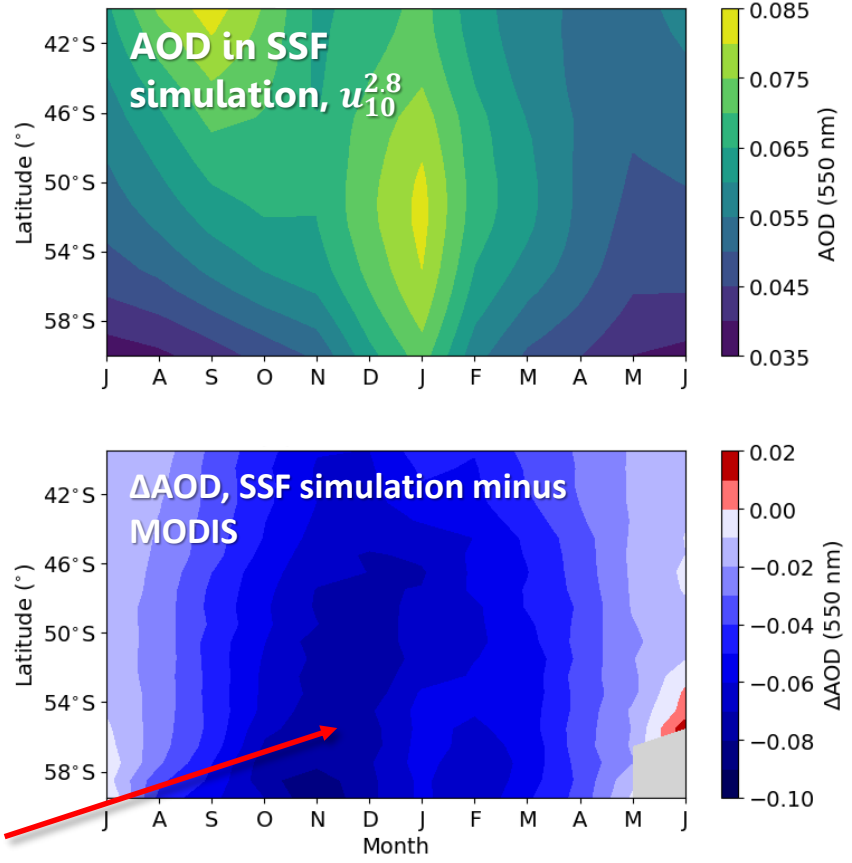
- Hartery *et al.* (2019) developed a power-law sea spray aerosol parameterisation similar in form to Gong (2003), but with $u_{10}^{2.8}$ instead of $u_{10}^{3.41}$ → see **SSF** simulation.
- Based on PCASP-100X optical particle counter measurements during a Tangaroa voyage from Wellington to the Ross Sea in Feb/March 2018.
- At high wind speeds the Hartery *et al.* (2019) sea spray aerosol parameterisation predicts up to 40% less aerosol than Gong (2003).

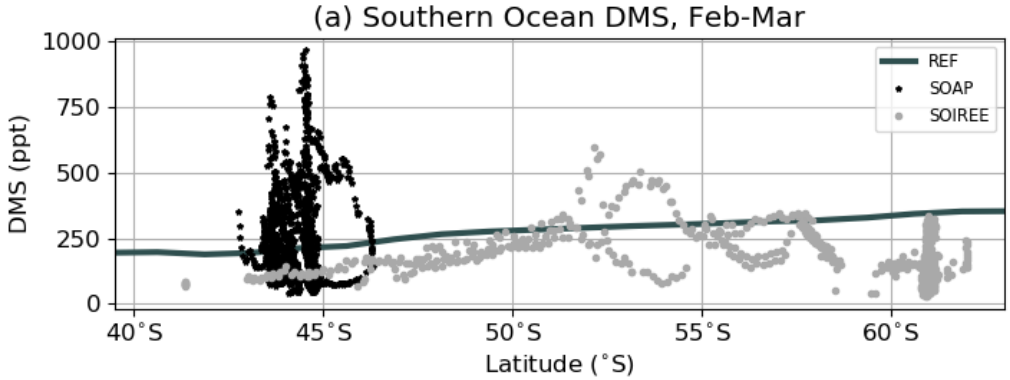


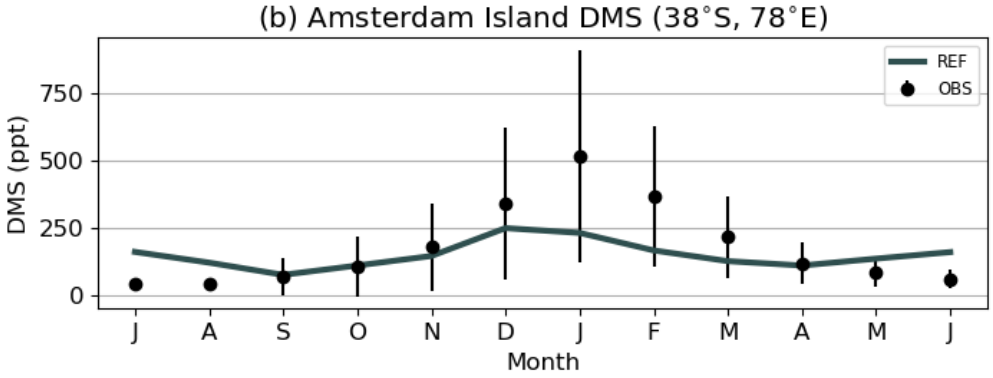
Hartery et al. (2019), J. Geophys. Res., in review

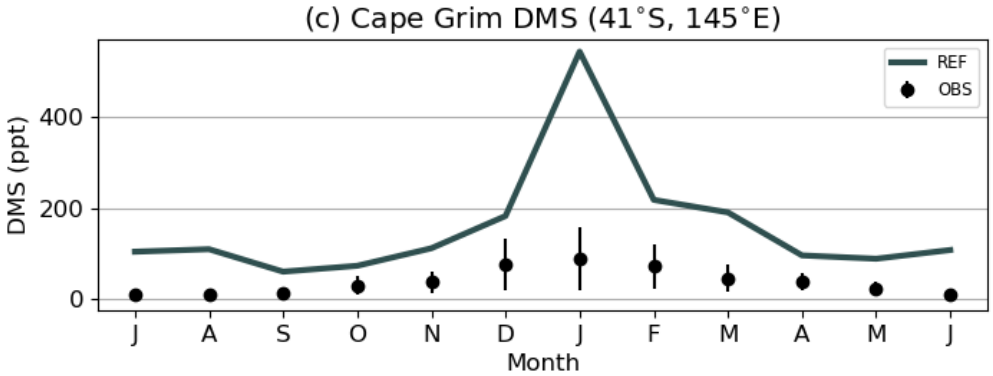
Simulated DMS

- Reasonable agreement between the model and measurements made during the Surface Ocean Aerosol Production campaign (SOAP, Feb-Mar 2012) and the Southern Ocean Iron Release Experiment (SOIREE, Feb 1999)

- Summertime atmospheric DMS concentrations are underestimated at Amsterdam Island but overestimated at Cape Grim.







The current DMS oxidation scheme in HadGEM3-GA7.1:

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Gas phase

$DMS + OH \rightarrow SO_2$ [DMS = dimethyl sulfide, CH_3SCH_3]

$DMS + OH \rightarrow MSA + SO_2$ [MSA = methane sulfonic acid, CH_3SO_2OH]

$DMS + NO_3 \rightarrow SO_2$

$DMS + O(^3P) \rightarrow SO_2$

$SO_2 + OH \rightarrow SO_3 + HO_2$

Aqueous phase

$HSO_3^- + H_2O_{2(aq)} + H^+ \rightarrow SO_4^{2-} + 2H^+ + H_2O_{(aq)}$

$HSO_3^- + O_{3(aq)} \rightarrow SO_4^{2-} + H^+ + O_{2(aq)}$

$SO_3^{2-} + O_{3(aq)} \rightarrow SO_4^{2-} + O_{2(aq)}$

We tested three DMS chemistry schemes

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	CHEM 1:	CHEM 2:	CHEM 3:
Gas phase reactions	DMS + OH	DMS + OH	DMS + OH
	DMS + OH	DMS + OH	DMS + OH
	DMS + NO_3	DMS + NO_3	DMS + NO_3
	DMS + $O(^3P)$	DMS + $O(^3P)$	DMS + $O(^3P)$
	SO_2 + OH	SO_2 + OH	SO_2 + OH
	DMS + BrO	DMS + BrO	DMS + BrO
	DMS + Cl	DMS + Cl	DMS + Cl
		DMSO + OH	DMSO + OH
		MSIA + OH	MSIA + OH
		MSIA + O_3	MSIA + O_3
		SO_2 + OH	SO_2 + OH
Aqueous phase reactions	HSO_3^- + $H_2O_{2(aq)}$ + H^+	HSO_3^- + $H_2O_{2(aq)}$ + H^+	$DMS_{(aq)}$ + $O_{3(aq)}$
	HSO_3^- + $O_{3(aq)}$	HSO_3^- + $O_{3(aq)}$	$MSIA_{(aq)}$ + $O_{3(aq)}$
	SO_3^{2-} + $O_{3(aq)}$	SO_3^{2-} + $O_{3(aq)}$	MSI^- + $O_{3(aq)}$
			HSO_3^- + $HOBr_{(aq)}$
			SO_3^{2-} + $HOBr_{(aq)}$
			HSO_3^- + $H_2O_{2(aq)}$
			HSO_3^- + $O_{3(aq)}$
			SO_3^{2-} + $O_{3(aq)}$

CHEM2 + CHEM3:
Chen et al., *Atmospheric Chemistry and Physics*, 2018

Changing the chemistry scheme alters the yield of DMS, SO_2 and H_2SO_4

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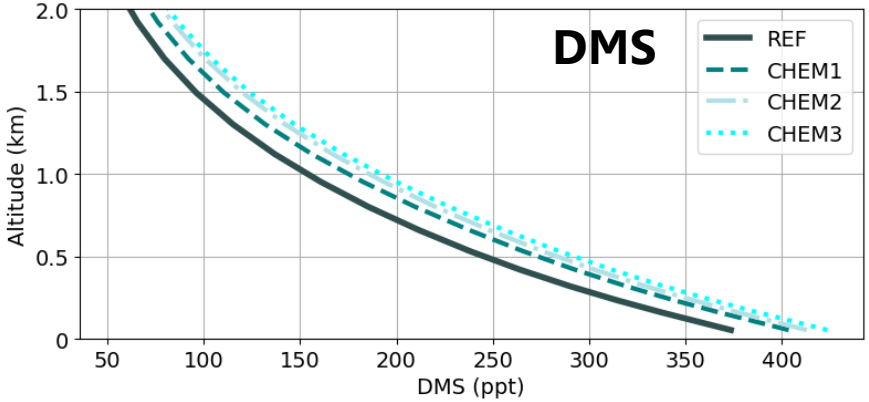
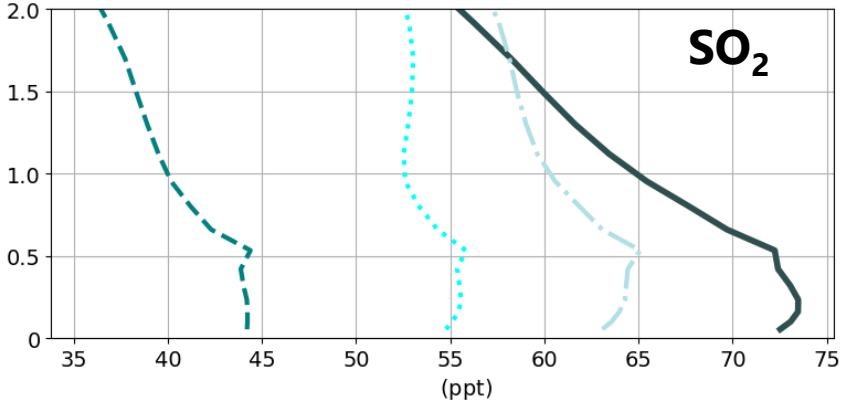
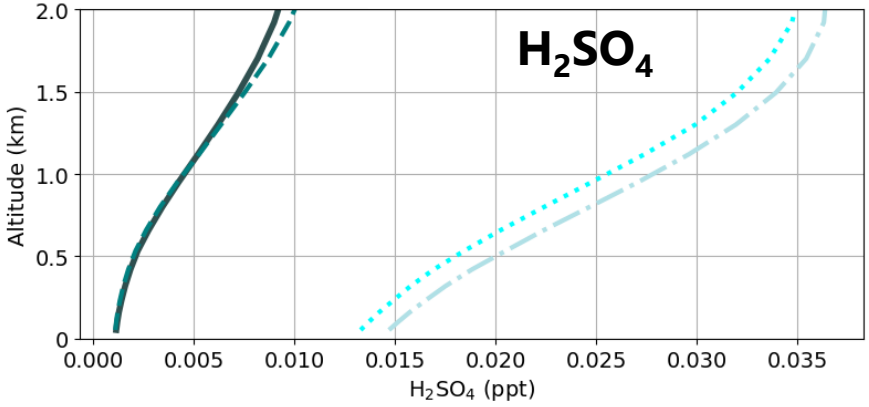
Concentrations in the CHEM sensitivity simulations, 40-60°S, DJF







Changes in aerosol mode number concentrations and particle diameters

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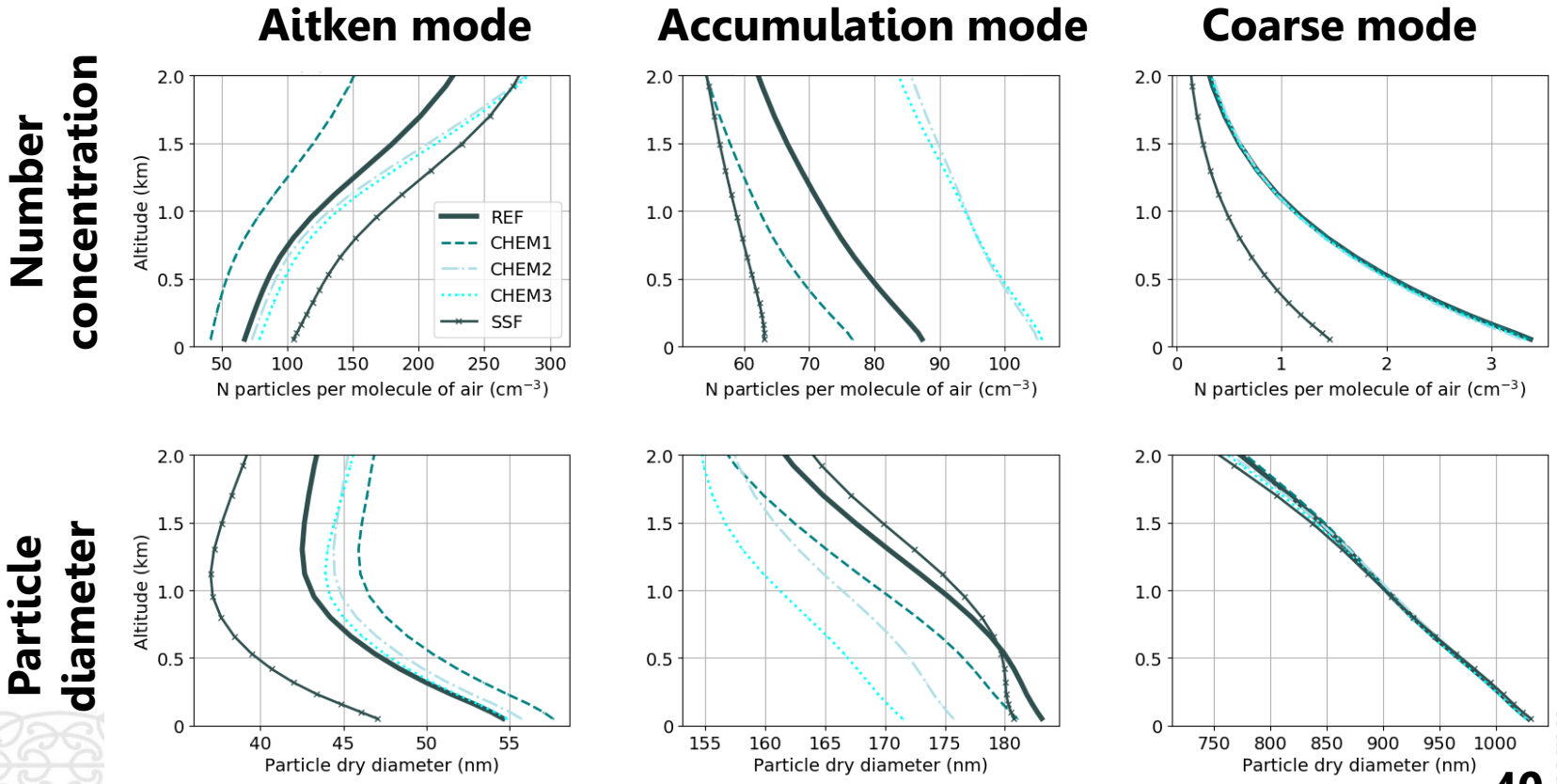

40-60°S, DJF

Improvement in cloud droplet number concentrations in the CHEM2 and CHEM3 simulations

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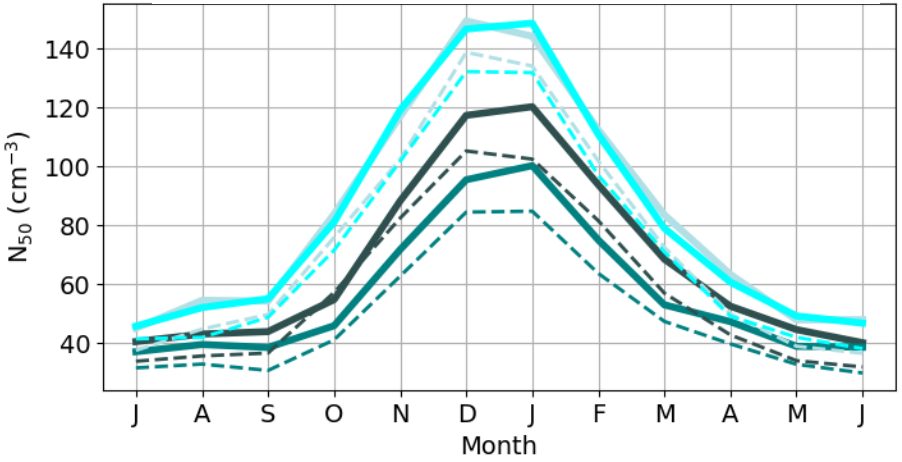
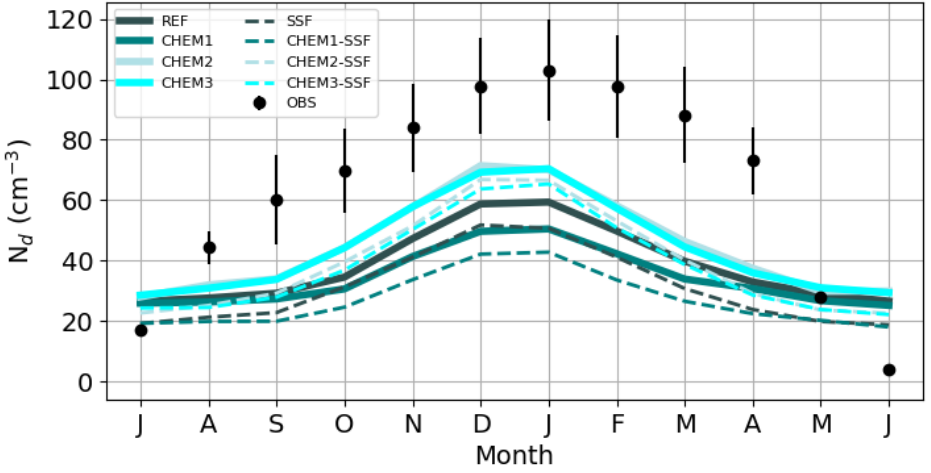
CCN concentration, 40-60°S (0.3% supersaturation)



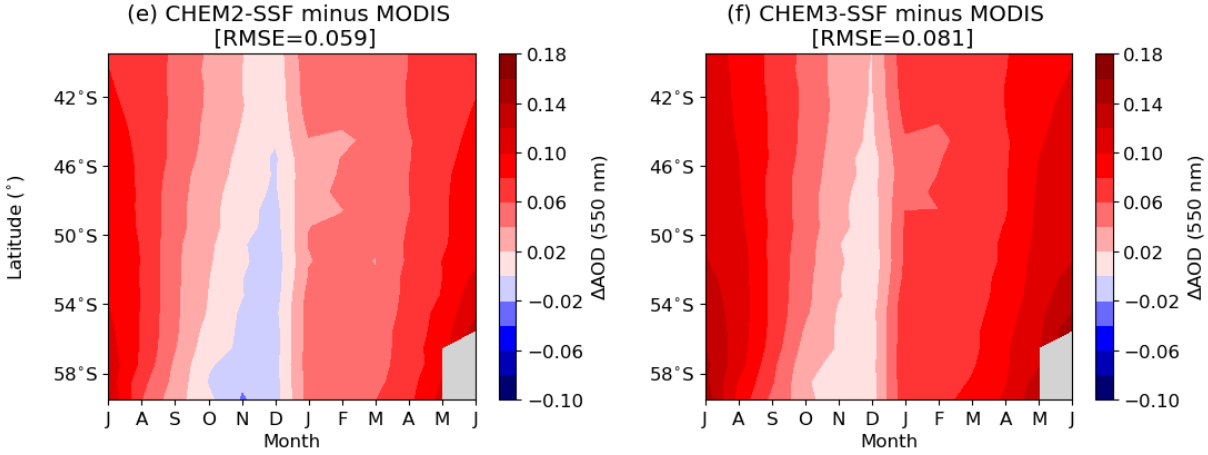
Cloud droplet number concentration



20% N_d increase in the CHEM2 and CHEM3 simulations compared with REF.

N_d observations: *Grosvenor & Wood, ACP, 2014*

Effects on AOD from combining the new sea salt aerosol parameterization and chemistry schemes



Conclusions

- Reducing the production of sea salt aerosol at high wind speeds improves the simulation of total AOD over the Southern Ocean during JJA.
- New DMS chemistry schemes improve the simulation of cloud droplet number concentrations relative to observations. Summertime AOD agrees better with MODIS observations but otherwise AOD is too high in the model.
- Seasonality must be taken into account in evaluating aerosols in climate models.
- For more detail, see:

Revell et al. (2019), Atmospheric Chemistry and Physics, 19, 15447-15466, https://doi.org/10.5194/acp-19-15447-2019

Or contact laura.revell@canterbury.ac.nz