

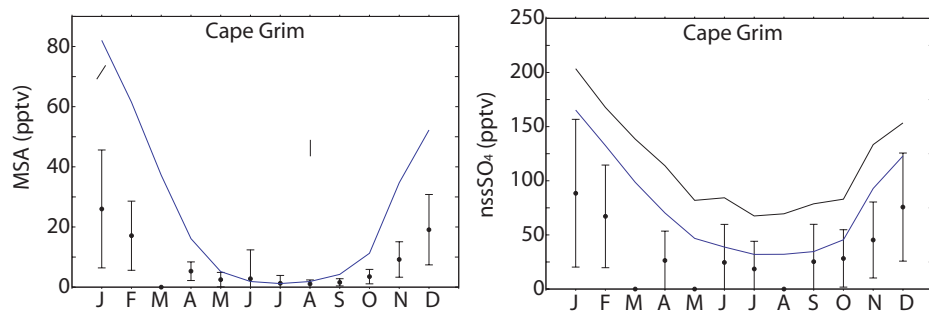
## **Supplementary material:**

# **A three-dimensional model study of methanesulphonic acid to non sea salt sulphate ratio at mid and high-southern latitudes**

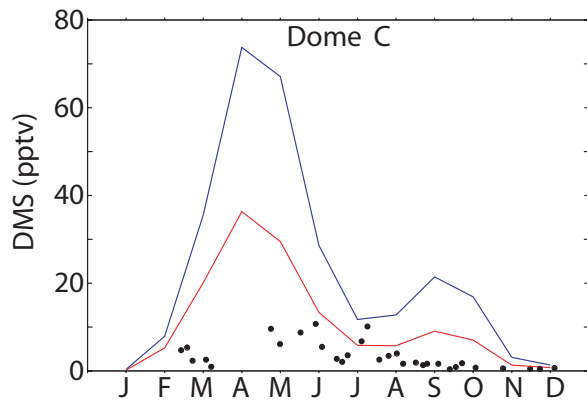
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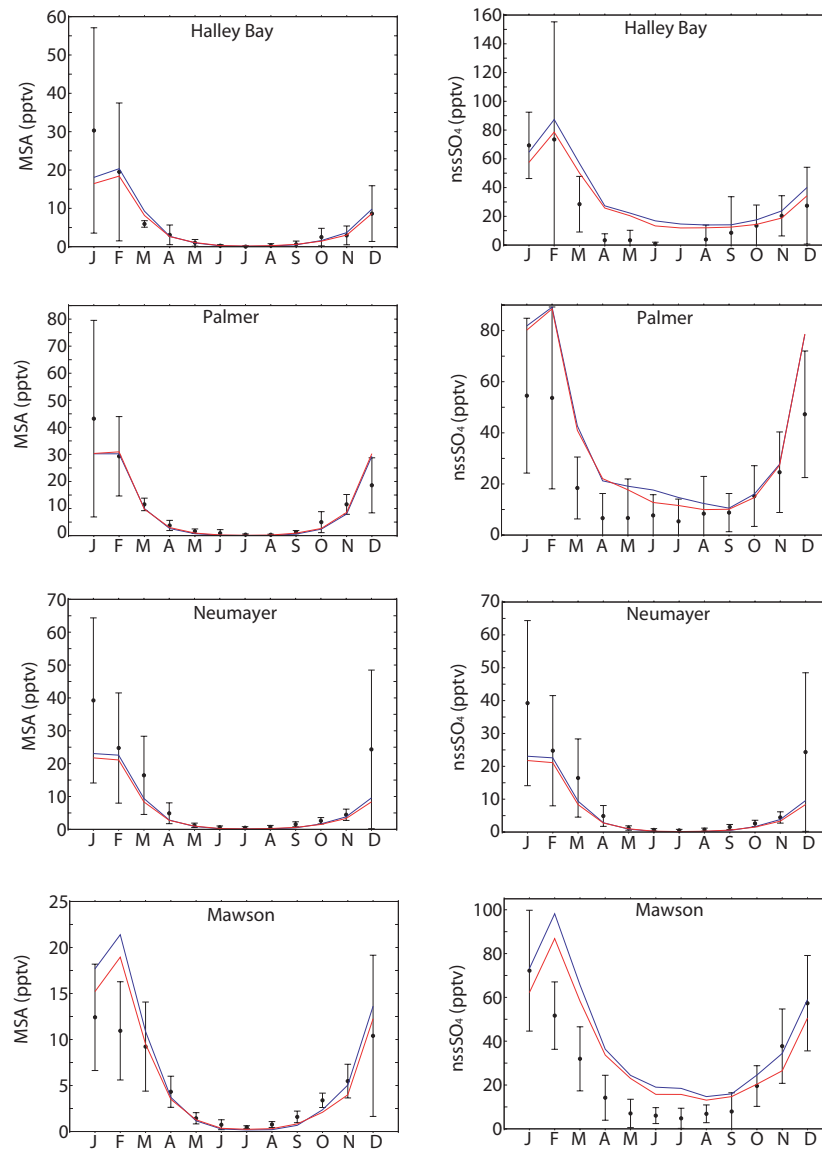
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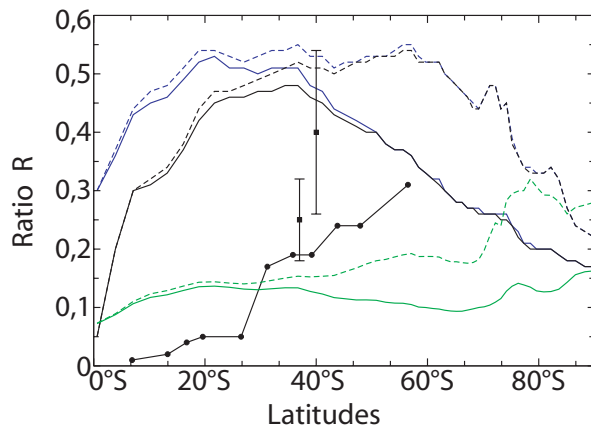
**Fig. 1.** Seasonal variations of MSA and nssSO<sub>4</sub> at Cape Grim station, for simulations REF (blue) and REF-ANTH (black). Observations (black circles) were provided by Dr. Keywood (personal communication) for 1988-2006. For MSA, the two lines corresponding to simulations REF and REF-ANTH are superimposed, as this species has no anthropogenic sources.



**Fig. 2.** Seasonal variations of DMS at Dome C, for simulations REF (blue) and TESTO<sub>3</sub> (red). Observations (black circles) were provided by CESOA (2007) for 2006.



**Fig. 3.** Observed (circles) and simulated seasonal variations of MSA (left panel) and nssSO<sub>4</sub> (right panel) at Halley Bay, Palmer, Neumayer and Mawson stations, for simulations REF (blue) and TESTO<sub>3</sub> (red). Inter-annual variability of measured species is represented (black bars).



**Fig. 4.** Modelled MSA to nssSO<sub>4</sub> ratio in the South Pacific Ocean. Annual and zonal mean modelled  $R_{sim}$  and  $R_{psim}$  are represented by solid and dashed lines respectively. Simulations results are represented in blue for REF, in black for REF-ANTH and in green for the simulation using the previous chemistry scheme (Cosme et al., 2002).

**Table 1.** Comparison of sea surface DMS concentrations from the NOAA-PMEL (2009) database with extrapolated data from Kettle et al. (1999) and Kettle and Andreae (2000). The NOAA-PMEL database contains regularly updated measurements of DMS concentrations in sea surface water. In order to evaluate the recent improvements of DMS oceanic concentrations database for the extra-tropical South hemisphere, data were extracted for years 1972-1998 (approximately corresponding to the data used by Kettle et al. (1999); Kettle and Andreae (2000)) and 1972-2009 (complete database). They are compared to mean DMS concentrations extrapolated to (largely dominant) areas where no data are available by Kettle et al. (1999) and Kettle and Andreae (2000), and interpolated to the LMD-ZT 4 model grid for the latitude bands: 65.4°S-79.5°S, 60.0°S-79.5°S and 30.0°S-79.5°S.

Month	Latitude obs. years	65°S-79°S		60°S-79°S		30°S-79°S	
		1972-1998	1972-2009	1972-1998	1972-2009	1972-1998	1972-2009
Jan.	NOAA-PMEL Nb data	119	345	195	578	295	783
	NOAA-PMEL [DMS]mean	30.9	17.6	20.2	12.9	14.3	10.5
	NOAA-PMEL [DMS]med.	4.9	5.8	2.3	5.0	2.0	3.6
	Kettle 1999 [DMS]mean	17.84		9.91		6.64	
	Kettle 2000 [DMS]mean	35.26		15.75		7.39	
Feb.	NOAA-PMEL Nb data	63	100	118	254	185	509
	NOAA-PMEL [DMS]mean	15.7	10.7	10.3	6.9	7.5	5.0
	NOAA-PMEL [DMS]med.	4.3	2.9	1.6	1.8	1.8	2.1
	Kettle 1999 [DMS]mean	13.34		7.57		4.54	
	Kettle 2000 [DMS]mean	8.72		4.48		3.10	
Mar.	NOAA-PMEL Nb data	58	58	127	131	271	2660
	NOAA-PMEL [DMS]mean	1.3	1.3	1.5	1.5	1.7	1.8
	NOAA-PMEL [DMS]med.	1.0	1.0	1.1	1.1	1.1	1.6
	Kettle 1999 [DMS]mean	1.87		1.14		1.72	
	Kettle 2000 [DMS]mean	1.44		0.99		1.33	
Apr.	NOAA-PMEL Nb data	50	50	194	194	410	1003
	NOAA-PMEL [DMS]mean	1.0	1.0	1.4	1.4	1.4	1.1
	NOAA-PMEL [DMS]med.	0.8	0.8	1.1	1.1	1.1	0.9
	Kettle 1999 [DMS]mean	1.76		1.03		1.06	
	Kettle 2000 [DMS]mean	1.26		0.81		0.87	
May.	NOAA-PMEL Nb data	2	2	2	2	6	21
	NOAA-PMEL [DMS]mean	1.5	1.5	1.5	1.5	0.8	0.7
	NOAA-PMEL [DMS]med.	1.2	1.2	1.2	1.2	0.5	0.6
	Kettle 1999 [DMS]mean	1.43		0.83		0.68	
	Kettle 2000 [DMS]mean	0.83		0.51		0.48	
Jun.	NOAA-PMEL Nb data	0	0	0	0	28	28
	NOAA-PMEL [DMS]mean	0	0	0	0	0.8	0.8
	NOAA-PMEL [DMS]med.	0	0	0	0	0.7	0.7
	Kettle 1999 [DMS]mean	1.25		0.73		0.69	
	Kettle 2000 [DMS]mean	0.26		0.16		0.25	
Jul.	NOAA-PMEL Nb data	1	1	1	1	6	6
	NOAA-PMEL [DMS]mean	0	0	0	0	0.4	0.4
	NOAA-PMEL [DMS]med.	0	0	0	0	0.3	0.3
	Kettle 1999 [DMS]mean	1.15		0.66		0.60	
	Kettle 2000 [DMS]mean	0.26		0.16		0.21	
Aug.	NOAA-PMEL Nb data	1	1	1	1	150	150
	NOAA-PMEL [DMS]mean	0	0	0	0	1.1	1.1
	NOAA-PMEL [DMS]med.	0	0	0	0	1.0	1.0
	Kettle 1999 [DMS]mean	1.05		0.61		0.59	
	Kettle 2000 [DMS]mean	0.42		0.25		0.38	
Sep.	NOAA-PMEL Nb data	14	14	34	34	57	57
	NOAA-PMEL [DMS]mean	0.9	0.9	0.7	0.7	1.1	1.1
	NOAA-PMEL [DMS]med.	0.3	0.3	0.3	0.3	0.4	0.4
	Kettle 1999 [DMS]mean	1.10		0.64		0.59	
	Kettle 2000 [DMS]mean	0.55		0.33		0.35	

**Table 1.** Continued.

Month	Latitude obs. years	65°S-79°S		60°S-79°S		30°S-79°S	
		1972-1998	1972-2009	1972-1998	1972-2009	1972-1998	1972-2009
Oct.	NOAA-PMEL Nb data	4	5	5	19	105	167
	NOAA-PMEL [DMS]mean	1.1	1.0	0.9	1.9	1.7	1.6
	NOAA-PMEL [DMS]med.	0.3	0.8	0.3	0.9	1.4	1.2
	Kettle 1999 [DMS]mean	1.81		1.07		0.78	
	Kettle 2000 [DMS]mean	0.35		0.24		0.33	
Nov.	NOAA-PMEL Nb data	155	363	196	469	1056	1428
	NOAA-PMEL [DMS]mean	3.5	7.3	3.1	6.1	2.1	3.2
	NOAA-PMEL [DMS]med.	2.1	3.2	1.8	2.1	1.3	1.4
	Kettle 1999 [DMS]mean	3.18		1.82		1.89	
	Kettle 2000 [DMS]mean	1.75		1.07		1.19	
Dec.	NOAA-PMEL Nb data	255	315	339	477	1327	1681
	NOAA-PMEL [DMS]mean	34.6	31.2	29.4	23.8	10.9	9.8
	NOAA-PMEL [DMS]med.	4.4	5.1	3.8	4.2	3.2	3.3
	Kettle 1999 [DMS]mean	13.98		7.63		4.67	
	Kettle 2000 [DMS]mean	46.27		20.47		9.64	

Improvements of NOAA-PMEL database since 1999: winter measurements are still very scarce - no data were added for the June to September months - even at mid-latitudes. For the summer season, observations from the highly productive Ross sea sector are included in the 1972-1998 dataset for the December to February period, whereas they were added after for November. With the addition of new data from other sectors, mean DMS concentrations at high latitudes in the overall database are lower than in the 1972-1998 data in December to February, whereas mean values nearly double in November. Thus the mean summer DMS concentrations around Antarctica may be very sensitive to the weight attributed to the Ross sea sector. The Kettle et al. (1999) extrapolated dataset was used in this study because in our model, the Kettle and Andreae (2000) dataset leads to too high DMS concentrations around Antarctica in summer.

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