Climate-driven chemistry and aerosol feedbacks in CMIP6 Earth system models

Gillian Thornhill[1], William Collins[1], Dirk Olivié[2], Alex Archibald[3,4], Susanne Bauer[5], Ramiro Checa-Garcia[6], Stephanie Fiedler[7], Gerd Folberth[8], Ada Gjermundsen[2], Larry Horowitz[9], Jean-Francois Lamarque[10], Martine Michou[11], Jane Mulcahy[8], Pierre Nabat[11], Vaishali Naik[9], Fiona M. O'Connor[8], Fabien Paulot[9], Michael Schulz[2], Catherine E. Scott[12], Roland Seferian[11], Chris Smith[12], Toshihiko Takemura[13], Simone Tilmes[10], James Weber[3],

[1]Department of Meteorology, University of Reading, Reading, RG6 6BB, UK
[2]Norwegian Meteorological Institute, Oslo, Norway
[3]Department of Chemistry, University of Cambridge, Cambridge, CB2 1EW, UK
[4]National Centre for Atmospheric Science, UK
[5]NASA Goddard Institute for Space Studies, 2880 Broadway
[6] IPSL/LSCE CEA-CNRS-UVSQ-UPSaclay UMR Gif sur Yvette, FRANCE
[7]Max-Planck-Institute for Meteorology, Hamburg, 20146, Germany
[8]Met Office Hadley Centre, Exeter, EX1 3PB, United Kingdom
[9] GFDL/NOAA, Princeton University, Princeton, NJ 08540-6649
[10] National Centre for Atmospheric Research, Boulder, CO, USA
[11] Centre National de Recherches Météorologiques, Meteo-France, Toulouse Cedex, France
[12]School of Earth and Environment, University of Leeds, Leeds, LS2 9JT
[13] Research Institute for Applied Mechanics, Kyushu University, Fukuoka, Japan
Correspondence to: Bill Collins (w.collins@reading.ac.uk)

S1 Model information

Earth System Model	**Species**	**Description of emission parameterisation** **(Dependence on wind, temperature, vegetation, soil moisture …)**	**References**
CNRM-ESM2	Sea Salt	three particle size bins (boundaries of 0.03–0.5, 0.5–5, 5–20 μm), desert dust also has three size bins (0.03–0.5, 0.5–0.9, 0.9–20 μm), and the boundaries given are for dry particles; however, the ambient humidity is taken into account in the computation of the aerosol optical properties.	(Grythe et al., 2014)

	Dust	Mineral dust is described by 6 bins modes from radius size of 0.05 um to 50 um.	(Michou et al., 2014)
	DMS	Prescribed	(Kettle et al., 1999)
	Vegetation VOC and OC	Climatology of biogenic secondary organic aerosol is taken from (Dentener et al., 2006) and doubled to give an overall source of 38 Tg SOA per year.	(Dentener et al., 2006)
	Marine VOC and OC		
UKESM1	Sea Salt	Primary emissions of sea-salt aerosols are calculated using the bin-resolved, windspeed-dependent flux parameterization.	(Gong, 2003)
	Dust	Mineral dust is described by a sectional/bin approach with 6 bins from size 0.0316 um to 31.6 um.	(Woodward, 2001)
	DMS	DMS are simulated interactively by the ocean biogeochemistry component, MEDUSA,	(Anderson et al., 2001)
	Vegetation VOC and OC	Emissions of monoterpenes and isoprene are generated by the interactive vegetation scheme. Monoterpene emissions are dependent on PAR and temperature whilst isoprene emissions are linked to photosynthesis rates. Monoterpenes are oxidised to generate condensable secondary organic material with a 13% molar yield; the yield is doubled to compensate for the lack of SOA produced by isoprene oxidation.	(Guenther, 1995; Pacifico et al., 2011)
	Marine VOC and OC	The organic mass fraction of the emitted sea spray aerosol,	(Gantt et al., 2011, 2012)

		is calculated as a function of the biological productivity (based on surface chlorophyll-a), the 10 m windspeed) and the sea-salt dry diameter.	
MIROC6	Sea Salt		(Monahan, 1986)
	Dust	6 radii from 0.1 to 10 μm	
	DMS	Dependence on downward solar flux	(Bates et al., 1987)
	Vegetation VOC and OC	Global Emissions Inventory Activity (GEIA)	(Guenther, 1995)
	Marine VOC and OC	Dependence on chlorophyll	(Gantt et al., 2012)
NorESM2	Sea Salt	Modal description of sea-salt with 3 modes (number median dry radii of 0.048, 0.30 and 0.75 um). Emissions depend on wind speed and sea-surface temperature.	(Kirkevåg et al., 2018; Salter et al., 2015)
	Dust	Modal description of mineral dust with 2 modes: accumulation and coarse (number mean dry radii of 1.59 and 2.0 um). Emissions based on DEAD model.	(Kirkevåg et al., 2018; Zender et al., 2003)
	DMS	DMS ocean concentration calculated by the ocean biogeochemistry module iHAMOCC.	Tjiputra et al., submitted

			(Nightingale et al., 2000)
	Vegetation VOC and OC	Emissions of monoterpenes and isoprene are generated interactively by the MEGAN algorithm within the Community Land Model (CLM5).	(Guenther et al., 2012; Kirkevåg et al., 2018)
	Marine VOC and OC	Primary organic upper ocean concentrations are based on a chlorofyl-a climatology.	(O'Dowd et al., 2008)
GFDL-ESM4	Sea Salt	Sea salt is described in 5 bins with the following radii (0.1-0.5, 0.5-1,1-2,2-5,5-10 um). Emissions are modulated by sea surface temperature.	(Jaeglé et al., 2011; Monahan and Muircheartaigh, 1980)
	Dust	Bin/modal scheme (Reference)	Horowitz et al. (in preparation)
	DMS	Prescribed sea water concentations	Horowitz et al. (in preparation)
	Vegetation VOC and OC	Emissions of isoprene and monoterpenes are calculated online in GFDL-ESM4 using the Model of Emissions of Gases and Aerosols from Nature (MEGAN; Guenther et al., 2006), as a function of simulated air temperature and shortwave radiative fluxes.	(Guenther et al., 2012)
	Marine VOC and OC		

S2 Figures in support of analysis in section 4 of the main text.

S2.1 Dust

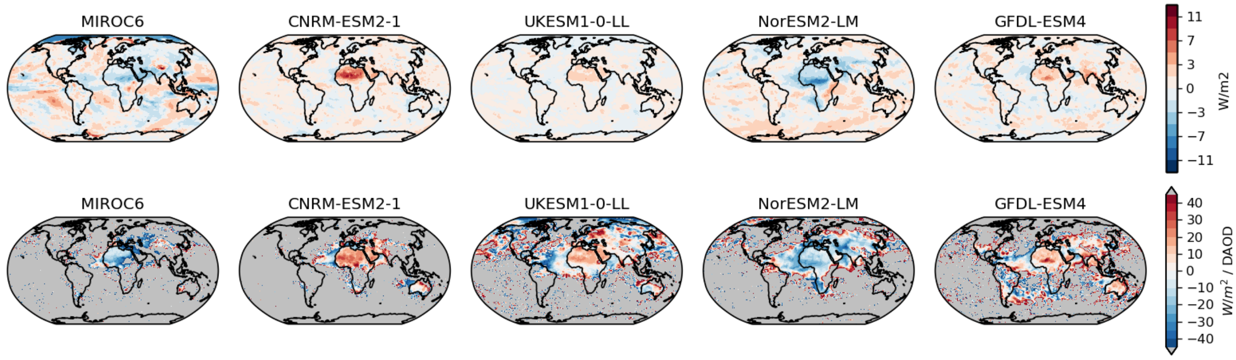

Figure S1: Effective radiative forcing from *2xdust* experiments. Top: ERF for each model. Bottom: ERF divided by AOD for each model.

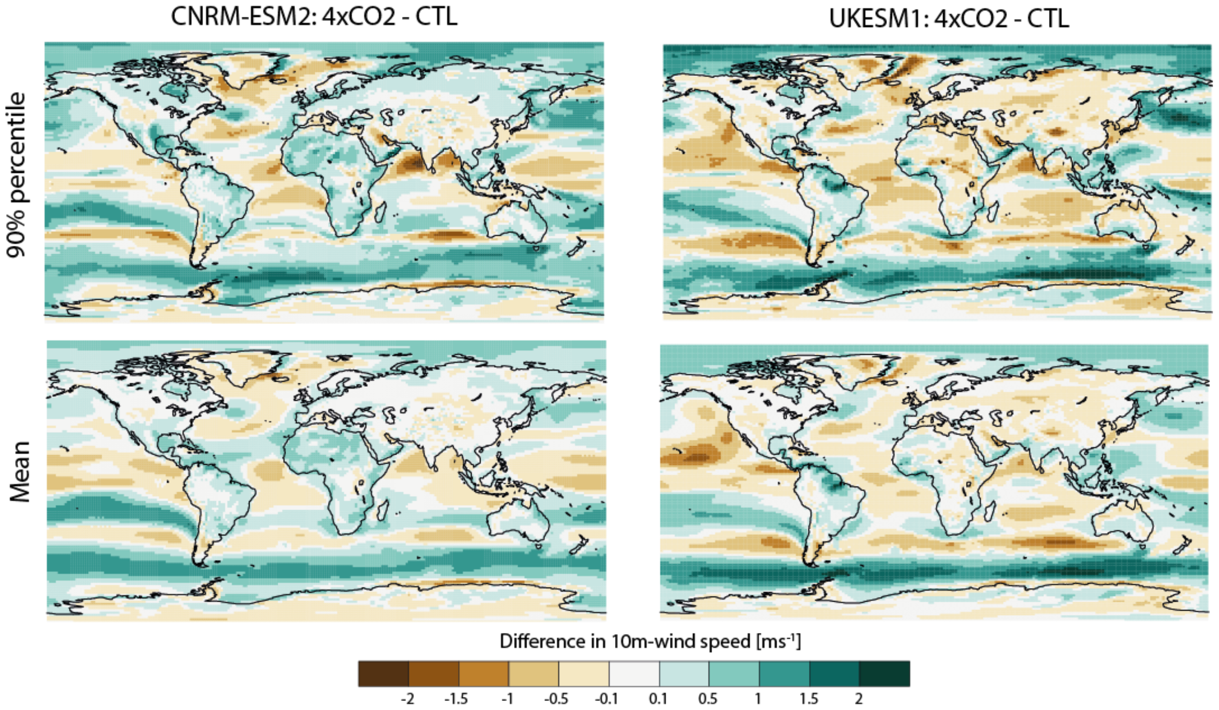

Fig S2: Difference in 10m-wind speeds for *4xCO2*. Shown are the difference for *4xCO2* against the pre-industrial climatology for (left) CNRM-ESM2 and (right) UKESM1 in the (top) 90% percentile and (bottom) mean of the monthly mean 10m-winds.

S2.2 Biogenic VOCs

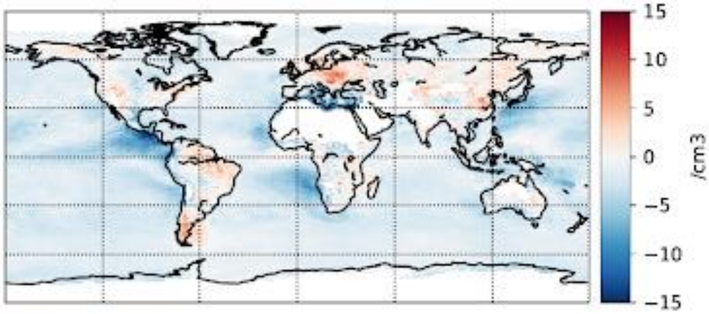

Figure S3: Multi-model mean difference in CDNC from *piClim-2xVOC* vs *piClim-control*.

References

Anderson, T. R., Spall, S. A., Yool, A., Cipollini, P., Challenor, P. G. and Fasham, M. J. R.: Global fields of sea surface dimethylsulfide predicted from chlorophyll, nutrients and light, J. Mar. Syst., doi:10.1016/S0924-7963(01)00028-8, 2001.

Bates, T. S., Charlson, R. J. and Gammon, R. H.: Evidence for the climatic role of marine biogenic sulphur, Nature, doi:10.1038/329319a0, 1987.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J. P., Textor, C., Schulz, M., Van Der Werf, G. R. and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., doi:10.5194/acp-6-4321-2006, 2006.

Gantt, B., Meskhidze, N., Facchini, M. C., Rinaldi, M., Ceburnis, D. and O'Dowd, C. D.: Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol, Atmos. Chem. Phys., doi:10.5194/acp-11-8777-2011, 2011.

Gantt, B., Johnson, M. S., Meskhidze, N., Sciare, J., Ovadnevaite, J., Ceburnis, D. and O'Dowd, C. D.: Model evaluation of marine primary organic aerosol emission schemes, Atmos. Chem. Phys., doi:10.5194/acp-12-8553-2012, 2012.

Gong, S. L.: A parameterization of sea-salt aerosol source function for sub- and super-micron particles, Global Biogeochem. Cycles, doi:10.1029/2003gb002079, 2003.

Grythe, H., Ström, J., Krejci, R., Quinn, P. and Stohl, A.: A review of sea-spray aerosol source functions using a large global set of sea salt aerosol concentration measurements, Atmos. Chem. Phys., doi:10.5194/acp-14-1277-2014, 2014.

Guenther, A.: A global model of natural volatile organic compound emissions, J. Geophys. Res., doi:10.1029/94JD02950, 1995.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K. and Wang, X.: The model of emissions of gases and aerosols from nature version 2.1 (MEGAN2.1): An extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., doi:10.5194/gmd-5-1471-2012, 2012.

Jaeglé, L., Quinn, P. K., Bates, T. S., Alexander, B. and Lin, J. T.: Global distribution of sea salt aerosols: New constraints from in situ and remote sensing observations, Atmos. Chem. Phys., doi:10.5194/acp-11-3137-2011, 2011.

Kettle, A. J., Andreae, M. O., Amouroux, D., Andreae, T. W., Bates, T. S., Berresheim, H., Bingemer, H., Boniforti, R., Curran, M. A. J., DiTullio, G. R., Helas, G., Jones, G. B., Keller, M. D., Kiene, R. P., Leek, C., Levasseur, M., Malin, G., Maspero, M., Matrai, P., McTaggart, A. R., Mihalopoulos, N., Nguyen, B. C., Novo, A., Putaud, J. P., Rapsomanikis, S., Roberts, G., Schebeske, G., Sharma, S., Simó, R., Staubes, R., Turner, S. and Uher, G.: A global database of sea surface dimethylsulfide (DMS) measurements and a procedure to predict sea surface DMS as a function of latitude, longitude, and month, Global Biogeochem. Cycles, doi:10.1029/1999GB900004, 1999.

Kirkevåg, A., Grini, A., Olivié, D., Seland, Ø., Alterskjær, K., Hummel, M., Karset, I. H. H., Lewinschal, A., Liu, X., Makkonen, R., Bethke, I., Griesfeller, J., Schulz, M. and Iversen, T.: A production-tagged aerosol module for earth system models, OsloAero5.3-extensions and updates for CAM5.3-Oslo, Geosci. Model Dev., doi:10.5194/gmd-11-3945-2018, 2018.

Michou, M., Nabat, P. and Saint-Martin, D.: Development and basic evaluation of a prognostic aerosol scheme in the CNRM Climate Model, Geosci. Model Dev. Discuss., doi:10.5194/gmdd-7-6263-2014, 2014.

Monahan, E. C.: The Ocean as a Source for Atmospheric Particles, in The Role of Air-Sea Exchange in Geochemical Cycling., 1986.

Monahan, E. C. and Muircheartaigh, I. O.: Optimal power-law description of oceanic whitecap coverage dependence on wind speed., J. PHYS. Ocean., doi:10.1175/1520-0485(1980)010<2094:opldoo>2.0.co;2, 1980.

Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S., Liddicoat, M. I., Boutin, J. and Upstill-Goddard, R. C.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, Global Biogeochem. Cycles, doi:10.1029/1999GB900091, 2000.

O'Dowd, C. D., Langmann, B., Varghese, S., Scannell, C., Ceburnis, D. and Facchini, M. C.: A combined organic-inorganic sea-spray source function, Geophys. Res. Lett., doi:10.1029/2007GL030331, 2008.

Pacifico, F., Harrison, S. P., Jones, C. D., Arneth, A., Sitch, S., Weedon, G. P., Barkley, M. P., Palmer, P. I., Serça, D., Potosnak, M., Fu, T. M., Goldstein, A., Bai, J. and Schurgers, G.: Evaluation of a photosynthesis-based biogenic isoprene emission scheme in JULES and simulation of isoprene emissions under present-day climate conditions, Atmos. Chem. Phys., doi:10.5194/acp-11-4371-2011, 2011.

Salter, M. E., Zieger, P., Acosta Navarro, J. C., Grythe, H., Kirkevåg, A., Rosati, B., Riipinen, I. and Nilsson, E. D.: An empirically derived inorganic sea spray source function incorporating sea surface temperature, Atmos. Chem. Phys., doi:10.5194/acp-15-11047-2015, 2015.

Woodward, S.: Modeling the atmospheric life cycle and radiative impact of mineral dust in the Hadley Centre climate model, J. Geophys. Res. Atmos., doi:10.1029/2000JD900795, 2001.

Zender, C. S., Bian, H. and Newman, D.: Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology, J. Geophys. Res. D Atmos., doi:10.1029/2002jd002775, 2003.