

Marine aerosol in CRESCENDO ESMs: evaluation and impact on radiative forcing

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What is marine aerosol?

Marine aerosol

- Sea salt aerosol (primary)
- Primary organic matter (primary)
- DMS emissions leading to SO_4 aerosol or MSA (secondary)

Non-marine natural aerosol

- Mineral dust, BVOCs (biogenic volatile organic compounds) leading to SOA, fire emissions
- SO_2 emissions from volcanoes leading to SO_4
- ...

Comparison with anthropogenic aerosol

- On the global scale, natural aerosol is more abundant. Locally, anthropogenic contribution might prevail.
- On a global scale : $\text{AOD}_{\text{nat}} \gg \text{AOD}_{\text{ant}}$
- Considerable contribution of PM_{10} (aerosol with diameter $> 10 \mu\text{m}$) is natural

Role of marine aerosol

Role in equilibrium state of the climate

Effective radiative forcing (ERF) from doubling natural emissions [Thornhill et al., 2021a]

		CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2-1	Multi-model
DMS	$[\text{W m}^{-2}]$		-1.22		-1.27		-0.61	-1.02 ± 0.29
Sea salt	$[\text{W m}^{-2}]$	-1.04	-1.27	-0.35	-2.28	-1.84	-1.30	-1.35 ± 0.61
OPOM	$[\text{W m}^{-2}]$				-0.54			

Role in climate change

Feedback factor [Thornhill, 2021a]

		CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2-1	Multi-model
DMS	$[\text{W m}^{-2} \text{K}^{-1}]$		0.0027		0.0125		-0.0006	0.005
Sea salt	$[\text{W m}^{-2} \text{K}^{-1}]$	-0.041	-0.0014	0.0004	-0.044	-0.084	0.0041	-0.027
OPOM	$[\text{W m}^{-2} \text{K}^{-1}]$							

Role in strength of anthropogenic ERF

Role as CCN or INP

Impact of natural background on indirect forcing [Carslaw et al., 2013]

Role in air-quality

Impact on $\text{PM}_{2.5}$ and PM_{10} from natural emissions [Allen et al., 2020]

What implies a good description of marine aerosol?

Emission strength/pattern – Atm. chemistry – Atm. transport – Deposition – Optical properties – Relation to cloud microphysics (CCN / INP)

Part 1

- ESMs and marine aerosol description
- Experimental setup
- Comparison with observations

Part 2

- Sensitivity simulations setup
- Impact of background natural emissions on anthropogenic ERF
- Factors determining natural aerosol ERF

Context

WP3 : Improving ESMs: natural aerosols and trace gases

WP6 : Evaluating natural aerosol and trace gas processes

Model overview

Earth System Models (ESMs)

	Horizontal resolution	#Levels	Tropospheric aerosol/chemistry
CNRM-ESM2-1	$1.4^{\circ} \times 1.4^{\circ}$	91	aer.
EC-Earth3-AerChem	$3^{\circ} \times 2^{\circ}$	34	chem. + aer.
IPSL-CM6-INCA5	$2.5^{\circ} \times 1.3^{\circ}$	79	chem. + aer.
NorESM1.2	$1.3^{\circ} \times 0.9^{\circ}$	30	aer.
UKESM1	$1.9^{\circ} \times 1.3^{\circ}$	85	chem. + aer.

Treatment of DMS

	Upper ocean DMS climatology	Flux parameterisation	Chemistry
CNRM	Kettle et al. [1999] (emitted as SO_2)	–	fixed lifetime of SO_2
EC-Earth	Lana et al. [2011]	Wanninkhof [2014]	full chemistry
IPSL			full chemistry
NorESM	Lana et al. [2011]	Nightingale et al. [2000]	prescribed oxidants
UKESM	Lana et al. [2011]	Nightingale et al. [2000]	full chemistry

Sea-salt

	Emission parameterisation	Representation of sea salt
CNRM (TACTIC)	Grythe et al. (2014)	3 bins
EC-Earth (M7)	Gong et al. [2003], Salter et al. [2015]	2 modes
IPSL (5 modes)		3 soluble modes
NorESM (10 modes)	Salter et al. [2015]	3 modes
UKESM (GLOMAP)	Gong et al. [2003]	2 modes

Experiment overview

Experimental setup (AMIP-type)

- Period 2000–2014
- Forcings (volcanoes, GHGs, landuse, ...) : follow 2000–2014 historical evolution
- SSTs : prescribed (follow historical evolution)
- **Free-running** and **nudged** (ERA-Interim)
- Emissions : present-day (PD), 1850, 1750 [allows to estimate ERF]

Simulations

			CNRM	CNRM-CRESC	EC-Earth	IPSL	NorESM	UKESM
(1)	Free running	PD emissions	x	x	x	x	x	x
(2)	Free running	1850 emissions	x	x	x	x	x	x
(3)	Free running	1750 emissions	x	x		x	x	
(4)	Nudged	PD emissions	x	x	x	x		
(5)	Nudged	1850 emissions	x	x		x		
(6)	Nudged	1750 emissions	x	x		x		

Model characteristics (for PD conditions) (1)

Emissions (global)

			CNRM	CNRM-CRESC1	EC-Earth	IPSL	NorESM	UKESM
DMS emission	[Tg yr ⁻¹]	Free running	(*)	(*)	51.5	40.8	67.8	32.7
		Nudged	(*)	(*)	51.8	41.1	65.7	
Sea-salt emissions	[Tg yr ⁻¹]	Free running		18342	5686	3950	2035	5595
		Nudged		10737	5728	3788	1916	

Life time (global)

			CNRM	CNRM-CRESC1	EC-Earth	IPSL	NorESM	UKESM
DMS	[day]	Free running	(*)	(*)	1.16	1.35	1.53	1.61
		Nudged			1.51	1.33	1.50	
Sea	[day]	Free running	0.32	0.39	0.38	2.65	0.97	0.53
		Nudged	0.24	0.30	0.37	2.68	1.00	
Sulphate	[day]	Free running	2.26	2.24	6.62	3.81	3.38	6.05
		Nudged	2.64	2.64	5.99	3.85	3.46	

Model characteristics (PD conditions) (2)

Dry deposition (% w.r.t. total deposition) (global)

			CNRM	CNRM-CRESC1	EC-Earth	IPSL	NorESM	UKESM
SO ₄ dry deposition	[%]	Free running	13	13	3	9	13	16
		Nudged	15	15		9	13	
Sea salt dry deposition	[%]	Free running	50	63	79	10	43	69
		Nudged	47	55		10	44	

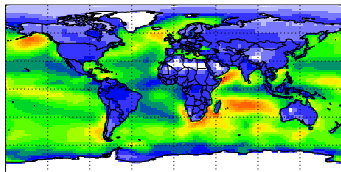
AOD (550 nm) (global)

			CNRM	CNRM-CRESC1	EC-Earth	IPSL	NorESM	UKESM
With PD emissions	[-]	Free running	0.098	0.108	0.129	0.119	0.123	0.177
		Nudged	0.083	0.085		0.117	0.130	
With 1850 emissions	[-]	Free running	0.080	0.092	0.083	0.090	0.098	0.114
PD anthropogenic contribution	[-]	Free running	0.017	0.016	0.046	0.029	0.025	0.063

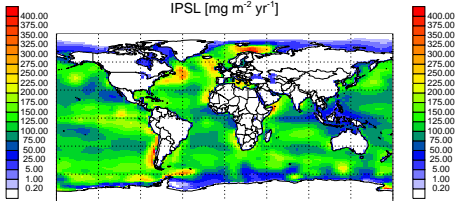
DMS emissions

Free running – 2000–2014 period – annual mean

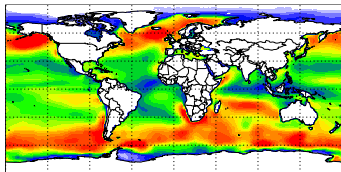
EC-Earth [$\text{mg m}^{-2} \text{yr}^{-1}$]



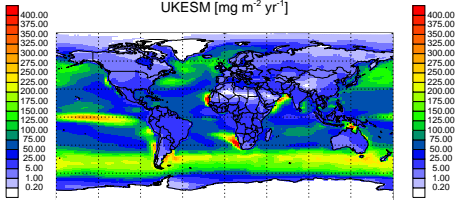
IPSL [$\text{mg m}^{-2} \text{yr}^{-1}$]



NorESM1.2 [$\text{mg m}^{-2} \text{yr}^{-1}$]



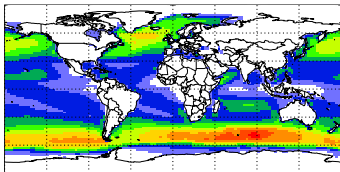
UKESM [$\text{mg m}^{-2} \text{yr}^{-1}$]



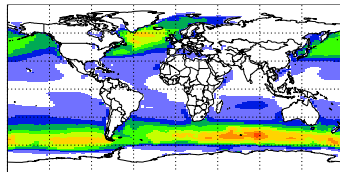
Sea salt emissions

Free running – 2000–2014 period – annual mean

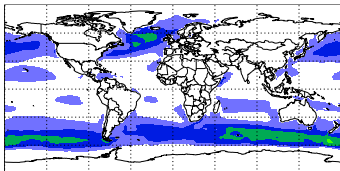
EC-Earth [$\text{g m}^{-2} \text{yr}^{-1}$]



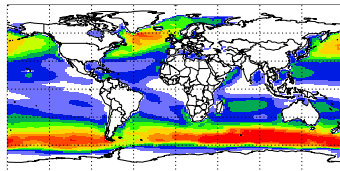
IPSL [$\text{g m}^{-2} \text{yr}^{-1}$]



NorESM1.2 [$\text{g m}^{-2} \text{yr}^{-1}$]



UKESM [$\text{g m}^{-2} \text{yr}^{-1}$]

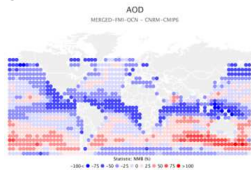


Comparison with observations

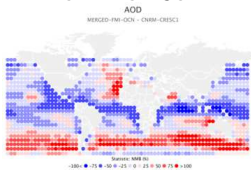
- AOD over the ocean
- DMS at Amsterdam Island
- SO₄ at remote locations
- Sea salt at remote locations

Comparison with AOD observations

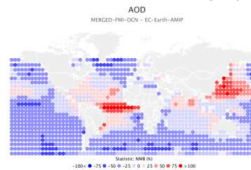
CNRM



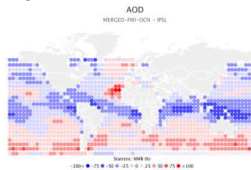
CNRM-CRESC



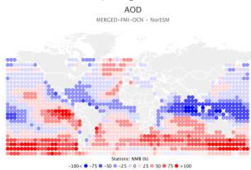
EC-Earth



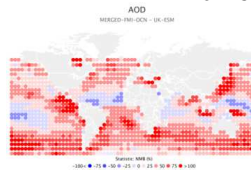
IPSL



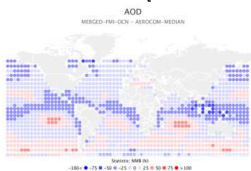
NorESM



UKESM

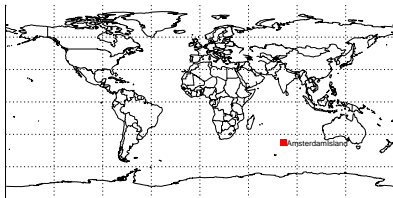


AeroCom-III median [Gliß et al., 2021]

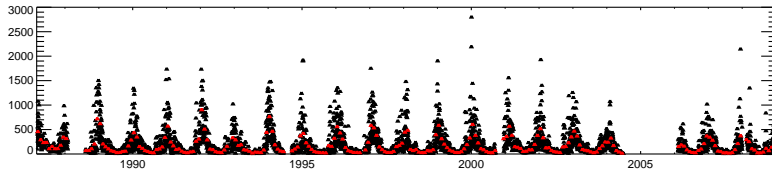


DMS observations at Amsterdam Island (1)

Amsterdam Island



Observations [Sciare et al., 2000; Sciare et al., 2001] (black = daily) (red = monthly)



Observation frequency

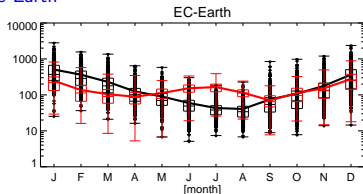
Observations done at 08:00 local time (estimated to be representative for daily mean)

DMS observations at Amsterdam Island (2)

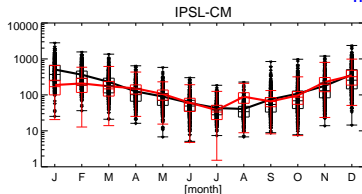
Seasonal cycle of atmospheric DMS concentrations at the surface [ppt]

Observations – Model results

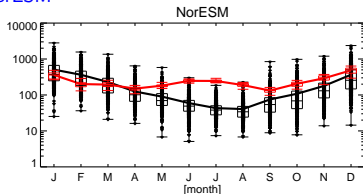
EC-Earth



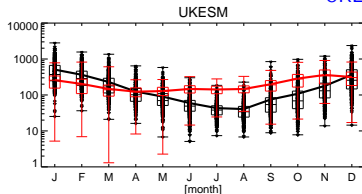
IPSL



NorESM

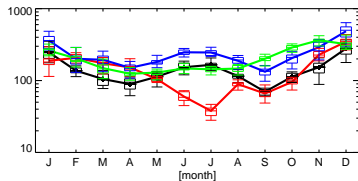


UKESM



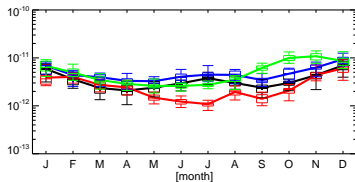
DMS observations at Amsterdam Island (3)

DMS concentration [ppt]

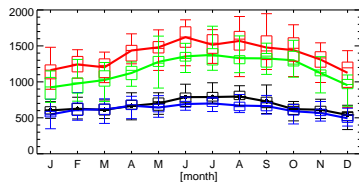


EC-Earth – IPSL – NorESM – UKESM

Local emission strength [$\text{kg m}^{-2} \text{s}^{-1}$]



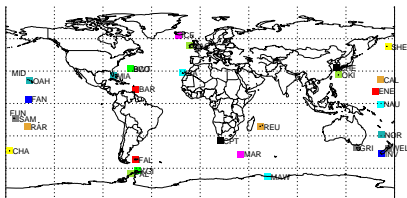
PBL height [m]



SO₄ observations at remote locations (1)

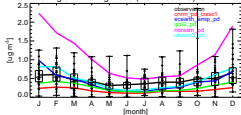
Observation period 1981–2002 [Savoie and Prospero, 1977]

Observation location

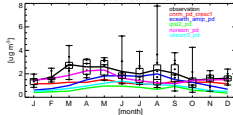


SO₄ observations at remote locations (2)

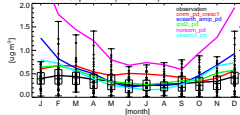
WEL Wellington / Baring Head (New Zealand) lon=174 lat=-41



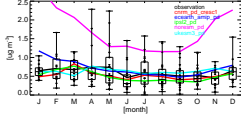
BWT Bermuda West lon=-64 lat=32



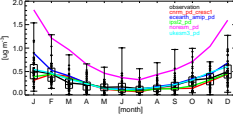
GRI Cape Grim (Tasmania) lon=144 lat=-40



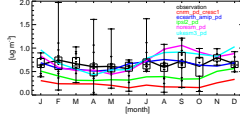
CPT Cape Point (South Africa) lon=18 lat=-34



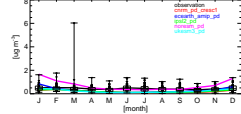
CHA Chatham Island (New Zealand) lon=-176 lat=-43



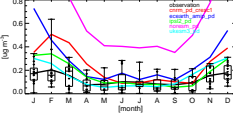
FAN Fanning Island lon=-159 lat=-3



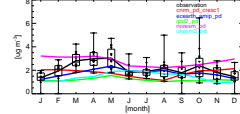
INV Invercargill (New Zealand) lon=168 lat=-46



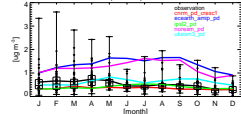
MAR Marion Island / Prince Edward Island lon=37 lat=-46



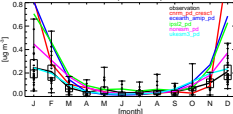
MIA Miami lon=-80 lat=25



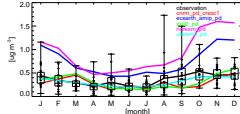
OAH Oahu (Hawaii) lon=-157 lat=21



PAL Palmer Station (Antarctica) lon=-64 lat=-64



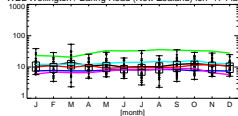
REU Reunion Island lon=55 lat=-21



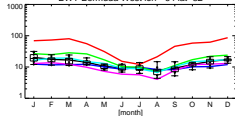
Observations – CNRM – EC-Earth – IPSL – NorESM – UKESM

Sea salt observations at remote locations

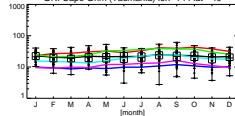
WEL Wellington / Baring Head (New Zealand) lon=174 lat=41



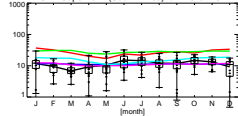
BWT Bermuda West lon=-64 lat=32



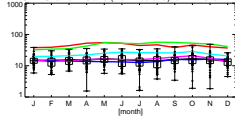
GRI Cape Grim (Tasmania) lon=144 lat=40



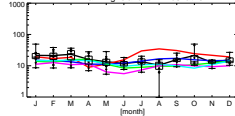
CPT Cape Point (South Africa) lon=18 lat=34



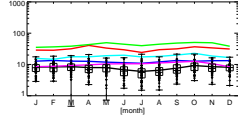
CHA Chatham Island (New Zealand) lon=-176 lat=-43



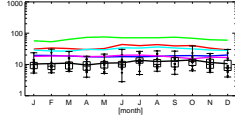
FAN Fanning Island lon=-159 lat=3



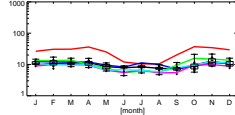
INV Invercargill (New Zealand) lon=168 lat=46



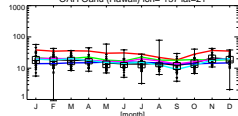
MAR Marion Island / Prince Edward Island lon=37 lat=-46



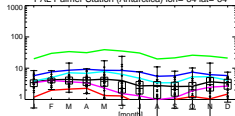
MIA Miami lon=-80 lat=25



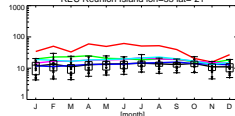
OAH Oahu (Hawaii) lon=-157 lat=21



PAL Palmer Station (Antarctica) lon=64 lat=-64



REU Reunion Island lon=55 lat=21



Observations – CNRM – EC-Earth – IPSL – NorESM – UKESM

Sensitivity experiments : motivation

Impact of natural emissions on anthropogenic ERF

- CMIP6 anthropogenic ERF estimates [Smith et al., 2020; Thornhill et al., 2021b]
- Are these estimates (very) sensitive to the natural aerosol background state?

Impact of anthropogenic emissions on natural aerosol ERF

- CMIP6 estimates for ERF of natural emissions for 100% → 200% in 1850 background [Thornhill et al., 2021a]
- Might result be sensitive to natural reference state : 0% → 50%, 50% → 100%, 100% → 200%?
- Might result be sensitive to anthropogenic background : 1850 versus PD?

Understanding the contribution from different natural aerosol

- Contribution from specific natural aerosol sources to AOD
- Contribution of DMS emissions to SO_4 surface concentration
- Contribution of OPOM emissions to OA surface concentration

Simulation setup

- For each natural aerosol components : removing (0%), halving (50%), or doubling (200%) of emissions
- Large number of simulations → short simulations → Nudged simulations
- Do free-running and nudged simulations behave similarly?

Sensitivity experiments : setup

Nudged simulations for year 2014

Emission	Scaling	CNRM 2014	CNRM-CRESC 2014	EC-Earth 2014	IPSL 2013–2014	NorESM 2012–2014	UKESM
DMS	×0.0			PD, 1850		PD, 1850, 1750	
	×0.5	PD		PD, 1850		PD, 1850, 1750	
	×2.0	PD		PD, 1850		PD, 1850, 1750	
Seasalt	×0.0			PD, 1850		PD, 1850, 1750	
	×0.5		PD	PD, 1850		PD, 1850, 1750	
	×2.0		PD	PD, 1850		PD, 1850, 1750	
OPOM	×0.0					PD, 1850, 1750	
	×0.5					PD, 1850, 1750	
	×2.0					PD, 1850, 1750	
Dust	×0.0			PD, 1850		PD, 1850, 1750	
	×0.5			PD, 1850	PD	PD, 1850, 1750	
	×2.0			PD, 1850	PD	PD, 1850, 1750	
BVOC	×0.0			PD, 1850		PD, 1850, 1750	
	×0.5			PD, 1850	PD	PD, 1850, 1750	
	×2.0	PD		PD, 1850	PD	PD, 1850, 1750	

Anthropogenic ERF

This study : 15-year period, nowadays SSTs

	Reference year		CNRM	CNRM-CRESC	EC-Earth	IPSL	NorESM	UKESM
Free running	1850	$[\text{W m}^{-2}]$	-0.86	-0.86	-1.00	-0.31	-1.08	1.44
Free running	1750	$[\text{W m}^{-2}]$	-0.88	-0.94			-1.07	1.37
Nudged	1850	$[\text{W m}^{-2}]$					-1.09	
Nudged	1750	$[\text{W m}^{-2}]$					-1.10	

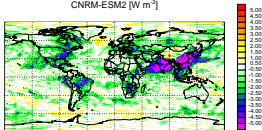
CMIP6 [Smith et al., 2020] : 30-year period, pre-industrial SSTs

		Aerosols	Anthropogenic
CNRM-CM6-1	$[\text{W m}^{-2}]$	-1.15	1.61
CNRM-ESM2-1	$[\text{W m}^{-2}]$	-0.74	1.66
EC-Earth3	$[\text{W m}^{-2}]$	-0.80	2.09
HadGEM3-GC31-LL	$[\text{W m}^{-2}]$	-1.10	1.81
IPSL-CM6A-LR	$[\text{W m}^{-2}]$	-0.63	2.32
NorESM2-LM	$[\text{W m}^{-2}]$	-1.21	2.06
NorESM2-MM	$[\text{W m}^{-2}]$	-1.26	
UKESM1-0-LL	$[\text{W m}^{-2}]$	-1.11	1.79
...	
Mean	$[\text{W m}^{-2}]$	-1.01	2.00

Distribution of ERF

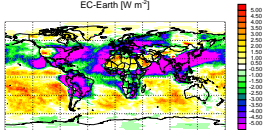
CNRM

CNRM-ESM2 [W m^{-2}]



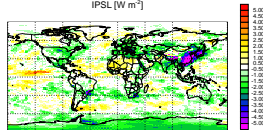
EC-Earth

EC-Earth [W m^{-2}]



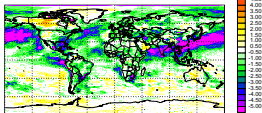
IPSL

IPSL [W m^{-2}]



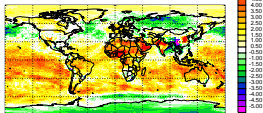
NorESM

NorESM1.2 [W m^{-2}]



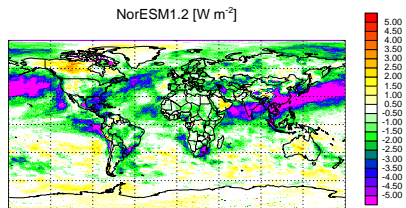
UKESM

UKESM [W m^{-2}]

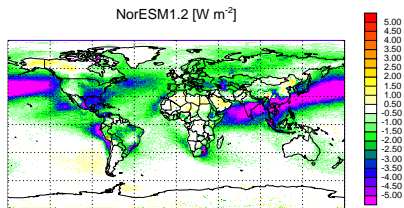


Anthropogenic ERF : sensitivitie to nudging

Free running (w.r.t. 1850)

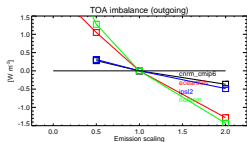


Nudging (w.r.t. 1850)



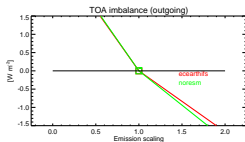
Impact of natural emissions on anthropogenic ERF (1)

$\Delta\text{TOA}(1850)$

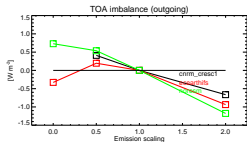
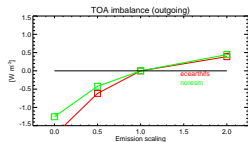


$\Delta\text{TOA}(\text{PD})$

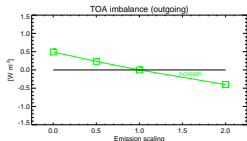
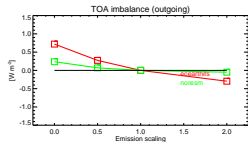
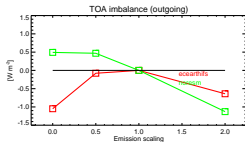
DMS



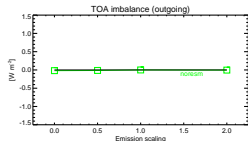
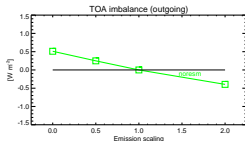
$\text{ERF} = \Delta\text{TOA}(\text{PD}) - \Delta\text{TOA}(1850)$



Sea salt



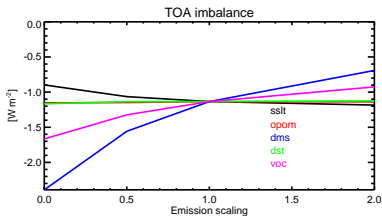
OPOM



CNRM – EC-Earth – IPSL – NorESM

Impact of natural emissions on anthropogenic ERF (2)

Sensitivity of anthropogenic ERF (NorESM)



Sea salt – OPOM – DMS – Dust – BVOCs

Sensitivity of natural emissions ERF

Forcing from doubling (100%→200%) in 1850 conditions [Thornhill et al., 2021a]

		CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2-1	Multi-model
DMS	[W m ⁻²]		-1.22		-1.27		-0.61	-1.02±0.29
Sea salt	[W m ⁻²]	-1.04	-1.27	-0.35	-2.28	-1.84	-1.30	-1.35±0.61
OPOP	[W m ⁻²]				-0.54			

Questions

- Role of PD or 1850 anthropogenic emissions?
- Is sensitivity equal : $2 \times (0\% \rightarrow 50\%) = 2 \times (50\% \rightarrow 100\%) = (100\% \rightarrow 200\%)$?

Sensitivity in natural ERF

		CNRM	EC-Earth	IPSL	NorESM
Present day					
DMS	[W m ⁻²]	-/-0.56/-0.37	-2.36/-2.10/-1.28	-/-0.62/-0.48	-4.98/-2.55/-1.43
Sea salt	[W m ⁻²]	-/-0.82/-0.66	1.05/-0.39/-0.94		-0.38/-1.08/-1.18
Ocean OM	[W m ⁻²]				-0.51/-0.47/-0.40
Dust	[W m ⁻²]		-0.28/-0.28/-0.26	-/-0.12/-0.09	0.67/0.32/0.12
BVOC	[W m ⁻²]	-/-/-0.23	0.69/0.84/0.38		-1.26/-0.70/-0.56
1850					
DMS	[W m ⁻²]		-4.65/-3.33/-1.67		-6.65/-3.39/-1.88
Sea salt	[W m ⁻²]		1.94/0.16/-0.64		-0.05/-0.94/-1.13
Ocean OM	[W m ⁻²]				-0.52/-0.50/-0.40
Dust	[W m ⁻²]		-0.06/-0.18/-0.21		0.61/0.32/0.11
BVOC	[W m ⁻²]		1.65/1.71/1.26		-1.95/-1.08/-0.77
$2 \times (0\% \rightarrow 50\%) / 2 \times (50\% \rightarrow 100\%) / 100\% \rightarrow 200\%$					

Marine aerosol modelling

- Emission parameterisations quite similar
- Large variation in emission strength (compared to anthropogenic emissions) : DMS and sea-salt emissions differ by factors of two
- Considerable differences in lifetime of DMS, SO₄ and sea salt
- Varying agreement with observations

Interactions between natural and anthropogenic aerosol

- Anthropogenic aerosol ERF strengthens for decreasing DMS emissions, but weakens for decreasing sea-salt emissions
- ERF of natural emissions is sensitive to anthropogenic (1850 versus PD) and natural background