Role of dimethyl sulfide on the formation and growth of aerosols, and its impact on liquid clouds in the Arctic summer

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EGU General Assembly
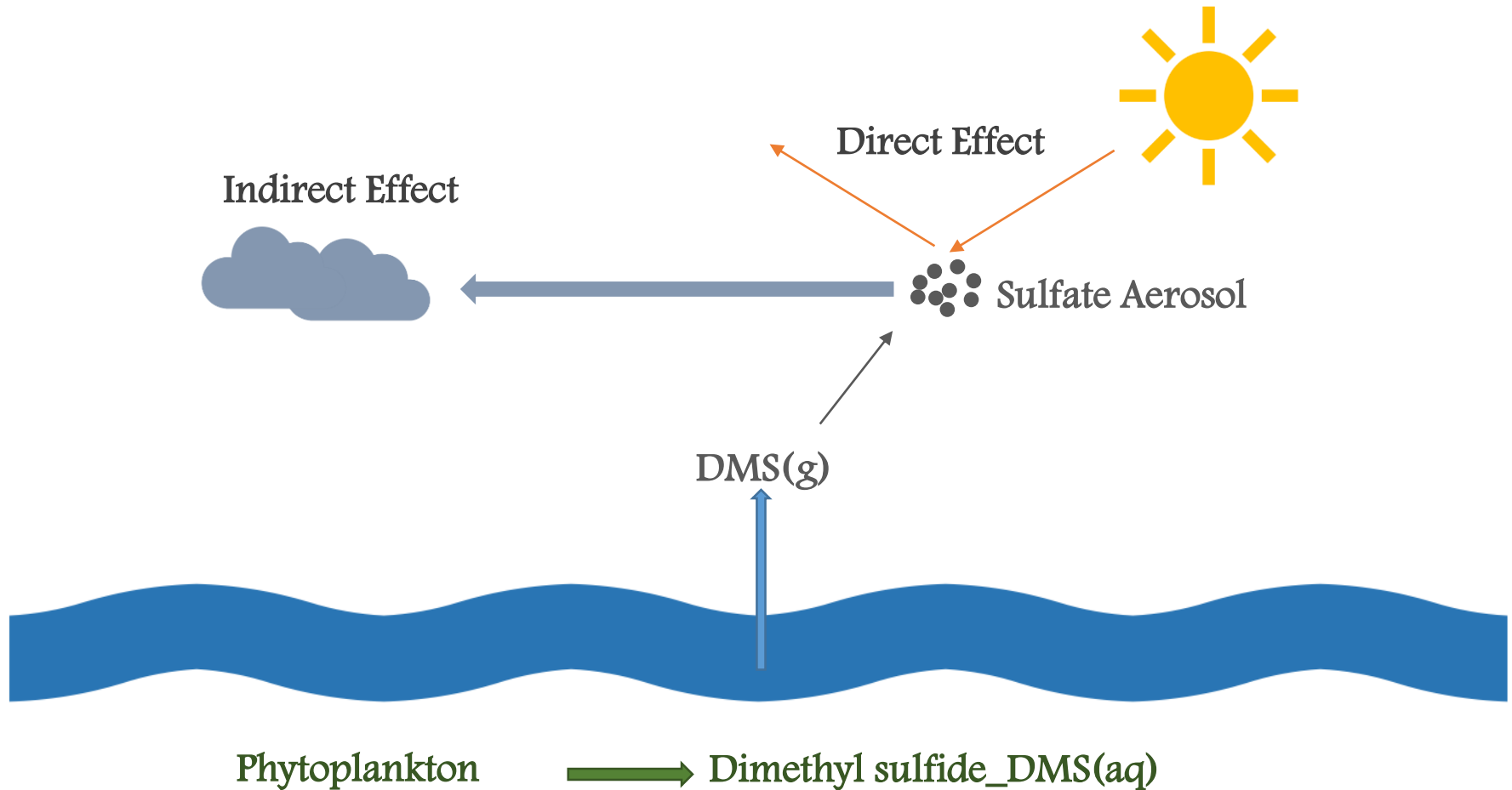
04-May-2020

Canada

Abstract

- Atmospheric dimethyl sulfide, DMS, is the main biogenic source of sulfate particles in the Arctic atmosphere. Sulfate particles have a net cooling effect, which can partially offset Arctic warming from absorbing aerosols, such as black carbon. As efficient cloud condensation nuclei (CCN), sulfate particles are also able to influence the cloud's microphysical properties.

- DMS production and emission to the atmosphere increase during the Arctic summer, due to a greater ice-free sea surface area and higher biological activity. In the model simulation of a field campaign conducted over the Canadian high Arctic during the summer of 2014 (NETCARE; Abbatt et al. 2019), the inclusion of DMS in the model, GEM-MACH, resulted in a significant increase, up to 100%, in the modelled atmospheric SO_2 in some regions of the Canadian Arctic. Analysis of the modelled size-segregated aerosol sulfate indicated that DMS has the most significant impact on particles in the size range of 50 – 200 nm in this case. Simulations have shown that localized regions of high seawater DMS can have a significant impact on atmospheric concentrations (Ghahremaninezhad et al., 2019).

- Further investigation of DMS impact on the Arctic summer cloud microphysics was carried out by using a fully coupled version of GEM-MACH. Overall, the model simulations show that the inclusion of DMS in model leads to an increase in cloud droplet number concentrations (CDNC), liquid water content (LWC) and a decrease in droplet mean mass diameters (MMD).

Outline

❑ Introduction

❑ DMS in GEM-MACH model

❑ Impact of DMS on sulfate aerosols in the Arctic

❑ Impact of DMS on liquid clouds in the Arctic summer

❑ Summary

DMS in the Atmosphere

DMS(g) the main biogenic source of sulfate particles in the Arctic

Direct Effect

Indirect Effect

Sulfate Aerosol

DMS(g)

Phytoplankton → Dimethyl sulfide_DMS(aq)

Why the Arctic & Why Summer

Climate change in the Arctic: Faster than lower latitudes

Motivation

Biogenic Sulfate:
15% of the tropospheric sulfur budget globally and
up to *100%* in some remote areas.
One example: July *2014 NETCARE* Campaign
More than 63% of sulfate fine aerosols (<0.49 μm) are from DMS
(Ghahremaninezhad et al., 2016).

DMS production and emission to the atmosphere increase during the
summer due to greater ice-free sea surface and higher biological
activity.

GEM-MACH

GEM-MACH, *ECCC*'s air quality prediction model; It consists of an online tropospheric chemistry module **MACH (M**odelling **A**ir quality and **CH**emistry) embedded within ECCC's numerical weather forecast model **GEM** (**G**lobal **E**nvironmental **M**ulti-scale model).

- GEM-MACH's chemistry module includes a comprehensive representation of air quality processes, such as gas-phase, aqueous-phase, heterogeneous chemistry; secondary organic aerosol formation; aerosol microphysics; sedimentation of particles; and dry deposition and wet removal of gases and particles.

- GEM-MACH uses a sectional representation and considers 8 chemical components (SO_4, NO_3, NH_4, EC, POC, SOC, CM, and sea-salt) for atmospheric aerosols.

Adding DMS(g) in GEM-MACH

Model: GEM-MACH V2, 12-bin (aerosol) configuration

Domain: Arctic 15-km

Chemical boundary conditions of DMS: MOZART-4 2014

Flux estimate: $F = -K_W \left(\dfrac{C_g}{K_H} - C_l \right)$ *(Johnson et al., 2010)*

K_H: "Dimensionless gas over liquid" form
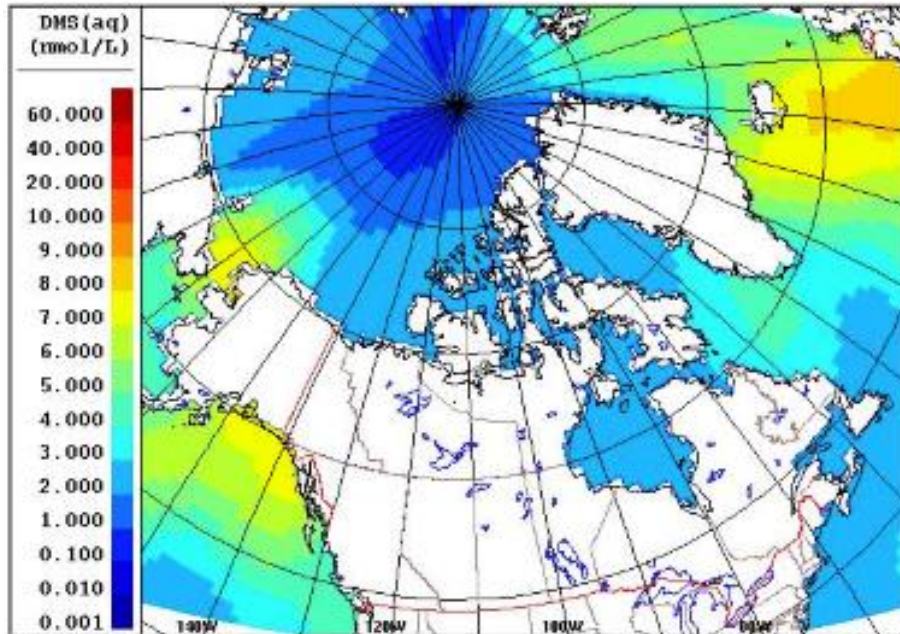of the Henry's law constant

K_W: Transfer velocity
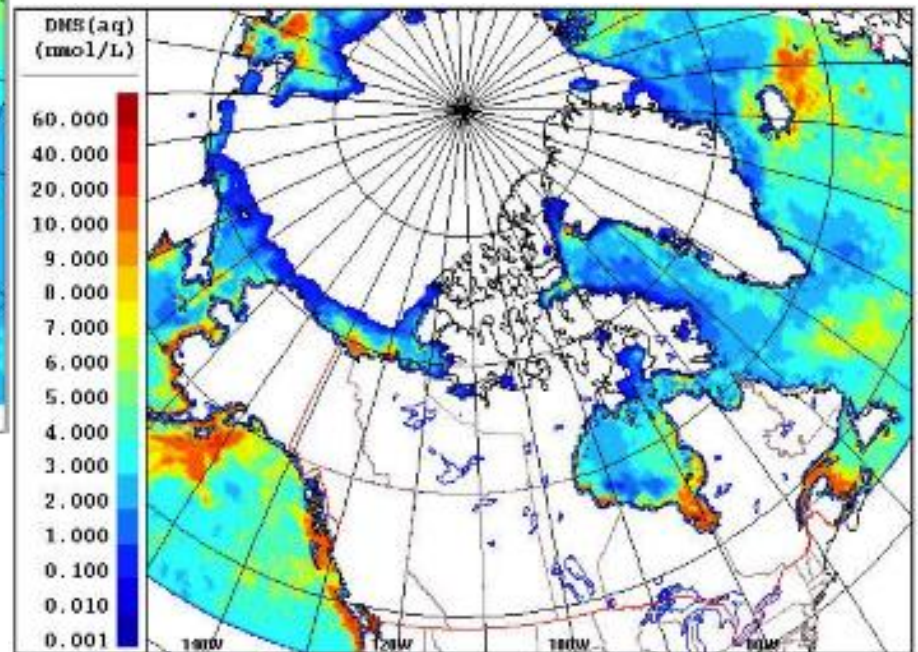
$\underline{C_l}$: DMS in water

1) DMS(aq) climatology dataset (Lana et al., 2011)
2) DMS(aq) satellite-based dataset (Galí et al., 2018).

Sea water DMS datasets
(Source of atmospheric DMS in the model)

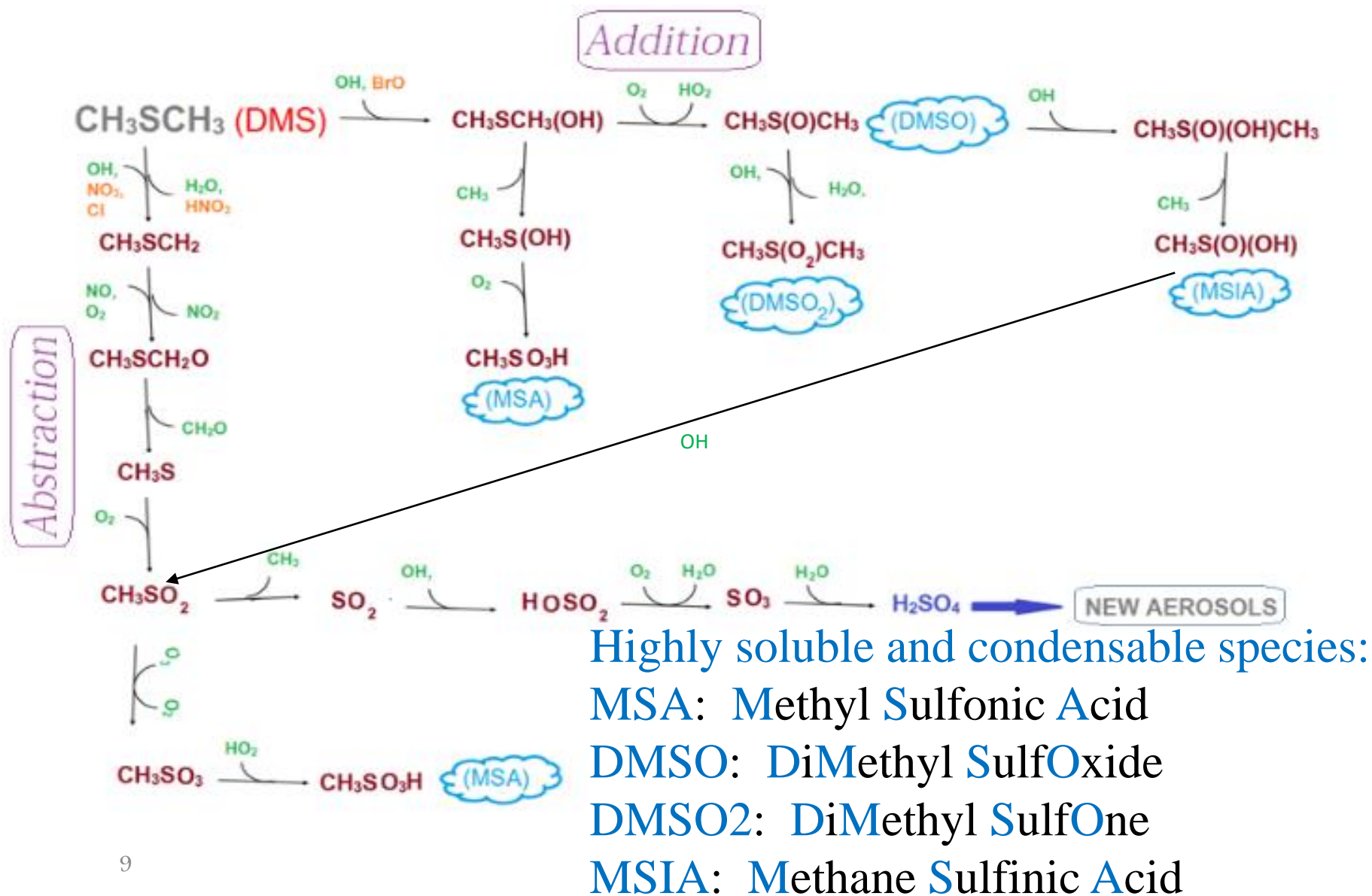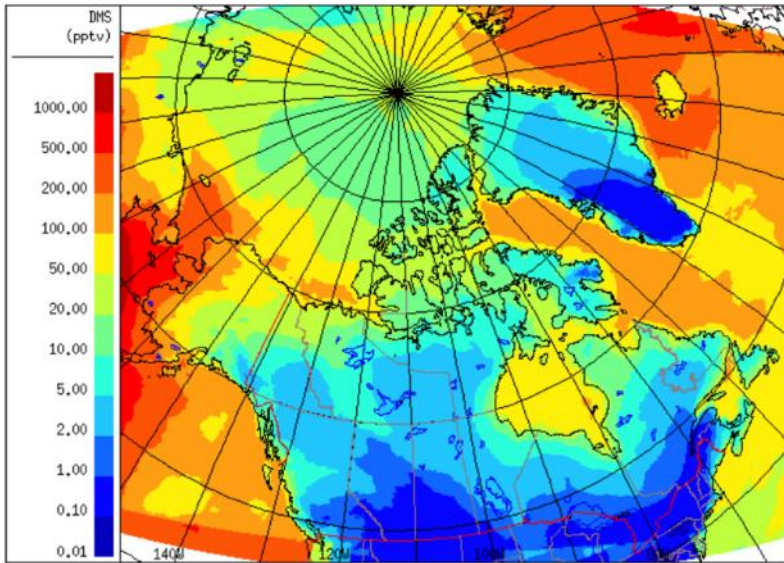### July average of the DMS(aq) concentrations (nmol/L)

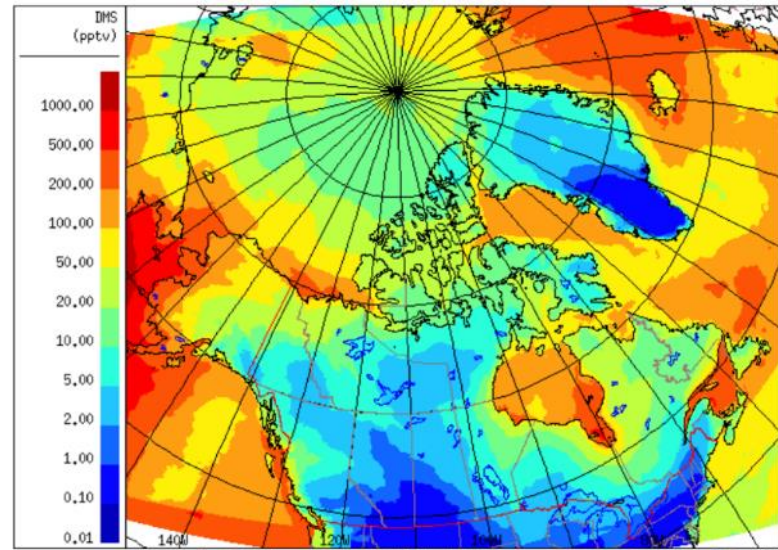Climatology DMS(aq)

Satellite DMS(aq)

DMS(g) reactions/chemistry added to GEM-MACH

Addition

OH, BrO

CH_3SCH_3 (DMS) → $CH_3SCH_3(OH)$ → O_2 HO_2 → $CH_3S(O)CH_3$ (DMSO) → OH → $CH_3S(O)(OH)CH_3$

Abstraction

OH, NO$_3$, Cl / H$_2$O, HNO$_3$

CH_3SCH_2

CH$_3$ → $CH_3S(OH)$

OH, / H$_2$O, → $CH_3S(O_2)CH_3$

CH$_3$ → $CH_3S(O)(OH)$

NO, O$_2$ / NO$_2$

CH_3SCH_2O

O$_2$ → CH_3SO_3H (MSA)

(DMSO$_2$)

(MSIA)

CH$_2$O

CH_3S

O$_2$

OH

CH_3SO_2 — CH$_3$ → SO_2 · — OH, → $HOSO_2$ — O$_2$ H$_2$O → SO_3 — H$_2$O → H_2SO_4 ⟹ NEW AEROSOLS

CH_3SO_3 — HO$_2$ → CH_3SO_3H (MSA)

Highly soluble and condensable species:
MSA: **M**ethyl **S**ulfonic **A**cid
DMSO: **Di**Methyl **S**ulf**O**xide
DMSO2: **Di**Methyl **S**ulf**O**ne
MSIA: **M**ethane **S**ulfinic **A**cid

Results: July 2014 averaged DMS(g)~pptv

Based on Climatology DMS(aq)

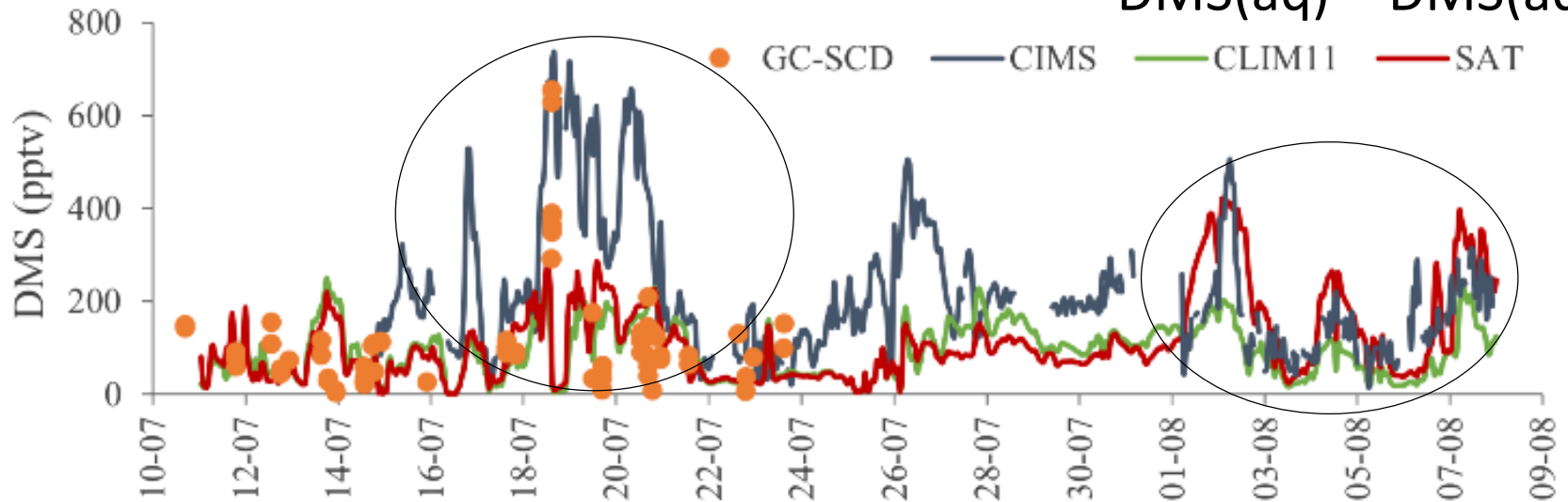Based on Satellite DMS(aq)

Amundsen: Observation vs GEM-MACH

GEM-MACH

Climatology Satellite

DMS(aq)   DMS(aq)

NETCARE Campaign-Summer 2014

 Observation:

GC-SCD *(Ghahremaninezhad et al., 2017)*
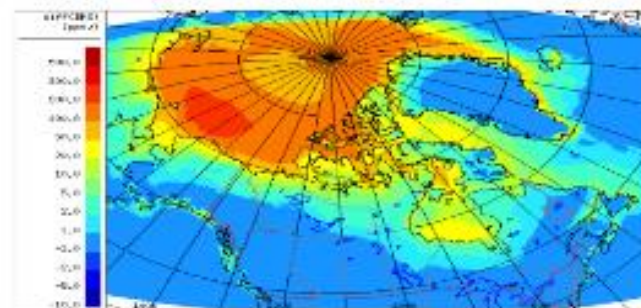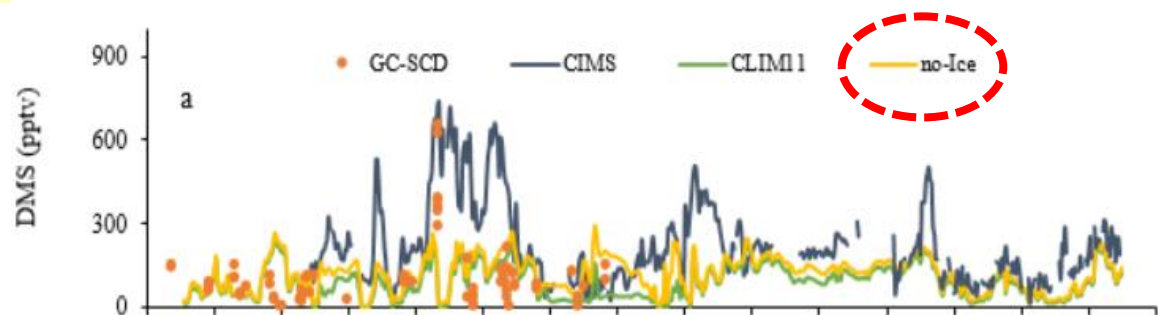
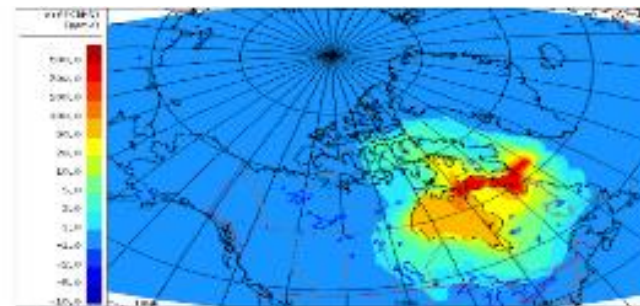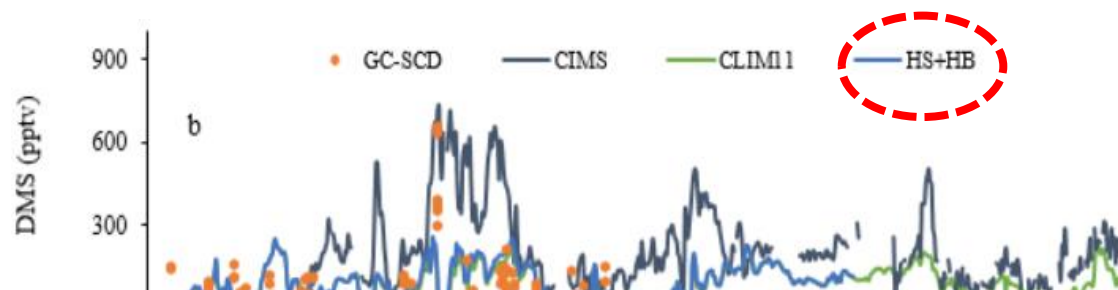CIMS *(Mungall et al., 2016)*

GEM-MACH:

CLIM11

SAT

Sensitivity Tests: what could account for the first DMS peak?
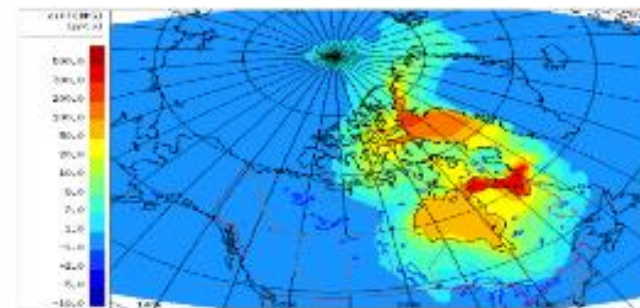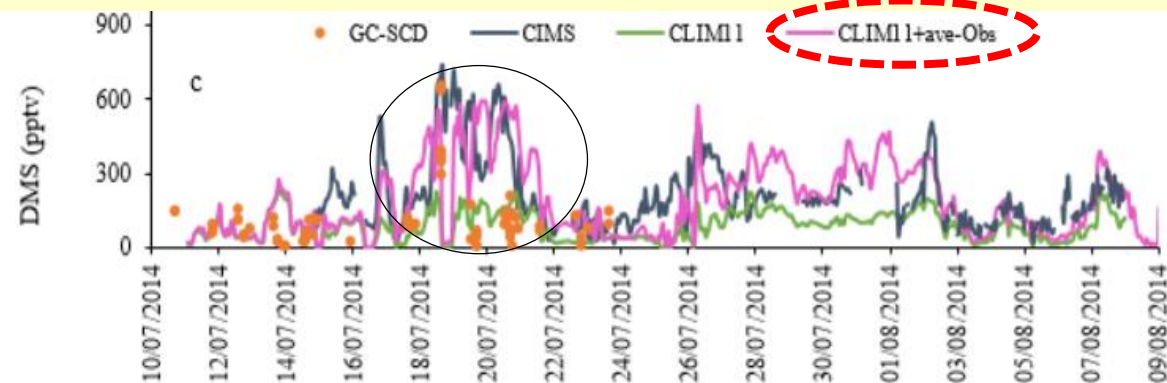
1. "No Ice" effect

Was DMS *transported* from these regions northwards? Apparently not – didn't get the first peak in the time series.

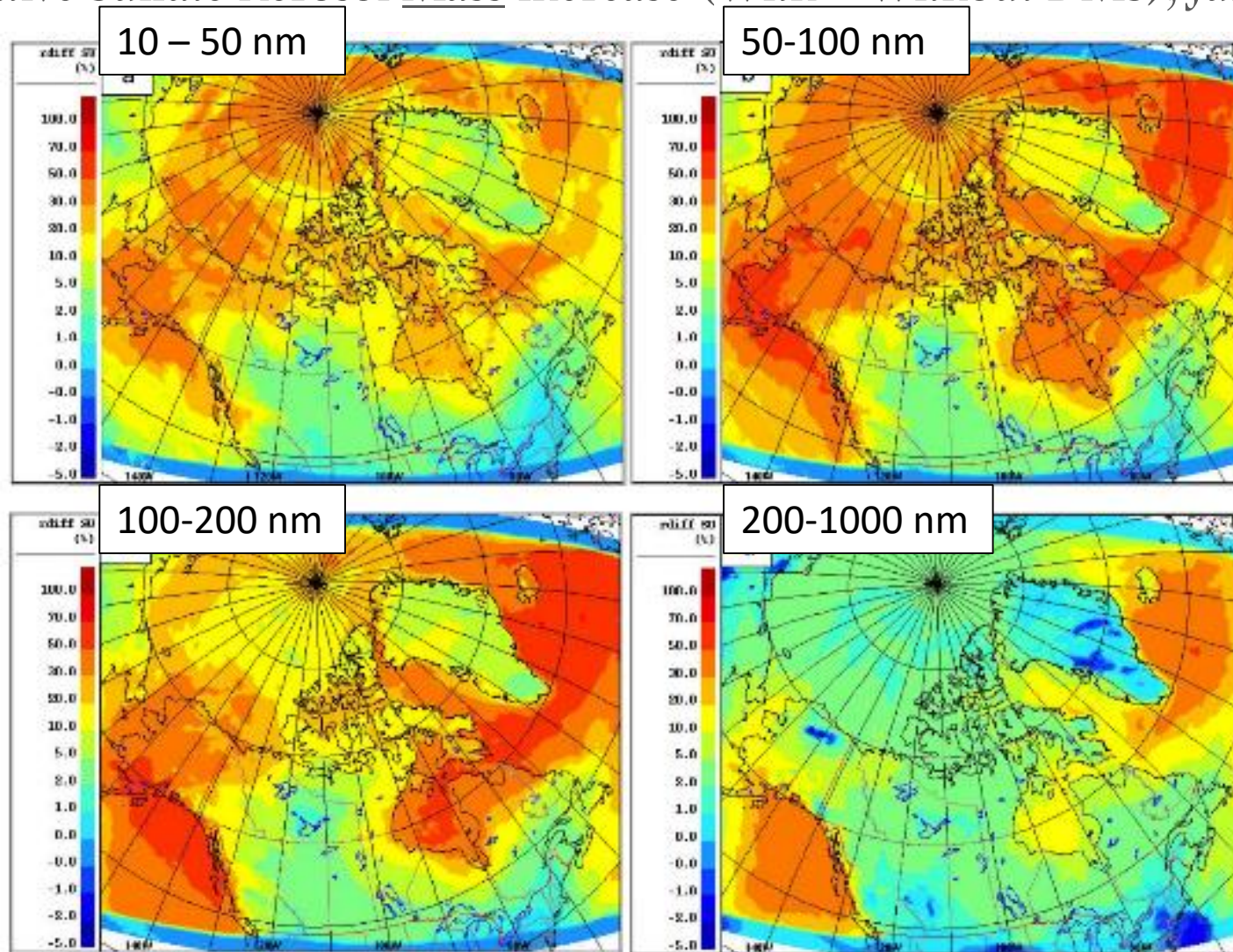## 2. Hudson Strait and Hudson Bay effect (HS~HB)

3. Updated DMS(aq) in Baffin Bay and Lancaster Sound with *in-situ* observations(CLIM11+ave~Obs)

In-situ values in Baffin Bay and Lancaster were required to get the observed peak. Also included HS-HB.

Relative Sulfate Aerosol Mass Increase (With – Without DMS), July 2014

10 – 50 nm | 50-100 nm | 100-200 nm | 200-1000 nm

The enhancement in sulfate particles is most significant in the size range of 50 to 100 nm at high latitudes (> 70° N), however, this enhancement is stronger in the 100~200 nm size range at lower latitudes (< 70° N).
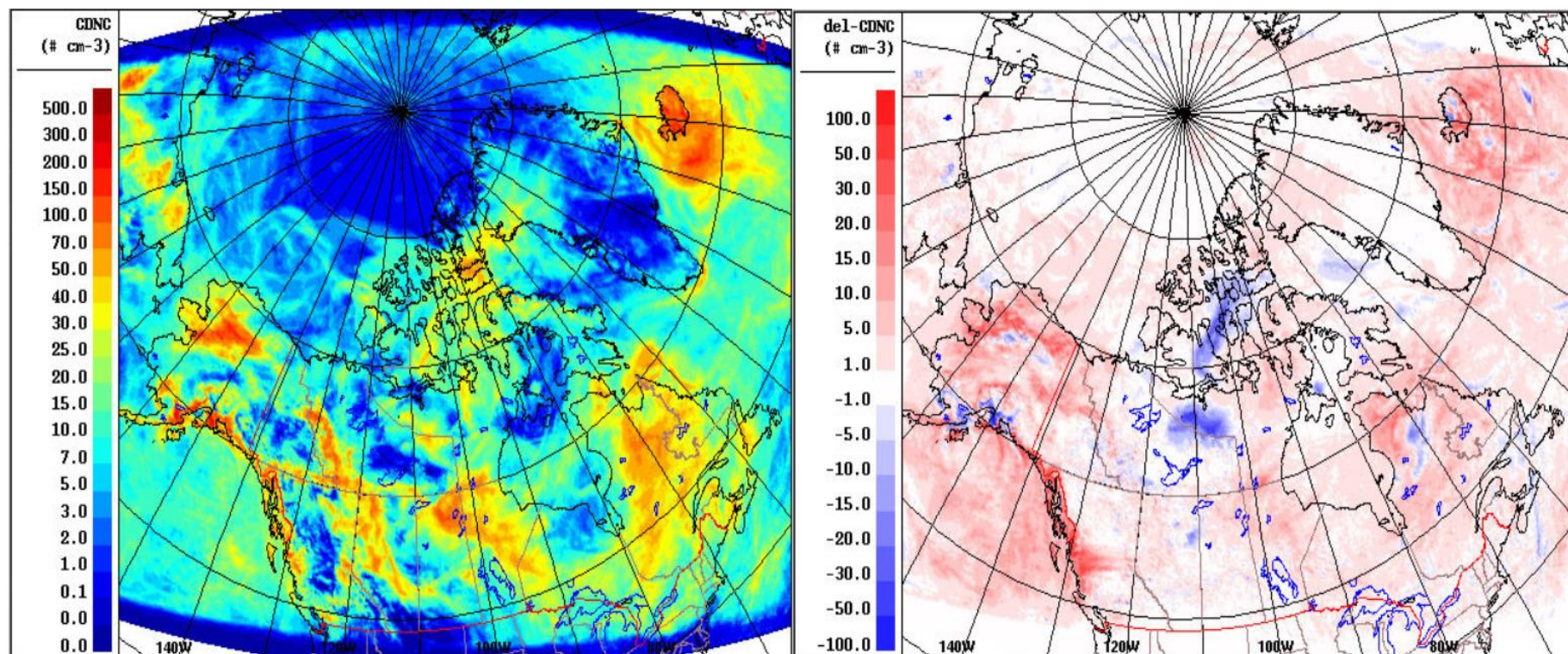
Next step: DMS(g) impact on CCN activation and Cloud Microphysics

Model simulations using <u>fully coupled GEM-MACH</u>, with and without DMS

- ❑ *Aerosol direct effect:* aerosol optical depth, single scattering albedo, and asymmetry factors, determined based on modelled size- and speciation-resolved aerosol, are fed into model radiative transfer code (Makar *et al.*, 2015a,b, AE).

- ❑ *Aerosol indirect effect:* droplet nucleation based on the sectional Abdul-Razzak & Ghan aerosol activation scheme, making use of the on-line size- and speciation-resolved aerosols, is implemented in the model's M-Y double moment microphysics scheme (Gong *et al.*, 2015, AE).

Cloud Droplet Number Concentration

July average of CDNC (# cm^{-3}) at GEM-MACH level 0.87 (~870 mb): Left panel GEM-MACH results without DMS, and right panel, absolute difference between runs with DMS and without DMS
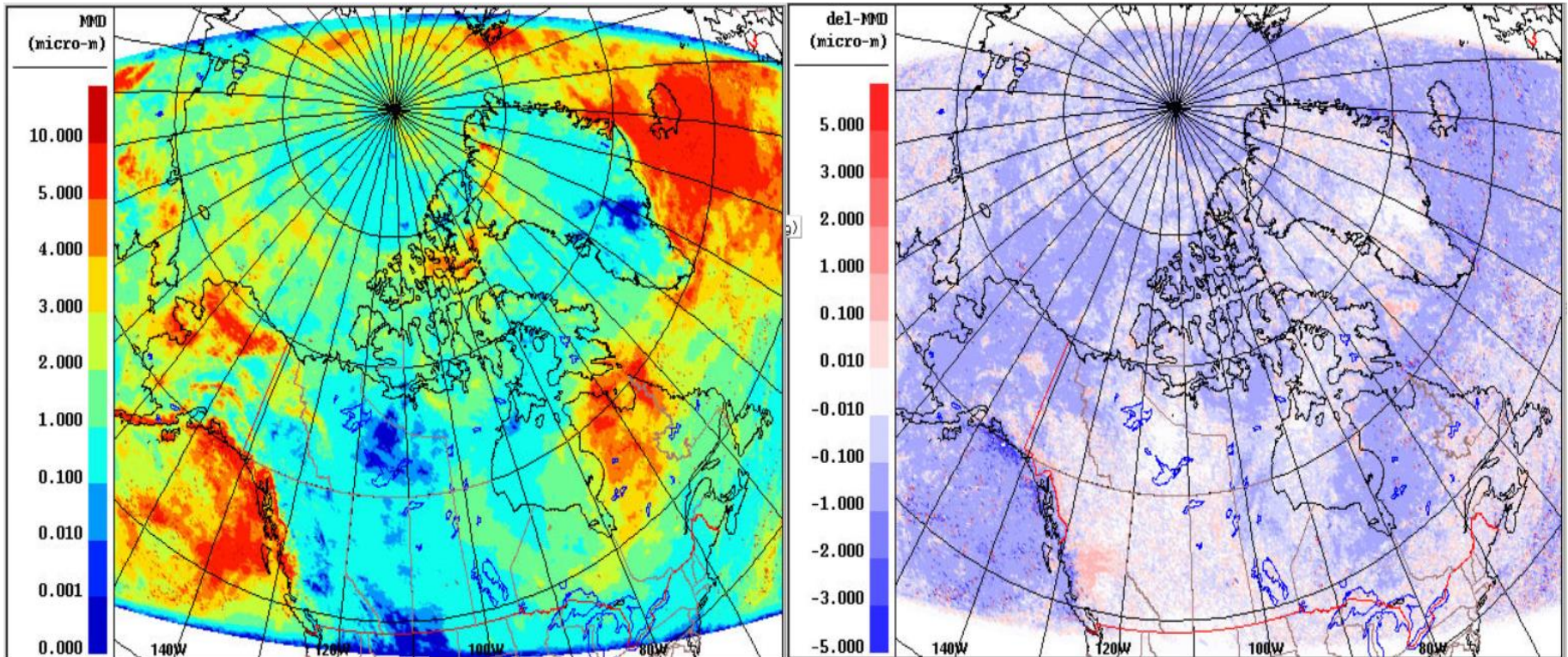

CDNC
(with no DMS)

ΔCDNC
{ (with DMS) −(noDMS) }

Cloud Droplet Mass Mean Diameter (MMD)

July average of MMD (µm) at GEM-MACH level 0.87 (~870 mb): Left panel GEM-MACH results without DMS, and right panel, absolute difference between runs with DMS and without DMS.

MMD
(with no DMS)

ΔMMD
{ (with DMS) −(noDMS) }

Liquid water content (LWC)

July average of LWC ($g\ m^{-3}$) at different GEM-MACH level 0.87 (~870 mb): Left panel GEM-MACH results without DMS, and right panel, absolute difference between runs with DMS and without DMS.
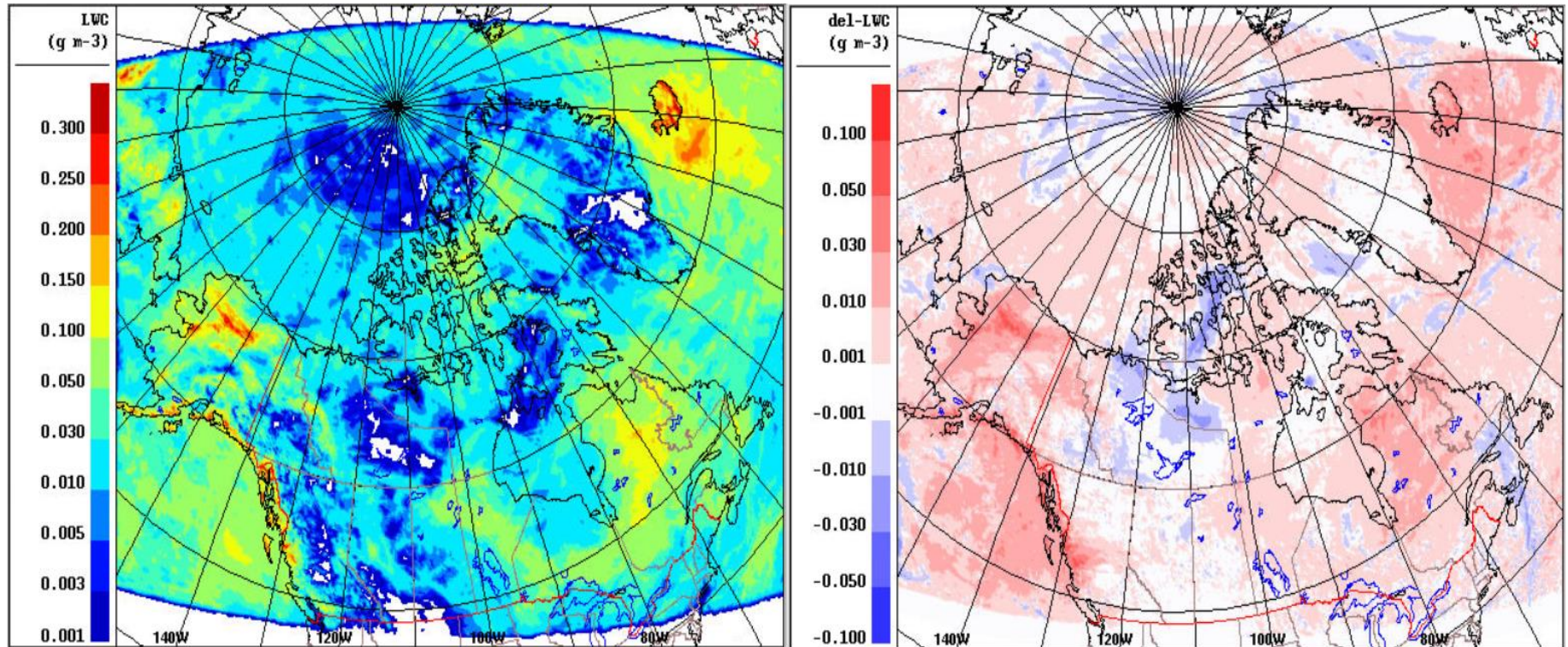

LWC
(with no DMS)

ΔLWC
{ (with DMS) − (noDMS) }

Summary

- Implementation of DMS(g) (as the main biogenic source of sulfate aerosol) in the GEM-MACH model.

- Agreement with the observations improved by replacing climatological values with *in-situ* measured DMS(aq).

- The enhancement in sulfate particles is most significant in the size range of 50 to 100 nm at high latitudes (> 70° N), however, this enhancement is stronger in the 200–1000 nm size range at lower latitudes (< 70° N).

- Implementation of DMS(g) in the model leads to increase of the CDNC with smaller size (MMD), and it is accompanied by the enhancement of LWC in the domain.

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Acknowledgments

Notes for slides:

- **Slide 4:** DMS(aq) is produced by the breakdown of dimethylsulfoniopropionate (DMSP), a compound synthesized by phytoplankton. DMSP breakdown is favoured by microbial interactions and environmental stressors, and is carried out by phytoplankton and bacterial DMSP-lyase enzymes. DMS produced in the upper mixed layer of the ocean is mostly removed by bacteria and photochemistry, and only 10–15% enters the atmosphere in the form of DMS(g) via processes such as turbulence, diffusion and advection. During summer, DMS(aq) production and emission to the atmosphere increase due to a larger ice-free sea surface and higher bioactivities.

- Atmospheric dimethyl sulfide, DMS, is the main biogenic source of sulfate particles in the Arctic atmosphere. Sulfate particles have a net cooling effect, which can partially offset Arctic warming from absorbing aerosols, such as black carbon. As efficient cloud condensation nuclei (CCN), sulfate particles are also able to influence the cloud's microphysical properties.

- **Slide 8:** Two seawater DMS datasets are used in this study: (1) a gridded global monthly climatology of the surface seawater DMS concentration at a 1deg x 1 deg resolution (Lana et al., 2011), (referred to as CLIM11), and (2) a new satellite-based sea surface DMS concentration dataset at a 28 km x 28 km spatial resolution and an 8 days temporal resolution (Galí et al., 2018) (referred to as SAT).

- **Slide 9:** The most important reaction of DMS(g) in the atmosphere is oxidation by hydroxyl and/or nitrate radicals. This addition pathway proceeds by adding the OH radical to DMS(g); its final products are dimethylsulfoxide (DMSO), dimethylsulfone (DMSO2) and methanesulfinic acid (MSIA), which are all highly soluble in water and readily condensable on existing aerosols. Oxidation of DMSO, DMSO2 and MSIA in the gas phase is not significant in the presence of clouds or high concentrations of aerosols. The abstraction pathway starts with the DMS(g) reaction with OH and NO3 radicals, and the main products are methanesulfonic acid (MSA) and SO2. The addition and abstraction pathways of DMS(g) oxidation with OH are temperature and light dependent. Reaction with halogens is a potential additional sink for DMS(g) in the remote marine atmosphere (not in GEM-MACH).

- Under *certain conditions* and in the presence of both amines and water vapour, MSA *may* form new particles (not in GEM-MACH).

Notes for slides:

- **Slide 10:** The modelled monthly averaged DMS(g) mixing ratios for July 2014, at the lowest model level (20 m), using the SAT and CLIM11 DMS(aq) datasets. Broadly speaking, the two model simulations show a similar geographical distributions of atmospheric DMS over the model domain.

- However, the two simulations do differ on a local scale, e.g. higher DMS(g) mean mixing ratio values are evident in the figure using the SAT DMS(aq) dataset for some regions such as Hudson Bay (up to 600 pptv using SAT vs. up to 75 pptv using CLIM11) and the Canadian Arctic Archipelago (up to 200 pptv using SAT vs. up to 100 pptv using CLIM11). The differences in modelled July-averaged DMS(g) between the two simulations largely reflect the differences between the two DMS(aq) datasets.

- **Slide 11:** The figure compares the time series of modelled DMS(g) mixing ratio values from the two simulations (using SAT and CLIM11) following the NETCARE 2014 Amundsen cruise track with the GC-SCD (Gas Chromatograph – Sulfur Chemiluminescence Detector) and CIMS (high resolution time-of-flight chemical ionization mass spectrometer, the Aerodyne HRToF-CIMS) measurements.

- Relatively high DMS(g) mixing ratios were observed during the biologically productive period of July; two main high DMS(g) concentration episodes were observed during the 18–21 and 26–27 July periods while Amundsen was traversing Lancaster Sound. The modelled DMS(g) from both simulations generally tracked the observations well but both missed the two abovementioned high DMS(g) episodes, although the simulation with SAT captured the higher DMS episodes in early August well. Overall the model simulations under-predicted the measurements.

- **Slide 12:** To find the reason of the discrepancy between model and observation, three sensitivity tests are discussed here: (1) a "no-Ice" model run where sea-ice cover is neglected; (2) a model run with enhanced DMS(aq) in Hudson Strait and Hudson Bay; and (3) a model run with further enhancements in DMS(aq) that makes use of the in situ DMS(aq) measurement from the NETCARE campaign. Results from our sensitivity tests suggest that the largest uncertainty arises from DMS(aq) concentrations in surface seawater.

- **Slides 15 to 17:** In general, implementation of DMS(g) in the model leads to increase of the CDNC with smaller size (MMD), and it is accompanied by the enhancement of LWC in the domain.