BACCHUS

Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding

Collaborative Project

SEVENTH FRAMEWORK PROGRAMME
ENV.2013.6.1-2

Atmospheric processes, eco-systems and climate change

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Executive summary:

In this deliverable the work is described that the different Earth System Models (ESM) undertook in order to include the potential role of organics as ice nucleating particles (INP) partially based on work that has been described in deliverable 2.4. The respected ESMs are the ECHAM-HAM-ESM that uses the Rinaldi et al. (2013) parameterization, the NorESM the O'Dowd et al. (2008) and Wilson et al. (2015) parameterizations and the MetOffice UM uses the DeMott (2010) parameterization. So far the results obtained with these models are rather diverse, pointing to a small impact of marine organic aerosols as INP in the ECHAM-HAM-ESM to a potentially large role in NorESM. The MetOffice results suggests that modifying the DeMott (2010) parameterization can reduce the bias in the reflected shortwave radiation over the Southern Ocean.

Summary of results

1. Marine organic aerosols

Freezing due to marine water insoluble organic matter (WIOM), referred to from here on as marine organic aerosols (MOA), has been implemented into the aerosol-climate model ECHAM6.3-HAM2.3-MOZ1.0-rc3 (for brevity from now on referred to as ECHAM6-HAM2).

1.1. Tracer

In order to more accurately keep track of the aerosols and due to the opposite dependence on wind speed when compared to sea salt emissions, MOA is traced separately in the model. Despite being insoluble in nature, it exists only in the soluble modes as we assume that MOA is already internally mixed with sea salt during emission. The species density is set to be 1 g/cm^3, following Vignati et al. (2010), and is non-hygroscopic. The latter is based on observations from e.g. Vaishya et al. (2013), who noted the low hygroscopicity of organic-rich sea spray. Due to the lack of measurements, the molecular weight and radiative properties of MOA are currently set to be the same as that of organic carbon. In terms of size, MOA is emitted into the accumulation mode ($0.05\mu m \leq \acute{r} \leq 0.5\mu m$) and is allowed to grow into the coarse mode ($0.5\mu m \leq \acute{r}$). The particles subsequently participate in all relevant aerosol growth and scavenging processes as dictated by their physical properties and as part of an internal mixture of species. Because there is no sea salt in the Aitken mode in ECHAM6-HAM2, there is also no MOA in the Aitken mode.

1.2. Emission

MOA is assumed to be co-emitted with sea salt (SS), such that the total sea spray emitted is a sum of the two, and the organic mass fraction (OMF) contained in the sea spray is

$$OMF = \frac{MOA}{MOA+SS}.$$

Various OMF parameterizations are available in the literature (e.g. Burrows et al. 2014, Vignati et al. 2010), which results in a wide range of OMF in some areas when applied to the global scale. Validations are, however, difficult due to limited observational data. For the current implementation, the parameterization from Rinaldi et al. (2013; also part of deliverable D2.1 in BACCHUS) is applied as it fits best to observations at Mace Head and Amsterdam Island when using our model wind speed, SS flux, and observed chlorophyll concentration data from GlobColour. The MOA mass flux is thus represented as

$$MOA_{massflux} = \frac{SS_{massflux} \times OMF}{1 - OMF},$$

where

$$OMF = 56.9 \times chl - 4.64 \times U_{10} + 40.9$$

with the chlorophyll concentration (chl) in mg/m^3 and the 10-meter wind speed (U_{10}) in m/s, valid for U_{10} > 4 m/s and OMF $\leq$ 78%. As the sea salt emission scheme in our model considers specifically the emission of NaCl particles, MOA is emitted as an additional mass flux such that the total mass flux from the ocean surface (sea spray) increases while the sea salt mass flux is kept untouched.

Due to the incomplete coverage of observational data and for applicability in future simulations, offline maps containing the averaged monthly mean values of chlorophyll concentration from 16 CMIP5 earth system models which include this variable in their outputs are used. Since we assume that MOA is co-emitted with sea salt, it is emitted directly into the internally mixed/soluble accumulation mode as a mass flux with no additional particle number flux.

It may be noted that the MOA flux depends strongly on the model sea salt flux. Our current model default uses a combination of the Monahan et al. (1986) and Smith and Harrison (1998) parameterizations, following Guelle et al. (2001), but other options (e.g. Gong 2003) are also available.

1.3. Freezing

The Wilson et al. (2015) parameterization that resulted from the BACCHUS project is used to describe immersion freezing of MOA. Dependent only on temperature and valid for temperature *T* in degrees centigrade not greater than -7.4 °C, the number of ice nucleating particles (INP) per mass of organic material is

$$INP = exp(11.2186 - 0.4459 \times T).$$

The active surface sites approach described in Connolly et al. (2009) and in Murray et al. (2012) is then extended to represent the ice active sites per mass ($n_{m,MOA}$) such that the fraction of droplets frozen (*FF*) is

$$FF = 1 - exp\left[-m_{MOA} \times n_{m,MOA}\right],$$

with m_{MOA} being the modal mean MOA mass per particle and $n_{m,MOA}$ represented as the INP number per mass from Wilson et al. (2015). The freezing per time step is thus

$$\Delta ICNC = N_{MOA,imm} \times FF,$$

where $\Delta ICNC$ is the change in the ice crystal number concentration per time step due to freezing, and $N_{MOA,imm}$ is a diagnostic value representing the number of immersed marine organic aerosols. This is defined as

$$N_{MOA,imm} = N_{TOT,act} \times \left(\frac{V_{MOA}}{V_{TOT}}\right)^{\frac{2}{3}},$$

where $N_{TOT,act}$ is the number concentration of activated aerosols in the internally mixed mode, and V_{MOA} and V_{TOT} are the volume of MOA and the volume of all aerosols in the mode, respectively, as diagnosed from mass and density. This is a surface area fraction representation where we approximate the ratio of MOA number to modal total number concentration with the ratio of MOA to total aerosol surface area.

In theory, in the model, all aerosols in the same internally mixed mode are distributed across the same number of aerosols, such that the number concentration of all species is the same for the same internally mixed mode. This is, however, unrealistic when we want to consider freezing of particles, as a small mass of MOA could then be spread across a large number of particles in the same mode in the model grid box, and as all of them contain a trace amount of MOA, all of them could potentially freeze and form ice crystals. Thus the above diagnostic is applied.

Another item of note is that aerosols are not directly removed due to droplet activation by acting as cloud condensation nuclei nor due to freezing as ice nucleating particle in ECHAM-HAMMOZ. Instead, in-cloud wet deposition is a separately parameterized process. This means that the same aerosols can potentially be repeatedly frozen across time steps, and to resolve this, the existing ICNC from the previous time step is subtracted from the current ΔICNC, with a minimum cutoff at zero—so no ICNC loss—to get the actual change in ice crystal number concentration.

A problem with the above described method for preventing re-freezing of the same aerosols is that not all ice crystals in the grid box are formed due to freezing of the ice nucleating particles. Other sources such as sedimentation from above could also lead to an influx of ice crystals. To prevent excessive suppression of heterogeneous freezing, a secondary method is thus applied whenever the current ΔICNC is less or equal to the previous ice crystal number concentration. This method more closely follows the ice active surface sites approach, which actually describes the total number of particles frozen when the temperature drops from 0°C to the stated temperature, and the change in ICNC is determined by subtracting the number of drops frozen at the previous temperature from that frozen under the current conditions. One downside to this approach is that it may fail to capture effects of an increase in MOA concentration in the grid box without a decrease in temperature. Also, only the temperatures at the previous time step and the current time step are considered, which would make this approach very sensitive to temperature changes on short time scales. Thus a combination of both methods (subtraction of the previous ICNC and that of the number frozen at the previous temperature is used to give the best estimate. In equation form, this can be summarized as follows:

At time t, the increase in ICNC due to freezing ($\Delta ICNC_{t,f}$) is: $\Delta ICNC_{t,f} = max\left[\Delta ICNC_{t,i} - ICNC_{t-1}, 0\right]$,

where

$$\Delta ICNC_{t,i} = N_{MOA,imm,t} \times \{1 - exp\left[-m_{MOA,t} \times n_{m,MOA}(T_t)\right]\}$$

and $ICNC_{t-1}$ is the ice crystal number concentration before the new freezing occurs.

Then if $\Delta ICNC_{t,f} = 0$,

$$\Delta ICNC_{t,f} = max\left[\Delta ICNC_{t,i} - \Delta ICNC_{t'}, 0\right],$$

where

$$\Delta ICNC_{t'} = N_{MOA,imm,t} \times \{1 - exp\left[-m_{MOA,t} \times n_{m,MOA}(T_{t-1})\right]\}$$

and $n_{m,MOA}(T_{t-1})$ is the number density of ice active sites calculated using the temperature at the previous time step.

Lastly, despite allowing MOA to grow from the accumulation mode to the coarse mode in the model, ice nucleation is only implemented for MOA particles in the accumulation mode. This is due to the lack of observational and experimental data examining the ice nucleating ability of larger coarse mode MOA. We do not expect such an exclusion to have a large impact on the results as the majority of MOA remains in the accumulation mode, but a sensitivity study will be performed.

1.4. Results

With the above described methodology, we obtain a global annual mean total MOA burden of 0.15 Tg and a global emission flux of 31 Tg/yr, which is much higher than the 5-8 Tg/yr range estimated in the literature (e.g. Gantt et al. 2011, Vignati et al. 2010). Further testing with different sea salt emission schemes and adjustments of the chlorophyll concentration maps will be needed to pinpoint the reason for such a bias. On the other hand, despite such a high amount of emitted MOA, no statistically significant differences in the top of atmosphere radiative balance and the aerosol optical depth were observed between simulations with and without added MOA.

Production of ice crystals by MOA in the model averages to a few crystals per cubic meter per day or less, as shown in Figure 1, resulting mainly from droplets freezing at a rate of a few hundred to a few thousand per cubic meter per day during less than 6% of the time steps (Figures 2 and 3). This is more significant than the mean freezing contribution of dust, the only other species capable of heterogeneous ice nucleation in our model, which, following the Niemand et al. (2012) parameterization, barely results in any freezing except for a narrow peak of a few droplets per m^3 per day in the northern hemisphere mid-latitudes during boreal autumn to spring. Nevertheless, no statistically significant differences can be found in the mean ice crystal number concentration of a multiyear model run with MOA compared to one without. This further supports previous observations that the current version of ECHAM-HAMMOZ is not sensitive to heterogeneous ice nucleation, but this might be specific to the ECHAM-HAMMOZ global climate model.

1.5. Outlook

For the next steps, sensitivity studies will be performed to examine the dependence of the results on the chlorophyll concentration, the chosen sea salt emission scheme, and the MOA freezing parameterization. For instance, MOA ice nucleation described by DeMott et al. (2016) will be tested in place of the Wilson et al. (2015) parameterization. Different scaling factors will be applied to investigate the sensitivity of the model to MOA, since there are still large uncertainties in both the global emission and the freezing ability of marine organic aerosols. More detailed analyses will also be performed to pinpoint the impact of MOA on a more regional scale and the relative importance of different ice formation mechanisms in the model. Once these further analyses have been completed, a paper on this topic will be submitted.

In addition a parameterization of INP based on different mineral dust samples based on the work by Boose et al. (2016) has been discussed in deliverable D2.4 and will be tested in the ECHAM-HAM global climate model.

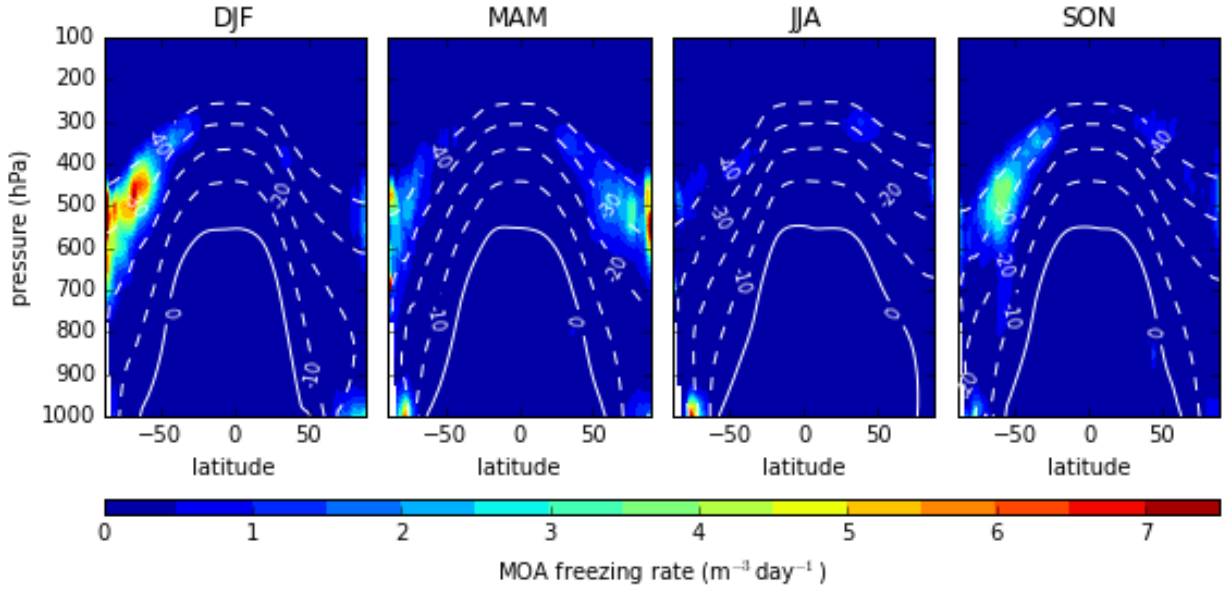

Figure 1: Seasonal and zonal mean freezing rate of marine organic aerosols (MOA) from a 7-year simulation using the newly implemented MOA freezing scheme. Only freezing that occurs more frequently than 1% of the time is shown. Contour lines denote the mean temperature in degrees centigrade.

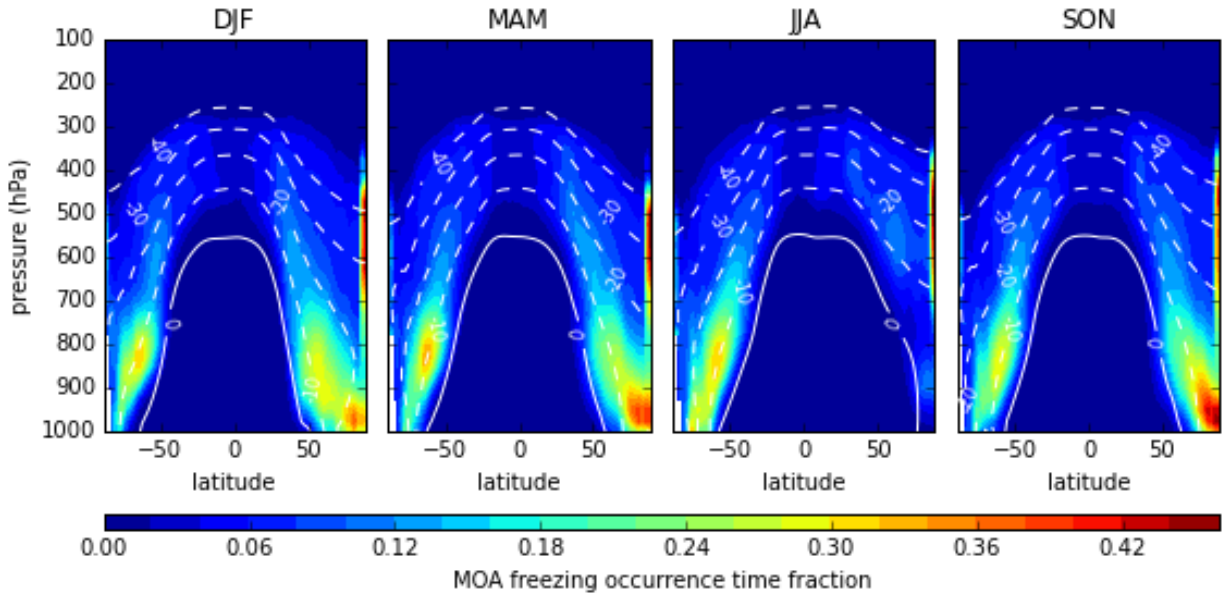

Figure 2: With the same setup as Figure 1 but showing the fraction of time when freezing of MOA occurs.

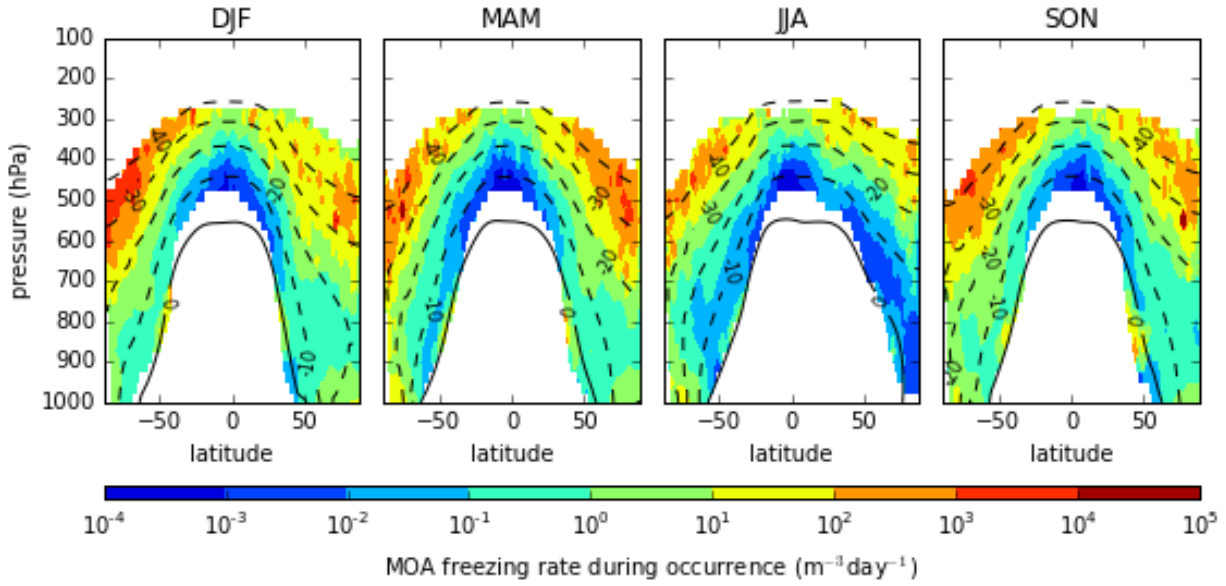

Figure 3: Same as Figure 1 but for the mean freezing rate when freezing actually occurs.

2. Marine organics as ice nucleating particles in the NorESM

The Norwegian Earth System Model (NorESM) has been modified to account for MOA as INP. Although the scientific basis for treating MOA emissions and freezing in the model is similar to the study with ECHAM6.3, a different aerosol scheme and a different approach for the ice nucleation parameterization can explain potentially varying results.

In NorESM, MOA emission parameterization is similar to ECHAM6.3, but with an organic mass fraction (OMF) given by O'Dowd et al. (2008). MOA is treated as a tracer, which is internally mixed with sea salt and is therefore not traced separately. The model separates between interstitial and cloud-borne aerosols, where the latter is used for immersion freezing. Data from Wilson et al. (2015) is used to derive a freezing parameterization based on classical nucleation theory (CNT). Here assumptions for the size of MOA were needed, and accumulation mode sea salt (r_N) of 300 nm from the NorESM is used for that. Together with the ice-nucleating particles per gram of total organic content (n_M) from Wilson et al. (2015), a 'measured' ice nucleation rate can be derived:

$$J_{imm,n_M} = \frac{\rho_P r_N n_M}{3\Delta t}$$

where ρ_P is the particle density and t the average time for freezing in the experiment presented in Wilson et al. (2015). The parameters for the simulated ice nucleation rate J_{imm} are derived by minimizing the root mean square error (RMSE) between both rates:

$$RMSE = \sqrt{\frac{1}{N}\sum_{1}^{N}\left(J_{imm} - J_{imm,n_M}\right)^2}$$

Comparing the ice-nucleating surface site density (n_S) calculated with this approach to measurements from DeMott et al. (2015) showed that the approach presented here gives values which are roughly two orders of magnitude higher (Figure 4). Further discussions about the different approaches are needed to fully understand this difference.

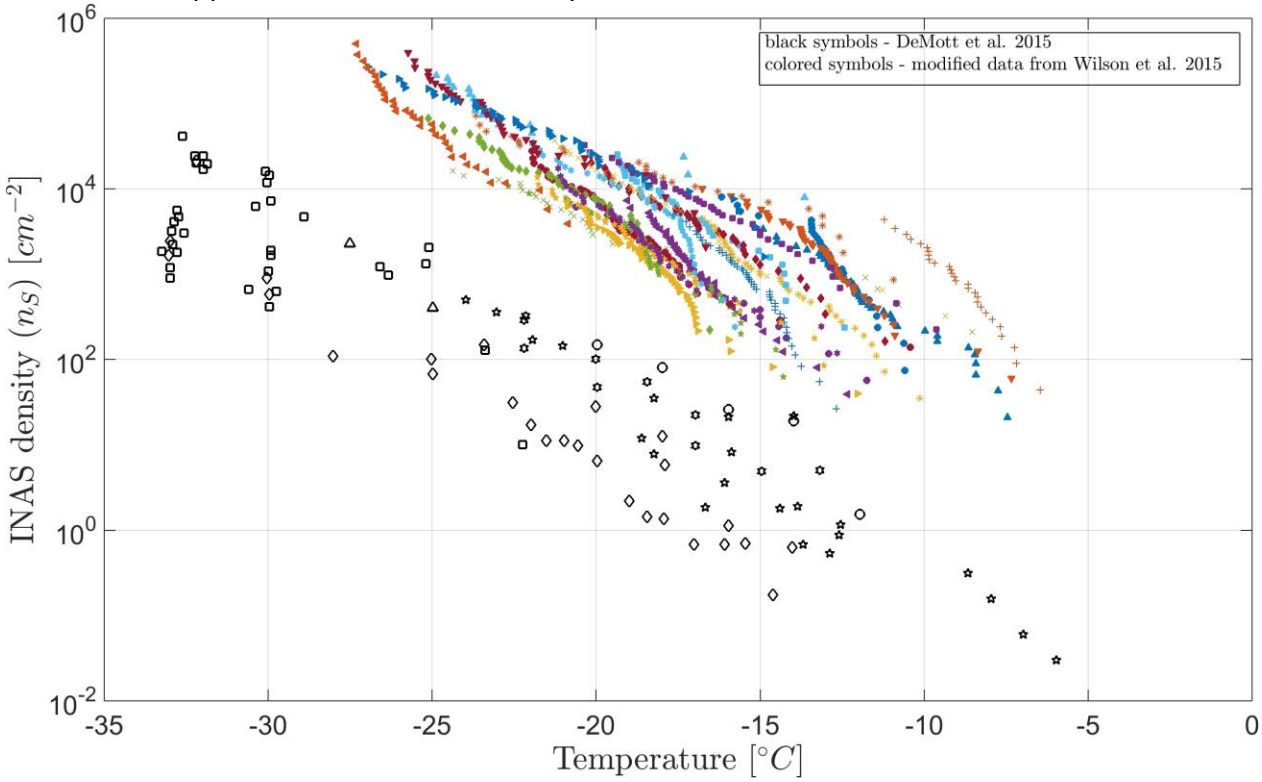

Figure 4: Ice-nucleating surface site (INAS) density calculated from Wilson et al. (2015) data for the cumulative INP per TOC by assuming $r_N = 0.3\ \mu m$ (colored symbols) in comparison with INAS density from DeMott et al. (2015) (black symbols).

First estimates of in-cloud INP concentrations match with results from Wilson et al. (2015), but indicate that the highest values in NorESM occur at slightly higher temperatures. This effect might result from differences in the freezing parameterizations: For the NorESM simulations, the logarithmic description of the ice-nucleating surface site density as a function of temperature has been extrapolated beyond the supporting data points to cooler temperatures until the homogeneous freezing threshold, whereas the CNT approach levels off and might even decrease slowly.

Further steps include a close comparison of INP concentrations to other model studies. The contribution of MOA to mixed-phase clouds will be evaluated by comparing to control simulation where both model runs are nudged to ERA-interim reanalysis data.

3. Defining INP concentrations and their impact in an ESM (Met Office Unified Model)

3.1. The importance of Southern Ocean clouds and their poor representation in models

The Southern Ocean is one of the regions of the world where climate models have some of the largest biases. In many climate models, the sea-surface temperature is biased by 2-3 degrees Celsius (Wang et al., 2014). This bias could be related to other well-known bias: clouds in the Southern ocean reflect too little solar radiation in models when compared with satellite data (Trenberth and Fasullo, 2010). Studies of cyclone composites in the Southern Ocean have identified mid and low-level clouds as the main contributors to this radiative bias (Bodas-Salcedo et al., 2012; Bodas-Salcedo et al., 2014).

These Southern Ocean clouds are typically within the mixed-phase range of temperatures (0 to -37^{o}C) where supercooled liquid cloud droplets can freeze to ice crystals by the action of an INP. Once a mixed-phase cloud starts to freeze, its albedo decreases substantially, as ice particles grow at the expense of liquid cloud droplets through the Wegener–Bergeron–Findeisen (WBF) process, creating less and larger hydrometeors. This implies less scattering of light, and hence less reflected solar radiation.

3.2. Hypothesis

The concentration of INPs differs in orders of magnitude from terrestrial environments to remote marine places (DeMott et al., 2016). Typically used parameterizations overestimate the concentration of ice nucleating particles in remote places (Vergara Temprado et al., 2016). This overestimation of INP could be making these clouds glaciate too quickly, being one of the causes of the shortwave radiative bias.

3.3. Methodology

In order to test the sensitivity of the reflected solar radiation from Southern Ocean cyclones to the INP concentration, we run some simulations using a high resolution model in the Southern

Ocean with resolved cloud microphysics, using two moments for the representation of ice particles (mass and number).
The domain of the simulation is centred at -52º latitude 0.02º longitude, and it is composed of 500x500 gridpoints with a resolution of 0.02º (~2km). We prepared the simulation for the 9th of December as a cyclone composite passed by.

3.4. Results

In our first run (ALL_ICE_PROC), we use DeMott's parameterization (DeMott, 2010) for heterogeneous ice nucleation. For the second run, we decrease the INP concentration by three orders of magnitude (3_ORD_LESS) in order to quantify the effect that correcting the INP bias could have in these type of clouds. Figure 5 shows the simulated reflected solar radiation (top of the atmosphere outgoing shortwave radiation) at 11:00h for both simulations. In the top panels, it can be seen that the reflected shortwave radiation effectively increases when the INP concentration is decreased three orders of magnitude.

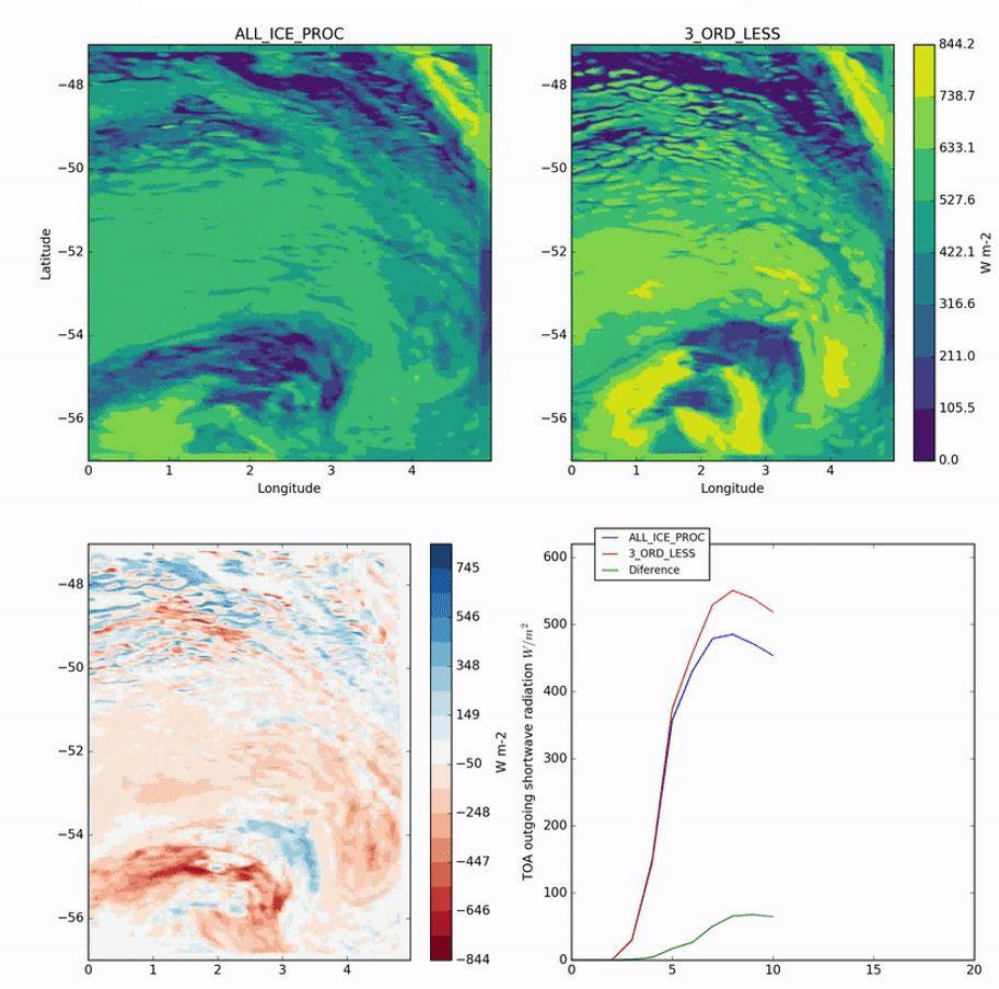

Figure 5. Top of the atmosphere outgoing shortwave radiation at 11:00 simulated using DeMott's parameterization (top left - ALL_ICE_PROC) and using the same parameterization decreased by 3 orders of magnitude (top right – 3_ORD_LESS). Bottom left shows the difference between ALL_ICE_PROC and 3_ORD_LESS. Bottom right shows the mean reflected shortwave radiation in the domain for every hour since the begging of the simulation (00:00 H to 11:00H).

3.5. Conclusions

We conclude that improving the representation of INP in climate models can help decreasing the shortwave bias in the Southern Ocean.

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Changes with respect to the DoW

With the sudden and unexpected death of Jon Egill Kristjansson, the UiO contribution did not advance as planned. Terje Berntsen has taken over as the PI at UiO and Matthias Hummel is now working on task 4.2.

Dissemination and uptake

The paper by Wilson et al. (2015) appeared in Nature and the Vergara Temprado et al. (2016) paper is in the discussion forum of ACP (see references). A paper on the implementation of marine organic aerosols as ice nucleating particles in the ECHAM-HAM global climate model by Katty Huang is in preparation. In addition the results described in this deliverable will be used in the ESMs for the simulations to be discussed in task 4.5 and deliverable 4.5.