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## **Deliverable D2.2 "Implementation and first simulations with improved aerosol-cloud interaction in COSMOS"**

### **1 INTRODUCTION**

Atmospheric aerosols play an important role in the climate system by altering the Earth's radiation budget through the so-called direct effect and by influencing the cloud properties by the so-called indirect effect [Twomey, 1999; Charlson et al., 1992]. However, the uncertainty of the radiative forcing of the direct and indirect effect is still very large [Forster et al., 2007; Denman et al., 2007], which makes it desirable to increase the understanding of the different interactions between aerosol particles and clouds. One aspect of these interactions is the removal of aerosol particles from the atmosphere by different scavenging processes in clouds. These mechanisms are important for the prediction of aerosol number and mass distribution in global climate models.

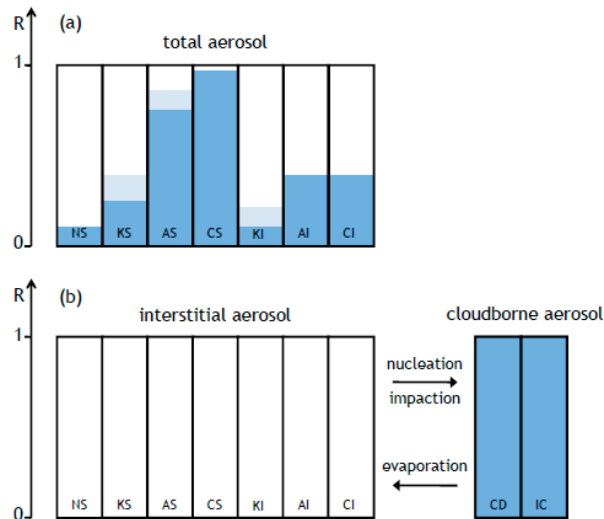
### **2. Model description: ECHAM5.5-HAM2**

The version of ECHAM5 used in this project is the fifth generation atmospheric general circulation model (GCM) developed at the Max-Planck Institute for Meteorology [Roeckner et al., 2003] with updates described in Lohmann and Hoose (2009) and Croft et al. (2010). The two-moment Hamburg Aerosol Model (HAM), which is coupled to ECHAM5.5, is used to describe the aerosol properties by predicting aerosol mixing state in addition to the aerosol mass and number concentrations [Stier et al., 2005]. The aerosol size-distribution is represented by a superposition of seven log-normal modes including the major global aerosol compounds sulphate, black carbon, organic carbon, sea salt, and mineral dust. The seven different modes are divided into four internally mixed/soluble modes nucleation (NS), Aitken (KS), accumulation (AS), and coarse (CS), - and three insoluble modes Aitken (KI), accumulation (AI), and coarse (CI). The count median of each mode is calculated from the information of aerosol mass and number concentrations. The transfer of mass and number between the insoluble and soluble modes and towards larger aerosol sizes is governed by condensation of sulfuric acid and coagulation between the aerosol particles.

### **3. Standard aerosol scavenging**

#### **3.1 Model description**

To parameterize the scavenging processes the standard version of ECHAM5.5-HAM2 uses prescribed aerosol scavenging fractions  $R$ , which depend on the aerosol size, solubility and cloud type (Fig 1a). These scavenging ratios are then used to determine the number of aerosol particles, which are removed from the atmosphere by wet deposition. The benefit of the usage of prescribed aerosol scavenging fractions is the low computational costs, which are markedly increased when more sophisticated approaches like prognostic aerosol cloud processing schemes [Ghan and Easter, 2006, Hoose et al., 2008a,b] are used.



**Figure 1:** Partitioning between interstitial and cloud-borne aerosol and scavenging ratios  $R$  for the seven plus two aerosol modes in the standard model (a) and with the new aerosol processing scheme (b). For the standard model (a), the light and dark blue indicates the values of  $R$  in liquid and mixed-phase clouds (from Hoose et al., 2008b)

One drawback of this method is that the scavenging ratios were taken from single measurements, but are prescribed globally in the model, which implies a source of uncertainty (see Section 3.2).

### 3.2 Sensitivity studies

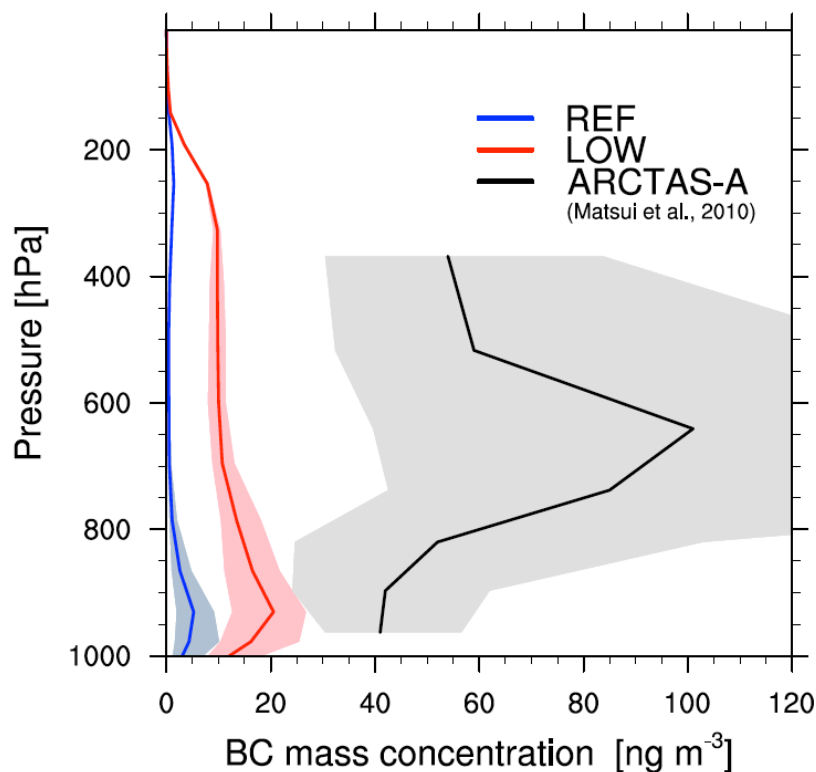
As showed by Bourgeois and Bey (2011) the transport of pollution from lower latitudes to the Arctic is strongly underestimated in ECHAM5-HAMMOZ due to an overestimated scavenging of aerosol particles. To estimate the effect of different prescribed scavenging ratios on the aerosol properties in the Arctic, simulations with the standard values of the scavenging ratios  $R$  and decreased values of  $R$  were compared with measurements of the ARCTAS-A campaign 2008 [Matsui et al, 2010]. The results presented in this study are from a one-year simulation including a spin-up time of three months and are nudged towards the meteorological conditions of the year 2008. The exact values of the scavenging ratios  $R$  can be seen in Table 1, following Bourgeois and Bey (2011).

Figure 2 shows the vertical profile of the monthly mean black carbon mass for the simulations of the standard and reduced scavenging ratios as well as the measurements in the area of the ARCTAS-A campaign. It can be clearly seen that results with ECHAM5.5-HAM2 using the standard values of the scavenging ratios show a strong underestimation of the black carbon mass in the polar region. By reducing the scavenging ratios the representation of the black carbon mass is improved, but still away from a good agreement (Fig. 2).

Mode	Liquid clouds		Mixed clouds		Ice clouds	
	REF	LOW	REF	LOW	REF	LOW
NS	0.10	<b>0.06</b>	0.10	<b>0.06</b>	0.10	<b>0.06</b>
KS	0.25	0.25	0.40	<b>0.06</b>	0.10	<b>0.06</b>
AS	0.85	0.85	0.75	<b>0.06</b>	0.10	<b>0.06</b>
CS	0.99	0.99	0.75	0.75	0.10	<b>0.06</b>
KI	0.20	0.20	0.10	<b>0.06</b>	0.10	<b>0.06</b>
AI	0.40	0.40	0.40	<b>0.06</b>	0.10	<b>0.06</b>
CI	0.40	0.40	0.40	0.40	0.10	<b>0.06</b>

**Table 1:** ECHAM5-HAM in-cloud scavenging parameters used in this study for the seven different aerosol modes, including four soluble modes (nucleation mode (NS), Aitken mode (KS), accumulation mode (AS), and coarse mode (CS)) and the three insoluble modes (Aitken mode (KI), accumulation mode (AI) and coarse mode (CI)). REF denotes the scavenging values in the standard version of ECHAM5-HAM, NEW denotes the values for the sensitivity study (bold values mark changes from standard version). Values for NEW are taken from Bourgeois and Bey, 2011.

The reduction of the scavenging ratios also showed a non-linear effect on the cloud properties. By reducing the scavenging of aerosol particles more cloud droplets were expected to nucleate due to the higher concentration of available cloud condensation nuclei. In contrast, the simulation with reduced scavenging ratios showed much fewer cloud droplets because of a decreased formation of new aerosol particles from the gaseous phase. This is compensated by an increase of condensation of  $\text{SO}_4$  on existing particles, leading to an overall reduction of cloud condensation nuclei.

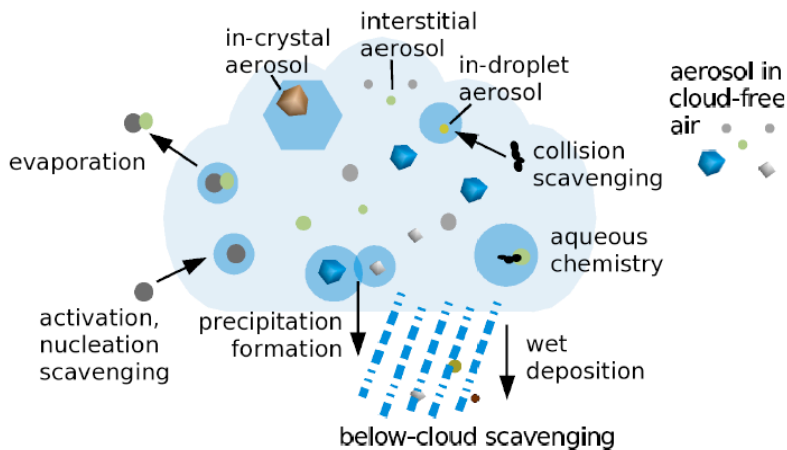


**Figure 2:** Vertical profile of the monthly mean black carbon (BC) mass concentration in April 2008 in every grid box between 60°N to 76°N and 196°E to 227°E for (blue) the standard version of ECHAM5-HAM2, (red) ECHAM5-HAM2 with the reduced scavenging ratios and (black) for the ARCTAS-A (spring 2008) campaign (Matsui et al., 2010) in the Arctic. The shaded areas are defined by the 25<sup>th</sup> and 75<sup>th</sup> percentiles for the respective cases.

## 4. Improved aerosol processing

### 4.1 Model description

The improved aerosol-cloud interaction scheme is based on Hoose et al (2008a,b) and Lohmann and Hoose (2009), as included in an older version of ECHAM5-HAM. The new scheme takes into account that aerosols are changed by clouds through different processes (Fig. 3). This scheme includes, among other things, the nucleation and impaction scavenging of aerosol particles and the addition of dissolved material on existing insoluble particles inside cloud droplets during the evaporation of the latter ones. These processes increase the size, reduce the number and change the chemical composition of aerosol particles and hence have an influence on subsequent cloud and ice nucleation events. For instance after such a cloud cycle the newly formed aerosol particles are larger and



**Figure 3:** Illustration of the processes involved in the aerosol processing in clouds, and of interstitial, in-droplet and in-crystal aerosols (from Hoose et al., 2008a).

more hygroscopic and therefore act more preferably as cloud condensation nuclei than before. Also, this cloud cycling permits a more realistic description of heterogeneous freezing in mixed-phase clouds, i.e. in clouds with temperatures between 0 and  $-35^{\circ}\text{C}$ , as it limits the number of available ice nuclei.

The aerosol processing scheme by Hoose et al. (2008a,b) includes two new aerosol modes in addition to the existing seven modes in the standard aerosol scheme in ECHAM-HAM. This allows the explicit treatment of in-droplet (CD) and in-crystal (IC) aerosol particles (Fig. 1b).

### 4.2 Current status of implementation

The improved aerosol processing is now technically integrated in ECHAM5.5-HAM2, but the results are not trustworthy yet. The reason, why the implementation of the aerosol processing took much longer than expected was the complete reorganization of the code compared to the version that was implemented in an earlier ECHAM5 version. This reorganization was heavy programming work, because the old version of the aerosol processing was deeply intertwined with the pure ECHAM5 code. For the future development of ECHAM-HAM it is demanded to

eliminate all HAM specific calculations from the pure ECHAM code. Therefore, the aerosol processing was organized in a more modular structure. For example, in the previous version of the aerosol processing, each process was called from a subroutine within the pure ECHAM code, which also creates much more communication and transfers between the individual subroutines. In the revised version, this is now done by a single call from ECHAM to an aerosol-processing interface. This interface is at the top of the aerosol-processing module and controls each process. Beside the reorganization also the readability of the code was much more improved, which facilitate possible future improvements of the aerosol processing. However, the results of the improved aerosol processing still show an unrealistic behaviour in the mass and number mixing ratios of the different aerosol modes pointing to a problem in the redistribution of the aerosol mass and number after the aerosol-processing module. Also the condensation of SO<sub>4</sub> on existing aerosol particles shows irregularities. These problems are under investigation right now.

## 5 OUTLOOK

The results of the sensitivity study with the different prescribed scavenging ratios show that the usage of prescribed aerosol particles is not desirable for a realistic description of the transport of aerosol particles and their interaction with clouds. Therefore we want to use the aerosol-processing scheme by Hoose et al. (2008a,b) to gain insight into the interactions between aerosol particles and clouds by investigating the transport of aerosol particles from lower latitudes to the Arctic. In a first step we hope to improve the performance of ECHAM for the reproduction of the black carbon mass in the region of the ARCTAS-A campaign. In a next step we want to study the influence of the aerosol processing on aerosol and cloud properties on a global scale.

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