Quantifying DMS-cloud-climate interactions using the ECHAM5-HAMMOZ model

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Ever since Charlson *et al*. [1] proposed a feedback loop between ocean ecosystems and the earth's climate (so called CLAW hypothesis), there has been considerable interest in understanding the role of dimethyl sulfide (DMS) in regulating the atmosphere over oceans. However, so for the exact contribution of each process in their proposed feedback loop is still uncertain. Here, we attempt to diagnose the localized aerosol-cloud climate feedback due to the DMS alone using the methods available. For this, first, we try to assess changes in cloud microphysical properties arising from prescribed perturbations to oceanic dimethyl sulphide (DMS) emissions in a present day climate scenario using a state of the art general circulation model, ECHAM5-HAMMOZ. ECHAM5-HAMMOZ consists of three interlinked modules, the atmospheric model ECHAM5, the aerosol module HAM and the tropospheric chemistry module MOZ. This study focuses on the atmosphere over the southern oceans where anthropogenic influence is minimal. We investigate changes in a range of aerosol and cloud properties to establish and quantify the linkages between them. We focus on changes in cloud droplet number concentration (CDNC), cloud droplet effective radii, total cloud cover and radiative forcing due to changes in DMS. Our preliminary results suggest that ECHAM5-HAMMOZ produces a realistic simulation of the first and second indirect aerosols effects over the Southern Ocean. The regions with higher DMS emissions show an increase in CDNC, a decrease in cloud effective radius and an increase in cloud cover. The magnitude of these changes is quantified with the ECHAM5-HAMMOZ model and will be discussed in detail.

[1] Charlson *et al.* (1987) *Nature* **326**, 655-661.

Al-Si complexing in high-*P* aqueous fluids: Implications for Al transport in subduction-zones

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Low solubility of Al in pure H_2O suggests that Al is immobile in metasomatic environments such as subduction zones [1]. However, Al-rich minerals in veins in the same settings imply significant Al mobility. To address this paradox, we investigated (1) the solubility of corundum in fluids of varying initial SiO_2/H_2O ratios at 800°C and 10 kbar, (2) the effect of P on corundum solubility at fixed SiO_2/H_2O ratio, and (3) the effect of temperature on the solubility of corundum at 15 kbar. Starting materials of synthetic corundum, natural high-purity quartz, and ultrapure H_2O were equilibrated using hydrothermal piston-cylinder methods [2]. Initial SiO_2 concentrations were varied between zero and quartz saturation by adding small quartz crystals. Solubility was determined by the weight loss from the corundum crystals. The solubility of Al in pure H_2O was 0.0028 molal, consistent with previous work [1]. We find that at 800°C, 10 kbar, the concentration of Al increases with increasing dissolved SiO_2, to a maximum of 0.0152 molal at quartz saturation. The data are described by the equation $m_{\mathrm{Al}} = 0.0065(m_{\mathrm{SiO2}})^2 - 0.0004(m_{\mathrm{SiO2}}) + 0.0032$, where m is the molality of the subscripted solute. The increase in Al concentration with increasing Si requires Al-Si complexing, consistent with recent predictions of Al-Si oligomerization in high P-T fluids [3,4]. The concave upward trend in the isothermal isobaric data indicates that on average, Al/Si in the oligomers is <1 [4]. In addition, Al solubility rises with increasing pressure at constant T and $m_{\mathrm{SiO2}} = 0.675 \pm 0.017$ molal, according to the equation $m_{\mathrm{Al}} = -0.0037P^2 + 0.0175P - 0.0069$. The Al solubility is ~2X higher than that of corundum in pure H_2O at each P. At 900°C, 15 kbar, the Al solubility is 33X higher than predicted values in pure H_2O, and at 950°C, 15 kbar the solubility is 100X higher. These results suggest that Al-Si complexing is a simple mechanism for enhancing Al solubility during subduction-zone metasomatism, as well as in other metamorphic settings.

[1] Tropper & Manning (2007) *Chem. Geol.* **240**, 54-60.
[2] Manning & Boettcher (1994) *Am. Min.* **79**, 1153-1158.
[3] Manning (2004) *Earth Planet. Sci. Lett.* **223**, 1-16.
[4] Manning (2007) *Geofluids* **7** 258-269.