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The sensitivity of Southern Ocean aerosol concentrations to sea spray and DMS emissions in the HadGEM3-GA7.1 chemistry–climate model

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With low concentrations of tropospheric aerosol, the Southern Ocean offers a "natural laboratory" for studies of aerosol–cloud interactions. Aerosols over the Southern Ocean are produced from biogenic activity in the ocean, which generates sulfate aerosol via dimethylsulfide (DMS) oxidation, and from strong winds and waves that lead to bubble bursting and sea spray emission. Here, we evaluate the representation of Southern Ocean aerosols in the Hadley Centre Global Environmental Model version 3, Global Atmosphere 7.1 (HadGEM3-GA7.1) chemistry–climate model. Compared with aerosol optical depth (AOD) observations from two satellite instruments (the Moderate Resolution Imaging Spectroradiometer, MODIS-Aqua c6.1, and the Multi-angle Imaging Spectroradiometer, MISR), the model simulates too-high AOD during winter and too-low AOD during summer. By switching off DMS emission in the model, we show that sea spray aerosol is the dominant contributor to AOD during winter. In turn, the simulated sea spray aerosol flux depends on near-surface wind speed. By examining MODIS AOD as a function of wind speed from the ERA-Interim reanalysis and comparing it with the model, we show that the sea spray aerosol source function in HadGEM3-GA7.1 overestimates the wind speed dependency. We test a recently developed sea spray aerosol source function derived from measurements made on a Southern Ocean research voyage in 2018. In this source function, the wind speed dependency of the sea spray aerosol flux is less than in the formulation currently implemented in HadGEM3-GA7.1. The new source function leads to good agreement between simulated and observed wintertime AODs over the Southern Ocean; however, it reveals partially compensating errors in DMS-derived AOD. While previous work has tested assumptions regarding the seawater climatology or sea–air flux of DMS, we test the sensitivity of simulated AOD, cloud condensation nuclei and cloud droplet number concentration to three atmospheric sulfate chemistry schemes. The first scheme adds DMS oxidation by halogens and the other two test a recently developed sulfate chemistry scheme for the marine troposphere; one tests gas-phase chemistry only, while the second adds extra aqueous-phase sulfate reactions. We show how simulated sulfur dioxide and sulfuric acid profiles over the Southern Ocean change as a result and how the number concentration and particle size

of the soluble Aitken, accumulation and coarse aerosol modes are affected. The new DMS chemistry scheme leads to a 20% increase in the number concentration of cloud condensation nuclei and cloud droplets, which improves agreement with observations. Our results highlight the importance of atmospheric chemistry for simulating aerosols and clouds accurately over the Southern Ocean.