



The impact of time-averaged volcanic sulphur emissions on the global cloud condensation nuclei budget in the pre-industrial era

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Volcanoes are a strong source of sulphur dioxide (SO₂) with time-averaged emission inventories (e.g. Andres and Kasgnoc, 1998) indicating that volcanoes account for around 40% of the total annual SO₂ flux in the pre-industrial atmosphere. We use a global aerosol microphysics model (GLOMAP-mode) to quantify the contribution of time-averaged volcanic sulphur emissions (from both continuous passive degassing and explosive volcanoes) on the global cloud condensation nuclei (CCN) budget. GLOMAP-mode is capable of simulating microphysical processes, such as binary homogeneous nucleation, hygroscopic growth, coagulation, condensation, cloud processing (oxidation of dissolved SO₂ to SO₄ in cloud droplets), as well as dry and wet deposition. For this study we use a sulphur chemistry scheme which includes 7 species (DMS, DMSO, MSA, SO₂, H₂SO₄, COS, CS₂). The runs were conducted using four internally mixed aerosol components, sulphate (SO₄), sea salt, black carbon (BC) and organic carbon (OC). We simulated the impact of volcanic degassing in a pre-industrial setting (i.e. using 1750 BC and OC emissions in the absence of any anthropogenic emissions) using the volcanic emission inventory by Dentener et al. (2006). This volcanic inventory is based on datasets by Andres and Kasgnoc (1998) and Halmer et al. (2002) and accounts for an annual flux of ~13 Tg(S) of volcanic SO₂.

Our simulations suggest that volcanic degassing contributes on average ~50 CCN (>35nm in radius) per cubic centimetre to the annual zonal mean CCN concentrations in the tropical boundary layer. The simulations also reveal complex changes in annual zonal mean total particle concentrations (CN). CN concentrations are more than double in large parts of the tropical boundary layer when comparing the unperturbed run (i.e. without volcanic degassing) to the run featuring time-averaged volcanic degassing. However, the simulations also reveal that the additional SO₂ and its subsequent conversion to sulphate aerosol enhances microphysical processes such as the coagulation of existing and/or entrained particles in upper parts of the atmosphere and in parts of the Southern Hemisphere boundary layer. Our model simulations suggest that volcanoes were a very important source of CCN in the pre-industrial atmosphere. Equatorwards of about 50° volcanoes can sustain CCN concentrations in excess of 50 cm⁻³, and up to 500 cm⁻³ over large tropical regions, even when the other main sulphur source (from DMS) is eliminated.

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