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Efficient formation of stratospheric aerosol for geoengineering though emission of low-volatility vapours in an aircraft plume

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Previous research that has explored the impacts of geoengineered stratospheric sulfate aerosol on climate and atmospheric chemistry has assumed that sulfur will be injected into the stratosphere as SO_2. SO_2, having a lifetime of about a month in the stratosphere, will be quite dispersed when it reacts with OH to form low-volatility H_2SO_4. As found by Heckendorn et al. (2009), much of the H_2SO_4 mass produced in geoengineering scenarios will go into increasing the size of existing aerosols, thus reducing aerosol lifetime and mass scattering efficiency. The overall effect is that continuous injection of SO_2 may be surprisingly ineffective; the analysis showed, for example, that a 10 Mt-S/yr injection rate produces a radiative forcing of only $\sim$1.7 Wm^{-2}, whereas previous calculations with fixed particle size predicted that 10 Mt-S/yr would produce a radiative forcing greater than 7 Wm^{-2}. In this talk we explore an alternative method of stratospheric sulfate aerosol geoengineering: the direct emission of H_2SO_4 (or SO_3) vapors. These vapors may be emitted from an aircraft by either boiling liquid H_2SO_4 or burning sulfur in the presence of a vanadium catalyst. We model the aerosol microphysics with an aircraft plume model coupled to a global stratospheric aerosol model. We find that the emission of H_2SO_4 yields a much narrower aerosol size distribution than emission of SO_2, thus yielding a 60-70% larger aerosol burden and 100% larger shortwave RF cooling in 5 MT S/yr cases. Because aerosol formation in an aircraft plume is dominated by homogeneous nucleation and fast condensation, the method is relatively insensitive the pre-existing aerosol distributions. For a given radiative forcing, less S injection is necessary through H_2SO_4 emissions, however, the total mass necessary to be lifted to the stratosphere depends on the injection method (e.g. boiling of H_2SO_4 versus combustion of S). Many of the negative unintended consequences of H_2SO_4 injection (eg. ozone depletion, effects on hydrological) will generally be similar to SO_2; however, H_2SO_4 injection would likely lead to less stratospheric warming from longwave absorption and less sulfate deposition. This method of the injection of low-volatility vapors to form aerosols is not limited to H_2SO_4, and materials could be tested for desired properties (e.g. low toxicity, high SW scattering efficiency, low LW absorption efficiency).

Heckendorn, P., et al., Env. Res. Let., 4 (2009) 045108