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Natural aerosol–climate feedbacks suppressed by anthropogenic aerosol

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[1] The natural environment is an important source of atmospheric aerosol such as dust, sea spray, and wildfire smoke. Climate controls many of these natural aerosol sources, which, in turn, can alter climate through changing the properties of clouds and the Earth's radiative balance. However, the Earth's atmosphere is now heavily modified by anthropogenic pollution aerosol, but how this pollution may alter these natural aerosol–climate feedbacks has not been previously explored. Here we use a global aerosol microphysics model to analyze how anthropogenic aerosol alters one link within these feedbacks, namely, the sensitivity of cloud albedo to changes in natural aerosol. We demonstrate that anthropogenic aerosol in the Northern Hemisphere has halved the hemispheric mean cloud albedo radiative effect that occurs due to changes in natural aerosol emissions. Such a suppression has not occurred in the more pristine Southern Hemisphere. **Citation:** Spracklen, D. V., and A. Rap (2013), Natural aerosol–climate feedbacks suppressed by anthropogenic aerosol, *Geophys. Res. Lett.*, *40*, doi:10.1002/2013GL057966.

1. Introduction

[2] The biosphere strongly influences atmospheric composition and climate [*Arneth et al*., 2010]. A key mechanism is through the emissions of natural aerosol to the atmosphere impacting climate through scattering and absorbing radiation and altering the properties of clouds [*Rap et al*., 2013]. Since many natural aerosol sources are themselves driven by climate, there is the potential for important climate feedbacks [*Carslaw et al*., 2010]. Best known of these natural aerosol–climate feedbacks is the CLAW hypothesis [*Charlson et al*., 1987], whereby climate change alters the emissions of dimethyl sulfide (DMS) from oceanic phytoplankton, modifying the amount of sulfate aerosol in the atmosphere, leading to changes in cloud reflectivity and, hence, climate. While recent work has questioned the strength of the CLAW climate feedback [*Woodhouse et al*., 2010; *Quinn and Bates*, 2011], many other natural aerosol–climate feedbacks are possible. For example, a warmer climate may increase the prevalence of wildfires [*Flannigan et al*., 2009], resulting in increased wildfire aerosol [*Spracklen et al*., 2009] which may then impact climate. Warmer temperatures may also result in increased emissions of biogenic volatile organic compounds (BVOCs) from vegetation, resulting in increased secondary organic aerosol (SOA) [*Heald et al*., 2008] with potential impacts on climate. This feedback mechanism between forests, emissions of BVOCs, formation of SOA, and climate was proposed by *Kulmala et al*. [2004] and has been recently explored through analysis of long-term observations of BVOCs and the number concentrations of aerosol particles [*Paasonen et al*., 2013].

[3] However, these natural aerosol–climate feedbacks now operate in an Earth system that is heavily altered by man; since the industrial revolution, there have been large increases in anthropogenic aerosol emissions [*Dentener et al*., 2006] which have greatly altered aerosol concentrations across the globe [*Forster et al*., 2007]. Because the impacts of aerosol on climate are nonlinear, especially aerosol indirect effects (AIEs) which operate through clouds [*Forster et al*., 2007; *Andreae and Rosenfeld*, 2008], it is likely that natural aerosol–climate feedbacks have changed greatly since the preindustrial period.

2. Methods

[4] To explore this possibility, we used the Global Model of Aerosol Processes (GLOMAP)–mode aerosol microphysics model [*Mann et al*., 2010] which is an extension of the TOMCAT global 3-D chemical transport model [*Chipperfield*, 2006]. The model is forced by analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF). We used a horizontal resolution of $2.8° \times 2.8°$ and 31 vertical levels between the surface and 10 hPa. We used the model to simulate the impact of changes in climatically driven natural aerosol sources both in a preindustrial (PI) and a present-day (PD) atmosphere. To isolate the impact of changing anthropogenic aerosol emissions between the two periods, we used the same meteorology for all simulations which we apply for the year 2000.

[5] The model simulates aerosol component mass and number concentration (two-moment modal) in five lognormal modes: hygroscopic nucleation, Aitken, accumulation, coarse, and nonhygroscopic Aitken modes. *Mann et al*. [2012] demonstrated that this modal version of GLOMAP simulates very similar aerosol compared to the sectional version of the same model [*Spracklen et al*., 2005]. GLOMAP includes representations of nucleation, particle growth via coagulation, condensation and cloud processing, wet and dry deposition, and in/below cloud scavenging. We used an identical model setup to that described in detail in *Mann et al*. [2010]. GLOMAP has been previously evaluated against observations of aerosol number concentration [*Spracklen et al*., 2010; *Mann et al*., 2010], aerosol number size distribution [*Spracklen et al*., 2007, 2008; *Mann et al*., 2010], and composition-resolved aerosol mass [*Spracklen et al*., 2011b]. Of particular importance for this study is the ability of the model to simulate observed concentrations of cloud condensation nuclei (CCN) and cloud droplet number. *Mann et al*. [2012] compared the model against an extensive data set of CCN observations

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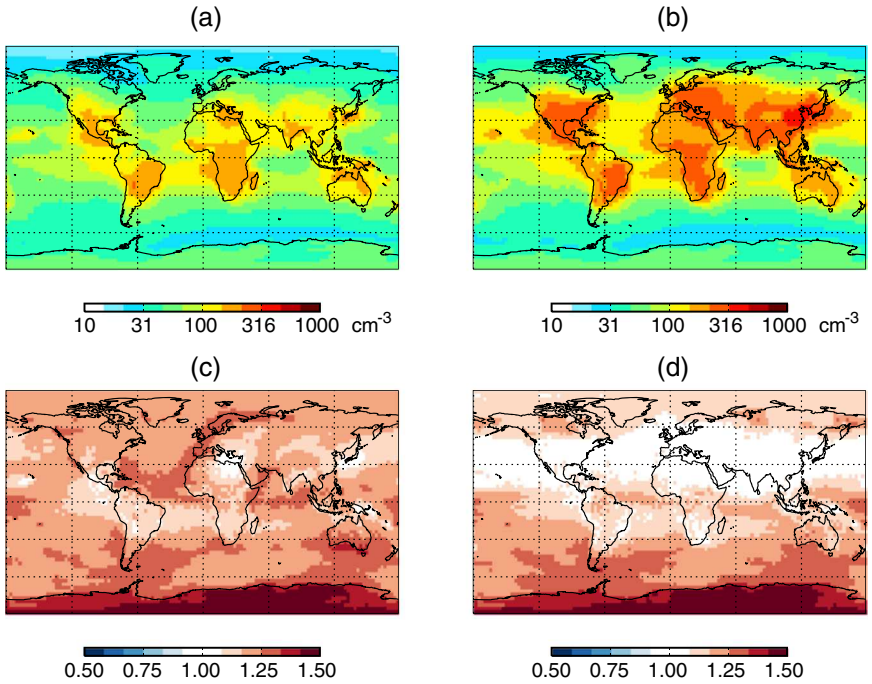

Figure 1. Cloud droplet number concentrations (CDNCs) in (a) preindustrial (PI) and (b) present day (PD). The fractional change in CDNC caused by a 100% increase in climatically driven natural aerosol sources in (c) PI and (d) PD.

synthesized by *Spracklen et al*. [2011a]. The normalized mean bias between model and observations was 12% [*Mann et al*., 2012], which is less than the stated observational error. The model has been also evaluated against observed cloud droplet number concentrations (CDNCs) [*Merikanto et al*., 2010].

[6] We used the model to simulate aerosol concentrations under both PI and PD conditions, accounting for the different anthropogenic aerosol emissions during the two periods [*Dentener et al*., 2006]. Anthropogenic aerosol emissions have increased greatly over this period, with anthropogenic sulfur emissions increasing from 0.1 Tg yr^{-1} in 1750 to 108.5 Tg yr^{-1} in 2000 [*Dentener et al*., 2006]. Our simulated global aerosol burden (excluding dust) increased from 11.0 mg m^{-2} in the PI to 13.6 mg m^{-2} in the PD. This increase (2.6 mg m^{-2}) matches previous studies that have been reported by the AeroCom multimodel exercise (3.7 ± 0.9 mg m^{-2}) [*Schulz et al*., 2006]. Natural aerosol emissions are as described in *Dentener et al*. [2006] and implemented in the model according to *Mann et al*. [2010]. Here we describe natural emissions that are of particular importance to this work. DMS emissions are calculated online in the model using ECMWF wind speeds in combination with the sea-air exchange parameterization from *Nightingale et al*. [2000] and monthly seawater DMS concentration fields from *Kettle and Andreae* [2000]. Wildfire emissions are from *van der Werf et al*. [2003]. Monoterpene emissions are taken from *Guenther et al*. [1995]. SOA is generated from the first-step oxidation of monoterpenes, with a fixed molar yield of 13%. Our treatment of SOA has been previously described in detail [*Mann et al*., 2010; *Scott et al*., 2013].

[7] To explore whether anthropogenic pollution has modified the first aerosol indirect effect that occurs due to changing natural aerosol emissions, we simulated identical increases to natural aerosol in both the PI and PD atmospheres. We selected three natural aerosol sources (oceanic DMS, wildfire, and BVOCs from vegetation) that are thought to be heavily influenced by climate [*Carslaw et al*., 2010] and simulated 25%, 50%, and 100% increases in emissions from these sources. The different sources are driven by diverse climatic factors [*Carslaw et al*., 2010]. Nevertheless, the increases we simulate are within the range of previous estimates of the impact of the 2000–2100 climate change on natural aerosol emissions [*Heald et al*., 2008; *Spracklen et al*., 2009; *Carslaw et al*., 2010; *Cameron-Smith et al*., 2011].

[8] We explored the impact of aerosols on clouds through the first aerosol indirect (cloud albedo) effect, which is the microphysical response of clouds to changes in CDNC [*Forster et al*., 2007]. While this metric does not capture the full range of aerosol-cloud interactions, it is widely used to estimate the impact of aerosols on clouds and climate [*Forster et al*., 2007]. We calculated CDNCs using the aerosol size distribution simulated by GLOMAP-mode and a mechanistic parameterization of cloud drop formation [*Nenes and Seinfeld*, 2003; *Fountoukis and Nenes*, 2005] for a cloud updraft velocity of 0.2 m s^{-1}. We then used a radiative transfer model [*Edwards and Slingo*, 1996] and a methodology described in previous studies [*Spracklen et al*., 2011a, 2011b; *Rap et al*., 2013] to calculate the impact of the changes in CDNC on cloud albedo and radiative balance. The radiative transfer model has six bands in the SW and nine bands in the LW, with a delta-Eddington 2 stream scattering solver at all wavelengths. We employed a monthly mean climatology for water vapor, temperature, and ozone based on ECMWF reanalysis data, together with surface albedo and cloud fields from the International Satellite Cloud Climatology Project D2 [*Rossow and Schiffer*, 1999] for the year 2000.

3. Results and Discussion

[9] The spatial distribution of simulated CDNCs has changed markedly from the PI (Figure 1a) to the PD (Figure 1b). In the PI atmosphere, the model simulates similar annual hemispheric mean CDNC in the Northern Hemisphere (NH, 85 cm^{-3})

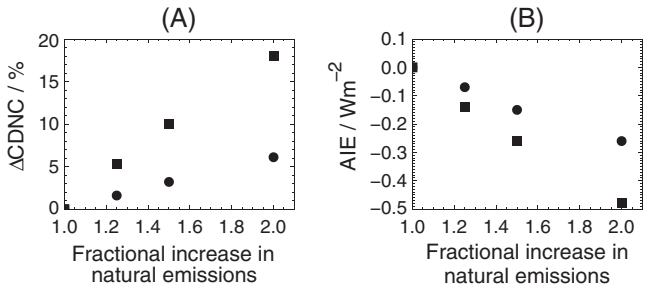

Figure 2. The impact of increased natural aerosol emissions on the Northern Hemisphere (a) mean cloud droplet number concentrations (CDNCs, 0.2 m s^{-1} updraft velocity) and (b) cloud albedo first aerosol indirect effect (AIE) in both present day (circles) and preindustrial (squares).

compared to the Southern Hemisphere (SH, 81 cm^{-3}). In the PD atmosphere, simulated hemispheric mean concentrations have increased by a factor of 1.75 in the NH (to 150 cm^{-3}) but by only a factor of 1.2 in the more pristine SH (to 96 cm^{-3}). This difference reflects the substantially greater anthropogenic aerosol sources in the NH [*Dentener et al*., 2006].

[10] We found that the simulated response of CDNC to changing natural aerosol emissions was substantially different in the PI (Figure 1c) compared to the PD (Figure 1d) atmosphere. In the PI atmosphere, doubling of natural aerosol emissions increased CDNC by 5–25% over much of the globe. In the PD atmosphere, the same increase in natural aerosol emissions resulted in a smaller change to CDNC, particularly over the NH where increases were typically less than 10%. The lower sensitivity in the PD atmosphere is due to anthropogenic emissions, increasing the baseline CDNC upon which changes to natural emissions operate. Concentrations of cloud droplets saturate at high aerosol number concentrations [*Andreae and Rosenfeld*, 2008; *Ramanathan et al*., 2001], so the CDNC in polluted regions is less sensitive to changes in natural emissions compared to pristine regions. This relationship between aerosol number concentration and CDNC has been carefully evaluated in our model [*Pringle et al*., 2009], giving us confidence that the response is well captured.

[11] Figure 2a shows the mean change in CDNC across the NH in response to changing natural emissions. We find that in the NH, the impact of changing natural aerosol emissions on CDNC is a factor of 3 smaller in the PD compared to the PI: doubling natural aerosol emissions leads to an 18% increase in the PI compared to a 6% increase in the PD. This behavior can be contrasted against the cleaner SH where doubling of natural aerosol has a very similar impact on CDNC in both the PI (19%) and the PD (16%).

[12] Figure 2b shows the first aerosol indirect effect due changes in natural aerosol sources in both the PD and PI atmospheres. In the PI atmosphere, doubling of natural aerosol sources results in a NH mean radiative effect of −0.51 W m^{-2}. In the PD atmosphere, the same increase in natural emissions results in a cooling effect which is a factor 2 smaller (−0.24 W m^{-2}). In the more pristine SH, the same fractional increases in natural aerosol emissions result in a hemispheric mean radiative effect of −0.65 W m^{-2} in the PI and −0.58 W m^{-2} in the PD. It is well known that pristine clouds are more susceptible to changing aerosol compared to polluted clouds due to the nonlinear response of CDNC to changing aerosol and because cloud albedo responds to fractional rather than absolute changes in CDNC [*Andreae and Rosenfeld*, 2008; *Ramanathan et al*., 2001]. Our results suggest that anthropogenic pollution aerosol is resulting in a substantial suppression of the sensitivity of cloud albedo to changing natural aerosol emissions across the Northern Hemisphere. This suppression has been recently observed at polluted locations [*Paasonen et al*., 2013] but has never been demonstrated at the global scale. Our results have implications for our understanding of the role of natural aerosol in climate change. We find that cloud albedo is more sensitive to changing natural aerosol emissions in the SH. Observed natural aerosol–climate feedbacks [*Paasonen et al*., 2013] are likely to have been more important in the prehuman atmosphere, before anthropogenic aerosol pollution.

[13] While we recognize that the magnitude of aerosol-cloud interactions is uncertain [*Forster et al*., 2007], our study indicates that interactions between natural aerosol and climate are likely to be weaker in today's polluted atmosphere than would have been the case before anthropogenic aerosol pollution. Our study may have implications for the role of other natural aerosol processes in climate. For example, interactions between cosmic rays, aerosol, clouds, and climate [*Carslaw et al*., 2002] which have been quantified as weak in today's atmosphere [*Pierce and Adams*, 2009; *Snow-Kropla et al*., 2011] may have also been more important prior to anthropogenic aerosol pollution. The reductions in anthropogenic aerosol emissions that are predicted over the next few decades [*van Vuuren et al*., 2011] may help to restore the role of natural aerosol–climate feedbacks in controlling future climate. Here we have used a global aerosol microphysics model to study one link within potential natural aerosol–climate feedbacks. Future studies need to explore the entire feedback cycle within fully coupled Earth system models.

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