

**Strong climate
warming from
pollution control**

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Air pollution control and decreasing new particle formation lead to strong climate warming

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Abstract

The number of cloud droplets determines several climatically relevant cloud properties. A major cause for the high uncertainty in the indirect aerosol forcing is the availability of cloud condensation nuclei (CCN), which in turn is highly sensitive to atmospheric new particle formation. Here we present the effect of new particle formation on anthropogenic aerosol forcing in present-day (year 2000) and future (year 2100) conditions. The total aerosol forcing (-1.61 W m^{-2} in year 2000) is simulated to be greatly reduced in the future, to -0.23 W m^{-2} , mainly due to decrease in SO_2 emissions and resulting decrease in new particle formation. With the total aerosol forcing decreasing in response to air pollution control measures taking effect, warming from increased greenhouse gas concentrations can potentially increase at a very rapid rate.

1 Introduction

Atmospheric new particle formation is a frequent phenomenon that enhances aerosol particle number concentrations almost everywhere in the troposphere (Kulmala et al., 2004; Spracklen et al., 2006, 2010; Yu et al., 2010). While the initial nucleation occurs between about 1–2 nm of particle diameter, nucleated particles are able to grow up to cloud condensation nuclei (CCN) sizes if sufficient amounts of condensable vapours are available (Lihavainen et al., 2003; Merikanto et al., 2009; Pierce and Adams, 2009). By this way, atmospheric new particle formation affects cloud droplet number concentrations (e.g. Kerminen et al., 2005; Makkonen et al., 2009; Merikanto et al., 2010) and contributes to indirect radiative effects (Wang and Penner, 2009; Kazil et al., 2010).

Sulphuric acid is thought to be the main precursor for atmospheric nucleation (Sipilä et al., 2010; Kulmala et al., 2006), hence the emissions of oceanic dimethyl sulphide (DMS) and both natural and anthropogenic sulphur dioxide (SO_2) are expected to dominate the new particle formation process. Anthropogenic SO_2 emissions have increased three orders of magnitude in the last 250 yr, from $0.06 \text{ Tg(S) yr}^{-1}$ (Dentener et al., 2006)

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in 1750 to 54 Tg(S) yr⁻¹ (Dentener et al., 2006) in 2000. Recently compiled “Representative Concentration Pathways” (RCPs) (Moss et al., 2010) project a significant decrease in global anthropogenic SO₂ emissions, with annual emissions of 6.5 Tg(S) (van Vuuren et al., 2007), 11 Tg(S) (Clarke et al., 2007; Smith et al., 2006; Wise et al., 2009) and 13 Tg(S) (Riahi et al., 2007) in year 2100. By contrast, the estimated present-day to future changes in oceanic DMS emissions are rather small: +6.5 % (), +2.4 % (Bopp et al., 2004) and –8.0 % (Stier et al., 2006). Most RCPs predict a continuous increase of ammonia emissions, at 2100 being 3 to 5 times those at 1850. Global emissions of fossil fuel organic and black carbon are predicted to peak before 2030 and then to decline rapidly, such that the levels at 2100 would be close to those at 1850. Future emissions of biomass burning aerosols behave very differently between the different RCPs in terms of both the sign of the trend and absolute emission levels.

Despite its importance for nucleation, sulphuric acid can explain only a fraction of the subsequent growth. Oxidation products of biogenic volatile organic compounds (BVOCs) may provide the required growth for aerosols to reach CCN sizes (Jimenez et al., 2009). In addition, certain organic compounds might also play a role in the first steps of nucleation (Metzger et al., 2010). However, the centennial-scale changes in BVOC emissions are highly uncertain, ranging from almost zero to increases by more than 70 %, depending on which processes are considered in the underlying emission algorithms and prognostic vegetation models (Arneth et al., 2007; Lathièrè et al., 2005).

Our aim is to study aerosol-cloud-climate interactions in three emission environments: pre-industrial (year 1750), present-day (year 2000) and future (year 2100). We provide a quantification of the impact of aerosol nucleation on particle number concentrations and anthropogenic aerosol forcing, accounting also for the possible impact of changes in biogenic precursor emissions (BVOCs and oceanic DMS).

2 Methods

We use the global climate model ECHAM5-HAM (Stier et al., 2005) with a novel description of boundary layer nucleation and particle growth by BVOC oxidation products (Makkonen et al., 2009). Aerosol model is coupled to a cloud microphysics model (Lohmann et al., 2007), and aerosols are activated as cloud droplets with a cut-off radius of 35 nm. The aerosol microphysics model M7 (Vignati et al., 2004) considers dust, sea salt, black carbon, particulate organic matter and sulfate. Particle formation is modeled with activation-type nucleation in the boundary-layer (Kulmala et al., 2006) and binary sulphuric acid-water nucleation throughout the atmosphere (Vehkamäki et al., 2002). Oxidation products from biogenic monoterpenes are assumed to condense on pre-existing aerosols in the boundary-layer. The sea-surface temperatures and sea ice extent are prescribed to present-day, and forcings are calculated as radiative flux perturbations (Haywood et al., 2009).

Anthropogenic aerosol and precursor emissions are taken from AeroCom for years 1750 (Dentener et al., 2006) and 2000 (Dentener et al., 2006). For the year 2100 we apply emissions of three RCPs: RCP 3-PD (van Vuuren et al., 2007), RCP 4.5 (Clarke et al., 2007; Smith et al., 2006; Wise et al., 2009) and RCP 8.5 (Riahi et al., 2007). The RCP 3-PD is an optimistic emission pathway corresponding to very low concentrations of greenhouse gases. Radiative forcing is expected to peak at 3.1 W m^{-2} and go down to 2.6 W m^{-2} until 2100. In RCP 4.5 total radiative forcing is stabilized to 4.5 W m^{-2} . RCP 8.5 presents a more pessimistic view of future where CO_2 concentration increases to 940 ppm until 2100 leading to total radiative forcing of about 8.5 W m^{-2} . Emitted primary aerosol size distributions are assumed identical in all simulations, and follow Stier et al. (2005). In simulations without nucleation, SO_2 is still able to condense on existing particles.

Natural emissions of sea salt and dust are modeled interactively in all simulations, hence their emission strength might differ between experiments e.g. in response to changes in wind speed. The emissions of volcanic sulphur are identical in all the sim-

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ulations performed corresponding to present-day emissions (Stier et al., 2005). For biogenic VOC emissions we apply monoterpene emission fields generated with the dynamic vegetation model LPJ-GUESS (Schurgers et al., 2009). Depending on whether the increasing atmospheric CO₂ concentration affect monoterpene production in the same way as it does for isoprene (Schurgers et al., 2009), these emissions may remain more or less unchanged, or they increase in response to warmer climate and higher vegetation productivity. To assess the responsiveness in the system we first assume almost constant BVOC emissions (Schurgers et al., 2009) and zero changes in DMS emissions, and then apply two additional future scenarios with an increase of 50 % in biogenic VOCs and an increase in oceanic DMS emissions by 10 % compared to present-day values.

3 Results and discussion

3.1 Particle number and CCN concentrations

Spatial distributions and temporal changes in CN (condensation nuclei, aerosol particles with diameter $d_p > 3$ nm) and CCN (cloud condensation nuclei, $d_p > 70$ nm) concentrations for the three years 1750, 2000 and 2100 are shown in Fig. 1, taken from simulations with new particle formation included. Pre-industrial primary emissions were low in terms of aerosol number: primary aerosols contributed significantly to aerosol number concentration only in regions with intensive wild-land fires (Africa, South America, North-East Eurasia). Pre-industrial anthropogenic sources of SO₂ were low, hence aerosol nucleation originated mostly from natural precursors. Some of these sources can be identified in upper panels of Fig. 1: volcanoes (Philippines, west of New Caledonia, and southern Japan) and DMS (band around 60° S). Human impact is clearly visible under present-day conditions. Increases in primary emissions and new particle formation lead to doubled CN concentrations in US, Europe, Middle East, India, China and South Africa. There are several large areas inside these regions where CN

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concentrations are up to five times above the pre-industrial values.

Figure 1 indicates significant reductions in CN concentration when moving from present-day to the year 2100: aerosol number concentrations are decreased by 50–90 % in North America, Europe, Eastern China and South Africa. Certain equatorial regions, such as Middle Africa, show increased CN concentrations due to increase in wild-fires. At global scale, only about 5 % of the future decrease in CN concentration can be explained by changes in primary particle emissions, the rest being due to a decrease in anthropogenic SO₂ emissions and the resulting decrease in atmospheric new particle formation.

For CCN, present-day emissions lead to doubled concentrations in almost all locations above land, compared to the pre-industrial values (Fig. 1, lower panels). CCN concentrations increase more than fivefold in North America, central Europe, India and China. Even with decreasing global emissions, CCN concentrations in year 2100 are still doubled to the pre-industrial values in India, Middle East, and some parts of Europe and Africa. Still, the simulated overall strong future decreases in the availability of condensation nuclei for cloud droplets are expected to lead to drastic changes in cloud properties.

The corresponding global 5-yr average cloud droplet number concentrations (CDNC) at cloud top height with and without nucleation are shown in Fig. 2. Including nucleation in the model increases the overall global CDNC by a factor of 2–3 compared to CDNC without nucleation. Our model shows an increase of 20 % in CDNC from year 1750 to 2000, which is lower than previously reported estimates: 66 % increase of CDNC between years 1850–2000 (Merikanto et al., 2010), 130 % increase in boundary layer CCN between years 1850 and 2000 (Wang and Penner, 2009) and 200 % increase in tropospheric CCN between years 1750–2000 (Pierce and Adams, 2009). This suggests that our model is less sensitive to the anthropogenic influence since pre-industrial times. When taking into consideration the uncertainty arising from future anthropogenic emission scenarios (red bar and corresponding shading), the introduction of air pollution control measures modifies future cloud droplet concentrations close

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to the pre-industrial state at global scale.

3.2 Forcing estimates

Figure 3 shows the total anthropogenic aerosol forcing for years 2000 and 2100 calculated as radiative flux perturbations (Haywood et al., 2009). The results clearly indicate how accounting for atmospheric nucleation is critical for aerosol forcing calculation. In the present atmosphere, including new particle formation increases the magnitude of total aerosol forcing from -1.03 W m^{-2} to -1.61 W m^{-2} , indicating more cooling from anthropogenic aerosols. More importantly, the change in total aerosol forcing from the year 2000 to the year 2100 is $+1.38 \text{ W m}^{-2}$ when nucleation is considered, while it is only $+0.64 \text{ W m}^{-2}$ without nucleation. Both of the present-day total aerosol forcing estimates are in the range of values reported by IPCC: a 90 % confidence range from -2.2 to -0.5 W m^{-2} (), which does not include the cloud lifetime effect. It should be noted that the minimum CDNC of $40 \# \text{ cm}^{-3}$ applied here can be considered rather high especially for pre-industrial clouds, hence causing an underestimation of the present-day forcing (Hoose et al., 2009).

The applied emission pathways show strong changes in anthropogenic emissions of black carbon (BC, 40–60 % reduction) and organic carbon (OC, 10–60 % reduction). It has been shown that a 50 % reduction in primary BC/OC mass and number emissions alone would lead to $+0.19 \text{ W m}^{-2}$ forcing in present-day climate (Chen et al., 2010), when taking into account both the cooling from reduced BC (-0.12 W m^{-2}) and simultaneous warming from decreased CDNC ($+0.31 \text{ W m}^{-2}$). Hence, the BC/OC reduction could explain a major part of the simulated future warming without nucleation ($+0.64 \text{ W m}^{-2}$). Kloster et al. (Kloster et al., 2008) showed that even a $+1.13 \text{ W m}^{-2}$ warming until 2030 from reduced aerosol load would be plausible under a “maximum feasible reduction” scenario. Although their present-day total aerosol forcing (-2.00 W m^{-2}) is stronger compared to our simulations, the applied 80 % reduction in anthropogenic SO_2 is comparable to future scenarios used here.

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While the simulated present-day total aerosol forcing (-1.61 W m^{-2}) is rather close (but of opposite signal) to the present-day forcing of CO_2 ($+1.66 \text{ W m}^{-2}$), the future warming from applied air pollution control and the resulting decrease in aerosol effects (a change of $+1.38 \text{ W m}^{-2}$ in total aerosol forcing) would accelerate the rate of climate warming in response to increasing greenhouse gas concentrations. For example, in the scenario of RCP 3-PD (van Vuuren et al., 2007) the CO_2 forcing is estimated to increase from $+1.5 \text{ W m}^{-2}$ (year 2000) to $+2.3 \text{ W m}^{-2}$ (year 2100), hence the warming from reduced aerosol forcing alone would be even 80 % more than the warming from increased CO_2 only.

Could changes in natural aerosol precursors counteract the effects of anthropogenic emission changes? This seems unlikely, as even in our simulation with +50 % BVOC the increased particle growth from oxidized organics introduce an extra cooling of -0.46 W m^{-2} and can not sustain the present-day level of aerosol negative forcing. Similarly, a dramatic increase of +10 % in DMS emission can only add an extra cooling of -0.12 W m^{-2} to the future aerosol forcing. It should be noted, however, that the effect of these additional emissions is not negligible, and that there might be additional processes and feedbacks involved that are not studied here, but that could act towards cooling in the future.

4 Conclusions

Our analysis provides evidence for how the present-day climate forcing by aerosol particles is controlled strongly by atmospheric new particle formation. This is due to a favourable combination of anthropogenic sulphur and biogenic organic vapour emissions, which together produce effectively new cloud condensation nuclei that enhance the negative cloud forcing. The current cooling effect by aerosol particles is reduced close to zero by 2100 regardless of exact time evolution of primary particle emissions, or the response of biogenic aerosol sources to the climate change. The suppression of future aerosol forcing, if true, would lead to even a larger warming of climate than

what is expected based on increased greenhouse gas concentrations (Andreae et al., 2005).

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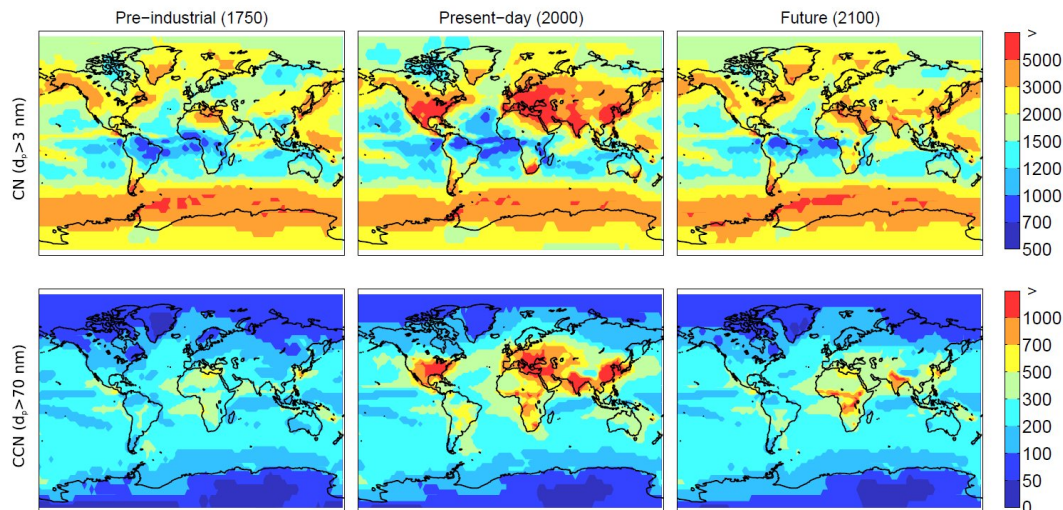


Fig. 1. Annual average concentration ($\# \text{cm}^{-3}$) of CN (condensation nuclei, aerosol particles with diameter $d_p > 3 \text{ nm}$) and CCN (cloud condensation nuclei, $d_p > 70 \text{ nm}$) in the boundary layer for year 1750 (left), 2000 (center) and 2100 (right). The results for the year 2100 are an average from all three future emission pathways: RCP 3-PD, RCP 4.5, RCP 8.5 (see Methods for details).

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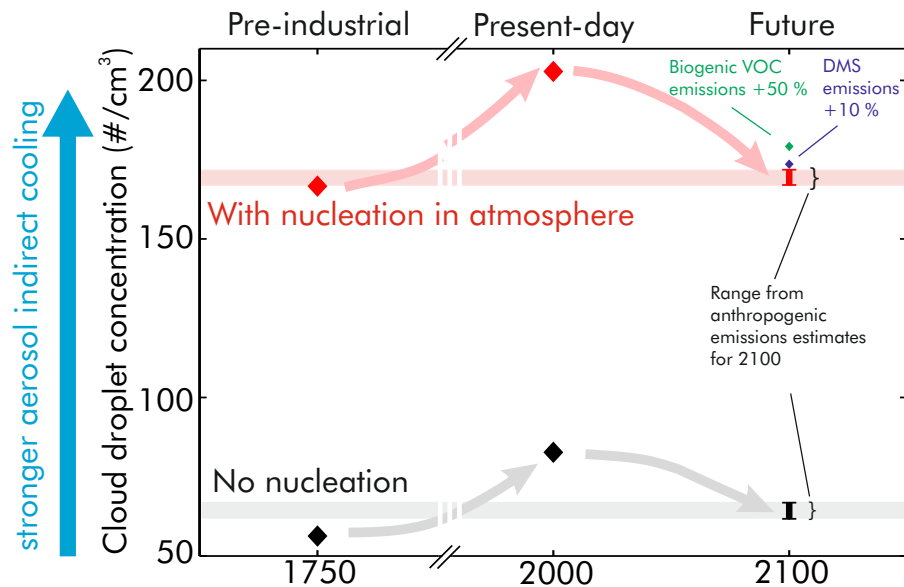


Fig. 2. Global 5-year average cloud droplet number concentration (\# cm^{-3}) for pre-industrial, present-day and future. Cloud droplet number concentration is sampled at cloud top height, from simulations with (red) and without (black) nucleation. Error bars and corresponding shading indicate the uncertainty in future anthropogenic emission pathway. Additional simulations with +50 % biogenic VOC emissions (green) and +10 % oceanic DMS emissions (blue) are simulated with the anthropogenic pathway RCP 4.5 (Clarke et al., 2007; Smith et al., 2006; Wise et al., 2009).

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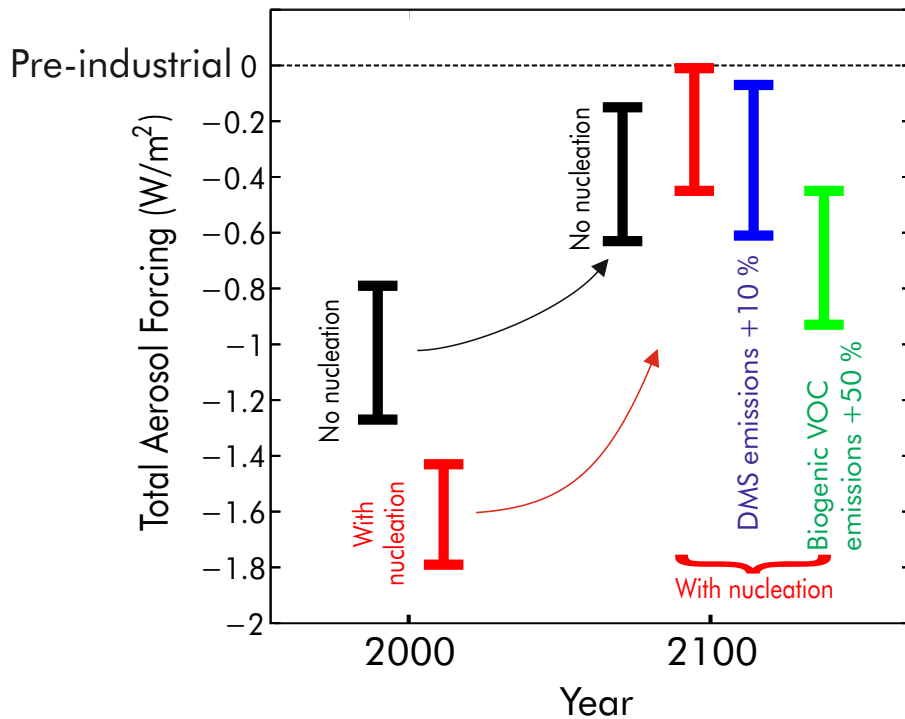


Fig. 3. Total aerosol forcing (W m^{-2}) for years 2000 and 2100. Additional simulations with +50% biogenic VOC emissions (green) and +10% oceanic DMS emissions (blue) are simulated with the anthropogenic pathway RCP 4.5 (Clarke et al., 2007; Smith et al., 2006; Wise et al., 2009).