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Long-term impacts of aerosols on the vertical development of clouds and precipitation

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Aerosols alter cloud density and the radiative balance of the atmosphere. This leads to changes in cloud microphysics and atmospheric stability, which can either suppress or foster the development of clouds and precipitation. The net effect is largely unknown, but depends on meteorological conditions and aerosol properties. Here, we examine the long-term impact of aerosols on the vertical development of clouds and rainfall frequencies, using a 10-year dataset of aerosol, cloud and meteorological variables collected in the Southern Great Plains in the United States. We show that cloud-top height and thickness increase with aerosol concentration measured near the ground in mixed-phase clouds—which contain both liquid water and ice—that have a warm, low base. We attribute the effect, which is most significant in summer, to an aerosol-induced invigoration of upward winds. In contrast, we find no change in cloud-top height and precipitation with aerosol concentration in clouds with no ice or cool bases. We further show that precipitation frequency and rain rate are altered by aerosols. Rain increases with aerosol concentration in deep clouds that have a high liquid-water content, but declines in clouds that have a low liquid-water content. Simulations using a cloud-resolving model confirm these observations. Our findings provide unprecedented insights of the long-term net impacts of aerosols on clouds and precipitation.

Aerosols, the tiny particles in the atmosphere produced by both natural processes and anthropogenic activities, impinge on Earth's climate by altering its energy balance and clouds[1,2]. More aerosols produce a higher number of smaller droplets, thus suppressing the warm rain-forming process[3–6]. On the other hand, delaying precipitation initiation to above the freezing level converts rain into ice hydrometeors. The release of extra latent heat could invigorate the vertical development of clouds and enhance precipitation[7–9]. Enhanced melting and evaporative cooling at lower levels can further invigorate convection[10,11], which may result in the enhancement of rainfall[10–15]. The opposite effects are dictated by microphysical, dynamic and thermodynamic conditions[13,16–18]. The overall net effects have yet to be identified, let alone quantified, owing to a lack of long-term observational data, a limited understanding of an overly complex problem, large model uncertainties and an excessive computation burden.

Since the late 1980s, the US Department of Energy's Atmospheric Radiation Measurements (ARM; refs 19,20) programme has provided extensive and accurate observations aimed at understanding and parameterizing atmospheric processes in climate models. The longest and most complete sets of measurements have been made at the Southern Great Plains (SGP), where a large array of state-of-the-art passive and active instruments has been deployed[21,22]. Continuous measurements of aerosol, cloud and meteorological variables are employed in this study. Cloud liquid-water path (LWP) is retrieved from microwave radiometer measurements[21]. Cloud geometry and phase are inferred from a suite of passive and active sensors such as millimetre cloud radars, laser ceilometers, lidars and microwave radiometers[22]. Both heights and temperatures of cloud bases and tops are used. Cloud-top temperature (CTT) helps identify the phase of a cloud, whereas cloud-base height (CBH) indicates the likelihood of interaction between clouds and aerosols measured near the ground. To better identify the aerosol effect, only single-layer clouds are considered.

Continuous cloud condensation nucleus (CCN) measurements were only available for a short period of time. Condensation nucleus measurements were used instead because of their longer record of collection and because the concentration of condensation nuclei is proportional to aerosol concentration. An analysis of three years' worth of concurrent condensation-nucleus and CCN data acquired at the SGP from 2006 to 2008 shows a sound correlation between them, with the regression line right along that derived from many previous observations[23], as shown in Supplementary Fig. S1. To avoid the influence of precipitation scavenging, condensation-nucleus measurements made 1.5 h before the onset of rain are used. As an indicator of convection, vertical wind data at 500 mb are also employed, together with other meteorological variables. Vertical wind data were derived from a variational analysis by taking advantage of extensive ARM measurements at the surface and at the top of the atmosphere, including precipitation, latent and sensible heat fluxes and radiative fluxes[24].

Aerosols and the vertical development of clouds

In light of the key factors influencing the aerosol invigoration effect[8,11], our analyses were carried out by differentiating clouds according to their base and top temperatures (CBT and CTT), CBH, phase and season. Clouds with CTT < −4 °C and CBT > 15 °C are considered as mixed-phase clouds, the most favourable condition for the aerosol invigoration effect postulated from theory[8]. To warrant statistical significance, all cloud and precipitation events that occurred over the 10-year period were analysed.

Figure 1 shows the variations of CTT with concentration of condensation nuclei, revealing the impact of various factors dictating aerosol–cloud interactions from the ten-year continuous

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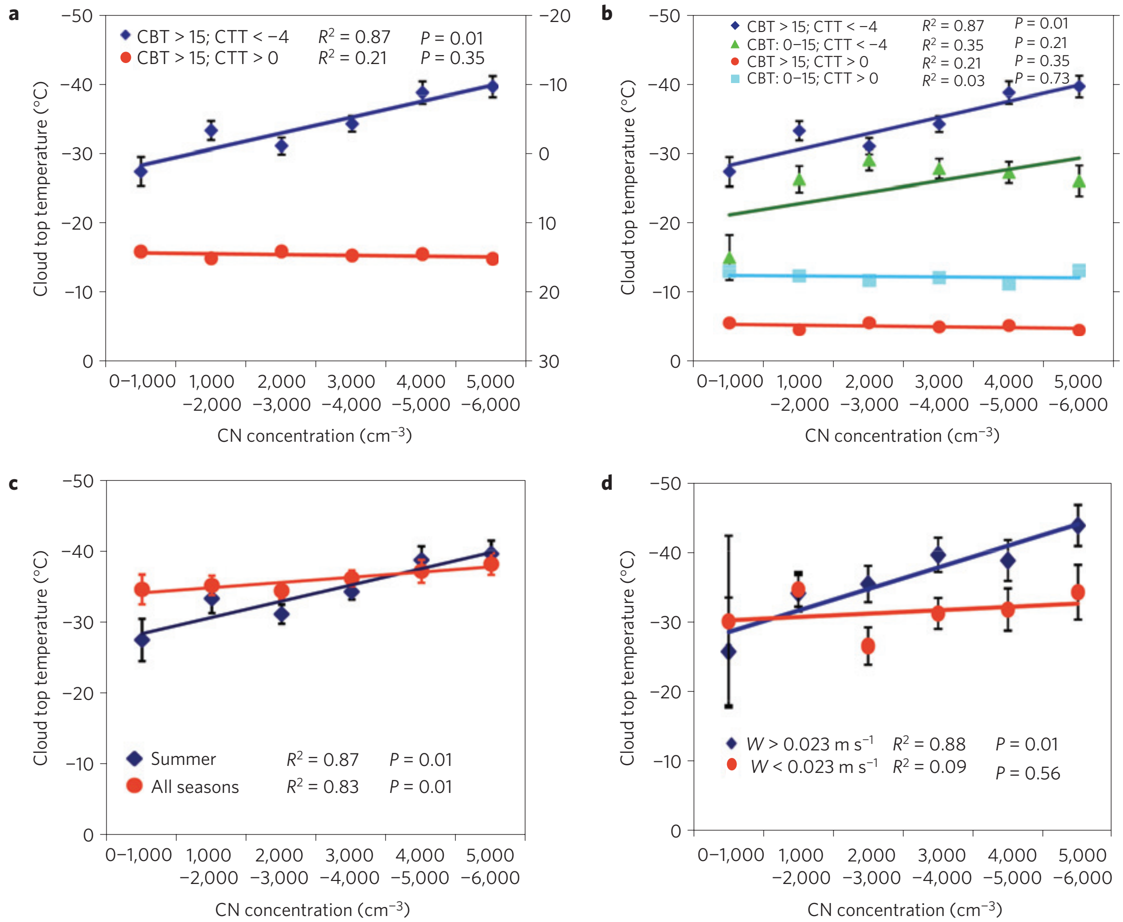

Figure 1 | Variations of cloud top temperature (CTT) with concentration of condensation nuclei (CN) for single-layer clouds. a,**b**, Summertime CTT for different ranges of cloud top and bottom temperatures. The left and right y axes correspond to the data of CTT $< -4\,°C$ and $> 0\,°C$ respectively. **c**, Single-layer clouds of CBT $> 15\,°C$ and CTT $< -4\,°C$ in summers and all seasons. **d**, The same clouds but differentiated by vertical velocities in summer. Error bars, s.e.m.; temperature unit, degrees Celsius.

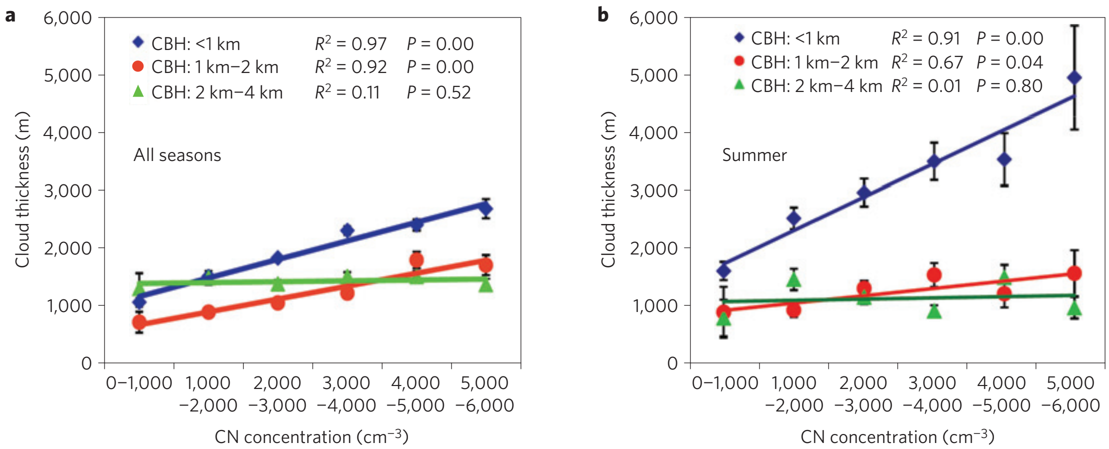

Figure 2 | Changes in cloud thickness with concentration of condensation nuclei (CN). a, Changes for all seasons. **b**, Changes in summers only. Clouds are divided into three ranges of CBH (<1, 1–2 and 2–4 km). No constraint is applied to CTH. Error bars, s.e.m.

measurements. Remarkable differences in the response of CTT to concentration of condensation nuclei exist between mixed-phase clouds containing liquid and ice particles and liquid-only clouds (Fig. 1a). The top of mixed-phase clouds increases significantly with increasing concentration of condensation nuclei, whereas there is no change for pure liquid clouds. The trend is statistically significant at the 95% confidence level. The sensitivity of the relationship between CTT and concentration of condensation nuclei is further demonstrated in Fig. 1b for four different combinations of CTT and CBT. There is little change in CTT with concentration of condensation nuclei when the CTT is above the freezing temperature. When CTT $< -4\,°C$, the sensitivity depends on the CBT. The effect is most significant for summer seasons, as the slope of the relationship for all seasons is considerably smaller than that in summer seasons

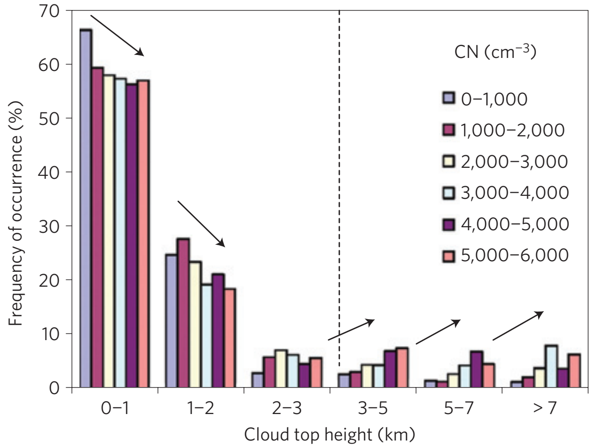

Figure 3 | Frequency of occurrence for six bins of cloud top height and six subsets of concentration of condensation nuclei (CN). The dashed line is the mean freezing level (∼3.3 km). Arrows indicate the trend of the frequency with increasing concentration of condensation nuclei. Except for 2–3 km and >7 km cloud top height bins, the trends are all significant at 95% significance levels.

(Fig. 1c), presumably because of less thermally driven convective clouds in colder seasons. As predicted by theory[8,17], invigoration occurs chiefly in warm-base convective clouds. The contrast between summer and all seasons may stem from a combination of factors. Further analyses of seasonal variations in various influential factors suggest that summer is favourable for the invigoration effect owing to strong convection, weak wind shear and high water-vapour content. The importance of convection is confirmed by partitioning the data into two equal-sized subsets differentiated by ARM observation-based vertical velocities at 500 hPa (Fig. 1d). For clouds developing in an atmosphere with a stronger vertical upward motion (>0.023 m s^{-1}), CTT decreases significantly with increasing concentration of condensation nuclei, but barely changes for clouds with weak updraughts and downdraughts.

Relating relationships between CTT and concentration of condensation nuclei to the aerosol effect is corroborated by the fact that the effect only occurs significantly for clouds with bases low enough to interact with boundary-layer aerosols measured near the ground. Figure 2 shows the dependence of cloud thickness on concentration of condensation nuclei for three ranges of CBH for all seasons (Fig. 2a) and for summer only (Fig. 2b). The response of cloud thickness to concentration of condensation nuclei is most significant for low-base clouds (CBH < 1 km). As the CBH increases, cloud thickness is gradually decoupled from the influence of ground-level condensation nuclei, as we would expect. For CBH greater than 2 km, the dependence disappears entirely. Yet, the sensitivity is significantly stronger in summer than in all other seasons. As the measurements were made under exactly the same ensemble conditions except for different CBH, there seems to be no other more plausible explanation than the aerosol effect.

The finding that invigoration only occurs for warm-base mixed-phase clouds is further reinforced by an analysis of cloud frequency for different ranges of concentration of condensation nuclei and cloud-top height (CTH), as shown in Fig. 3. CTHs are divided into six bins. In each height bin, there are six condensation nucleus subsets and the frequency of occurrence is calculated in each bin and subset. As the concentration of condensation nuclei increases, deep clouds occur more frequently whereas shallow clouds occur less frequently; the transition takes place right around 3.3 km, the mean freezing level as determined from ARM data. This illustrates that aerosols tend to inhibit the development of low thin clouds and foster the development of deep thick clouds.

Aerosols and precipitation

The significant impact of aerosols on cloud vertical development probably induces changes in precipitation with aerosol loading as well[10–13]. To test this, we first examine the influence of aerosols on rainfall frequency by counting all individual raining events that occurred over the 10-year period and then associating them with the concentration of condensation nuclei measured before the onset of rain. Rainfall frequency is calculated as the ratio of the number of rain events divided by the total number of observations, regardless of rain duration. As rain frequency is highly correlated with LWP, the data are grouped into two categories based on LWP to constrain its dominant influence. The categories are LWP > 0.8 mm and LWP < 0.8 mm, which correspond to deep and shallow clouds, respectively. As seen in Fig. 4a, rainfall frequency increases with increasing concentration of condensation nuclei for high LWP but decreases for low LWP. The regression relations are statistically significant at a 95% confidence level.

Aerosols can also alter the rainfall regime, as revealed by the contrast in the frequency distribution of rain amount per rain

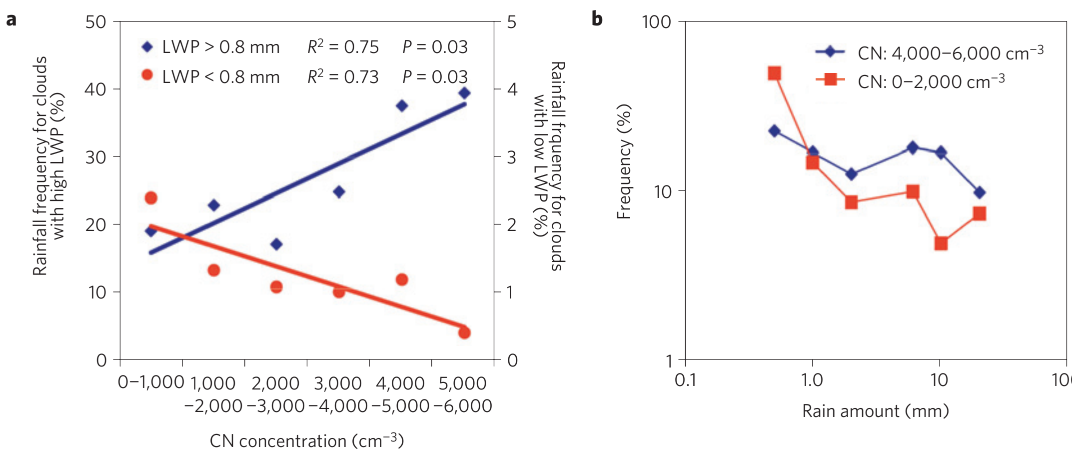

Figure 4 | Changes in rainfall frequency and rain rate distribution with concentration of condensation nuclei (CN). a, Rainfall frequency as a function of concentration of condensation nuclei for different LWP bins at the SGP site during all summer seasons. Clouds are grouped into two categories: LWP > 0.8 mm and LWP < 0.8 mm. Note that different ranges of rainfall frequencies are used, as indicated by the two *y* axes. **b**, Frequency of occurrence of rain amount per rain event during all summer seasons. The rain events are grouped into two categories (clean, CN < 2,000 cm^{-3}, and dirty, 4,000 < CN < 6,000 cm^{-3}) on the basis of concentration of condensation nuclei measured 1.5 h before the rain event.

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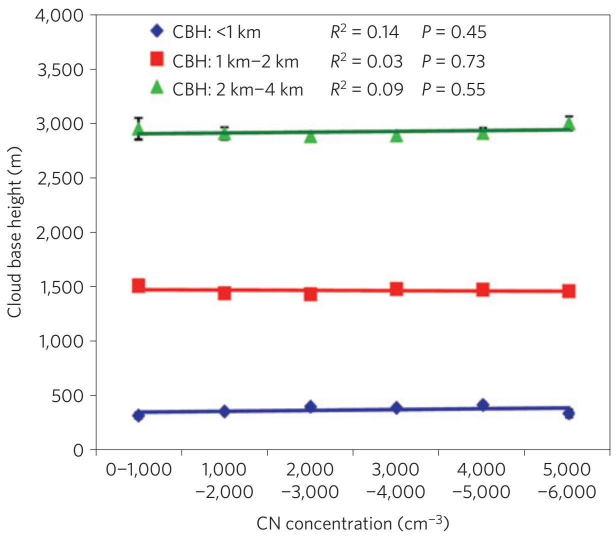

Figure 5 | Cloud base height (CBH) as a function of concentration of condensation nuclei (CN) for single-layer clouds during all summer seasons. No constraint is applied to cloud top height. Error bars, s.e.m.

event for summer between clean (concentration of condensation nuclei, CN < 2,000 cm^{-3}) and dirty (4,000 < CN < 6,000 cm^{-3}) conditions as shown in Fig. 4b. Overall, more than 50% of rain events precipitate less than 1 mm per rain event. Heavy rains occur more frequently and light rains occur less frequently under polluted conditions than under clean conditions. This is probably because aerosols suppress light warm rain processes[25] but invigorate intense ice precipitation processes that can lead to the formation of more hail[26], which when melting aloft produces high rain intensities. Note that changes in rain frequency can also be affected by gustiness due to aerosol-induced increases in evaporation and downdraughts[11,13].

Real or false relationships?

Given the long-term data used, the above findings attest to the climatological significance of the impacts of aerosols on clouds and precipitation. Whereas it has always been a big challenge to obtain direct evidence of these effects, the following analyses may be construed as indirect evidence. First, by cloud formation theory, cloud condensation determines the CBH, which is dictated by atmospheric thermodynamics, that is temperature and humidity profiles. As shown in Supplementary Fig. S2, neither profile is affected by the concentration of condensation nuclei. It is thus expected that CBH is independent of the concentration of condensation nuclei. This is confirmed in Fig. 5. In sharp contrast to the dynamic variations of CTT with concentration of condensation nuclei, CBH is not affected by concentration of condensation nuclei at all for any range of CBH. This lack of response to the concentration of condensation nuclei also supports our argument that changes induced by atmospheric dynamics/thermodynamics are effectively removed by using an exceptionally large number of samples in this study.

However, the argument would not be valid if the concentration of condensation nuclei were the proxy of a meteorological variable. Both clouds and precipitation are strongly affected by a large number of meteorological variables. There is no doubt that, for each individual cloud and rain event, the influence of meteorology is so overwhelming that it can overshadow aerosol effects. To test if the concentration of condensation nuclei is correlated with any meteorological variables influencing cloud development, we examined the relationships between the concentration of condensation nuclei and four sets of meteorological variables: (1) surface temperature, pressure, wind and humidity; (2) the profiles of temperature (T), dew-point temperature (T_d) and wind shear; (3) atmospheric stability indices and (4) surface flux. The results are presented in Supplementary Figs S2–S5.

There is no significant relationship between any of the surface meteorological variables and the concentration of condensation nuclei, except for a weak relation with wind speed (Supplementary Fig. S3). More condensation nuclei accumulate under calm conditions than under windy conditions, which fails to explain the above finding because calm atmospheric conditions correspond to a stable atmosphere. The profiles of T and T_d overlap tightly between low and high aerosol loadings (Supplementary Fig. S2), as is also indicated by the virtually equal values of the lower-tropospheric static stability[27]. Mean lower-tropospheric static stability values are 14.71 °C and 14.93 °C under the cleanest and dirtiest conditions, implying that the concentration of condensation nuclei is independent of atmospheric thermodynamics. Looking more closely, the boundary layer is slightly more stable under dirty atmospheric conditions than under clean ones (Supplementary Fig. S2), and thus cannot explain the systematic differences in CTT and rainfall properties. As the driving factor for thermally induced convection, surface-sensible heat fluxes show no obvious trend with the concentration of condensation nuclei (Supplementary Fig. S4). There is a weak trend of wind shear increasing with concentration of condensation nuclei (Supplementary Fig. S5). Model studies showed that the aerosol invigoration effect depends on wind shear. For a single cloud, weak wind shear favours the effect[17]. For cloud systems, aerosols may intensify secondary clouds under strong wind shear[28], whereas our model simulations described below reveal a clear invigoration effect for a cloud system under weak wind shear.

Model simulations

A decade seems to be a sufficiently long period to effectively minimize the influence of meteorological variability. To provide further support and insight to our observation-based findings, we conducted model simulations using a full-fledged cloud-resolving model[16,17] for two cloud systems over the SGP. The two cases (2 April 2006 and 8 July 2008) are typical of a warm-base (CBT of about 19 °C) convective cloud in summer and a cool-base (CBT of about 11 °C) frontal cloud in spring. Wind shear is stronger in the spring case than in the summer case, also typical for these seasons. Model simulations were conducted over nested domains of 1,656 × 1,608 km^2 and 770 × 746 km^2 for the outer and inner domains at resolutions of 12 and 2.4 km, respectively. The three-hourly National Centers for Environmental Prediction North American Regional Reanalysis dataset (32 km) is used to provide initial and boundary conditions for the coarse domain. Sensitivity tests were conducted for changing CCN concentrations only while other conditions remained the same for the inner domain. The simulated range of CCN (280–1,680 cm^{-3}) corresponds to a condensation nucleus range of 1,037–6,222 cm^{-3}, which is very close to the observed range. Simulated changes in cloud properties and precipitation in response to changes in CCN concentration are similar to the observational findings for both clouds and precipitation (Fig. 6). As CCN increases, cloud thickness, top height and rain frequency and amount increase for the warm-base cloud (CBT > 15 °C). No significant changes are seen in cloud properties and a decreasing trend in rain frequency and amount is suggested for the cool-base cloud (CBT < 15 °C). These model results support our arguments for the causes of the observed trends. In theory, the enhancement of rain frequency by CCN could result from enhanced gustiness[11,13] and droplet freezing[10,17]. To examine the relative contributions of these two factors, all simulations for the two cases were rerun without considering ice processes. In general, the rain frequency decreases slightly with increasing CCN (Supplementary Fig. S6), suggesting that gustiness is not the main factor responsible for enhanced rain frequency due to increasing CCN.

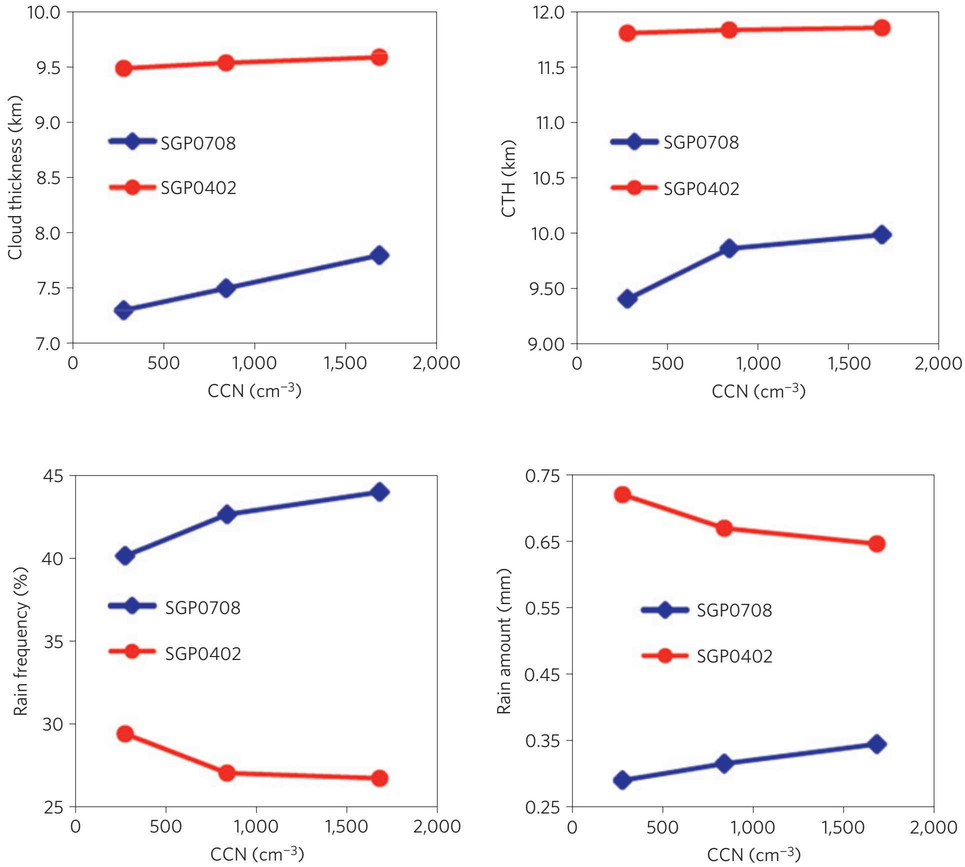

Figure 6 | Modelled changes in cloud thickness, CTH, rain frequency and rain amount with CCN. SGP0708 indicates the summertime warm-base (CBT of about 19 °C) convective cloud case and SGP0402 indicates the springtime cool-base (CBT of about 11 °C) convective cloud case. Cloud thickness and top height are averaged over the grid points where LWP > 0.8 mm. The rain frequency is the number of grid points with a rain rate greater than 0.01 mm h^{-1} divided by the total number of grid points (as a percentage). The rain amount is averaged over the domain that covers the convective system but excludes the effect from the boundaries.

Findings and implications

Aerosols are known to have a variety of effects on clouds and precipitation, contingent on meteorological conditions and aerosol properties. The vast majority of previous observational studies are based on limited cases from which it is hard to determine which effects are more significant or dominant than others and thus their long-term implications remain unknown. Using an unprecedented set of extensive measurements collected over a 10-year period at the ARM SGP site, strong long-term aerosol effects are revealed. A strong aerosol invigoration effect on convection is observed in summer, leading to higher cloud tops for mixed-phase clouds with low bases. The precipitation frequency is found to increase with increasing concentration of condensation nuclei for clouds with high water contents but decreases for clouds with low water contents. The findings concerning the effects of aerosols on both clouds and precipitation have numerous implications for climate studies, and even have economic consequences.

The invigoration-induced upward motion can change regional circulation patterns[29], which can potentially alter larger-scale circulations and affect global climate[30]. The delayed onset of precipitation and stronger updraughts could result in more aerosol particles and water transported into the upper troposphere and even the lower stratosphere. It also suppresses the wet scavenging of aerosols, creating a positive feedback[8].

The significant increase in CTH induced by the aerosol invigoration effect reduces the long-wave emission by clouds owing to lowered temperatures[31], a warming effect. On the other hand, an increase in cloud thickness can enhance solar reflection, a cooling effect. Because the two effects offset each other[10,11], their net effect is uncertain because their responses to changing cloud properties are different and depend on cloud thickness and top temperature.

The findings presented here also have important implications for the redistribution, availability and usability of water resources in different regions of the world. Pollution would have a net suppressing effect on precipitation from clouds that form in relatively dry environments, hence exacerbating aridity. Conversely, aerosols present in moist climates are likely to fuel convective clouds and worsen flooding in the summertime owing to the invigoration effect[16,32]. The strong signal of the human impact on nature emerging from long-term observations thus has social and economic consequences.

The clear observational evidence with the support of model simulation results of aerosols affecting convective clouds and precipitation is a testimony to the fact that human activities can impinge on the natural system of our planet by altering cloud development, precipitation and latent heating profiles to a much greater extent than previously thought. Although the effects for different kinds of cloud are of opposite signs and partially buffer each other[33,34], changes in the vertical and spatial distribution of heating can still have a substantial impact on the climate system. Incorporation of these effects into climate models may reveal significantly different impact than considered until now.

Economic development is often accompanied by increases in aerosol emissions, especially in developing countries[35–37]. It is worth noting that similar findings emerge from an analysis of global multiple A-train satellite products[38], attesting to the ubiquity of the effects. The findings presented here imply a potentially adverse impact on sustainable development over regions vulnerable to extreme meteorological events such as drought or flooding. Even if total rainfall amounts remain intact, changes in the frequency of light and heavy rains as found here would have consequences in terms of water usage efficiency, a key factor for life and agriculture.

Methods

Cloud-resolving model. Simulations have been carried out using the Weather Research and Forecasting model[39] coupled with a spectral-bin microphysics (SBM; ref. 40). The SBM solves a system of kinetic equations for the size distribution functions of water drops, ice crystals (plate, columnar and branch types), snow/aggregates, graupel and hail/frozen drops, as well as aerosols. Each size distribution is represented by 33 mass-doubling bins; that is, the mass of a particle m_k in the k th bin is determined as $m_k = 2m_{k-1}$. All relevant microphysical processes/interactions including droplet nucleation, primary and secondary ice generation, condensation/evaporation of drops, deposition/sublimation of ice particles, freezing/melting and mutual collisions between the various hydrometeors are calculated explicitly. We employed a fast version of SBM called 'Fast-SBM' (ref. 32) in which all ice crystals and snow (aggregates) are calculated on one size distribution. Smaller ice particles with sizes less than 150 μm are assumed to be crystals, whereas larger particles are assigned to snow. Similarly, graupel and hail are also combined into one size distribution. No changes in the microphysical processes have been made, compared with the full SBM. As a result, the number of size distributions decreases from eight to four (aerosols, water drops, low-density ice, high-density ice). The Fast-SBM used in this study has been updated with a new remapping scheme applied to diffusion growth/evaporation and an updated melting scheme[41].

Weather Research and Forecasting simulations were carried out for two deep convective cloud cases over the SGP. One occurred on 8 July 2008, a typical summertime warm-base convective cloud with weak wind shear (referred to as SGP0708). The other case is a cool-base convective cloud occurring on 2 April 2006, representing a typical springtime frontal-like system with stronger wind shear (referred to as SGP0402). Simulations are carried out using realistic boundary conditions. Two nested domains with a horizontal resolution of 12 km and 2.4 km are used with 51 vertical levels. The numbers of horizontal grid points for domain 1 (coarse-grid domain) and domain 2 (fine-grid domain) are 138 × 134 and 321 × 311, respectively. The three-hourly National Centers for Environmental Prediction North American Regional Reanalysis data on the Eta 221 grid (32 km) are used to provide initial and boundary conditions for domain 1. To reduce computational time, we used the 'nest down' approach, in which simulations for the finer grid are carried out separately with initial and lateral boundary conditions obtained from the coarse-grid runs. Whereas the coarse-grid runs were carried out using the two-moment bulk scheme[42], the 'nest down' finer-grid runs were carried out using SBM.

Aerosol effects are examined by running three sensitivity simulations with a total CCN concentration of 280, 3 × 280 and 6 × 280 cm^{-3}, respectively. The simulated range of the CCN (280–1,680 cm^{-3}) corresponds to a condensation nucleus range of 1,037–6,222 cm^{-3}, which is very close to the observed range. For each cloud case, simulations are run for 36 h. To avoid washout of aerosols by incoming air from the lateral boundaries, aerosol sources are set up at the lateral boundaries that include the outer five grid cells on each side of the domain. CCN and cloud drop concentration are diagnosed rather than predicted for clouds at the lateral boundaries. The Goddard shortwave radiation scheme and rapid radiative transfer model longwave radiation scheme are used in this study.

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Author contributions

Z.L. initiated the project, led the study and wrote the manuscript. F.N. carried out data analyses, prepared the figures and documented the study. J.F. conducted model simulations. D.R. and Y.L. participated in science discussions and suggested analyses. Y.D. helped generate some supplementary figures.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturegeoscience. Reprints and permissions information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to Z.L.