Vol 461|1 October 2009|**doi:10.1038/nature08281** nature

REVIEWS

Untangling aerosol effects on clouds and precipitation in a buffered system

Bjorn Stevens[1,2] & Graham Feingold[3]

It is thought that changes in the concentration of cloud-active aerosol can alter the precipitation efficiency of clouds, thereby changing cloud amount and, hence, the radiative forcing of the climate system. Despite decades of research, it has proved frustratingly difficult to establish climatically meaningful relationships among the aerosol, clouds and precipitation. As a result, the climatic effect of the aerosol remains controversial. We propose that the difficulty in untangling relationships among the aerosol, clouds and precipitation reflects the inadequacy of existing tools and methodologies and a failure to account for processes that buffer cloud and precipitation responses to aerosol perturbations.

Precipitation-mediated aerosol–cloud relationships are often called 'lifetime effects', and stem from the hypothesis that changes in the aerosol lead to changes in the precipitation efficiency (or colloidal stability) of clouds[1], which in turn changes cloud amount. The potential importance of such effects for cloud radiative forcing is especially evident in shallow marine cloud systems[2–4]. The clouds in such systems have an albedo tenfold larger than that of the ocean surface. By virtue of their shallowness, however, they radiate energy at an effective temperature commensurate with that of the surface. Hence, shallow maritime clouds effectively modulate the net solar radiation entering the climate system, with no compensating effects on the budget of outgoing terrestrial radiation[5]. Less appreciated is that shallow clouds (particularly trade-wind clouds) are also especially effective at mixing moisture away from the surface, thus enhancing surface evaporation and moistening the lower troposphere[6]. Because they affect both the hydrological cycle and the balance of radiant energy (that is, the net radiative forcing), understanding how shallow maritime clouds respond to perturbations in anthropogenic aerosol is key to establishing the extent to which changes in the aerosol may offset or counteract the warming effects of anthropogenic greenhouse gases[7]. Interest in cloud lifetime effects is motivated by these considerations.

The complexity of the climate system, and the limitations of the tools we have to study it, have led to the development of two schools of scientific enquiry into the lifetime hypothesis described above. The first takes the lifetime effect as given and attempts to measure its trace statistically, either in observations or by enforcing relationships among the aerosol, clouds and precipitation, in large-scale models. The second takes the effect as hypothetical and endeavours to test it or the various assumptions upon which it is based, for instance through dedicated field or modelling studies that explore how individual clouds or fields of clouds respond to changes in the ambient aerosol. The two approaches naturally differ in scale, but also conceptually. Large-scale modelling studies are often premised on clouds and precipitation being strongly sensitive to the aerosol. However, such premises ignore mechanisms that, according to small- or regime-scale studies, absorb (or offset) some of the effects of aerosol perturbations and, hence, buffer the system. This disjunction, combined with a growing understanding of other limitations of the tools employed in large-scale surveys (that is, satellite observations and large- or global-scale models), explains why the statistical effect of the aerosol on clouds and precipitation remains so controversial.

Here we propose that the sensitivity of clouds and precipitation to changes in the aerosol is regime dependent, and that although we expect lifetime effects to be on average weaker than implied by simple arguments (that is, buffered), substantial effects may still emerge in specific circumstances or regimes. Hence, research aimed at untangling the effects of the aerosol on clouds and precipitation should intensify efforts to understand those cloud and precipitation regimes in which the signature of lifetime effects is likely to be clearest.

Cloud lifetime hypotheses

What has come to be known as the cloud lifetime effect was put forward twenty years ago[3]. Initially it was formulated in specific terms for one particular cloud regime, as illustrated in Fig. 1. The basic idea is that the aerosol, through its effect on the ability of clouds to form precipitation, can alter cloudiness. It has since been extended to incorporate a variety of mechanisms or regimes[8], some having little to do with the concept of a cloud lifetime[4]; hence the tendency to speak of cloud lifetime hypotheses (or effects) rather than a single 'hypothesis'. The absence of observations of cloud life cycles that show precipitation reducing cloud lifetimes underscores how loosely the term 'lifetime' has come to be applied[9].

The distinguishing quality of lifetime hypotheses is instead the idea that the macrostructure of the cloud (such as its spatial extension or liquid-water content) is determined by the efficiency with which precipitation develops, which is in turn regulated (at least in part) by the aerosol. This differentiates 'cloud lifetime effects' from the 'cloud albedo effect'[10], for which aerosol perturbations lead to more reflective clouds given a fixed cloud macrostructure.

Lifetime hypotheses encapsulate three basic ideas: that the number concentration of cloud droplets increases with anthropogenic aerosol burden; that the precipitation efficiency of shallow clouds decreases monotonically as a function of the concentration of cloud droplets; and that cloud albedo is a strictly decreasing function of precipitation efficiency. The first idea is also the basis for the albedo effect, which rests on the radiative signature of such changes alone. The second idea has a long history in the literature[1] and has served as the basis for much research, and even more speculation, on the efficacy of weather modification by glaciogenic or hygroscopic cloud seeding[11]. The

---

[1]Max-Planck-Institut für Meteorologie, KlimaCampus, 20251 Hamburg, Germany. [2]Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles, California 90095-1565, USA. [3]NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, Colorado 80305, USA.

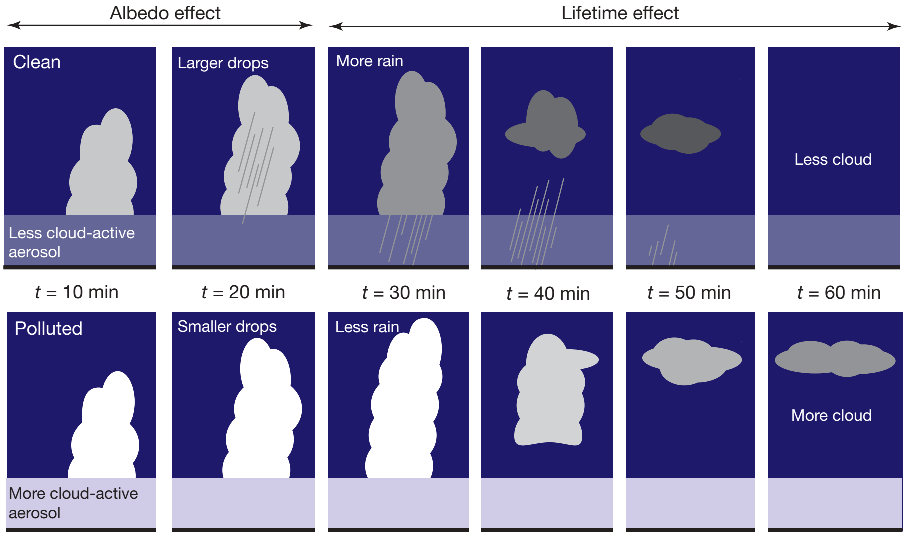

Figure 1 | The lifetime (and albedo) effect as originally proposed. In polluted air masses, clouds consist of more droplets that coalesce into raindrops less effectively, leaving longer-lived clouds (*t*, time). Here and in Figs 3 and 4, a single cloud is meant to represent the average response of a field of clouds.

third idea provides the essential link to the radiative forcing of the climate system. Our exploration of these ideas, and lifetime effects as a whole, focuses on warm clouds, as these have historically been at the centre of the lifetime hypothesis. Many of our findings are, however, also relevant to emerging studies of lifetime effects in cold or ice-bearing clouds[8,12].

Satellite surveys

Observations of the manifestations of a lifetime effect can be striking. Photographs of the collapse of clouds in shafts of precipitation that readily form in aerosol-poor environments are common. Equally evocative is the apparent transition from low-albedo (open-cellular) shallow-cumulus cloud regimes to high-albedo (closed-cellular) stratocumulus cloud regimes as a result of aerosol infusions from passing ships[13]. An example of the latter is shown in Fig. 2. Such features differ from ship tracks (which are often taken as a signature of the cloud albedo effect)[14] because the open-cellular structure in the low-albedo region is associated with regions of precipitation, whereas the closed-cellular structure in the high-albedo region is usually observed to be non-precipitating[13,15,16].

Large-scale satellite surveys provide a detailed view of the aerosol–cloud–precipitation system, particularly with the emergence of a new generation of active remote sensors[17]. Correlative studies based on the satellite record have found intriguing relationships between proxies for cloud-active aerosol and cloud microstructure the senses of which are consistent with the cloud albedo and lifetime effects. For example, cloud droplets are on average smaller in the presence of more aerosol particles. Although evidence of this kind is quite common, estimates of its global magnitude are sensitive to methodological details[18–21]. Cloud fraction also tends to be larger, on average, in the presence of more aerosol particles[20–23]. Such correlations have been taken as evidence in support of a strong cloud lifetime effect, whereby changes in cloud forcing as large as $-8\ \mathrm{W\ m^{-2}}$ have been attributed to lifetime effects on ocean-basin scales[22]. Global surveys, on the other hand, suggest somewhat weaker effects, with a radiative forcing of between 0 and $-1\ \mathrm{W\ m^{-2}}$ (refs 20, 24).

Some examples of aerosol-induced microphysical changes suppressing the development of precipitation have also been reported[25,26], but a robust demonstration of aerosol suppression of surface precipitation remains lacking[11].

The correlation–causation conundrum

Two lines of argument challenge the interpretation of satellite-derived correlations between cloud amount and aerosol optical thickness as evidence in support of a lifetime effect. The first contends that aerosol–cloud correlations are prone to measurement artefacts and thus are not reliable. The second contends that to the extent such correlations are reliable, they can be amply explained by alternative (and, as argued below, in many cases simpler) ideas[21].

The artefact argument is germane because to observe correlations between clouds and the aerosol, it must first be possible to distinguish between them. However, this proves difficult, especially at coarse resolution and from space. Although cloud-active aerosol particles differ from cloud droplets thermodynamically, they can be difficult to distinguish radiatively[27,28]. Optically thick aerosol layers can often take on a milky, cloud-like appearance[29] and be interpreted as cloud. Likewise, subvisible cloud layers will by definition be classified as clear sky by human observers. To the extent that situations with high aerosol loading are more likely to admit the false detection of clouds, it is expected that spurious correlations between satellite-based retrievals of cloud amount and ambient aerosol will be found.

Three-dimensional radiative effects can also introduce measurement artefacts. By conditioning clear-sky measurements on the sun–satellite geometry and distance from the nearest cloud, recent work has shown that the scattering of photons from cloud edges can lead to significant overestimates in retrievals of aerosol optical depths in cloud-free pixels as far as 15 km away. Because such effects increase as cloud distance decreases, they can also produce spurious correlations between aerosol optical depth and cloud amount[30].

Deficiencies in the data record, and a poor understanding of what processes regulate the behaviour of cloud regimes, frustrate attempts to attribute real correlations among the aerosol, clouds and precipitation to lifetime effects. Examples of common deficiencies in the data record include poor or absent vertical and temporal resolutions and missing measurements of key variables, such as relative humidity or even precipitation. Vertical resolution is necessary to assess whether or not cloud and aerosol layers are intermingled[31]. Temporal resolution is necessary to assess causality. However, the most advanced (active and multispectral) sensors are mounted on polar-orbiting satellites, which allow return times that range (depending on the sensor footprint) from days to weeks. Humidity regulates both cloud amount and aerosol optical depth and may be a non-trivial source of correlation between the two quantities[23]. Likewise, the absence of precipitation measurements in many records makes interpretation of precipitation-mediated correlations between aerosol and cloud amount ambiguous. A recent field experiment[32], for example, showed (as did others[21]) that the aerosol, relative humidity and cloud amount are positively correlated in regions of

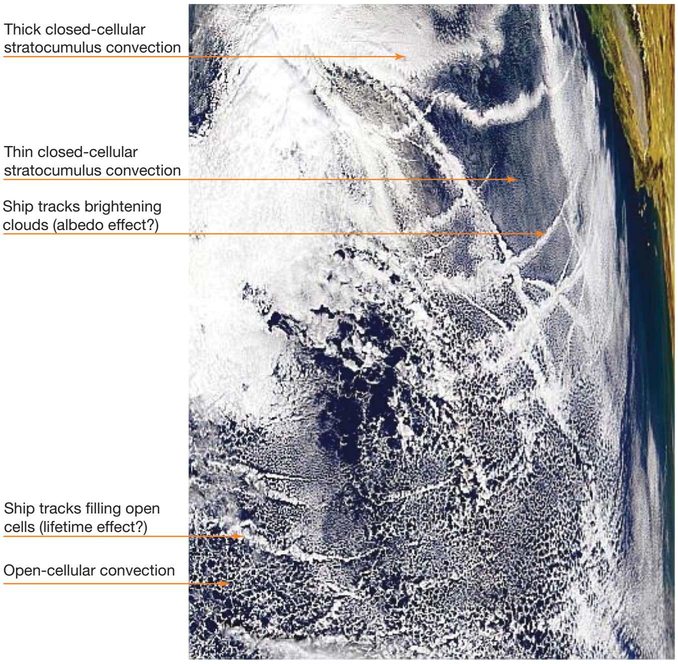

Figure 2 | Satellite image of the northeast Pacific Ocean showing ship tracks, both in thin closed-cellular stratocumulus regions and in open-cellular regions. The southern Californian coastline is visible in the upper-right corner. Open-cellular regions are characterized by clouds in a lace-like network. Closed-cellular regions have a higher albedo and a finer granulation. Recent work has shown regions of open cells to be associated with precipitation and depleted aerosol, although the origins of such features are unclear. At times, closed cells can be embedded in broader regions of open cells, in which case they are referred to as pockets of open cells. Such features, although not clearly evident here, suggest that both states are possible for a given large-scale meteorological environment. Adapted with permission from NASA.

homogeneous trade-wind convection. However, those studies also found that in conditions with higher aerosol optical depth the clouds rain more, not less, and that the ambient low-level winds are stronger. The interpretation is that stronger surface winds favour more surface evaporation and sea spray, and a deeper and more humid cloud layer[21,23,33]. The deeper clouds favour the production of more rain, whereas the more humid layer, combined with the greater sea-spray aerosol, leads to a higher aerosol optical depth.

Many of the above issues are being addressed, but the uncertain relationship between the statistics of the cloud field and the ambient meteorological environment (which has come to be known as the 'cloud problem'[34]) confounds attempts to interpret the data. The cloud problem means that, when it comes to clouds, we do not know what the implications of a small change are and, hence, cannot control for meteorological effects on clouds[35]. This problem is compounded by the tendency of the aerosol to correlate strongly with meteorological conditions[36]. Correlations arise because the meteorology and the aerosol burden depend strongly on the air-mass history[37], because radiative effects of the aerosol can alter the meteorological properties of the air mass in which it is embedded and because cloud processes exert a strong control on the aerosol[38,39].

Global modelling

We now consider what global climate models teach us about the interaction between clouds and the aerosol on large scales. Attempts to quantify cloud-mediated aerosol effects using global models date back more than a decade[40], with estimates of the radiative forcing ranging from near zero to as much as $-4\,\mathrm{W\,m^{-2}}$. Separating lifetime effects from other cloud-mediated aerosol effects is difficult but studies that attempt to do so suggest that lifetime effects alone are responsible for a forcing of between -0.3 and $-1.3\,\mathrm{W\,m^{-2}}$ (ref. 41). Such estimates, however, should not be interpreted as a manifestation of physical constraints, both because processes that may generate positive forcing (usually involving ice) are only beginning to be incorporated[12,41] and because models are tuned to preclude estimates of the cloud-mediated cloud radiative forcing that are too high[42].

By using satellite data to rescale relations that emerge from an ensemble of models several studies have argued for weaker cloud-mediated aerosol effects[43]. Such arguments are consistent with recent (purely observational) estimates that limit the total aerosol forcing to $-1.2\,\mathrm{W\,m^{-2}}$ (ref. 44), which is significantly smaller than previous estimates[45], but they have not advanced our understanding of aerosol–cloud interactions, nor reduced our level of uncertainty.

The main challenge for model-based estimates of cloud lifetime effects is to extract a credible prediction from models whose representations of clouds, and cloud microphysical processes, are known to have serious deficiencies. Regional and global models systematically misrepresent the distribution of clouds, especially shallow maritime clouds. This, again, is the cloud problem, as even after averaging across models that have been tuned to satisfy global energy constraints, systematic regional biases in cloud radiative forcing remain and can be as large as $50\,\mathrm{W\,m^{-2}}$ across planetary (1,000-km) scales. Furthermore, studies so far incorporate only a portion of our understanding of cloud microphysical processes[42,46], and the choice of which physical processes to model often reflects what can practicably be introduced into the model as much as it reflects our understanding of what is physically important[12,42]. For these reasons, and because lifetime effects depend critically on the interplay of uncertainly parameterized physical processes, global model-based estimates of lifetime effects remain controversial, and very likely underestimate our true level of uncertainty[47].

A buffered system

The effect of the anthropogenic aerosol on clouds and precipitation often proves difficult to establish. To find out why this is, considerable effort has been devoted to understanding the chain of events encompassed by lifetime hypotheses (see, for example, Fig. 3). As we review below, evidence continues to mount that couplings necessary to evaluate lifetime effects are sensitive to the details of their representation as well as to the state of the system as mediated by other couplings, which themselves are often poorly understood[48]. This means that changes in the system in isolation may be cancelled, or compensated for, by an opposing change that becomes evident when the system is looked at as a whole.

The tendency for a change in one process in a complex system to be compensated for by the response of another is often described as a negative feedback on the system scale. Formally, however, the concept of feedback implies that the output of a system modifies the input. Because this is not the case in many of the examples we cite, we prefer to speak of buffering, wherein different paths to a specific end buffer the system against disruptions to any particular path. Our use of the term is consistent with its colloquial meaning, as it conveys the sense that the response of a system to a forcing is weaker than would have been expected had internal mechanisms—which absorb the impact of the forcing, and hence buffer the system—not been accounted for.

Microphysical buffers. On the scale of individual cloud droplets (the microphysical scale), buffering is a-priori expected to reduce the strength of the coupling between the aerosol and cloud microstructure on the one hand, and between cloud micro- and macrostructure on the other. Microphysical buffering in the first sense is familiar in terms of the process of activation of aerosol particles to form cloud droplets. It has long been understood that changes in the size distribution or composition of cloud-active aerosol, which in the absence of other effects lead to fewer cloud droplets, also result in locally higher supersaturation. This tends to counter, or partially compensate for, the initial changes by allowing smaller condensation nuclei to be activated[49]. Such effects are evident in the sublinear dependence of cloud droplet concentrations on aerosol number[50–52]. Conversely,

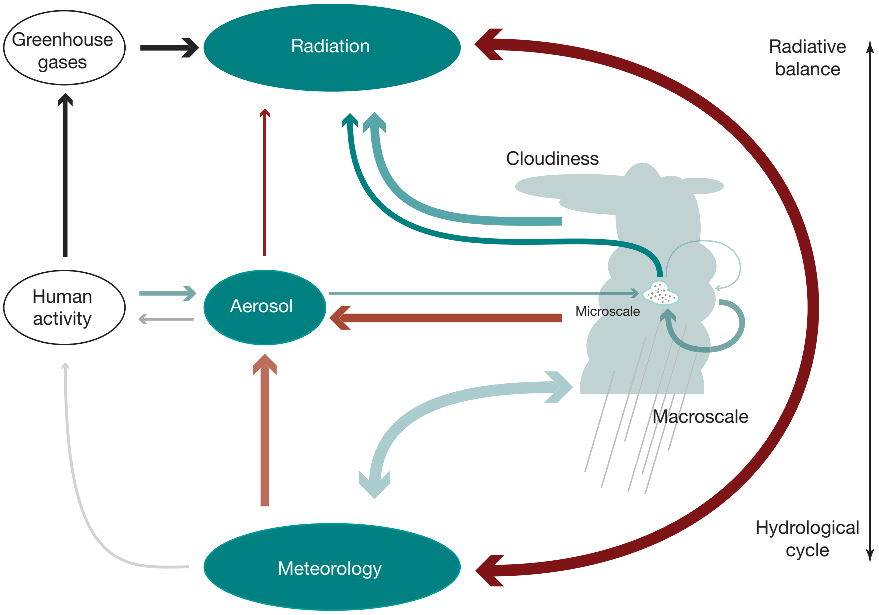

Figure 3 | Aerosol–cloud interactions in the context of the atmospheric component of the Earth system. The response of the system to changes in the aerosol is buffered by, or depends on, the strong (as measured by arrow thickness) and in places very uncertain (as measured by arrow transparency) coupling between clouds (microscale, C_m; macroscale, C_M) and other components of the cloud system (human activity, H; aerosol, A; radiation, R). Unlike the cloud albedo effect, which is expressed in the H–A–C_m–R pathway, cloud lifetime effects follow H–A–C_m–C_M–R and other components of the system. Our discussion is principally concerned with the blue-green component of the system, although interactions in brown-red also help determine the behaviour of the physical system.

higher wind speeds tend to produce more sea-salt aerosol, inhibiting the development of precipitation, while also producing more sea-spray and large cloud condensation nuclei, which efficiently suppress activation of some of the smaller particles[51], seed the coalescence process and help to maintain or even enhance the efficiency of rain production[53].

Recent work also suggests that the precipitation susceptibility of shallow clouds, β (Box 1), which provides an indication of the sensitivity of rain to changes in the cloud droplet number concentration, is less than predicted by simple scaling arguments. These arguments, which form the basis of parameterizations in large-scale models, yield $\beta = 2$ and imply a strong sensitivity of rain to droplet number[54]. In stratocumulus, observations suggest that $\beta = 1$ and, therefore, that the sensitivity of rain to droplet number is weaker[55–57]. Even this value may be an overestimate, as such measurements fail to account for the dependence of the aerosol (and, hence, drop number) on the precipitation, through the precipitation scavenging or washout of the cloud-active aerosol[58]. Indeed, recent analyses of satellite-based cloud radar (NASA's CloudSat mission) data yield yet weaker susceptibilities ($0.1 < \beta < 1.1$)[59]. Idealized modelling studies of cumulus-like drafts, which emphasize the importance of accretion processes (that is, the collection of cloud droplets by precipitation embryos)[60], also result in weaker susceptibilities ($\beta \leq 1/2$)[61].

Box 1 | Precipitation susceptibility

In ice-free clouds, the primary process for the formation of precipitation is through the collision and coalescence of drops falling at different velocities. Faster-falling large drops collect smaller drops in their paths and grow even larger, increasing their chances of surviving the fall to the surface as raindrops. The efficiency of coalescence increases with increasing cloud water content and/or decreasing drop concentration. Thus, aerosol perturbations, which increase drop concentrations, are likely to reduce precipitation. The precipitation susceptibility[44], $\beta = -\mathrm{d}(\ln R)/\mathrm{d}(\ln N_\mathrm{d})$, has been proposed as a measure of how the rain rate, R, depends on the cloud droplet number concentration, N_d. Like so many aspects of such systems, β is probably regime or state dependent. Larger values of β imply a greater sensitivity of rain to changes in N_d. The precipitation susceptibility provides a useful framework within which to identify the regions on Earth and the cloud types that are likely to suffer most from reductions in precipitation resulting from natural and anthropogenic aerosol perturbations. First results[58] suggest that these regions are, as expected, characterized by large aerosol perturbations, but also that they fall into a limited range of cloud conditions. Clouds that have low liquid-water contents generate little precipitation and therefore are not susceptible; clouds that have very high liquid-water contents generate precipitation efficiently, and therefore become increasing less susceptible. Clouds in some intermediate range of water contents are likely to be the most susceptible. Identifying the bounds of these regimes is an area of ongoing research.

The inertial clustering of cloud droplets, which is associated with intense turbulent accelerations within clouds, may also have a role in setting the rate of rain formation in warm clouds[62]. Because the turbulence intensity depends on the cloud macrostructure, such processes may further buffer the system against changes in cloud microstructure due to the changing aerosol.

Macrophysical buffers. Macrophysical (cloud-dynamical) effects may also buffer the response of the system to aerosol perturbations. A number of studies have shown that the flux of liquid water through the cloud layer is critical to the growth rate of the layer[63–66] and, accounting for this process, can produce unexpected results[67]. For cumulus clouds, the mechanism is easy to understand. Less precipitation means more liquid is lofted to the cloud-top region, where it evaporates. The associated cooling destabilizes the environment, making it conducive to the growth of deeper clouds (Fig. 4). However, deeper clouds produce more rain, which can more than compensate for the initial (or upstream) suppression of precipitation[61,66]. In such cases, increases in the aerosol can be expected to produce more rain, not less. Variants of this idea have also been shown to invigorate convection and precipitation in ice clouds (by making more lofted condensate available for freezing)[8,68]. However, for ice-bearing systems the complexity of microphysical processes leads to the possibility of a much richer set of behaviours[69–72].

Effects such as these are not just limited to the interplay between mixing of surface moisture up into the cloud layer and cloud-top evaporation. In cases of weak precipitation, fine-scale simulations

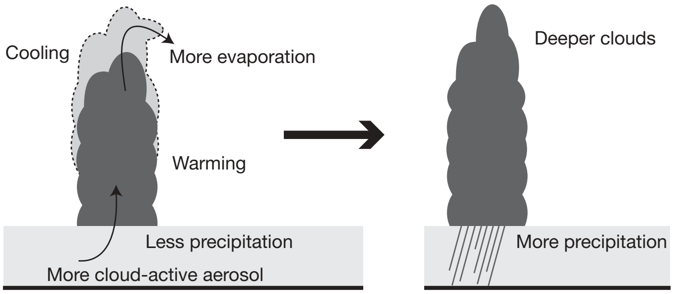

Figure 4 | The deepening effect. The local inhibition of precipitation helps precondition the environment for deeper convection, which then rains more.

that resolve the turbulent dynamics of stratocumulus show that the evaporation of drizzle just below the cloud base can destabilize the subcloud layer and enhance the mixing of moisture into the cloud, thus increasing cloudiness[73]. Related effects are evident in simulations and observations of shallow cumulus, for which studies suggest that an increase in the aerosol increases droplet surface area and, hence, the rate of droplet evaporation as clouds mix with subsaturated environments. Increasing the rate of evaporation reduces cloudiness[74,75].

The tendency of precipitation to reduce the growth (or cloud-top entrainment) rate of the stratocumulus-topped boundary layer on the one hand, and to enhance subcloud mixing on the other, can produce surprising results. Reduced cloud-top entrainment at night (due to precipitation) maintains a shallower, moister boundary layer, and weak precipitation during the day serves to keep the cloud layer strongly coupled to the surface. The net result is that less aerosol promotes more cloud, not less, in contradiction to what would have been expected had cloud dynamics been neglected[76,77]. Processes such as these are probably responsible for satellite observations that run counter to conventional wisdom[78,79].

The idea that precipitation may enhance cloudiness is familiar to students of deep convection. In environments with vertical shear of the horizontal wind, it is well understood that the longevity of cloud systems is intimately tied to the development of precipitation and the pools of cold air that it produces[80]. Recent field work suggests that for shallow convection over the ocean, cloud activity also organizes along the outflow of cold air associated with showers from previous convection, which suggests that here, too, precipitation creates clouds[32,81]. Finally, for weakly precipitating clouds, larger drops have more thermal inertia and thus linger in subsaturated environments, leading to effectively more cloud, not less, in the presence of precipitation[61,82].

Radiative processes, through their ability to change the large-scale thermodynamic environment in which clouds form, may also act to buffer lifetime effects. Because cloud-active aerosol tends to correlate with absorbing aerosol, clouds that form in layers of increased aerosol loading may be suppressed through radiative heating (stabilization) of the cloud layer[83,84]. Perhaps even more effective is that both enhanced scattering and absorption by the aerosol increase the total extinction, thereby acting to cool the surface, further stabilizing the layer and reducing cloudiness[85]. Here again a mechanism emerges whereby an increase in aerosol amount may reduce, rather than enhance, cloudiness, hence reinforcing the concept of a well-buffered system.

This is not to say that lifetime effects are entirely without merit. Pockets of open cells (Fig. 2) provide dramatic support for the conventional wisdom that precipitation reduces cloud amount[13]. Rather, our argument is that the sensitivity of clouds and precipitation to changes in the aerosol is, on average, weaker than implied by simple arguments and is regime or state dependent. This capacity of the system to respond differentially to change in the aerosol buffers the global system, by endowing it with the ability to offset positive responses within some regimes with negative responses in others.

Why it makes sense to keep searching

The possibility of significant, but regime- (or regionally) specific[86,87], responses is one reason to intensify research efforts—even if the diversity of responses and regimes makes it likely that lifetime effects are negligible (or at least not discernible given current approaches) on global scales. Another compelling reason for intensifying our efforts to understand lifetime effects is because doing so can help solve the cloud problem, namely that of relating the statistics of cloud fields to their meteorological environment. This is a vital issue.

Because clouds and precipitation and the effect that the aerosol has on them are almost certainly regime dependent, future work must identify how the trace of such interactions depends on the state of the system, and improve the representation of such cloud regimes in models. Absent some new organizing principle, this means that the most fruitful approach is to work through the issues regime by regime. It remains to be seen just how many regimes carry the cloud, or aerosol–cloud–precipitation, signal and its imprint on the global scale.

Given our present understanding, certain cloud regimes can already be identified as worthy of closer scrutiny, irrespective of whether the interest is in clouds and precipitation or the effects of the aerosol thereon. These include shallow maritime clouds (including trade-wind cumulus and stratocumulus), tropical deep convection over land and stratiform cloud regimes in the polar regions. Trade-wind cumulus are crucial because they prevail over the global ocean, are at the centre of the initial lifetime hypothesis and are known to play an important role both in the current climate and in estimates of climate change. Stratocumulus are important because observations suggest that they carry a pronounced signature of changing aerosol, and because they have been identified by geoengineering proposals[88] as a possible pathway through which one could regulate the planetary albedo. Tropical deep convection over land should be considered both because of its role in the hydrological cycle and because preliminary work suggests a pronounced susceptibility of rainfall to aerosol infusions (which, on the regime scale, can be enormous). Stratiform clouds in the arctic warrant attention because they may mediate interactions over a part of the earth system that appears to be unusually sensitive to the changing climate.

Shifting our emphasis to regime-centred studies also offers methodological advantages. Because of their limited spatial scale, studies of particular regimes are well suited to fine-scale models, which are capable of resolving (rather than parameterizing) the multitude of interactions ranging from the cloud microscale to the cloud macroscale. To the extent that fine-scale models identify (and observations support) a robust sensitivity of a cloud regime to the aerosol, this sensitivity can be incorporated into the parameterizations used by large-scale models. Then, rather than attempting to mediate among fine (or subgrid) scale processes for which information is always lacking, large-scale models can focus on aspects of the problem to which they are well suited, that is, exploring the interplay between a robustly parameterized effect of the aerosol on clouds and the nature of the large- (or resolved-) scale circulation in which it is embedded.

Localization of the problem also makes it more straightforward to augment and enrich current[17] and future space-based measurements[89,90] The most effective way of doing so would be through the deployment of arrays of ground-based remote sensors that can both vertically and temporally resolve the aerosol, clouds, precipitation and the meteorological state. Finally, regime-centred studies also offer the opportunity to take advantage of novel experimental strategies. One possibility is through the use of natural experiments, accidental or otherwise (for example large-scale biomass burnings or aerosol infusions from ships). Other possibilities include the development of new observational platforms, for instance high-altitude airships capable of deploying heavy payloads (comprising, perhaps, active and hyperspectral remote sensors such as those now only deployed on polar-orbiting satellites) to geostationary positions in the lower stratosphere for periods of months[91].

Elements of the above approach are evident in some recent initiatives[32,92], although in most cases they favour the study of processes operative on timescales of minutes to hours, in slowly changing large-scale environments. Comprehensive data sets capable of documenting the behaviour of cloud regimes on timescales of days to seasons[93] are still relatively rare. However, it is precisely data of this kind that are necessary if we are to develop our understanding of the character of regimes as a whole, and over a broad range of environmental conditions. Thus, if we wish to make significant strides in understanding the interplay among the aerosol, clouds and precipitation, we consider it imperative to launch significant new international initiatives, with comprehensive, coordinated and enduring measurements, targeting specific regimes and coupled to state-of-the-art modelling.

1. Squires, P. The microstructure and colloidal stability of warm clouds. I. The relation between structure and stability. *Tellus* **10,** 256–271 (1958).
   This paper shows that clouds that form in clean marine air are more apt to precipitate than clouds forming in air containing a high aerosol burden.
2. Liou, K.-N. & Ou, S.-C. The role of cloud microphysical processes in climate: an assessment from a one-dimensional perspective. *J. Geophys. Res.* **94,** 8599–8607 (1989).
3. Albrecht, B. A. Aerosols, cloud microphysics and fractional cloudiness. *Science* **245,** 1227–1230 (1989).
   This study postulates that by suppressing precipitation, the aerosol might increase cloud lifetime and thus enhance radiative forcing.
4. Pincus, R. & Baker, M. B. Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer. *Nature* **372,** 250–252 (1994).
5. Hartmann, D. L. & Doelling, D. On the net radiative effectiveness of clouds. *J. Geophys. Res.* **96,** 869–891 (1980).
6. Tiedtke, M. A comprehensive mass flux scheme for cumulus parameterization in large-scale models. *Mon. Weath. Rev.* **117,** 1779–1800 (1989).
7. Bony, S. & Dufresne, J.-L. Marine boundary layer clouds at the heart of tropical cloud feedback uncertainties in climate models. *Geophys. Res. Lett.* **32,** L20806 (2006).
   This paper shows that the sensitivity of shallow marine clouds to changing environmental conditions is the main source of uncertainty in tropical cloud feedbacks simulated by climate models.
8. Rosenfeld, D. *et al.* Flood or drought: how do aerosols affect precipitation? *Science* **321,** 1309–1313 (2008).
9. Jiang, H., Xue, H., Teller, A., Feingold, G. & Levin, Z. Aerosol effects on the lifetime of shallow cumulus. *Geophys. Res. Lett.* **33,** doi:1029/2006GL026024 (2006).
10. Twomey, S. The influence of pollution on the shortwave albedo of clouds. *J. Atmos. Sci.* **34,** 1149–1152 (1977).
    This was the first study to point out the potential for the aerosol to brighten clouds, leading to the concept of cloud-mediated (or indirect) aerosol effects.
11. Levin, Z. & Cotton, W. (eds) *Aerosol Pollution Impact on Precipitation* (Springer, 2008).
    This book provides a comprehensive review of aerosol effects on clouds and precipitation.
12. Storelvmo, T. *et al.* Modeling the Wegner-Bergeron-Findeisen process—implications for aerosol indirect effects. *Environ. Res. Lett.* **3,** 045001 (2008).
13. Stevens, B. *et al.* Pockets of open cells and drizzle in marine stratocumulus. *Bull. Am. Meteorol. Soc.* **86,** 51–57 (2005).
14. Radke, L. F., Coakley, J. A. Jr & King, M. D. Direct and remote sensing observations of the effects of ships on clouds. *J. Appl. Meteorol.* **246,** 1146–1149 (1989).
15. Comstock, K. K., Bretherton, C. S. & Yuter, S. E. Mesoscale variability and drizzle in southeast Pacific stratocumulus. *J. Atmos. Sci.* **62,** 3792–3807 (2005).
16. Sharon, T. M. *et al.* Aerosol and cloud microphysical characteristics of rifts and gradients in maritime stratocumulus clouds. *J. Atmos. Sci.* **63,** 983–997 (2006).
17. Stephens, G. *et al.* The CloudSat mission and the A-TRAIN: a new dimension to space-based observations of clouds and precipitation. *Bull. Am. Meteorol. Soc.* **83,** 1771–1790 (2002).
18. Coakley, J. A., Bernstein, L. & Durkee, A. Effect of ship-stack effluents on cloud reflectivity. *Science* **237,** 1020–1022 (1987).
19. Han, Q., Rossow, W. B. & Lacis, A. A. Near-global survey of effective droplet radii in liquid water clouds using ISCCP data. *J. Clim.* **7,** 465–497 (1994).
20. Sekiguchi, M. *et al.* A study of the direct and indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters. *J. Geophys. Res.* **108,** doi:10.1029/2002JD003359 (2003).
21. Loeb, N. G. & Schuster, G. L. An observational study of the relationship between cloud, aerosol and meteorology in broken low-level cloud conditions. *J. Geophys. Res.* **113,** D14214 (2008).
    The introduction to this study provides a salient overview of many of the challenges of using satellite observations to relate clouds to the aerosol.
22. Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D. & Rudich, Y. The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean. *Proc. Natl Acad. Sci. USA* **102,** 11207–11212 (2005).
23. Matheson, M. A., Coakley, J. A. & Tahnk, W. R. Aerosol and cloud property relationships for summertime stratiform clouds in the northeastern Atlantic from Advanced Very High Resolution Radiometer observations. *J. Geophys. Res.* **110,** D24204 (2005).
24. Nakajima, T., Higurashi, A., Kawamoto, K. & Penner, J. E. A possible correlation between satellite-derived cloud and aerosol microphysical parameters. *Geophys. Res. Lett.* **28,** doi:10.1029/2000GL012186 (2001).
25. Rosenfeld, D. TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall. *Geophys. Res. Lett.* **26,** 3105–3108 (1999).
26. Andreae, M. O. *et al.* Smoking rain clouds over the Amazon. *Science* **303,** 1337–1342 (2004).
27. Charlson, R., Ackerman, A., Bender, F.-M., Anderson, T. & Liu, Z. On the climate forcing consequences of the albedo continuum between cloudy and clear air. *Tellus B* **59,** doi:10.1111/j.1600–0889.2007.00297.x (2007).
28. Koren, I., Remer, L. A., Kaufman, Y. J., Rudich, Y. & Martins, J. V. On the twilight zone between clouds and aerosols. *Geophys. Res. Lett.* **34,** L08805 (2007).
29. Anderson, J. B. Observations from airplanes of cloud and fog conditions along the southern Californian coast. *Mon. Weath. Rev.* **59,** 264–270 (1931).
30. Várnai, T. & Marshak, A. MODIS observations of enhanced clear sky reflectance near clouds. *Geophys. Res. Lett.* **36,** L06807 (2009).
    This study shows that three-dimensional radiative interactions between clouds and their surroundings extend up to 15 km beyond the boundaries of the clouds.
31. Avey, L., Garrett, T. J. & Stohl, A. Evaluation of the aerosol indirect effect using satellite, tracer transport model, and aircraft data from the International Consortium for Atmospheric Research on Transport and Transformation. *J. Geophys. Res.* **112,** doi:10.1029/2006JD007581 (2007).
32. Rauber, R. *et al.* Rain in (shallow) cumulus over the ocean–the RICO campaign. *Bull. Am. Meteorol. Soc.* **88,** 1912–1928 (2007).
33. Nuijens, L., Stevens, B. & Siebesma, A. P. The environment of precipitating shallow cumulus convection. *J. Atmos. Sci.* **66,** 1962–1969 (2009).
34. Arakawa, A. in *The Physical Basis of Climate and Climate Modelling* 181–197 (GARP Publ. Ser. 16, ICSU/WMO, 1975).
    This prescient study was perhaps the first to appreciate the singular problem clouds pose for the modelling of the general circulation of the atmosphere.
35. Stevens, B. & Brenguier, J.-L. in *Clouds in the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics and Precipitation* (eds Heintzenberg, J. & Charlson, R. J.) Ch. 8 (MIT Press, 2009).
36. Brenguier, J. L., Pawlowska, H. & Schüller, L. J. Cloud microphysical and radiative properties for parameterization and satellite monitoring of the indirect effect of aerosol on climate. *J. Geophys. Res.* **108,** doi:10.1029/2002JD002682 (2003).
37. Mauger, G. S. & Norris, J. R. Meteorological bias in satellite estimates of aerosol-cloud relationships. *Geophys. Res. Lett.* **34,** doi:10.1029/2007GL029952 (2007).
38. Hoppel, W. A., Fitzgerald, J. W., Frick, G. M., Larson, R. E. & Mack, E. J. Aerosol size distributions and optical properties found in the marine boundary layer over the Atlantic Ocean. *J. Geophys. Res.* **95,** 3659–3686 (1990).
39. Hegg, D., Majeed, R., Yuen, P., Baker, M. & Larson, T. The impacts of SO_2 oxidation in cloud drops and in haze particles on aerosol light scattering and CCN activity. *Geophys. Res. Lett.* **23,** 2613–2616 (1996).
40. Boucher, O. & Lohmann, U. The sulfate-CCN-cloud albedo effect: a sensitivity study with two general circulation models. *Tellus B* **47,** 281–300 (1995).
41. Hoose, C., Kristjánsson, J. E., Kirkevåg, A., Seland, Ø. & Gettelman, A. Constraining cloud drop number concentration in GCMs suppresses the aerosol indirect effect. *Geophys. Res. Lett.* **36,** doi:10.1029/2009GL038568 (2009).
42. Lohmann, U. & Feichter, J. Global indirect aerosol effects: a review. *Atmos. Chem. Phys.* **5,** 715–737 (2005).
    This paper provides a comprehensive overview of aerosol–cloud interactions (indirect effects) in global climate models and suggest required improvements.
43. Quaas, J. *et al.* Aerosol indirect effects — general circulation model intercomparison and evaluation with satellite data. *Atmos. Chem. Phys. Discuss.* **9,** 12731–12779 (2009).
44. Murphy, D. M. *et al.* An observationally based energy balance for the Earth since 1950. *J. Geophys. Res.*. (in the press).
45. Knutti, R., Stocker, T. F., Joos, F. & Plattner, G.-K. Constraints on radiative forcing and future climate change from observations and climate model ensembles. *Nature* **416,** 719–723 (2002).
46. Rotstayn, L. D. Indirect forcing by anthropogenic aerosols: a global climate model calculation of the effective-radius and cloud-lifetime effects. *J. Geophys. Res.* **104,** 9369–9380 (1999).
47. Lohmann, U. & Feichter, J. Impact of sulfate aerosols on albedo and lifetime of clouds: a sensitivity study with the ECHAM GCM. *J. Geophys. Res.* **102,** 13685–13700 (1997).
48. Feingold, G. & Siebert, H. in *Clouds in the Perturbed Climate System: Their Relationship to Energy Balance, Atmospheric Dynamics and Precipitation*, (eds Heintzenberg, J. & Charlson, R. J.) Ch. 14 (MIT Press, 2009).
49. Twomey, S. A. The nuclei of natural cloud formation, part II: the supersaturation in natural clouds and the variation of cloud droplet concentration. *Pure Appl. Geophys.* **43,** 243–249 (1959).
50. Martin, G. M., Johnson, D. & Spice, A. The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds. *J. Atmos. Sci.* **51,** 1823–1842 (1994).
51. O'Dowd, C. D., Lowe, J. A., Smith, M. H. & Kaye, A. D. The relative importance of non-sea sulphate and sea-salt aerosol to the marine cloud condensation nuclei

population: an improved multi-component aerosol-cloud droplet parameterization. *Q. J. R. Meteorol. Soc.* **125,** 1295–1313 (1999).
52. Ramanathan, V., Crutzen, P., Kiehl, J. & Rosenfeld, D. Aerosols, climate and the hydrological cycle. *Science* **294,** 2119–2124 (2001).
53. Johnson, D. B. The role of giant and ultragiant aerosol particles in warm rain initiation. *J. Atmos. Sci.* **39,** 448–460 (1982).
54. Seifert, A. & Beheng, K. D. A double-moment parameterization for simulating autoconversion, accretion and self collection. *Atmos. Res.* **59–60,** 265–281 (2001).
55. Pawlowska, H. & Brenguier, J.-L. An observational study of drizzle formation in stratocumulus clouds for general circulation model (GCM) parameterization. *J. Geophys. Res.* **33,** L19810 (2003).
56. Comstock, K. K., Wood, R., Yuter, S. E. & Bretherton, C. S. Reflectivity and rain rate in and below drizzling stratocumulus. *Q. J. R. Meteorol. Soc.* **130,** 2891–2918 (2004).
57. vanZanten, M., Stevens, B., Vali, G. & Lenschow, D. Observations of drizzle in nocturnal marine stratocumulus. *J. Atmos. Sci.* **62,** 88–106 (2005).
58. Petters, M. D. *et al.* Accumulation mode aerosol, pockets of open cells and particle nucleation in the remote subtropical pacific marine boundary layer. *J. Geophys. Res.* **111,** D02206 (2005).
59. Sorooshian, A., Feingold, G., Lebsock, M. D., Jiang, H. & Stephens, G. L. On the precipitation susceptibility of clouds to aerosol perturbations. *Geophys. Res. Lett.* **36,** doi:10.1029/2009GL038993 (2009).
60. Wood, R. Rate of loss of cloud droplets by coalescence in warm clouds. *J. Geophys. Res.* **111,** D21205 (2006).
61. Stevens, B. & Seifert, A. Understanding the macrophysical outcomes of microphysical choices in simulations of shallow cumulus convection. *J. Meteorol. Soc. Jpn* **86A,** 141–163 (2008).
62. Ayala, O., Rosa, B., Wang, L.-P. & Grabowski, W. Effects of turbulence on the geometric collision rate of sedimenting droplets. Part 1. Results from direct numerical simulation. *N. J. Phys.* **10,** 075015 (2008).
63. Stevens, B., Cotton, W. R., Feingold, G. & Moeng, C.-H. Large-eddy simulations of strongly precipitating, shallow, stratocumulus-topped boundary layers. *J. Atmos. Sci.* **55,** 3616–3638 (1998).
64. Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E. & Toon, O. B. The impact of humidity above stratiform clouds on indirect aerosol climate forcing. *Nature* **432,** 1014–1017 (2004).
65. Bretherton, C., Blossey, P. & Uchida, J. Cloud droplet sedimentation, entrainment efficiency and subtropical stratocumulus albedo. *Geophys. Res. Lett.* **34,** doi:10.1029/2006GL027648 (2007).
66. Stevens, B. On the growth of layers of non-precipitating cumulus convection. *J. Atmos. Sci.* **64,** 2916–2931 (2007).
67. Wood, R. Cancellation of aerosol indirect effects in marine stratocumulus through cloud thinning. *J. Atmos. Sci.* **64,** 2657–2669 (2007).
68. Koren, I., Kaufman, Y. J., Rosenfeld, D., Remer, L. A. & Rudich, Y. Aerosol invigoration and restructuring of Atlantic convective clouds. *Geophys. Res. Lett.* **32,** L14828 (2005).
69. Tao, W.-K. Cloud resolving modeling. *J. Meteorol. Soc. Jpn* **85B,** 305–330 (2007).
70. Khain, A. P., BenMoshe, N. & Pokrovsky, A. Factors determining the impact of aerosols on surface precipitation from clouds: an attempt at classification. *J. Atmos. Sci.* **65,** 1721–1748 (2008).
71. Kay, J. E. & Wood, R. Timescale analysis of aerosol sensitivity during homogeneous freezing and implications for upper tropospheric water vapor budgets. *Geophys. Res. Lett.* **35,** L10809 (2008).
72. Lee, S. S., Donner, L. J., Phillips, V. T. J. & Ming, Y. Examination of aerosol effects on precipitation in deep convective clouds during the 1997 ARM summer experiment. *Q. J. R. Meteorol. Soc.* **134,** 1201–1220 (2008).
73. Feingold, G., Stevens, B., Cotton, W. R. & Frisch, A. S. On the relationship between drop in-cloud residence time and drizzle production in stratocumulus clouds. *J. Atmos. Sci.* **53,** 1108–1122 (1996).
74. Wang, S., Wang, Q. & Feingold, G. Turbulence, condensation and liquid water transport in numerically simulated nonprecipitating stratocumulus clouds. *J. Atmos. Sci.* **60,** 262–278 (2003).
75. Small, J. D., Chuang, P. Y., Feingold, G. & Jiang, H. Can aerosol increase cloud lifetime? *Geophys. Res. Lett.*. (in the press).
76. Lu, M.-L. & Seinfeld, J. Study of the aerosol indirect effect by large-eddy simulation of marine stratocumulus. *J. Atmos. Sci.* **62,** 3909–3932 (2005).
77. Sandu, I., Brenguier, J.-L. & Geoffroy, O. Aerosol impacts on the diurnal cycle of marine stratocumulus. *J. Atmos. Sci.* **65,** 2705–2718 (2008).
78. Han, Q., Rossow, W. B., Zeng, J. & Welch, R. Three different behaviors of liquid water path of water clouds in aerosol-cloud interactions. *J. Atmos. Sci.* **59,** 726–735 (2002).
79. Matsui, T. *et al.* Satellite-based assessment of marine low-cloud variability associated with aerosol, atmospheric stability and the diurnal cycle. *J. Geophys. Res.* **111,** doi:10.1029/2005JD006097 (2006).
80. Houze, R. Jr. Mesoscale convective systems. *Rev. Geophys.* **42,** doi:10.1029/2004RG000150 (2004).
81. Xue, H., Feingold, G. & Stevens, B. Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection. *J. Atmos. Sci.* **65,** 392–406 (2008).
82. Xue, H. & Feingold, G. Large eddy simulations of tradewind cumuli: investigation of aerosol indirect effects. *J. Atmos. Sci.* **63,** 1605–1622 (2006).
83. Ackerman, A. S. *et al.* Reduction of tropical cloudiness by soot. *Science* **288,** 1042–1047 (2000).
84. Koren, I., Kaufman, Y. J., Remer, L. A. & Martins, J. V. Measurement of the effect of Amazon smoke on inhibition of cloud formation. *Science* **303,** 1342–1345 (2004).
85. Feingold, G., Jiang, H. & Harrington, J. Y. On smoke suppression of clouds in Amazonia. *Geophys. Res. Lett.* **32,** doi:10.1029/2004GL021369 (2005).
86. Lau, K.-M., Kim, M.-K. & Kim, K.-M. Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau. *Clim. Dyn.* **26,** 855–864 (2006).
87. Baker, M. & Charlson, R. J. Bistability of CCN concentrations and thermodynamics in the cloud-topped boundary layer. *Nature* **345,** 142–145 (1990).
This paper postulated the existence of two stable states for shallow clouds that have since been verified by observation.
88. Latham, J. *et al.* Global temperature stabilization via controlled albedo enhancement of low-level maritime clouds. *Phil. Trans. R. Soc. A* **366,** 3969–3987 (2008).
89. Heliere, A., Lefebvre, A., Wehr, T., Bezy, J.-L. & Durand, Y. in *Proc. Geosci. Remote Sensing Symp. 2007* 4975–4978 (IEEE, 2007).
90. Henson, R. (ed.) *Satellite Observations to Benefit Science and Society: Recommended Missions for the Next Decade Committee on Earth Science and Applications from Space: A Community Assessment and Strategy for the Future* 10 (US National Academies Press, 2008).
91. Jamison, L., Sommer, G. & Porche, I. R. III. *High-Altitude Airships for the Future Force Army*. Tech. Report TR-234 (RAND Corporation, 2005).
92. Wood, R., Mechoso, C., Bretherton, C., Huebert, B. & Weller, R. The VAMOS ocean-cloud-atmosphere-land study (VOCAL). *U.S. CLIVAR Var.* **5,** 1–5 (2007).
93. McComiskey, A. M. *et al.* An assessment of aerosol-cloud interactions in marine stratus clouds based on surface remote sensing. *J. Geophys. Res.* **114,** doi:10.1029/2008JD011006 (2009).

Acknowledgements We thank M. O. Andreae, T. Garrett, J. Marotzke, L. Nuijens, R. Pincus, I. Sandu, A. Seifert and R. Wood for comments on an earlier draft of this manuscript. We also wish to acknowledge contributions to this document arising through the course of many discussions with members of the Aerosol, Clouds, Precipitation and Climate Initiative planning team.

Author Contributions The ideas presented here were developed jointly by B.S. and G.F., and both authors participated actively in the writing of the manuscript and the drafting of the figures.

Author Information Reprints and permissions information is available at www.nature.com/reprints. Correspondence should be addressed to B.S. (bjorn.stevens@zmaw.de).