

independently simulated Jurassic and Cretaceous  $p\text{CO}_2$  levels using a model based on the  $^{87}\text{Sr}/^{86}\text{Sr}$  record that accounts for the effect of weathering of young volcanic rocks on the strontium records. This also results in lower than expected  $p\text{CO}_2$  values during the late Jurassic and early Cretaceous periods, in agreement with earlier modelling efforts<sup>10,11</sup>. Of the existing  $p\text{CO}_2$  records, those from liverwort most closely match the simulation, although a reconstruction of  $\text{CO}_2$  from the pores of fossil leaves also yields largely similar results<sup>5</sup>.

The liverwort  $\text{CO}_2$  reconstruction, when converted into temperature, is broadly consistent with the carbonate-based surface temperatures that had sparked earlier debate<sup>5</sup>, and it shows a positive correlation with surface temperatures derived from pH-corrected  $\delta^{18}\text{O}$  values measured in marine carbonate fossils<sup>4,9</sup> (Fig. 1). This correlation might be fortuitous, or it may suggest that ancient  $p\text{CO}_2$  records are better preserved in liverworts than in other archives such as fossilized soils and leaves that have previously been used to reconstruct atmospheric compositions<sup>3</sup>.

Carbon isotope reconstructions from liverworts yield much lower  $p\text{CO}_2$  values and lower surface temperatures than

from fossilized soils over the Jurassic to Cretaceous periods. If they prove to be a reliable archive for ancient  $p\text{CO}_2$  values, the new results raise questions about the validity of the previously used proxies.

Deviations between reconstructions from liverwort and fossil soils could, however, be due to rapid changes in  $p\text{CO}_2$  that are as yet beyond the temporal resolution of the records. For example, the Jurassic and Cretaceous periods could have generally experienced a warm greenhouse climate that was punctuated by multiple discrete cooling events, probably accompanied by rapid changes in  $p\text{CO}_2$  (ref. 3). More liverwort data need to be generated to cross-check the new method with other more established proxies.

Fletcher and co-workers' reconstructions suggest that the Earth's long-term climate history is indeed controlled by atmospheric  $\text{CO}_2$  concentrations. However, the various proxies for both  $\text{CO}_2$  levels and temperatures over million-year timescales do not yet provide an unambiguous picture, leaving models of the global carbon cycle poorly constrained. This is not necessarily surprising; after all, even the modern global carbon cycle is not fully understood. Important feedbacks that may

have far reaching consequences for our understanding of past and future climate change<sup>12</sup> are still being discovered.

It is important for geoscientists to continue to improve proxies and models that help reconstruct the ancient global carbon cycle, while taking into consideration the results of those working on modern carbon cycling and future climate change.

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## ATMOSPHERIC SCIENCE

# Black carbon and brown clouds

In aerosol hot spots around the globe, solar radiation is dimmed down on its way to the Earth's surface. The resulting surface cooling turns out to be almost in balance with heating of the atmosphere due to black carbon.

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**H**ow atmospheric particles — generally termed aerosols — affect air temperatures depends on altitude: they reflect some of the incoming solar radiation back to space, cooling the Earth's surface, and at the same time absorb some of the sun's energy, heating the atmosphere around them. But the magnitude of aerosol energy absorption on the global scale and its contribution to global warming are uncertain. Two related papers by Ramanathan *et al.*<sup>1</sup> and Stone *et al.*<sup>2</sup> tackle the sources of that uncertainty. The first study pulls together a global suite of data and simulations of the

atmosphere to characterize the extent and distribution of atmospheric brown clouds, which are areas of high aerosol concentration. The second paper concentrates on one specific site in the Indian Ocean, seeking to explain the origin of the absorbing aerosols in the brown clouds there.

Black carbon, typically emitted from combustion processes, is the main light-absorbing component of aerosols. The radiative properties of individual aerosol particles depend on the extent to which black carbon is mixed with primarily scattering aerosol components, such as sulphates and organics<sup>3</sup>. The amount of light absorbed by each particle is measured by its single scattering albedo — the ratio between the light extinction due to scattering alone and the total light extinction from both scattering

and absorption. If the single scattering albedo lies below a critical value, the combined aerosol–Earth system reflects less energy back to space than the Earth's surface alone, leading to a net warming of the Earth. But this critical single scattering albedo depends strongly on the Earth's local albedo (that is, its reflectivity) — the more reflective the background to the aerosol, the higher the critical value. Optically thick clouds below an aerosol layer effectively serve as a highly reflective background<sup>4</sup>, so absorption by aerosols is highly sensitive to the vertical distribution of particles relative to clouds.

The effect of anthropogenic aerosols on the global energy budget can be estimated by comparing two climate model simulations — one for present-day and one for pre-industrial conditions — using

prescribed aerosol emissions. A recent study<sup>5</sup> comparing predictions of nine different global climate models resulted in an average estimate of anthropogenic aerosol radiative forcing at the top of the atmosphere of  $-0.22 \text{ W m}^{-2}$ . This number reflects the sum of surface cooling and atmospheric heating. However, individual simulations gave values ranging from  $0.04$  to  $-0.41 \text{ W m}^{-2}$ . Differences between the predictions of these models reflect, in part, the uncertainty of aerosol sources. Averaged over the nine models, the simulated increase in heating between pre-industrial and present-day conditions within the atmosphere is  $0.82 \text{ W m}^{-2}$ . To put these numbers in context, the anthropogenic radiative forcing of carbon dioxide in 2005 was  $1.66 \text{ W m}^{-2}$  (ref 6). There is little disagreement that the rise of aerosols since pre-industrial times has led to both a substantial reduction in solar radiation at the surface and increased solar heating of the atmosphere itself. But global models disagree as to the magnitude of these effects.

Ramanathan *et al.*<sup>1</sup> use a range of satellite observations and surface measurements, together with models simulating radiation and atmospheric transport, to characterize brown clouds and their radiative effects worldwide. Radiative effects of aerosols can be expressed in terms of the aerosol optical depth, which is a measure of the amount of light airborne particles prevent from passing through the atmosphere. An optical depth of less than 0.1 corresponds to a crystal clear sky, whereas at an optical depth of 1, 37% of the sunlight is prevented from reaching the surface. Ramanathan *et al.* define regional hot spots as areas that fulfil two criteria: an anthropogenic aerosol optical depth larger than 0.3, and an absorption aerosol optical depth — that is, light extinction due to absorption only — exceeding 0.03. Such conditions require concentrations of anthropogenic aerosols that are 3 to 4 times greater than the hemispheric average. According to these criteria, east Asia, the Indo-Gangetic Plains in south Asia, the Indonesian region, southern Africa and the Amazon Basin emerge as hotspots.

Like all aerosols, black carbon scatters a portion of an incoming solar beam back to space, which leads to a reduction in solar radiation reaching the Earth's surface. A portion of the incoming solar beam is also absorbed by the black carbon. The black carbon absorbs radiation yet again from the diffuse upward beam of scattered sunlight, reducing the solar radiation reflected back to space. (This effect is particularly accentuated when absorbing aerosols lie above clouds.) With a sufficiently absorbing aerosol, the cooling caused by radiation scattering back to space can be compensated for by the heating



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**Figure 1** Megacity aerosols. Los Angeles smog layer on 29 January 2004. The satellite-derived annual mean aerosol optical depth over Los Angeles from 2001–2003 was about 0.3, classifying the city as a regional aerosol hot spot. Los Angeles was one of 26 megacities evaluated by Ramanathan and colleagues<sup>1</sup>.

caused by absorbed radiation, leading to no net change in the radiative balance at the top of the atmosphere. Over the hot spot regions, aerosol pollution decreases the annual mean surface radiation by  $10 \text{ W m}^{-2}$  or more, but this is accompanied by a comparable amount of atmospheric aerosol solar heating through absorption.

Of 26 megacities evaluated by Ramanathan *et al.*, 13 were identified as hot spots (Fig. 1), where the reduction in annual mean solar radiation at the surface is in the range of 20 to  $60 \text{ W m}^{-2}$  and is again accompanied by substantial solar heating of the atmosphere. In these areas, surface dimming is nearly balanced by solar heating of the atmosphere by absorbing aerosols. Moreover, atmospheric solar heating from aerosols roughly equals that from greenhouse gases in these regions.

Stone *et al.*<sup>2</sup> investigate the source of aerosol black carbon in the Indian Ocean brown clouds through surface measurements at two sites in the Maldives, located in the northern Indian Ocean. Using a combination of molecular markers and detailed organic speciation, they determine that biofuel combustion and fossil-fuel burning are the main sources of the black carbon, although establishing the relative contributions of these two sources proved to be difficult. The importance of biofuel combustion in this region, as opposed to biomass burning, is a significant finding. Extensive source profiling for south and southeast Asian biofuels is still needed in order to gain a more quantitative breakdown of the black carbon sources.

Observations of recent climatic change in the Asian region include a tendency

towards increased summer floods in south China, increased drought in north China, moderate cooling in China and India, and decreased surface evaporation and summer monsoon rainfall over India<sup>7,8</sup>. Moreover, an increase in atmospheric stability — which implies relative cooling of the surface compared with higher in the atmosphere — and a decrease in the sea surface temperature gradients of the Indian Ocean have been documented<sup>8</sup>. Estimates of the climatic impact of the widespread layers of aerosols over south Asia and the Indian Ocean generally predict similar effects<sup>7–9</sup>. Future work is needed to investigate possible climatic effects of regional brown clouds in other areas of the earth.

It has been suggested that reducing black carbon emissions could be a strategy for counteracting global warming<sup>10,11</sup>. Accurate knowledge of such emissions is therefore essential in assessing the potential effectiveness of this approach for buffering climate change.

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