

# Uncertainty in climate sensitivity: Causes, consequences, challenges

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Fossil fuels supply about 85% of the world's primary energy, and future use would not appear limited by availability of reserves, especially of coal. Rather, future use of fossil fuels will likely be limited by controls on the emission of carbon dioxide into the atmosphere that are agreed to by the nations of the world. The increase in atmospheric CO<sub>2</sub> over the past 200 years, mainly from fossil fuel combustion, is confidently thought to have increased global temperatures and induced other changes in Earth's climate, with the prospect of much more severe consequences from projected future emissions. Limiting such changes in Earth's climate would place major constraints on the combustion of fossil fuels and/or the emission of CO<sub>2</sub> into the atmosphere. Developing effective and cost-effective strategies for limiting CO<sub>2</sub> emissions requires the confident ability to project the changes in climate that would result from a given increase in atmospheric CO<sub>2</sub>. However, even the change in global mean surface temperature (GMST), the single most important index of climate change, that would result from a given increase in atmospheric CO<sub>2</sub> remains uncertain to a factor of 2 or more, largely because of uncertainty in Earth's climate sensitivity, the change in GMST per change in radiative flux. This uncertainty in climate sensitivity, which gives rise to a comparable uncertainty in the shared global resource of the amount of fossil fuel that can be burned consonant with a given increase in global mean surface temperature, greatly limits the ability to effectively formulate strategies to limit climate change while meeting the world's future energy requirements. Key limits on determining climate sensitivity are the small change in downwelling longwave irradiance, less than one percent, that would give rise to changes in climate that reach the level of concern, the complexity of cloud processes and the difficulty of representing them in climate models, and limited understanding of the processes that control the radiative influences of atmospheric aerosols. A recent empirical calculation of Earth's climate sensitivity as the quotient of the relaxation time constant of GMST upon the effective heat capacity characterizing climate change on the multidecadal time scale points to a possible alternative approach to determining Earth's climate sensitivity. While improved knowledge of Earth's climate sensitivity is essential to development of optimal energy strategies, even for climate sensitivity at the low end of the range of present estimates, substantial reductions in CO<sub>2</sub> emissions from their present values would be required to avert dangerous anthropogenic interference with the climate system that would otherwise occur well before the end of the present century.

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## Introduction

Earth's present temperate climate depends on the presence in the atmosphere of infrared-active gases which absorb thermal infrared radiation emitted from Earth's surface and re-emit much of this radiation in the downward direction, thereby increasing surface temperature over that which would obtain in the absence of these gases. This phenomenon, which is commonly denoted the greenhouse effect, is a well understood feature of Earth's climate system. It is well established also that the amount of carbon dioxide in Earth's atmosphere has increased over the past 250 years and is continuing to increase, in large part as a consequence of fossil fuel combustion for energy production. It is widely accepted, on the basis of theoretical understanding and much observational evidence, that Earth's climate has changed as a consequence of increases in CO<sub>2</sub> and other atmospheric constituents over the industrial period, and that continued climate change may be expected in the future as a consequence of future emissions. A comprehensive review of research pertinent to climate change is provided by the several recent (2007) assessment reports<sup>1-4</sup> of the Intergovernmental Panel on Climate

Change (IPCC), which organization was awarded the Nobel Peace Prize in 2007 (along with former US Vice-President Al Gore) for its work in documenting climate change, the science that has been conducted to characterize climate change and to understand the controlling processes, and approaches to adapt to and mitigate climate change. Despite extensive research, however, major uncertainties remain in the quantitative relations between changes in atmospheric composition and climate change.

Climate change and its relation to controlling factors such as CO<sub>2</sub> are of great consequence to the inhabitants of Earth and their governments in large part because of the intrinsic connection between increased amounts of atmospheric CO<sub>2</sub> and production of energy from combustion of fossil fuels. Thus planning for the ways in which the nations of the world will meet their energy requirements is dependent on knowledge of the climatic consequences of alternative energy and emissions strategies. However, present uncertainties in prospective climate change that would result from a given change in atmospheric composition limit effective planning.

Climate and climate change can be characterized in many ways—temperature: annual mean (spatially, globally), seasonal variation, diurnal range; hydrological cycle: evapotranspiration, precipitation (local, seasonal, episodic), snow and ice cover; and further ramifications involving the biosphere. However, the single key index of climate change, and perhaps the best quantified and understood measure of climate change, is the change in annual and global mean near-surface air temperature. Specifically, for characterization of temperature change the quantity that is most useful is the temperature anomaly, defined as the difference in temperature at a given observation site relative to a climatological mean temperature at that site over an arbitrary but suitably long period (the same for all sites). Expressing temperature change as change in temperature anomaly accounts for local differences in temperature due to altitude, proximity to large bodies of water, and other local and regional climate-influencing geographical features. Temperature anomaly is shown to exhibit strong spatial coherence (auto-correlation distance of order 1000 km; ref. 5), permitting confident spatial averaging, identification of outlier stations, and the like. Use of temperature anomaly makes it possible to speak confidently about changes in global mean surface temperature and the sensitivity of such changes to increased concentrations of greenhouse gases (GHGs) and other climate-influencing variables.

A major objective of current climate research is determination of Earth's climate sensitivity, the change in global mean surface temperature, GMST, that would result from a given sustained perturbation in Earth's radiation budget; in consideration of temperature changes over time, the "anomaly" can be disregarded as the climatological mean is the same for all times under consideration and thus cancels out. The underlying assumption in examination of Earth's climate sensitivity is that the equilibrium change in GMST  $\Delta T_s$  that would result from a given change in a radiative flux component of Earth's climate system due to a given change in atmospheric composition or other radiation influencing component of the climate system (forcing,  $F$ ) would be proportional to the forcing according to

$$\Delta T_s = SF, \quad (1)$$

where  $S$  is the equilibrium sensitivity of the climate or often simply "climate sensitivity." An inherent assumption of the forcing-response paradigm is that change in GMST is proportional to the imposed forcing and that forcings that are different in kind but equal in magnitude would effect equal change in GMST. Neither of these assumptions appears to be strictly correct. Earth's climate system is an inherently nonlinear, chaotic system, which in principle might exhibit large and/or apparently nondeterministic changes in response to small perturbations,<sup>6</sup> as manifested, for example in the large swings between temperate and glacial periods that appear to have resulted from rather small triggering forcings. Additionally, one might expect departures from linearity due to second-order effects, such as melting of ice sheets increasing the absorption of solar radiation. Thus the climate sensitivity might be thought of as the derivative  $S = d\Delta T_s/dF$  evaluated at some initial climate state, but which might vary with change in climate state. Experiments with climate models suggest an approximate linearity between forcing and change in GMST at least for small changes in GMST that would support the use of climate sensitivity to estimate the magnitude of future change of GMST that would result from a sustained forcing, albeit with the caveat that sensitivity might itself change as GMST changes (non-zero second derivative). Climate model studies suggest as well that the changes in GMST for forcings of differing nature (different greenhouse gases, aerosols) are similar (ref. 1, § 2.8.4–5). It might be noted that the climate sensitivity, as defined, would not encompass feedbacks on the forcing itself, such as a change in uptake of CO<sub>2</sub> by vegetation with change in GMST that would effectively change the forcing per emitted CO<sub>2</sub>.

While the emphasis of the present paper is on climate sensitivity of the planet as a whole, that is, the change in global mean surface temperature that would result from a given sustained forcing, this emphasis should not be taken to mean that the change in temperature resulting from a given forcing would be expected to be spatially (or seasonally) uniform. Both observations and climate model studies suggest that sensitivity at high latitudes (especially in the Northern Hemisphere) to forcing from spatially uniform increments of greenhouse gas mixing ratios may be substantially greater than the global average sensitivity.<sup>1,7</sup> There is also substantial uncertainty in climate sensitivity in the tropics.<sup>7</sup> The spatial distribution of temperature change in response to spatially nonuniform forcings, such as by atmospheric aerosols, which are concentrated mainly in industrial regions of the Northern Hemisphere, is indicated in model studies to be nonuniform, greatest in regions where the forcing is greatest,<sup>8,9</sup> consistent with the spatial distribution of observed temperature change over the twentieth century.<sup>10</sup> Considerations such as these certainly limit the applicability of global climate sensitivity as a predictor of change in average temperature at any given location. Nonetheless, global climate sensitivity remains the foremost single indicator of climate response to forcing.

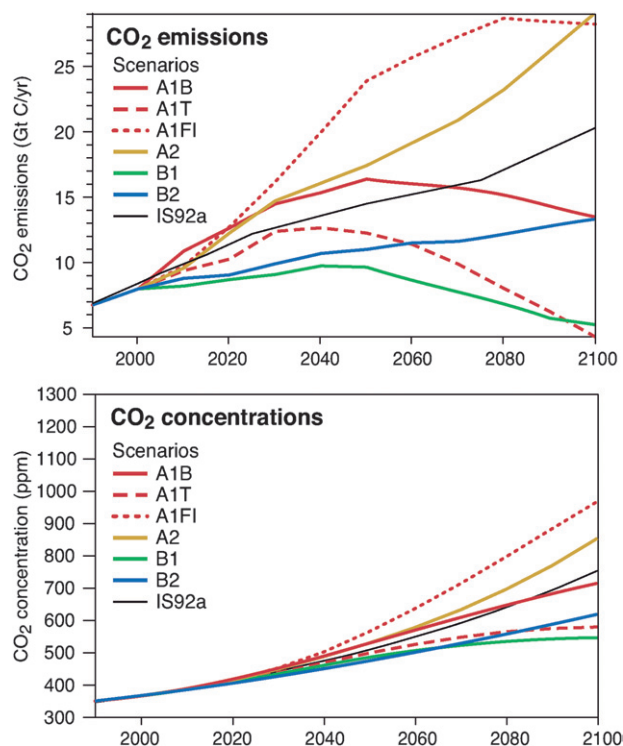
Because of historical precedent Earth's climate sensitivity is commonly expressed as a change in GMST that would result from a doubling of the amount of CO<sub>2</sub> in Earth's atmosphere  $\Delta T_{2\times}$ , but as discussed below, in view of uncertainty associated with the change in flux that would result from a doubling of atmospheric CO<sub>2</sub>, climate sensitivity might better be expressed simply as change in GMST that would result from a given change in

a specified radiative flux component, normalized to that change; the unit for this sensitivity would be  $\text{K}/(\text{W m}^{-2})$  or equivalently  $\text{K W}^{-1} \text{m}^2$ . Temperature change for doubled  $\text{CO}_2$  was employed in early climate model studies solely as a measure of climate sensitivity; however, with the increase in fossil fuel combustion in recent decades it is clear that a mixing ratio double that of the preindustrial atmosphere, 278 ppm (parts per million or  $\mu\text{mol}$  per mole of dry air) is likely to be reached well within the present century, absent major changes in  $\text{CO}_2$  emissions from currently projected growth profiles, Fig. 1, and thus that the amount by

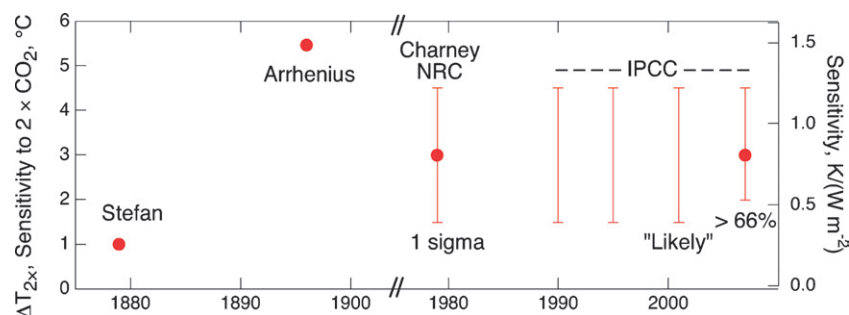
which GMST would increase as a consequence of a doubling of  $\text{CO}_2$  assumes a significance to the people of the world that is far greater than simply a measure of climate sensitivity.

In view of the importance of Earth's climate sensitivity to formulating energy policy and more broadly to understanding climate and climate change generally, there has been substantial effort to determine this quantity over an extended period, as reflected in Fig. 2. The lowest and earliest value shown, that denoted "Stefan," for a black-body radiator at Earth's mean surface temperature 288 K, is for a planet without any feedback processes. Positive feedback processes operating in Earth's climate system such as increased water vapor in a greenhouse-warmed world would increase climate sensitivity over that of a black body radiator. Svante Arrhenius<sup>12</sup> carried out what today would be called a spreadsheet calculation (except that the numerical entries were calculated by hand, not a computer) examining the feedbacks due to changes in atmospheric water vapor and in snow and ice cover as a function of latitude and season. The several other values shown in Fig. 2 reflect broad assessments of scientific understanding based on empirical inference and climate model studies, as carried out by a panel of the US National Research Council,<sup>13</sup> and more recently in four major reports conducted under the auspices of the Intergovernmental Panel on Climate Change. The first three reports of that series declined to present a best estimate for climate sensitivity, indicating only an estimated range; the most recent (2007) assessment report<sup>1</sup> presented a best estimate value and narrowed the range slightly, while also more precisely defining the meaning of the uncertainty range. As is shown in the figure, despite extensive research neither the best estimate nor the estimated range for Earth's climate sensitivity has changed markedly in the last 39 years. At present, this sensitivity is uncertain to at least a factor of 2 between the low and high ends of the uncertainty range (66% likelihood that the actual sensitivity is within the indicated uncertainty range; ref. 1, Summary for Policymakers) and perhaps more. This multiplicative uncertainty expressed as the ratio between sensitivity at the high and low ends of the range,  $U_S \geq 2$ , has major implications on developing strategies to limit changes in Earth's climate.

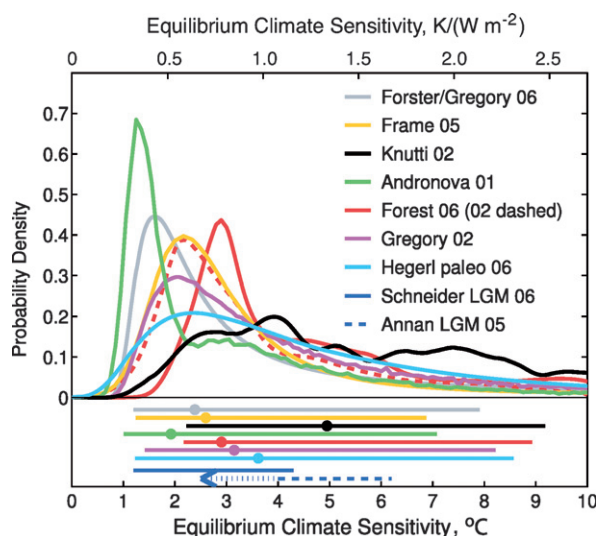
An alternative means of presenting estimated climate sensitivity and associated uncertainty is by means of probability



**Fig. 1** Potential scenarios for emission of  $\text{CO}_2$  from fossil fuel combustion over the twenty first century,  $\text{Pg}$  ( $10^{15} \text{ g}$ )  $\text{C yr}^{-1}$  and corresponding projections of  $\text{CO}_2$  mixing ratios, parts per million (ppm) or  $\mu\text{mol}$  per mole of dry air. From ref. 11, Summary for Policymakers, Fig. 5, in which the several emissions scenarios are identified and discussed.



**Fig. 2** Estimates of Earth's climate sensitivity, expressed as the increase in global mean surface temperature (GMST) that would result from a doubling of the amount of  $\text{CO}_2$  in the atmosphere  $\Delta T_{2\times}$ , left axis, or as the change in GMST that would result from a change in radiative flux of  $1 \text{ W m}^{-2}$ , right axis; the conversion from  $\Delta T_{2\times}$  to  $\text{K W}^{-1} \text{m}^2$  assumes a forcing of doubled  $\text{CO}_2$   $F_{2\times}$  equal to  $3.7 \text{ W m}^{-2}$ . The point noted "Stefan" is the sensitivity that would apply to a black body radiator at Earth's mean surface temperature, 288 K. The notations "1 sigma", "Likely", and "> 66%" denote the likelihood that the actual climate sensitivity lies within the uncertainty range indicated. Charney, National Academy of Sciences Report, ref. 13; IPCC, ref. 1 and earlier assessments in this series.



**Fig. 3** Estimates of the probability distribution function (PDF) for climate sensitivity ( $^{\circ}\text{C}$ ) corresponding to a doubling of  $\text{CO}_2$ ,  $\Delta T_{2\times}$ . PDFs are scaled to integrate to unity between  $0^{\circ}\text{C}$  and  $10^{\circ}\text{C}$ ; the bars show the respective 5 to 95% ranges, dots the median estimate. PDFs from Andronova, Forest (dashed line, anthropogenic forcings only; solid, anthropogenic and natural forcings) Gregory, Knutti, Frame, and Forster and Gregory are based on instrumental temperature data. Hegerl is based on multiple paleoclimatic reconstructions of Northern Hemisphere mean temperatures over the last 700 years. Also shown (Annan, Schneider) are the approximate 5 to 95% ranges for estimates from the last glacial maximum (LGM); ranges extending beyond the published range in Annan are indicated by dots and an arrow. Modified, by addition of auxiliary scale at top, from ref. 1, Figure 9.20, which gives references; after ref. 14.

distribution functions (PDFs), Fig. 3, which depicts the results of several current approaches to determine sensitivity, expressed as temperature increase corresponding to a doubling of  $\text{CO}_2$ ,  $\Delta T_{2\times}$ . Although median values of sensitivity are mainly in the range  $\Delta T_{2\times} = 2\text{--}4\text{ K}$ , each of the PDFs exhibits a substantial contribution to values as great as  $\Delta T_{2\times} = 6\text{ K}$  or more.

The value of climate sensitivity and its uncertainty have important implications. The nations of the world are committed through the United Nations Framework Convention on Climate Change<sup>15</sup> to “achieve... stabilization of greenhouse gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system.” While the nature and extent of climate change that would constitute “dangerous” interference with the climate system are not settled, and while the threshold for the onset of such dangerous interference is undoubtedly not characterized by a single unique quantity or value of that quantity, most analyses characterize this threshold by an increase in GMST, with values in the range 1 to 3 K above present (*e.g.*, ref. 2, p. 38; ref. 16–19). Here it should be noted that GMST increased over the twentieth century by 0.6 to 0.8 K. To lend context to such values of threshold temperature change one might note that the temperature change between the present and the last glacial maximum (21 ka before present), during which kilometre thick ice sheets covered much of the Northern Hemisphere continental land masses, is estimated to have been 4 to 7 K (ref. 1, §6.4.1.2).

For the mixing ratio of atmospheric  $\text{CO}_2$  reaching double its preindustrial value, which, according to the several scenarios in Fig. 1 would occur by the middle to the end of this century, the equilibrium increase in GMST, relative to its preindustrial value, due to this increase in  $\text{CO}_2$  alone (not accounting for time lags in the climate system, additional warming influences on climate, as from other greenhouse gases, countervailing cooling influences, such as from atmospheric aerosols, or possible changes in sensitivity with increasing GMST) for the present central value estimate of climate sensitivity given in Fig. 2 would be 3 K, and for the lower or higher ends of the 66% probability range for that estimate, 2 K or 4.5 K, respectively. Substantially improved knowledge of Earth’s climate sensitivity is thus essential to knowing the extent of increase in GMST that might be expected for a given emission scenario, or alternatively expressed, as input to making the collective decision of which temporal profile of future emissions would be consonant with a given target maximum increase in GMST.

More quantitatively, if it is posited that the increase in GMST shall not exceed a target value, denoted  $\Delta T_s^*$ , then for a given climate sensitivity  $S$  it is possible to calculate the maximum allowable forcing  $F^*$  consonant with that value of  $\Delta T_s^*$  as

$$F^* = \Delta T_s^* / S, \quad (2)$$

from which it is seen that the multiplicative uncertainty that characterizes the allowable forcing is the same as that for climate sensitivity; *i.e.*,  $U_{F^*} = U_S$ . This consideration can be extended to an allowable incremental amount of a climate-forcing agent  $\Delta x^*$ . Clearly, if the forcing is linear in the atmospheric amount of the forcing agent, then  $U_{\Delta x^*} = U_S$ . As it turns out, because the infrared absorption spectrum of atmospheric  $\text{CO}_2$  is highly saturated, the forcing by incremental  $\text{CO}_2$  is not linear in the amount of atmospheric  $\text{CO}_2$  but is approximately proportional to the logarithm of the mixing ratio of atmospheric  $\text{CO}_2$ . However, when considering the forcing due to an incremental mixing ratio of  $\text{CO}_2$   $\Delta x_{\text{CO}_2}$ , within the approximation  $\ln(1 + \Delta) \approx \Delta$  the forcing is approximately proportional to the incremental mixing ratio, and hence the multiplicative uncertainty in maximum allowable incremental mixing ratio of  $\text{CO}_2$   $U_{\Delta x_{\text{CO}_2}^*}$  is, to good approximation, equal to the multiplicative uncertainty in sensitivity. Finally, as the increase in the amount of  $\text{CO}_2$  in the atmosphere in a given year is approximately proportional to the emissions, and as the time constant characterizing the decrease of incremental  $\text{CO}_2$  is long relative to energy planning horizons, the multiplicative uncertainty in the allowable mass of future emissions of  $\text{CO}_2$  consonant with a given allowable change in GMST  $M_{\text{CO}_2}^*$  is likewise equal to the multiplicative uncertainty in climate sensitivity; *i.e.*,  $U_{M_{\text{CO}_2}^*} = U_S$ . From this argument it may be seen that the multiplicative uncertainty in the shared global resource of future emissions of  $\text{CO}_2$  that would be consonant with any agreed upon allowable increase in GMST is currently uncertain to at least a factor of 2. This uncertainty may be seen as greatly limiting the ability of the nations of the world to effectively formulate strategies to meet their energy needs consonant with meeting their obligation to prevent dangerous anthropogenic interference with the climate system.

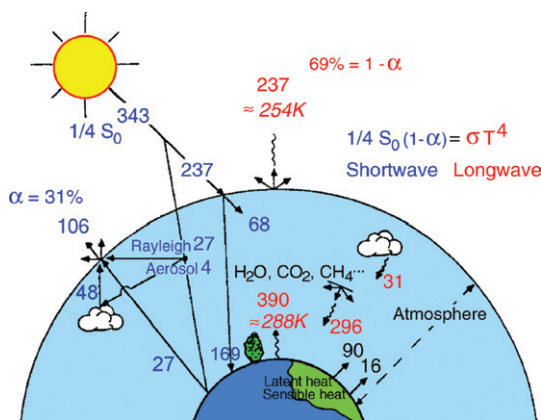
This paper examines approaches to determining Earth’s climate sensitivity, points to the reasons why determining this



sensitivity is so difficult, examines the consequences of uncertainty in Earth's climate system, and outlines approaches to reducing this uncertainty.

## Background

Examination of the consequences of a perturbation to Earth's radiation budget is usefully informed by consideration of the overall radiation budget that drives Earth's climate system, Fig. 4, which provides context for any perturbation. Earth's climate system consists of a delicate balance between absorbed incident solar (shortwave) radiation and emitted thermal infrared (longwave) radiation. From this perspective, Earth is in near radiative "equilibrium" or steady-state. Average incoming solar irradiance is about  $343 \text{ W m}^{-2}$ , one-fourth of the solar constant, the factor of 4 accounting for the area of the planet relative to that of the subtended disk. The fraction of this incoming shortwave irradiance that is reflected or scattered is denoted the planetary albedo  $\alpha$  (so-called "Bond albedo," the average fraction of shortwave radiation that is reflected by the entire illuminated hemisphere). This albedo is not known *a priori* but must be measured; measurements from satellites have established  $\alpha$  to be about 30% ( $0.293 \pm 0.010$ , ref. 22). Clouds are

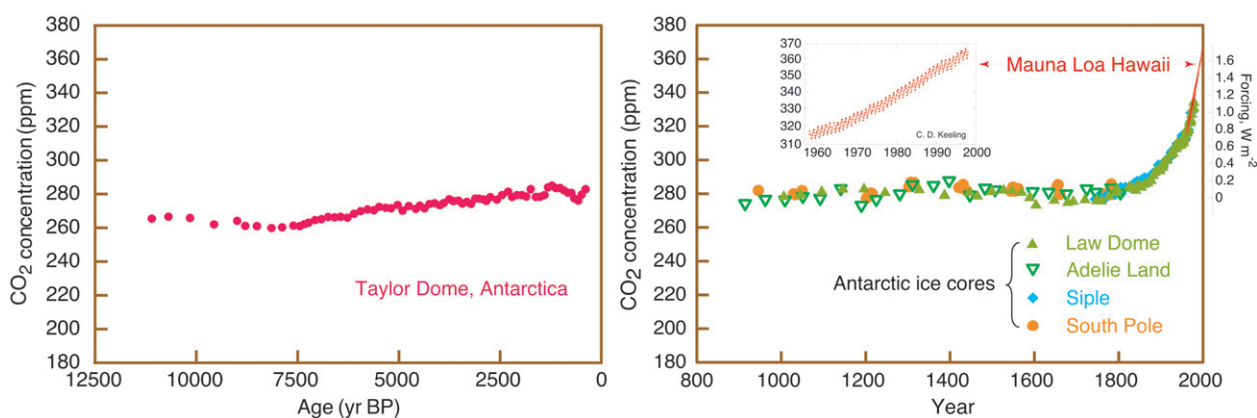


**Fig. 4** Earth's radiation balance, the global-annual average energy budget, in units of watts per square meter,  $\text{W m}^{-2}$ . The global average incident shortwave (solar) power is the solar constant divided by 4. Of this incident power, (shortwave fluxes are shown in blue) roughly 30% (shortwave albedo,  $\alpha$ ) is reflected to space mainly by clouds ( $48 \text{ W m}^{-2}$ ), Rayleigh scattering ( $27 \text{ W m}^{-2}$ ), or reflectance from the surface ( $27 \text{ W m}^{-2}$ ); the balance is absorbed in the atmosphere ( $68 \text{ W m}^{-2}$ ) or at the surface ( $169 \text{ W m}^{-2}$ ). The surface of the planet radiates in the thermal infrared (longwave fluxes are shown in red); the global mean surface temperature of approximately  $288 \text{ K}$  ( $15^\circ \text{C}$ ) corresponds by the Stefan-Boltzmann equation for a black-body radiator to a flux of  $390 \text{ W m}^{-2}$ . Heat is transferred from the surface to the atmosphere also by convection (sensible heat,  $16 \text{ W m}^{-2}$ ) and through evaporation and transpiration of water which subsequently condenses in the atmosphere (latent heat,  $90 \text{ W m}^{-2}$ ). Much of the thermal infrared radiation emitted at the surface is absorbed in the atmosphere and re-radiated downwards by clouds ( $31 \text{ W m}^{-2}$ ) and infrared active gases, mainly water vapor  $\text{H}_2\text{O}$ , carbon dioxide  $\text{CO}_2$ , and methane  $\text{CH}_4$ , ( $296 \text{ W m}^{-2}$ ). The longwave power emitted at the top of the atmosphere ( $237 \text{ W m}^{-2}$ , corresponding to a radiative temperature of  $254 \text{ K}$  or  $-19^\circ \text{C}$ ) is equal to the absorbed shortwave power. Values shown are averages of quantities that vary substantially with location and time. Modified from Schwartz,<sup>20</sup> after Ramanathan.<sup>21</sup>

a major contributor to shortwave reflectance; other contributions include the surface, Rayleigh scattering from air, and scattering by aerosol particles (microscopic and submicroscopic particles suspended in the air). The complement of this incident energy is absorbed by the Earth (including the atmosphere). To a very good approximation, Earth's energy budget is balanced, so the absorbed shortwave energy is emitted to space as longwave infrared radiation.

As a consequence of absorption of solar radiation the land and oceans are warmed and emit longwave infrared radiation. For a global average surface temperature of about  $15^\circ \text{C}$  or  $288 \text{ K}$  the corresponding black body irradiance is  $390 \text{ W m}^{-2}$ , in excess even of the shortwave radiative flux incident at the top of the atmosphere. This can be understood by the fact that much of this infrared energy is returned to the surface by emission from infrared active molecules in the atmosphere (water vapor, carbon dioxide, methane, nitrous oxide, chlorofluorocarbons) and by clouds, which absorb and re-radiate in the infrared—the so-called greenhouse effect (a misnomer<sup>23</sup>). Of particular importance in the context of anthropogenic perturbations to the greenhouse effect is the magnitude of downwelling infrared radiation due to molecular absorption and re-emission, approximately  $300 \text{ W m}^{-2}$ . Here it should be emphasized that all numbers appearing in the figure are global averages of highly variable quantities—variable with latitude and season, and at any location over a variety of time scales. Especially variable are the downwelling emissions from water vapor and from clouds as a consequence of the variability of the amount and vertical distribution of water vapor and clouds in the atmosphere. Completing the picture are contributions from latent heat ( $90 \text{ W m}^{-2}$  corresponding to a global average precipitation of  $\sim 1 \text{ m y}^{-1}$ ) and sensible heat, estimated as about  $16 \text{ W m}^{-2}$ . The emitted longwave flux at the top of the atmosphere,  $237 \text{ W m}^{-2}$ , is well less than that at the surface,  $390 \text{ W m}^{-2}$ , another manifestation of the greenhouse effect. The equality of net downwelling, shortwave irradiance is confirmed by satellite measurements of albedo and upwelling longwave irradiance within a fairly large instrumental bias of about  $5 \text{ W m}^{-2}$  (ref. 22).

At issue at present are the consequence of increases in  $\text{CO}_2$  and other greenhouse gases and other forcing agents, importantly anthropogenic aerosols, which alter Earth's energy balance (*i.e.*, exert a forcing on the climate system) and thereby give rise to change in global mean temperature and to other changes in Earth's climate system (response of the climate system). The mixing ratio of atmospheric  $\text{CO}_2$ , as determined from air trapped in ice cores in Antarctica, Fig. 5, was quite constant over the past 10 000 years ( $260\text{--}280 \text{ ppm}$  [ref. 11, Fig. 3.2]) until beginning to increase strongly after about 1800. The right-hand axis of Fig. 5 gives the calculated increase in the downwelling longwave irradiance from the atmosphere to the surface due to the increase in  $\text{CO}_2$  over the industrial period, reaching about  $1.6 \text{ W m}^{-2}$  by the year 2000. It is this forcing, together with comparable but lesser forcings due to increases in other greenhouse gases, that constitutes the anthropogenic enhancement of Earth's natural greenhouse effect that is the basis for the present concern over climate change. Comparison with the natural greenhouse effect of about  $300 \text{ W m}^{-2}$  (Fig. 4) shows that this enhancement is well less than 1%. Thus the question naturally arises whether such a slight change could give rise to climate change whose



**Fig. 5** Atmospheric mixing ratio of CO<sub>2</sub>, parts per million (ppm) or  $\mu\text{mol}$  per mole of dry air, as determined from air trapped in ice cores in Antarctica and by contemporary measurements at Mauna Loa, Hawaii. Scale on right hand axis gives forcing relative to preindustrial CO<sub>2</sub>, 278 ppm. Modified from ref. 11, Fig 3.2 by inclusion of Mauna Loa data in inset and by addition of scale for forcing at right; Mauna Loa data from Keeling and Whorf.<sup>24</sup>

magnitude would be of any appreciable concern. From eqn (1), for values of sensitivity (as given by ref. 1 with  $F_{2\times}$   $3.7 \text{ W m}^{-2}$ ) 0.54 to  $1.21 \text{ K W}^{-1} \text{ m}^2$ , the resultant temperature increase is calculated as 0.86 to 1.94 K. Such a temperature increase from CO<sub>2</sub> alone is comparable to the range of concern noted above. Comparison of the enhanced greenhouse forcing of incremental CO<sub>2</sub> compared to the natural greenhouse effect also reveals a major source of the difficulty associated with determining the climate change due to incremental CO<sub>2</sub>, namely that the climate system must be understood sufficiently well that the effect of an increase of less than one percent in global mean downwelling longwave irradiance, itself a quantity that is highly variable spatially and temporally, can be determined with sufficient accuracy and confidence that this determination can be useful in formulating strategies to control climate change. This is the great challenge that faces the climate change research community.

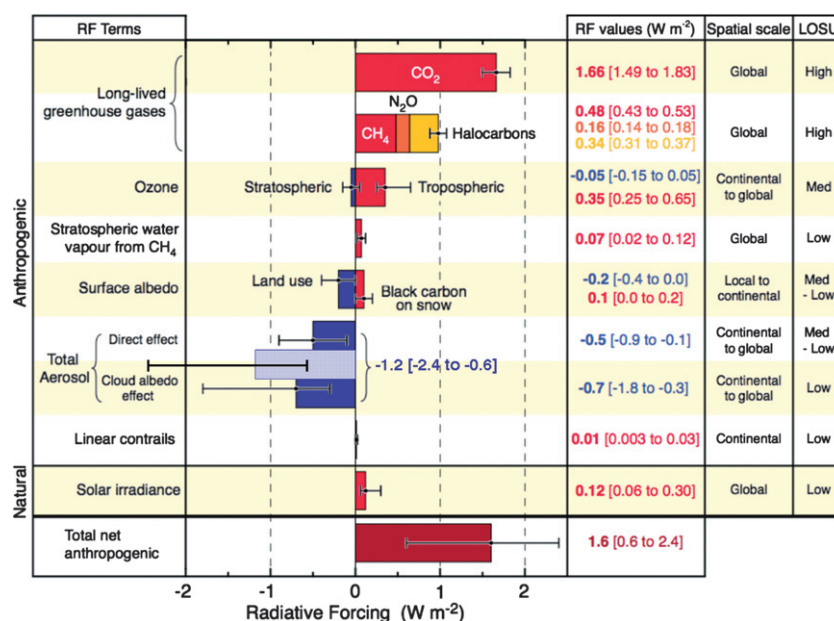
The importance of the present continuing increase in atmospheric CO<sub>2</sub> indicated in Fig. 5 may be assessed by the following back-of-the-envelope calculation. Consider the equilibrium temperature change due to an increment of CO<sub>2</sub> mixing ratio, which is proportional to the forcing and thus to the logarithm of the mixing ratio,

$$\begin{aligned}\Delta T_s(\Delta x_{\text{CO}_2}) &= \frac{\Delta T_{2\times}}{\ln 2} \ln \frac{x_{\text{CO}_2}}{x_{\text{CO}_2}^0} \\ &= \frac{\Delta T_{2\times}}{\ln 2} \ln \left( 1 + \frac{\Delta x_{\text{CO}_2}}{x_{\text{CO}_2}^0} \right) \approx \frac{\Delta T_{2\times}}{\ln 2} \frac{\Delta x_{\text{CO}_2}}{x_{\text{CO}_2}^0},\end{aligned}$$

where  $x_{\text{CO}_2}^0$  and  $x_{\text{CO}_2}$  are the initial and final mixing ratios and  $\Delta x_{\text{CO}_2}$  is the increase in CO<sub>2</sub> mixing ratio. For current observed values of CO<sub>2</sub> mixing ratio, 380 ppm, and annual increase in this mixing ratio,  $1.8 \text{ ppm y}^{-1}$ , the resultant equilibrium change in GMST, for  $\Delta T_{2\times}$  (2, 3, 4.5) K, Fig. 2, the rate of equilibrium temperature increase is, respectively, (0.014, 0.020, and 0.031)  $\text{K y}^{-1}$ , and the time for GMST to increase by 1 K is, respectively, (73, 49, and 33) years. Given that an increase in GMST of 1 K is important in considerations of global climate change, it is seen that the rate of increase of GMST obtained by this calculation for the present rate of increase of atmospheric CO<sub>2</sub> is important even for climate sensitivity at the low end of

the range given by the 2007 IPCC assessment report<sup>1</sup> and quite important for climate sensitivity at the high end of that range. The same calculation for the total increase of CO<sub>2</sub> mixing ratio of 100 ppm observed over the industrial period (Fig. 5) indicates equilibrium temperature increase of (0.7, 1.0, and 1.5) K. The actual temperature increase over this period would depart from such values because of the influence of other forcings, time lag in reaching equilibrium temperature change, climate sensitivity having a value other than those used here, and/or failure of the linear forcing response model. Quantitative understanding of all these effects is of course a major objective of the large current research endeavor directed to climate change.

In addition to CO<sub>2</sub>, there have been several other important influences on atmospheric radiation over the industrial period whose influence on climate change must be determined in order to gain confident understanding of climate change over this period. These influences are summarized in Fig. 6, modified from the 2007 IPCC assessment report.<sup>1</sup> Here the forcings are meant to represent the changes in longwave or shortwave flux over the industrial period. In addition to the longwave forcing due to the increase in CO<sub>2</sub>, there is longwave forcing from increases in atmospheric methane, nitrous oxide, and chlorofluorocarbons and from increases in tropospheric ozone that is produced photochemically from anthropogenic nitrogen oxides and hydrocarbons, which in the aggregate,  $1.33 \text{ W m}^{-2}$ , approach the forcing of CO<sub>2</sub>. The I-beams in the figure represent estimated uncertainties in the several forcings (5–95% confidence range). The decrease in stratospheric ozone due to decomposition catalyzed by chlorine compounds from chlorofluorocarbons results in a slight negative (cooling) forcing. Changes in surface albedo due to land-use changes and due to black carbon (soot) on snow are thought to induce slight forcings in the shortwave. The next important class of forcings is that due to anthropogenic tropospheric (lower atmosphere) aerosols. Anthropogenic aerosols scatter light, effectively increasing Earth's albedo, and absorb light, decreasing albedo if the aerosols are above bright surfaces such as snow or clouds; light scattering dominates, so the net effect is a cooling influence or negative forcing, denoted direct aerosol forcing. Aerosol particles also serve as the seed particles



**Fig. 6** Global average radiative forcing (RF) estimates and uncertainty ranges (5–95% confidence interval) in 2005, relative to the preindustrial climate, for anthropogenic carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), halocarbons (mainly chlorofluorocarbons CFCs) and aerosols and for other important identified agents and mechanisms, together with the typical geographical extent (spatial scale) of the forcing and the assessed level of scientific understanding (LOSU). Forcings are expressed in units of watts per square metre, W m<sup>-2</sup>. The total anthropogenic radiative forcing and its associated uncertainty are also shown. The figure is modified from ref. 1, Figure SPM-2, by addition of a bar for total aerosol forcing (light blue) representing the sum of aerosol direct and indirect forcings, and associated uncertainty.

for cloud droplet formation (cloud condensation nuclei, CCN); other things being equal, a greater number concentration of aerosol particles results in a greater concentration of cloud-droplets and in turn in enhanced multiple scattering within the cloud and a resultant increase in cloud albedo, a further cooling forcing, denoted indirect aerosol forcing. Both of these effects are highly uncertain; the light blue bar and associated I-beam have been added to the figure to represent the total aerosol forcing and associated uncertainty. The uncertainty associated with the aerosol forcing is the dominant uncertainty of the several forcings.

The total forcing over the industrial period, evaluated as the sum of the several forcings, is shown in the bar at the bottom of Fig. 6, together with its associated uncertainty, which is dominated by the uncertainty in aerosol forcing. If the actual aerosol forcing is at the low (negative) end of its estimated uncertainty range then the aerosol forcing offsets only a small fraction of the greenhouse gas forcing, and the total forcing is at the high end of its uncertainty range, 2.4 W m<sup>-2</sup>. On the other hand, if the aerosol forcing is offsetting a major fraction of greenhouse gas forcing, and the total forcing is at the low end of its range, 0.6 W m<sup>-2</sup>. The resultant multiplicative uncertainty in total forcing over the industrial period  $U_F = 4$  has important implications for empirically inferring climate sensitivity from the temperature increase that has occurred over this period. A total forcing at the high end of the uncertainty range would imply a rather low climate sensitivity, and *vice versa*, as the following calculation suggests; see also Gregory *et al.*<sup>25</sup> If the increase in GMST over the twentieth century is taken as 0.6 K (ref. 26) and if the forcing during the twentieth century is taken as 75% of

that given in Fig. 6 for 1750–2005, then the climate sensitivity is 0.3 K W<sup>-1</sup> m<sup>2</sup>, equivalent, for  $F_{2\times}$  taken as 3.7 W m<sup>-2</sup>, to  $\Delta T_{2\times} = 1.2$  K. A total forcing at the low end of the uncertainty range would imply a climate sensitivity 4 times as great, 1.3 K W<sup>-1</sup> m<sup>2</sup>, equivalent to  $\Delta T_{2\times} = 5$  K. These estimates assume that the climate system has responded completely to the applied forcing; that is, that the temperature change over the twentieth century is close to its equilibrium value for the forcing applied over that period. Several investigators<sup>27–30</sup> have suggested that further increase in temperature beyond that realized thus far might be expected in response to forcing due to changes in atmospheric composition that have already occurred—so-called “unrealized” or “committed” warming, or warming that is “in the pipeline” and whose delay in being realized is attributed to “thermal inertia”. Such additional temperature increase would result in an equilibrium sensitivity that is greater than the values just presented; the question of such committed warming is examined further below. However irrespective of that question, it is clear the multiplicative uncertainty of a factor of 4 in climate forcing over the industrial period greatly limits the ability to empirically infer climate sensitivity by this approach. This uncertainty also has implications on the use of global climate models to simulate climate change over this period, as discussed below.

One final point that should be noted in conjunction with Fig. 6 has to do with the spatial scale that characterizes the several forcing agents, which is a consequence of their atmospheric residence times. While there are various means of characterizing these residence times, one measure that is especially pertinent for CO<sub>2</sub> is based on the observation that the increase in present atmospheric CO<sub>2</sub> mixing ratio relative to its preindustrial value is equal to 26 years of emissions of fossil fuel CO<sub>2</sub> at the present

emission rate (or because of increasing emissions over this time period, equal to 45 years of integrated fossil fuel CO<sub>2</sub> emissions). As such a time period greatly exceeds the time required for mixing of the atmosphere, this excess CO<sub>2</sub> is uniformly distributed in the atmosphere, and the forcing is likewise relatively uniformly distributed globally. In contrast, the residence time of tropospheric aerosols responsible for the aerosol forcing is about a week, so that in conjunction with the highly nonuniform distribution of sources, these aerosols and the resultant forcings are quite inhomogeneously distributed spatially and temporally. These differences have implications both for characterizing the forcing and for understanding of climate change and in developing strategies to control it.

### Single-compartment energy balance model of Earth's climate system

Considering Earth's atmosphere-ocean-land system as a single compartment immediately leads to an expression for the rate of change of the global heat content that serves as the basis of energy balance models of Earth's climate system. While such models are highly simplified representations of the climate system and clearly cannot represent any of the vertical, horizontal, or seasonal fine structure of the climate system, they are useful to illustrate important features of Earth's climate system such as sensitivity and feedbacks and thus lead to considerable insight. According to such a model, the rate of change of the heat content of Earth's climate system is given by

$$\frac{dH}{dt} = Q - E + P \quad (3)$$

where  $Q$  is the rate of absorption of solar (shortwave) energy,  $E$  is the rate of emission of thermal (longwave infrared) radiation at the top of the atmosphere, and  $P$  denotes the rate of energy production by other processes, principally residual energy from planetary formation and decay of natural radionuclides; fossil fuel combustion, and nuclear energy production. These additional sources of energy, about 0.1 W m<sup>-2</sup> in the aggregate (Table 1), are quite small relative to the radiative energy terms and are neglected in further discussion here, although they become important in consideration of the imbalance in Earth's energy budget associated with the secular increase in GMST (ref. 22,30). Hence to good approximation

$$\frac{dH}{dt} = Q - E \quad (4)$$

**Table 1** Nonradiative contributions to Earth's energy budget

| Energy contribution              | Average energy flux/mW m <sup>-2</sup> | Data source  |
|----------------------------------|--|--|
| Conduction from Earth's interior | 90                                     | Pollack <i>et al.</i> <sup>31</sup> ;<br>Jaupart <i>et al.</i> <sup>32</sup> |
| Fossil fuel combustion           | 25                                     | BP <sup>33</sup>   |
| Nuclear energy                   | 2                                      | BP <sup>33</sup>   |
| TOTAL                            | 120                                    |  |

Fossil fuel and nuclear energy production rates are converted to global average heat flux.

Eqn (4) is the basis for the energy balance model of Earth's climate system that leads to a derivation of the climate sensitivity of the planet as a whole in terms of pertinent "whole earth" variables.

Derivation of an expression for Earth's climate sensitivity in the single-compartment model (*e.g.*, ref. 34,35) assumes that the system is initially in steady-state,

$$Q_0 - E_0 = 0, \quad (5)$$

where the subscript 0 denotes the initial state. Following imposition of a forcing (taken here to be positive), the energy balance of the climate system is restored as the surface temperature increases, increasing outgoing longwave radiation, thereby limiting the resulting increase in temperature rise, and the climate system relaxes to a new steady-state. Conventionally for small perturbations, a linear relation, eqn (1), is assumed between steady-state change in  $T_s$ ,  $\Delta T_s(\infty)$ , and the imposed forcing  $F$ . The equilibrium climate sensitivity  $S$  is equal to the change in temperature at the new steady-state after the change in a radiative flux, that is a forcing  $F$ , has been imposed on the system, divided by the forcing. At the new steady-state

$$Q_0 + \Delta Q - E_0 - \Delta E + F = 0 \quad (6)$$

whence

$$\Delta E - \Delta Q = F \quad (7)$$

From the definition of sensitivity (eqn (1))

$$S = \frac{\Delta T_s}{F} = \frac{\Delta T_s}{\Delta E - \Delta Q} = \frac{1}{\left. \frac{dE}{dT_s} \right|_0 - \left. \frac{dQ}{dT_s} \right|_0} \quad (8)$$

where the subscripts 0 on the derivatives indicate that they are to be evaluated at the initial state; the entire model in fact consists of exploration of the consequences of a small perturbation on the initial state, with retention only of first-order terms. Expressing the rate of absorption of solar (shortwave) energy as

$$Q = \gamma J_s/4 \quad (9)$$

where  $\gamma \equiv 1 - \alpha$  is the planetary coalbedo (complement of albedo) and  $J_s$  is the solar constant, and the rate of emission of longwave radiation according to the Stefan-Boltzmann radiation law in terms of the global mean surface temperature  $T_s$  and an effective planetary emissivity  $\varepsilon$  as

$$E = \varepsilon \sigma T_s^4 \quad (10)$$

yields

$$\varepsilon_0 \sigma T_{s0}^4 = \gamma_0 J_s/4 \quad (11)$$

Here the subscript 0 denoting the initial state has been added not just to the surface temperature but also to the emissivity and coalbedo in the expectation that these quantities may also change as the planetary temperature reaches its new steady-state value, for example by a change in cloudiness or atmospheric water vapor content.

If it is assumed that neither  $\varepsilon$  nor  $\gamma$  depends on  $T_s$ , then  $dE/dT_s = 4\varepsilon \sigma T_s^3 = \gamma J_s/T_s$  and  $dQ/dT_s = 0$  and



$$S = \frac{T_s}{\gamma J_s} \equiv S_{\text{SB}}, \quad (12)$$

which is denoted the Stefan–Boltzmann climate sensitivity. More generally

$$\left. \frac{dE}{dT_s} \right|_0 = 4\epsilon_0 \sigma T_{s0}^3 + \sigma T_{s0}^4 \left. \frac{d\epsilon}{dT_s} \right|_0 = \frac{\gamma_0 J_s}{T_s} \left( 1 + \frac{1}{4} \left. \frac{d \ln \epsilon}{d \ln T_s} \right|_0 \right) \quad (13)$$

and

$$\left. \frac{dQ}{dT_s} \right|_0 = \frac{J_s}{4} \left. \frac{d\gamma}{dT_s} \right|_0 = \frac{\gamma_0 J_s}{T_{s0}} \frac{1}{4} \left. \frac{d \ln \gamma}{d \ln T_s} \right|_0 \quad (14)$$

whence

$$\begin{aligned} S &= \frac{1}{\frac{\gamma_0 J_s}{T_{s0}} \left( 1 + \frac{1}{4} \left. \frac{d \ln \epsilon}{d \ln T_s} \right|_0 - \frac{1}{4} \left. \frac{d \ln \gamma}{d \ln T_s} \right|_0 \right)} \\ &= S_{\text{SB}} \frac{1}{1 - \left( \frac{1}{4} \left. \frac{d \ln \gamma}{d \ln T_s} \right|_0 - \frac{1}{4} \left. \frac{d \ln \epsilon}{d \ln T_s} \right|_0 \right)} \end{aligned} \quad (15)$$

where the two derivatives in the denominator represent physical feedbacks in the climate system, that is, changes in the properties of the climate system that further influence the absorption of solar radiation or the emission of infrared radiation by the climate system, respectively, beyond the imposed radiative perturbation. Note that a decrease in emissivity with increasing surface temperature, as would result from an increase in atmospheric water vapor with increasing surface temperature (as by Clausius Clapeyron) would decrease the denominator and increase climate sensitivity; this would be a positive feedback in the climate system. A decrease in cloudiness with increasing surface temperature would increase shortwave coalbedo, again resulting in increased sensitivity (positive feedback) whereas an increase in cloudiness would decrease coalbedo and decrease sensitivity (negative feedback). It is convenient to denote

$$f \equiv \frac{1}{1 - \left( \frac{1}{4} \left. \frac{d \ln \gamma}{d \ln T_s} \right|_0 - \frac{1}{4} \left. \frac{d \ln \epsilon}{d \ln T_s} \right|_0 \right)} = \frac{1}{1 - \mathcal{F}} \quad (16)$$

the climate feedback factor and

$$\mathcal{F} \equiv \frac{1}{4} \left. \frac{d \ln \gamma}{d \ln T_s} \right|_0 - \frac{1}{4} \left. \frac{d \ln \epsilon}{d \ln T_s} \right|_0 \quad (17)$$

the climate feedback strength, with the sign convention that positive  $\mathcal{F}$  denotes a positive feedback. For climate feedback strength  $\mathcal{F}$  approaching unity the feedback factor becomes quite large and the feedback factor and climate sensitivity become quite sensitive to the value of  $\mathcal{F}$  (ref. 34,36,37).

Evaluation of the Stefan–Boltzmann sensitivity for global mean surface temperature  $T_s = 288$  K, solar constant  $J_s = 1370 \text{ W m}^{-2}$  (ref. 22), and planetary coalbedo 0.71 (ref. 22) yields  $S_{\text{SB}} = 0.30 \text{ K W}^{-1} \text{ m}^2$ ; for a forcing due to doubling of  $\text{CO}_2$   $F_{2\times} = 3.7 \text{ W m}^{-2}$ , the corresponding doubling temperature is  $\Delta T_{2\times} = 1.10$  K. This sensitivity is of comparable magnitude to, but considerably lower than, current estimates for climate sensitivity shown in Fig. 2 and 3, indicative of substantial positive feedback

in the climate system. For the central value and 17–83% uncertainty range of climate sensitivity given by the 2007 IPCC report<sup>1</sup>  $\Delta T_{2\times} = (2, 3, 4.5)$  K, for which the multiplicative uncertainty  $U_S = 2.25$ , the feedback factor  $f = (1.8, 2.7, 4.1)$ , and the feedback strength  $\mathcal{F} = (0.45, 0.63, 0.76)$ . Such a strong dependence of climate sensitivity on feedback strength—an increase in  $\mathcal{F}$  of 19% resulting in an increase in  $S$  of 50%—places severe requirements on the accuracy of  $\mathcal{F}$  needed to determine climate sensitivity from climate models and is thus a major challenge to the climate modeling research community.

## Global climate models

Global climate models, GCMs, are sets of difference equations representing the properties and processes that comprise Earth's climate system: transfer, scattering, reflection absorption, and emission of short- and longwave radiation; horizontal and vertical transport of air and water and their heat content (and in the air, water vapor and clouds), evaporation and condensation of water, precipitation, and the like under the constraints of conservation of matter, energy, and momentum that are solved through computer-intensive forward finite-difference treatment from specified initial conditions. While the acronym GCM originally referred to a general circulation model of the global atmosphere or the global ocean, it now refers more generally to coupled atmosphere–ocean global climate models. Global climate models are viewed as the principal tool by which climate scientists and policymakers can determine the consequences of alternative prospective future scenarios of emissions of  $\text{CO}_2$  and changes in other influences on Earth's climate. A key challenge of global climate modeling is determining the consequence of small perturbations in Earth's radiative balance. GCMs must represent current climate with sufficient accuracy and fidelity that changes resulting from small perturbations can be calculated with an accuracy that is useful for effectively developing energy strategies.

In principle, the underlying physics that must be represented in a global climate model is considered well understood; however, in practice the wide variety of scales of important processes that must be represented in the model—from submicrometre, for aerosol and cloud processes, to thousands of kilometres for large-scale circulations—makes it computationally impossible to solve the finite difference equations on global scales with the accuracy and precision that would characterize solving the fluid flow problem, say, for an airfoil, for which grid sizes may be made sufficiently small to guarantee accurate convergence. Consequently, fairly large grid sizes must be used in GCMs, typically about 100 km in the horizontal and 1 km in the vertical, with so-called sub-grid processes, physical processes on smaller scales and associated spatial inhomogeneities, being represented through parameterizations. Important subgrid processes involve transfer of short- and longwave radiation in inhomogeneous cloudy atmospheres, vertical transfer of air in up- and down-drafts and associated transfer of heat and water, cloud formation and dissipation, and precipitation development. The need for parameterization also arises in describing the size distributions of cloud drops and aerosol particles, as clearly models are incapable of following the dynamics of individual drops or particles; for the most part even the size distributions are assumed, rather than

evolved. A similar situation arises also in treating atmospheric radiation transfer, in which the highly structured wavelength dependence of molecular absorption features necessitates parameterized treatment. Thus, there is much skill involved in developing GCMs and much effort required to test them against observations on various temporal and spatial scales.

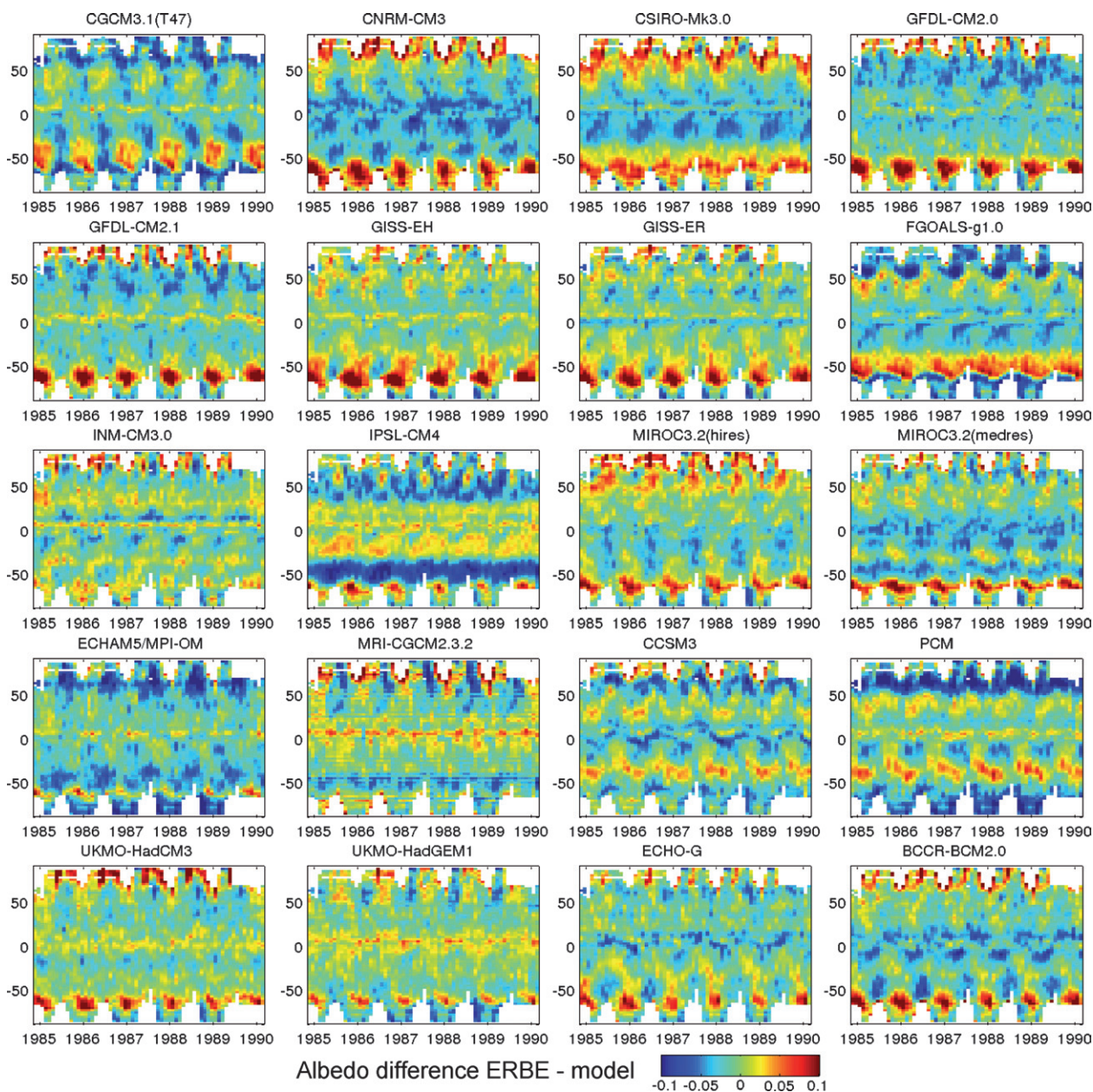
In addition to issues of parameterization of subgrid processes, there are important subgrid processes that are not well understood but which are thought to exert major influence on Earth's climate that must be represented in climate models. Key among these are the coalescence of cloud droplets to form precipitation and a variety of processes involving atmospheric aerosols as discussed below. Differences in treatment of these processes can lead to substantial differences in modeled climate sensitivity and to other changes in climate that would result from a given perturbation in atmospheric composition. Developing improved understanding of these processes continues to be a key objective of atmospheric science research.

Thus far no approach has emerged by which subgrid processes can be uniquely or optimally represented in GCMs. Consequently a situation has arisen in which some fifteen research groups globally are engaged in developing and testing more or less independent GCMs (or in some instances multiple models within the same group), the differences to great extent reflecting different approaches to representing subgrid processes involving radiation, clouds, and aerosols. Much effort has gone into evaluation of the accuracy of GCMs in reproducing Earth's present climate, especially temperature, cloudiness, and precipitation as a function of location and season. Accuracy in treatment of cloudiness and its latitudinal and seasonal dependence is especially important because of the importance of clouds in affecting short- and longwave radiation. As may be seen from Fig. 4, a 10% error in treatment of clouds in the climate model would result an error of some  $4.8 \text{ W m}^{-2}$ . Errors of comparable magnitude in longwave radiation can arise from errors in the vertical distribution of clouds. Errors in the seasonal or latitudinal dependence of cloudiness could similarly exert substantial influences on large scale circulations. Accurate representation of the seasonal and latitudinal dependence of clouds is also important because these dependences might serve as indicators of how cloudiness and cloud properties might change in response to temperature change in a greenhouse warmed world affecting cloud feedbacks. A recent comparison, Fig. 7, of shortwave albedo in 20 current climate models with satellite measurements shows major model-to-model differences in the seasonal and latitudinal dependence; these differences are due almost entirely to treatment of clouds in the models. The consequences of errors in modeled albedo of the magnitudes shown here can be substantial; for a mean incident shortwave flux at the top of the atmosphere of  $243 \text{ W m}^{-2}$ , an error in albedo as low as 0.01 results in an error in absorbed solar irradiance of  $2.4 \text{ W m}^{-2}$ . To some extent the error in absorbed shortwave energy is compensated in the longwave, but such compensation applies only to the total energy balance, and not to the vertical distribution of heating rate, which affects circulations on a variety of scales. Here it should be borne in mind that the quantities plotted are zonal monthly means; shorter term temporal and spatial (longitudinal) differences would be expected to be even greater.

## Climate sensitivity from global climate models

A major impetus for development of global climate models has been to examine the consequences of a given historical or assumed prospective change in atmospheric composition (or other influences on climate change such as change in surface albedo or evapotranspiration associated with land use changes). These consequences might be examined by running the climate model with and without the imposed changes sufficiently long to reach an equilibrium state, and sufficiently long to obtain accurate temporal statistics over multiple years, perhaps 30 to 100 or more. Often such models are run to equilibrium using only a mixed-layer ocean to decrease computer time needed to reach the new equilibrium, but with attendant concern over the accuracy of ocean temperature distribution and how errors might feed back into atmospheric circulation and other atmospheric properties. Alternatively the time-dependent coupled atmosphere–ocean equations might be solved, integrating the equations over a time period during which concentrations of  $\text{CO}_2$  and other GHGs and aerosols are increased, to ascertain the time-dependent climate change. Central to evaluating the performance of climate models is to examine their performance over the twentieth century, for which there are reliable instrumental measurements against which to compare changes in temperature and precipitation to assess confidence in the model. Because of inherent variability both in Earth's actual climate and in modeled climate, it has become standard practice to run a given climate model multiple times with somewhat different initial conditions to obtain variability statistics necessary to assess model accuracy in comparison with observations—so-called ensemble runs. A major application of the models is to run them for future emissions scenarios to assess the consequences of different prospective future emissions, again perhaps as an ensemble of runs for each of the several models.

Because of differences in model formulations there is a substantial range in model sensitivities, as summarized in Table 2 for 18 current climate models. Attention is called to the multiplicative uncertainty, evaluated between the 5th and 95th percentiles, for the equilibrium sensitivity  $\Delta T_{2\times}$ , 2.1. A slightly lower multiplicative uncertainty, 1.7, is indicated for the transient sensitivity. Transient sensitivity is determined by increasing the  $\text{CO}_2$  mixing ratio at 1% per year (compounded) so that by 70 years  $\text{CO}_2$  mixing ratio has doubled; the transient sensitivity is evaluated as the average increase in GMST in years 60–80 relative to the initial temperature. Transient sensitivity defined and determined in this way is less than the equilibrium sensitivity because of time lag in reaching the new steady-state climate. Transient sensitivity is thought to be a more accurate measure of climate system response to increasing concentrations of greenhouse gases such as has occurred over the past century than equilibrium sensitivity. Time-dependent runs are sensitive to the representation of the role of ocean currents in latitudinal distribution of heating and transfer of heat to the deep ocean. The value and uncertainty range in modeled climate sensitivity are a major input to present assessments of Earth's climate sensitivity and uncertainty range reflected in Fig. 2 and 3 above. Understanding the reasons for the large model-to-model differences in sensitivity may help identify where research efforts might



**Fig. 7** Difference between planetary albedo as determined by satellite measurements (Earth Radiation Budget Experiment, ERBE) and twenty global climate models as a function of latitude and time (November 1984–February 1990). Positive anomalies, where albedo determined by ERBE is higher, are indicated with red, and negative anomalies, where ERBE albedo is lower, with blue colors. Modified from Bender *et al.*<sup>38</sup>; courtesy of F. Bender, University of Stockholm.

most usefully be directed to reduce the uncertainty range attached to estimated climate sensitivity.

Also shown in Table 2 is the forcing corresponding to a doubling of atmospheric  $\text{CO}_2$   $F_{2\times}$ , which exhibits a range among the several models of almost  $1 \text{ W m}^{-2}$ . The spread in this quantity, which serves as the basis for characterizing model sensitivity expressed as  $\Delta T_{2\times}$  can potentially result in a misleading picture of the level of agreement of model sensitivity. As the statistics characterizing the spread in equilibrium sensitivity  $S$  expressed in units of  $\text{K W}^{-1} \text{ m}^2$  are somewhat greater than those for  $\Delta T_{2\times}$ , there appears to be a slight anticorrelation between model sensitivity and the value of  $\text{CO}_2$  doubling forcing

$F_{2\times}$  employed in the several models; that is models having a greater sensitivity employed a lower value of  $F_{2\times}$ . Such an anticorrelation has been noted previously by Webb *et al.*<sup>39</sup> using a somewhat different set of models. Attention has previously been called to the spread in values of  $F_{2\times}$  as calculated by the radiation codes of various GCMs even for cloud-free skies<sup>40</sup> for which differences in  $F_{2\times}$  cannot be attributed to differences in the cloud properties of the several models but must be ascribed to differences in treatment of the radiative transfer. Substantial systematic differences were also shown between climate models and much more accurate line-by-line radiation codes. These findings suggest that the effect of model-to-model variation in the

**Table 2** Equilibrium sensitivity, transient sensitivity, and CO<sub>2</sub> doubling forcing from 18 current global climate models<sup>a</sup>

| Model                              | Equilibrium sensitivity $\Delta T_{2\times}/K$ | Transient sensitivity/K | CO <sub>2</sub> Doubling forcing $F_{2\times}/W\ m^{-2}$ | Equilibrium sensitivity $S/K\ W^{-1}\ m^2$ |
|------------------------------------|--|-------------------------|--|--|
| IPSL-CM4                           | 4.4  | 2.1                     | 3.48   | 1.26                                       |
| UKMO-HadGEM1                       | 4.4  | 1.9                     | 3.78   | 1.16                                       |
| MIROC3.2(hires)                    | 4.3  | 2.6                     | 3.14   | 1.37                                       |
| MIROC3.2(medres)                   | 4.0  | 2.1                     | 3.09   | 1.29                                       |
| CGCM3.11(T47)                      | 3.4  | 1.9                     | 3.32   | 1.02                                       |
| ECHAM5/MPI-OM                      | 3.4  | 2.2                     | 4.01   | 0.85                                       |
| GFDL-CM2.1                         | 3.4  | 1.5                     | 3.50   | 0.97                                       |
| UKMO-HadCM3                        | 3.3  | 2.0                     | 3.81   | 0.87                                       |
| ECHO-G                             | 3.2  | 1.7                     | 3.71   | 0.86                                       |
| MRI-CCGCM2.3.2                     | 3.2  | 2.2                     | 3.47   | 0.92                                       |
| CSIRO-MMK3.0                       | 3.1  | 1.4                     | 3.47   | 0.89                                       |
| GFDL-CM2.0                         | 2.9  | 1.6                     | 3.50   | 0.83                                       |
| CCSM3                              | 2.7  | 1.5                     | 3.95   | 0.68                                       |
| GISS-EH                            | 2.7  | 1.6                     | 4.06   | 0.67                                       |
| GISS-ER                            | 2.7  | 1.5                     | 4.06   | 0.67                                       |
| FGOALS-g1.00                       | 2.3  | 1.2                     | 3.71   | 0.62                                       |
| INM-CCM3.0                         | 2.1  | 1.6                     | 3.71   | 0.57                                       |
| PCM                                | 2.1  | 1.3                     | 3.71   | 0.57                                       |
| Average                            | 3.2  | 1.8                     | 3.64   | 0.89                                       |
| Standard deviation                 | 0.7  | 0.4                     | 0.29   | 0.25                                       |
| Relative standard deviation        | 0.23   | 0.21                    | 0.08   | 0.28                                       |
| Range                              | 2.3  | 1.4                     | 0.97   | 0.80                                       |
| Relative Range                     | 0.72   | 0.79                    | 0.27   | 0.90                                       |
| Multiplicative uncertainty U(5/95) | 2.1  | 1.7                     | 1.3  | 2.3  |

<sup>a</sup> From ref. 1, Table 8.2 and Table S8.1.

value of  $F_{2\times}$  on reported model sensitivities might be avoided by expressing sensitivities in units of  $K\ W^{-1}\ m^2$  rather than as  $\Delta T_{2\times}$ . Still, as the multiplicative uncertainty in  $F_{2\times}$  is well less than that in model sensitivities, uncertainty in  $F_{2\times}$  cannot be the major cause of model-to-model variation in sensitivity. Nonetheless, because of model-to-model differences in CO<sub>2</sub> doubling forcing, expressing model sensitivity as the change in GMST normalized to a specified change in a radiative flux component, rather than change in GMST due to doubling of CO<sub>2</sub>, would seem to eliminate this contribution to model-to-model differences in sensitivity.

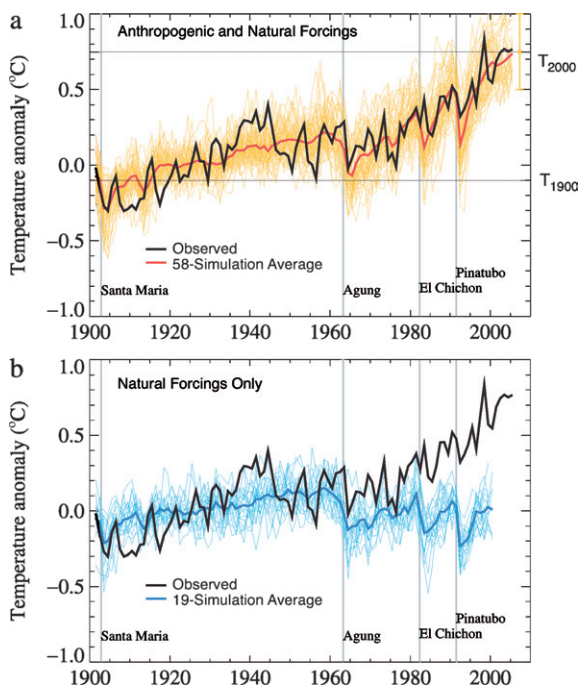
## Performance of climate models over the twentieth century

Just as with the actual climate, perhaps the most important attribute of any GCM is its climate sensitivity, the equilibrium change in global mean surface temperature that would result from, and normalized to, a given change in a radiative flux component such as might result from increase in mixing ratio of CO<sub>2</sub> or other GHGs or from increased amounts of atmospheric aerosols. The intent, of course, is that the model provides a representation of the climate sensitivity of Earth's climate system that is sufficiently accurate to be used with confidence in developing strategies for managing the planet's energy economy consonant with an acceptable degree of climate change. While climate sensitivity is by no means the only pertinent index of climate change, it is arguably the single most important such index for purposes of planning energy futures. Furthermore, this index, being a measure of global climate response, is arguably more likely than any other modeled quantity to be an accurate

estimator of actual climate response. Appropriately, therefore, this quantity is the first quantity that is used to compare the response of a given model to forcings that are different in nature, for example response to forcings by different GHGs or between forcings by GHGs and aerosols; most models exhibit linearity in response to a given forcing and a sensitivity to different forcing agents that is the same within  $\pm 20\%$  or so (ref. 1, § 2.8.4–5). Sensitivity is also perhaps the key model attribute that is used to compare and contrast climate change calculated by different models.

A key test of the accuracy with which climate models might be expected to determine future climate change that would result from for a given scenario of emissions or atmospheric composition (Fig. 1) is their ability to represent past climate change. An exercise was reported in the 2007 IPCC Working Group I Assessment<sup>1</sup> in which the results of 58 model runs obtained with 14 different GCMs were compared with observations of GMST anomalies over the twentieth century, for which there are instrumental measurements of high quality and spatial coverage over most of the planet (Fig. 8); the study also reported the results of 19 model runs using 5 different models for natural forcings only. In the model runs conducted, each modeling group used its own representation of anthropogenic forcings over the modeled time period. As well, the simulations used models with different climate sensitivities and rates of ocean heat uptake. As seen in the figure, the model runs that did not include anthropogenic forcings did not exhibit the systematic increase in GMST that is characterized by the observations, but when anthropogenic forcings were included, the modeled temperature changes  $\Delta T_s = 0.85 \pm 0.25\ K$  agreed fairly closely with the observations. This agreement led the IPCC Working Group to conclude that





**Fig. 8** Global mean surface temperature anomalies ( $^{\circ}\text{C}$ ) from observations (black) and coupled atmosphere-ocean global climate model simulations forced with (a) both anthropogenic and natural forcings and (b) natural forcings only. All data are shown as GMST anomalies relative to the period 1901 to 1950, as observed (black, Hadley Centre/Climatic Research Unit gridded surface temperature data set, HadCRUT3; ref. 41) and, in (a) as obtained from 58 simulations produced by 14 models with both anthropogenic and natural forcings. The multi-model ensemble mean is shown as the thick red curve, and individual simulations are shown as thin yellow curves. Vertical gray lines indicate the timing of major volcanic events, which would exert a cooling influence on climate by injecting aerosol particles into the stratosphere. The simulated global mean temperature anomalies in (b) are from 19 simulations produced by five models with natural forcings only. The multi-model ensemble mean is shown as a thick blue curve, and individual simulations are shown as thin blue curves. Each simulation was sampled so that coverage corresponds to that of the observations. Modified from ref. 1, Figure 9.5, by addition of horizontal lines for temperatures at 1900 and 2000 and uncertainty range for temperature at 2000.

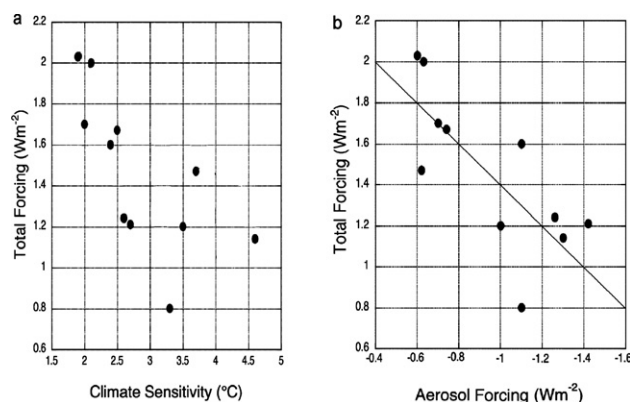
“simulations that incorporate anthropogenic forcings, including increasing greenhouse gas concentrations and the effects of aerosols, and that also incorporate natural external forcings, provide a consistent explanation of the observed temperature record” (ref. 1, § 9.1.4.2).

As noted by Schwartz *et al.*,<sup>41</sup> for the increase in modeled GMST over the time period  $\Delta T_s = 0.85\text{ K}$ , the range of modeled  $\Delta T_s$ ,  $\pm 0.25\text{ K}$  corresponds to a multiplicative uncertainty  $U_{\Delta T_s} = 1.1\text{ K}/0.6\text{ K} \leq 2$ . While such a multiplicative uncertainty associated with the modeled increase in GMST is comparable to that characterizing the sensitivities of current GCMs (2.1 for equilibrium sensitivity; 1.7 for transient sensitivity; Table 2), it is well less than the multiplicative uncertainty in total forcing, 4, Fig. 6. These multiplicative uncertainties suggest that if a single temporal profile of forcing were used in the suite of GCM calculations, the multiplicative uncertainty in modeled  $\Delta T_s$  would be a factor of 1.7 to 2.1; in contrast, if a set of runs were

carried out with a single GCM for the full range of forcings, the multiplicative uncertainty would be a factor of 4. The effect of combining the uncertainties in both model sensitivity and forcing would be expected to yield an uncertainty well greater than that in model sensitivity and certainly not well less than that in the forcing, as is the case with the modeled global temperatures shown in Fig. 8. Schwartz *et al.*<sup>42</sup> suggested that a possible explanation for this might be that the forcings employed in the model runs were anticorrelated with the sensitivities of the models; that is, that models with high sensitivities used low forcings and *vice versa*. Subsequently, Kiehl<sup>43</sup> showed for a subset of the models employed that model sensitivity and forcing were indeed anticorrelated and that the spread in the forcings employed in the several models was due mainly to the choice of the aerosol forcings employed, Fig. 9. It would thus seem that a more accurate picture of the ability to represent twentieth century climate with current climate models would be obtained by exercising each of the models over the uncertainty range of forcing and then examining the envelope of modeled change in GMST, and it seems likely that such a picture would indicate a much greater uncertainty than is indicated in Fig. 8.

### Clouds: the major source of uncertainty in GCM climate sensitivity

Clouds play a central role in Earth's climate system, and it is thus to be expected that modifications of cloud properties in conjunction with greenhouse warming would exert important feedbacks on the climate system affecting temperature sensitivity. Clouds reflect shortwave radiation and absorb and emit long-wave radiation. Low clouds, *e.g.*, stratus, are efficient shortwave reflectors, but because their temperatures are close to those of the surface, they do not exert a strong influence on longwave fluxes. In contrast, high clouds, *e.g.*, cirrus, do not reflect much short-wave radiation but exert a strong influence on the longwave by absorbing and re-radiating terrestrial radiation that would otherwise escape to space. The aggregate clouds are shown, by



**Fig. 9** (a) Total anthropogenic forcing *versus* equilibrium climate sensitivity  $\Delta T_{2\times}$  from nine coupled climate models and two energy balance models that were used to simulate the climate of the 20th century. (b) Total anthropogenic forcing *versus* aerosol forcing; note reverse sense of the abscissa scale; slope of diagonal corresponds to  $\Delta(\text{total forcing})/\Delta(\text{aerosol forcing})$  equal to unity. Modified from ref. 43.

satellite measurements, to exert a global average decrease in absorbed shortwave irradiance of  $48 \text{ W m}^{-2}$  and a decrease in emitted longwave radiance of  $31 \text{ W m}^{-2}$ . Hence even a rather small error in the representation of clouds in the present climate can result in an error in radiative fluxes that is substantial in the context of flux changes associated with increases in  $\text{CO}_2$  and other climate-forcing agents. The difficulty in representing clouds in GCMs has been noted above. Equally important in the context of determining Earth's climate sensitivity is the change in cloud properties that would result from a change in GMST that would either augment (positive feedback) or diminish (negative feedback) the temperature change resulting from the forcing by the increase in  $\text{CO}_2$  or other forcing agent.

In view of the importance of clouds in the climate system much attention has been paid to the examination of the representation of clouds in climate models and to the assessment of cloud feedbacks in these models. As shown almost two decades ago,<sup>35</sup> most of the then threefold variation in modeled climate sensitivity was attributable to differing parameterizations of clouds that resulted in different cloud feedbacks, changes in net short- or longwave radiation due to changes in cloud amount or properties with changing GMST. Despite much work in developing and testing cloud parameterizations in the intervening years, a strong variation of GCM sensitivity is still manifested in current climate models resulting from differences in cloud feedback strength, Fig. 10. These differences in cloud feedbacks are not tuned in the models *per se*, but result, rather, from differences in the treatment of the processes that govern the amount and properties of clouds and the dependence on properties of the climate system such as humidity, temperature lapse rate, and vertical velocities, that govern cloud properties and that co-vary with GMST. Understanding these processes and accurately parameterizing them in global-scale climate models remain

major challenges to the atmospheric research and climate modeling communities.

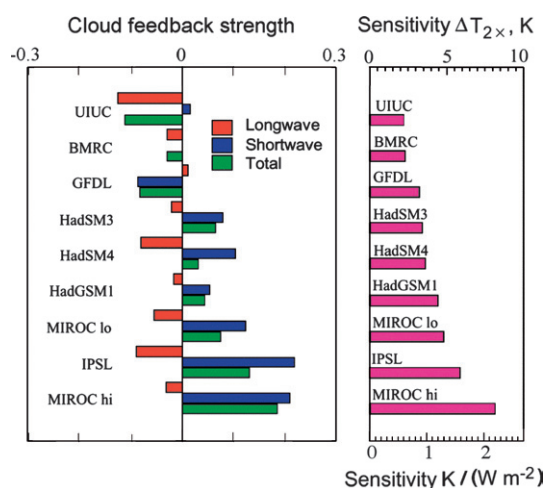
## Aerosols: the major source of uncertainty in radiative forcing

As noted above, accurate knowledge of radiative forcing of climate change is essential to evaluating the performance of climate models over the period for which there are reliable instrumental measurements. While there is confident knowledge of increases in the atmospheric mixing ratios of the long-lived GHGs from contemporary measurements or from ice cores, knowledge of aerosol forcing is much more uncertain, at present, and all the more so historically over the industrial period. Here the mechanisms of aerosol forcing are reviewed, the reasons for the uncertainty in aerosol forcing examined, and approaches to decreasing this uncertainty outlined.

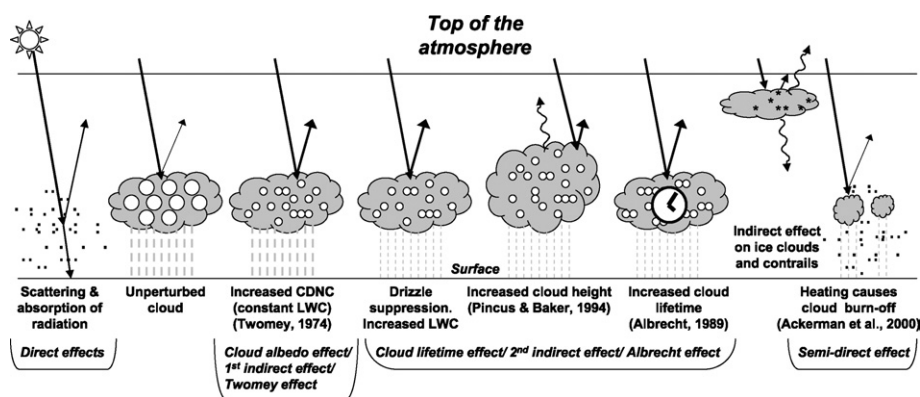
Atmospheric aerosols influence Earth's radiation budget, directly and indirectly, in a variety of ways. They scatter and absorb short- and longwave radiation and serve as the seed particles on which cloud drops and cloud ice particles form (cloud condensation nuclei, CCN; ice forming nuclei, IFN). Consequently, enhancement of the amount of atmospheric aerosols and modification of their properties by human activities have the potential for modification of climate by modifying the amount and locus of absorption of shortwave radiation (aerosol direct forcings), the properties of clouds, affecting their reflectance and persistence (aerosol indirect forcings), and the nature and locus of precipitation. These aerosol influences on radiation, clouds, and precipitation are illustrated schematically in Fig. 11.

As noted above, aerosols exhibit much greater spatial and temporal variability than greenhouse gases. Additionally, there is much heterogeneity in key aerosol properties that influence climate forcing: particle size, composition, and size-distributed composition, affecting light absorption and scattering, hygroscopic growth, and cloud nucleating ability; and particle morphology, affecting optical properties. In contrast to GHGs important aerosol properties affecting Earth's radiation budget evolve as a consequence of atmospheric processes. Removal processes are likewise highly variable in space and time, being governed mainly by precipitation for particles that are important contributors to radiative and cloud modifying effects. All of these differences make it much more difficult to characterize aerosol influences on radiation than is the case with greenhouse gases.

Approaches to estimating aerosol influences on Earth's radiation budget are based on observations, model calculations, or both. Recognition of the importance of aerosol forcing has led to enhanced measurement programs, mainly by remote sensing from the surface<sup>45</sup> and satellites.<sup>46,47</sup> Such remote sensing does not, however, immediately yield the aerosol properties needed to calculate forcing. Surface-based remote sensing yields an accurate measure of aerosol optical depth (column extinction attributable to aerosols) and, depending on the measurement approach, some measure of the angular distribution of light scattering necessary for calculating forcing, but forcing calculations still require assumptions on aerosol optical properties. Thus far as well, measurements are geographically quite limited, confined mainly to continental locations, and further question



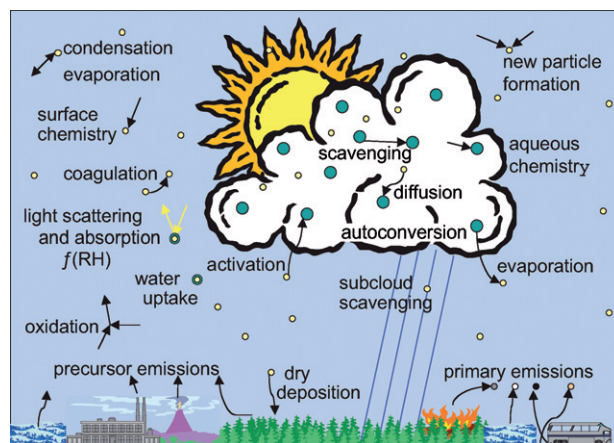
**Fig. 10** Influence of cloud feedback strength on sensitivity of current GCMs. Left panel shows total cloud feedback strength and long- and shortwave components in nine current climate models normalized to the inverse of the Stefan-Boltzmann sensitivity, taken as  $0.3 \text{ K W}^{-1} \text{ m}^2$ . Right panel shows modeled climate sensitivity in units of  $\text{K W}^{-1} \text{ m}^2$ , and as the equilibrium increase in global mean surface temperature that would result from a doubling of  $\text{CO}_2$ ,  $\Delta T_{2x}$ , evaluated for the  $\text{CO}_2$  doubling forcing  $F_{2x}$  taken as  $3.7 \text{ W m}^{-2}$ . Modified from ref. 39.



**Fig. 11** Schematic diagram showing the various mechanisms by which aerosols are thought to influence Earth's radiation budget. The small black dots represent aerosol particles; the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger drops, as only natural aerosols are available to serve as cloud condensation nuclei (CCN), whereas the perturbed cloud contains a greater number concentration of smaller cloud drops, as both natural and anthropogenic aerosols are available as CCN. The vertical gray dashes represent rainfall; LWC refers to the liquid water content. Source, ref. 1, Figure 2.10; modified from ref. 44.

can be raised as to whether the networks are sufficiently dense to accurately account for strong spatial gradients. Satellite measurements provide much more spatial coverage, but retrievals are mainly limited to radiance, which must be converted, through a model-based approach, to irradiance. Satellite measurements are often limited to one or two overpasses a day, limiting and perhaps biasing estimates of diurnal variability, as from relative humidity, for example. Inevitably there are issues of estimating forcing in partly cloudy scenes. A more intrinsic limitation to the observation-based approach is the inability to distinguish between natural and anthropogenic contributions, although some headway can be made from consideration of the size distribution (smaller aerosol particles ascribed to anthropogenic sources), where the size distribution is inferred from the wavelength dependence of the extinction and scattering. Measurement-based inferences of aerosol indirect effects are quite limited.

Model-based approaches in principle afford the possibility of determining forcing by anthropogenic aerosols as the difference between irradiance calculated with the total aerosol and with natural aerosol only. The approach is based on representing the processes that govern the geographical and temporal distribution of aerosols and the pertinent chemical and microphysical properties required to calculate aerosol forcing. Achieving this in turn requires that these processes and their dependence on controlling variables such as precursor gas concentrations, atmospheric photochemistry, and aerosol microphysics, be understood and that these processes and their consequences be accurately represented in cloud models, radiation transfer models, and ultimately in climate models examining changes in Earth's climate over the industrial period. The pertinent processes are shown schematically in Fig. 12. Aerosol particles are directly emitted as primary particles and are also formed secondarily by oxidation of emitted gaseous precursors. The production of low-volatility materials in this way results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does



**Fig. 12** Important aerosol processes that influence climate and which must be accurately represented in climate models. Modified from ref. 48.

not result in new particle formation. Particle composition and size distribution evolve by condensation and by surface and volume reactions and by coagulation. With increasing relative humidity, particles may accrete water vapor by deliquescence and further hygroscopic growth; with decreasing relative humidity, water is lost and ultimately particles may effloresce to the dry state. The uptake of water increases particle size, affecting also the particle optical properties. During cloud formation some fraction of aerosol particles serve as cloud condensation nuclei by becoming activated, that is, overcoming a free-energy barrier to form cloud droplets. Within clouds interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. Cloud droplets take up soluble gases which can react in solution to form nonvolatile products. If cloud droplets evaporate the nonvolatile material again becomes an aerosol, but with modified composition and size distribution; if the cloud precipitates the material is carried below the cloud and reaches the surface unless the precipitating drops completely



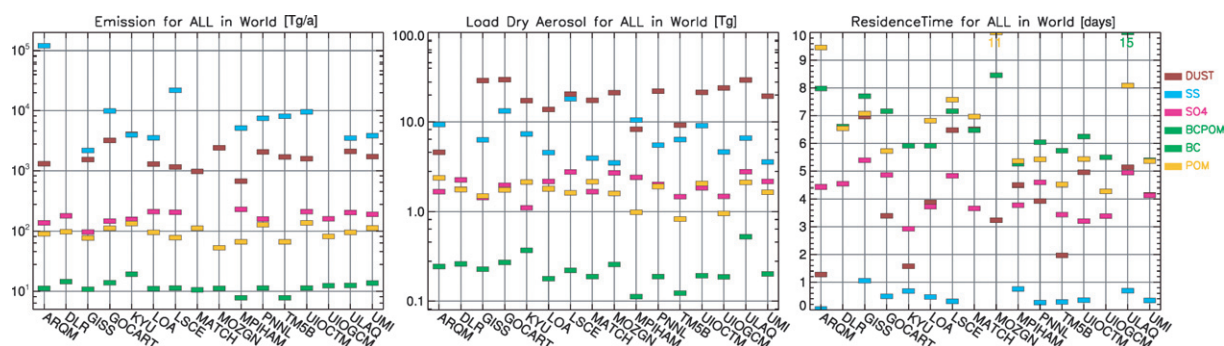
evaporate. Precipitation development in warm clouds occurs by autoconversion—coalescence of cloud droplets to form drops that are sufficiently large to fall by gravitational settling, initiating precipitation. Aerosol particles below precipitating clouds can also be removed from the atmosphere by impaction by precipitating drops and by dry deposition to the surface. These processes must be understood and their dependence on controlling variables identified in order to represent them initially in chemical transport models, to assess the accuracy of the model-based representation, and ultimately in climate models, to include these influences in assessments of climate change.

While the processes governing aerosol properties can be reasonably delineated, understanding is not sufficiently advanced that the aerosol properties can be modeled with confidence, for several reasons. There remain substantial uncertainties in the emissions of anthropogenic and natural primary aerosols and precursor gases. Atmospheric transformation and removal processes are likewise uncertain. Representations of aerosol size distribution and size distributed composition are highly parameterized, and calculation of aerosol optical and cloud nucleating properties from these parameterized distributions is therefore model dependent. These uncertainties are to some extent revealed in intercomparisons of chemical transport models. For example, a study carried out under the auspices of the AeroCom project<sup>49</sup> shows substantial model-to-model variation in global emissions, burdens, and turnover times of the several major aerosol components, Fig. 13. Intercomparisons of this sort, not just of extensive properties which vary with the amount of material but especially of intensive properties such as turnover time, forcing per optical depth, extinction per mass burden are particularly valuable diagnostics of the treatment of atmospheric processes and aerosol properties that can lead to model improvement. Comparisons of modeled aerosol concentrations with observations, especially those made in intensive campaigns in which detailed measurements are made of key aerosol properties and processes over large spatial domains are likewise essential and often reveal departures of up to several fold for major aerosol components, *e.g.*, ref. 50

Secondary organic aerosol, that is organic aerosol formed by atmospheric chemical reactions, has received much attention in

the past several years, as field studies<sup>51–54</sup> have shown formation rates to be as much as an order of magnitude greater than had been previously recognized and represented in models that had been developed from laboratory-based studies of aerosol production. Newly developed aerosol mass spectrometers have led to wide improvement in the ability to distinguish different classes of organic substances, and in particular to identify the amounts of primary (directly emitted) and secondary organic aerosol. These mass spectrometric measurements have shown secondary organic aerosols to be much more prevalent than had been previously recognized, comprising a major fraction of total submicrometre aerosol mass concentration at many locations throughout the Northern Hemisphere.<sup>55</sup> Laboratory studies identify the formation mechanisms, the rates of production of the materials, and the dependence of these rates on controlling variables. Importantly, recent laboratory studies<sup>56</sup> have identified previously unrecognized formation of organic aerosol from isoprene, a low molecular weight hydrocarbon emitted by deciduous vegetation; while the yield of aerosol per reacted isoprene is low, the high abundance of this precursor gas can result in substantial aerosol formation. One model study<sup>57</sup> indicates that the resulting aerosol may actually be the dominant aerosol species in the middle troposphere over a wide area globally. In sum, while these studies indicate major progress in understanding the processes responsible for aerosol forcing, they also suggest that this understanding is still in an exploratory stage that cannot yet be represented confidently in assessments of climate forcing.

Major advances are also being made in characterizing the microphysical dynamics that governs aerosol size distribution and size-distributed composition. Instruments that can measure the concentrations of newly formed particles down to diameters of 3 nm have shown that formation of new particles, which had previously been thought to be fairly rare, especially in and downwind of urban areas where there is much aerosol surface area to serve as a sink to newly produced condensable matter, occurs in bursts that can increase the total particle number concentration by an order of magnitude in two or three hours. Newly developed mass spectrometers that can determine the composition of these particles have shown that their composition can be dominated by organics.<sup>58</sup>



**Fig. 13** (a) Global, annual average emissions in 16 chemical transport models for dust, sea salt (SS), sulfate (SO<sub>4</sub>), black carbon (BC), and particulate organic matter (POM); for sulfate the sum of direct emission and chemical production is shown. (b) The global, annual average aerosol mass burden of the five aerosol species. (c) Tropospheric residence times (turnover times), evaluated as mass burden divided by removal rate. Modified from ref. 49, which identifies the several models.



Understanding and quantifying the dynamics that control aerosol size distribution is especially important to accurately evaluating aerosol indirect effects, which depend on the number concentration of aerosol particles affecting cloud microphysics.

Reducing the uncertainty in modeled aerosol forcing will require substantial effort in characterizing the emissions of primary particles, including their size-dependent composition, and precursor gases, understanding the atmospheric chemical reactions responsible for gas-to-particle conversion, the processes whereby aerosol composition and size distribution evolve, principally condensation and coagulation, and the processes whereby aerosol particles are removed from the atmosphere, mainly involving precipitation. It will be necessary also to develop confident relations between size-dependent composition and aerosol optical properties, including influence of relative humidity, and, similarly, confident understanding of the influence of the size-dependent composition of aerosol particles on the properties of clouds. Effort must be directed as well to developing improved understanding and numerical representation of the processes responsible for precipitation development governing, importantly, aerosol indirect influences on short- and longwave radiation and effects of aerosols on precipitation. A systematic approach is required<sup>48</sup> to develop this understanding and model-based representation that includes field measurements of aerosol properties and evolution and laboratory studies. Only after confident understanding is gained of these processes can they be confidently represented in hemispheric or global-scale models to permit evaluation of aerosol effects. Calculating aerosol forcing over the twentieth century necessary to evaluate the performance of climate models over this time period requires knowledge of space- and time-dependent emissions. Ultimately it would seem that aerosol processes need to be modeled interactively in climate models in order to account for feedbacks whereby aerosol processes influence radiation and hydrology that affect evolution of the climate system. For all of these reasons developing the understanding of these aerosol processes and representing them in climate models must be seen as a major challenge to understanding climate change, to determining Earth's climate sensitivity, and more broadly to developing the capability of determining the multiple responses of Earth's climate to prospective changes in atmospheric composition.

In view of the large uncertainty associated with estimates of aerosol forcing by observations or by chemical transport modeling, an alternative approach, inferring aerosol forcing by difference between GHG forcing and total forcing that is consistent with observed temperature change and an assumed climate sensitivity (so-called inverse calculation<sup>59</sup>) has gained favor among some investigators and has generally led to estimates of global mean aerosol forcing that are substantially less (in magnitude) than estimates by calculations based on process modeling or that are rooted in observed aerosol concentrations, so-called forward calculations, as described above. While inverse calculations are useful to assess the aerosol forcing that would be consistent with a given climate sensitivity, it needs to be stressed that the use of a forcing obtained in this way to infer climate sensitivity would constitute circular reasoning.<sup>60</sup>

## Empirical determination of climate sensitivity by single-compartment energy balance model

A new alternative empirical approach to the determination of Earth's climate sensitivity introduced by Schwartz<sup>61,62</sup> is based on the single-compartment energy balance model introduced above. This approach is briefly reviewed here. The basis of this approach is that the rate of change of the heat content of the climate system  $dH/dt$  may be related to the change in GMST as

$$\frac{dH}{dt} = C \frac{dT_s}{dt} \quad (18)$$

where  $C$  is the pertinent heat capacity. Here it must be stressed that  $C$  is an effective heat capacity that reflects only that portion of the global heat capacity that is coupled to the perturbation on the time scale of the perturbation. In the present context of global climate change induced by changes in atmospheric composition on the decade to century time scale the pertinent heat capacity is that which is subject to change in heat content on such time scales. Eqn (4), (9), (10), and (18) yield an equation solely in GMST:

$$C \frac{dT_s}{dt} = \gamma J_s - \varepsilon \sigma T_s^4 \quad (19)$$

For a small step-function radiative forcing  $F$  imposed at time, linearization of eqn (19) yields for the time-dependent change in global mean surface temperature normalized to the forcing

$$\Delta T_s(t)/F = S(1 - e^{-t/CS}), \quad (20)$$

where  $S$  is the equilibrium climate sensitivity given by eqn (15). It is seen that GMST approaches its new steady-state with time constant

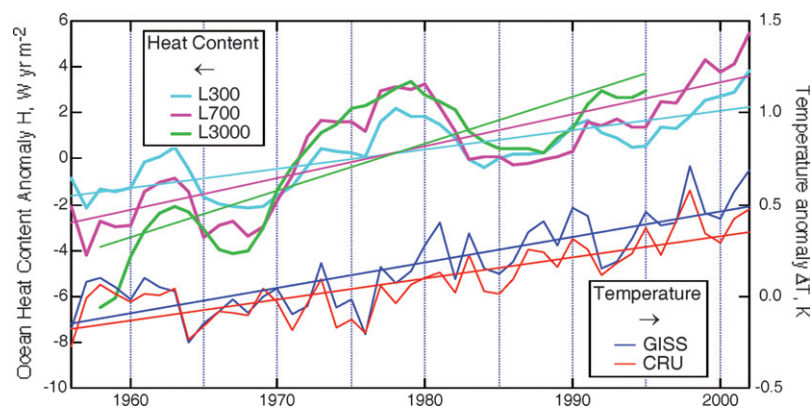
$$\tau = CS. \quad (21)$$

or alternatively that the climate sensitivity is related to the effective heat capacity of the system and the relaxation time constant as

$$S = \tau/C. \quad (22)$$

Eqn (22) suggests determining the climate sensitivity from the effective heat capacity of the climate system and the time constant characterizing relaxation of perturbations of global mean surface temperature provided that these quantities can be independently and empirically determined.

The effective heat capacity of the climate system was determined<sup>61</sup> as  $C = (dH/dt)/(dT_s/dt)$  (eqn (18)), where the time rate of change of global heat content and global mean surface temperature were obtained from measurements. The rate of change of global heat content was based on ocean temperature measurements as a function of depth over the past 50 years, as converted by Levitus *et al.*<sup>63</sup> into heat content anomaly of the global ocean to depths of 300 m, 700 m, and 3000 m, Fig. 14. Also shown are measurements of global mean surface temperature as tabulated by the Goddard Institute of Space Studies (GISS, NASA, USA, ref. 64) and the Climatic Research Unit (CRU, University of East Anglia, UK, ref. 43). An effective heat capacity, expressed per square metre of Earth surface, was derived for each depth



**Fig. 14** Determination of effective heat capacity of the global ocean as  $C = (dH_0/dt)/(dT_s/dt)$  where  $H_0$  is ocean heat content,  $T_s$  is global mean surface temperature, and  $t$  is time. L300, L700, and L3000 represent ocean heat content anomaly to depth of 300, 700, and 3000 m, respectively, from the compilation of Levitus *et al.*<sup>63</sup> GISS and CRU, respectively, represent global mean surface temperature from the compilations of the Goddard Institute for Space Studies<sup>64</sup> (updated at <http://data.giss.nasa.gov/gistemp/>) and the Climatic Research Unit of the University of East Anglia UK<sup>42</sup> (updated at <http://cdiac.esd.ornl.gov/trends/temp/jonescr/jones.html>). After Schwartz.<sup>61</sup>

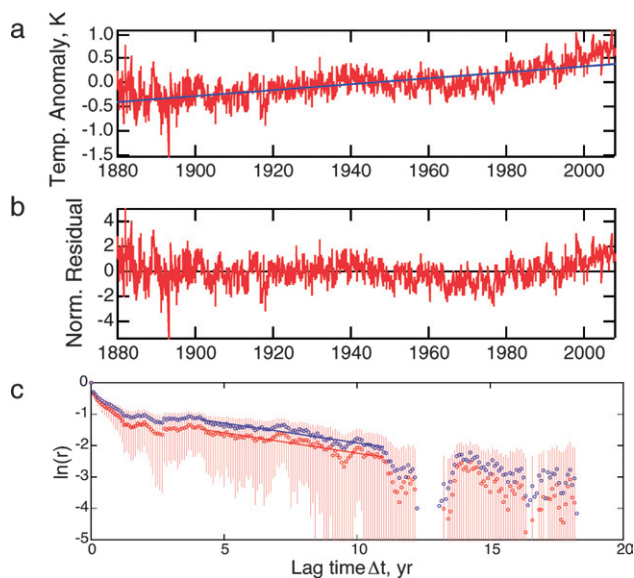
range:  $6.5 \pm 1.9$ ,  $10.4 \pm 3.4$  and  $14.0 \pm 5.9$   $\text{W y m}^{-2} \text{K}^{-1}$ ; almost half of the uptake of heat during the 50 year period is in the upper 300 m of the ocean, with relatively little heat penetrating below 700 m. Levitus *et al.*<sup>63</sup> estimate that heat uptake by the world's ocean comprises 84% of global heat uptake, with the balance comprised of roughly equal contributions from heating of terrestrial land masses, heating of the atmosphere, and melting of continental glaciers. Accounting for these further increases in Earth's heat content raises the estimate of the effective global heat capacity pertinent to increasing global temperature over the 50 year period to  $C = 17 \pm 7$   $\text{W y m}^{-2} \text{K}^{-1}$ .

The climate system time constant was determined<sup>61,62</sup> from analysis of the decrease in autocorrelation of time series of GMST as a function of lag time, Fig. 15; the relationship between autocorrelation and time constant, which follows from Einstein's fluctuation–dissipation theorem<sup>65,66</sup> can be written as

$$\tau = -1/[\text{dln}r(\Delta t)/\text{d}\Delta t], \quad (23)$$

where  $r(\Delta t)$  is the autocorrelation as a function of lag time  $\Delta t$ , the Pearson product-moment correlation coefficient of the time series with a copy of itself lagged by time  $\Delta t$ . Here, the temperature time series is the deseasonalized monthly mean GMST anomaly. As pointed out by Scafetta,<sup>67</sup> there is an initial rapid decrease in autocorrelation on a time scale of about half a year; the slower decrease in autocorrelation over a period of a decade or more, which is the time scale pertinent to climate change on the multidecadal time scale, exhibits a longer time constant  $\tau = 8.5 \pm 2.5$  years, where the estimate accounts for a slight correction due to the shortness of the time series, 128 years, and where the uncertainty estimate was derived from use of several data sets and several approaches to evaluation of  $\tau$ .

The empirically determined values of climate system heat capacity  $C$  and time constant  $\tau$  permit evaluation of climate sensitivity by eqn (22) as  $0.51 \pm 0.26$   $\text{K W}^{-1} \text{m}^2$ , corresponding for doubled  $\text{CO}_2$  forcing taken as  $F_{2\times} = 3.7$   $\text{W m}^{-2}$ , to an equilibrium temperature increase for doubled  $\text{CO}_2$   $\Delta T_{2\times} = 1.9 \pm 1.0$  K, somewhat lower than the range given in the IPCC 2007 assessment report,<sup>1</sup> but consistent within the mutual



**Fig. 15** Determination of time constant of Earth's climate system. (a) Time series of global monthly mean surface temperature data [GISS Land-Ocean Temperature Index data set] (ref. 64, updated at <http://data.giss.nasa.gov/gistemp/>); blue line denotes linear fit. (b) Normalized residual to fit in (a). (c) Semi-logarithmic plot of autocorrelation of normalized residual  $r$  as a function of lag time  $\Delta t$  and associated uncertainties as evaluated conventionally (red) and correcting for bias due to the short duration of the time series (blue), and associated slopes over the indicated range of lag time. Climate system time constant was evaluated from slope as  $\tau = -1/\text{dln}r(\Delta t)/\text{d}\Delta t$  is  $7.7 \pm 0.4$  and  $7.9 \pm 0.3$  years for the conventional and bias corrected data, respectively. After Schwartz.<sup>61,62</sup>

uncertainties. Additionally, the rather short time constant implies that global mean surface temperature is in near equilibrium with the applied forcing; that is, that there is little additional heating in the pipeline that would result from incremental  $\text{CO}_2$  now in the atmosphere.

While the simplicity of the approach to determining climate sensitivity by a single-compartment energy balance makes it seemingly attractive, this approach has been criticized as yielding

a time constant of the climate system that greatly underestimates the actual time scale of a climate system response to forcing, as not supported when applied to the output of climate models, and, ultimately, as implausible.<sup>68,69</sup> While large-scale climate models are essential to provide more refined projections of climate change than would be available from a single-compartment energy balance model, it would seem that an empirical approach such as this might usefully constrain climate models and reduce the uncertainty associated with estimates of global climate sensitivity.

## Discussion

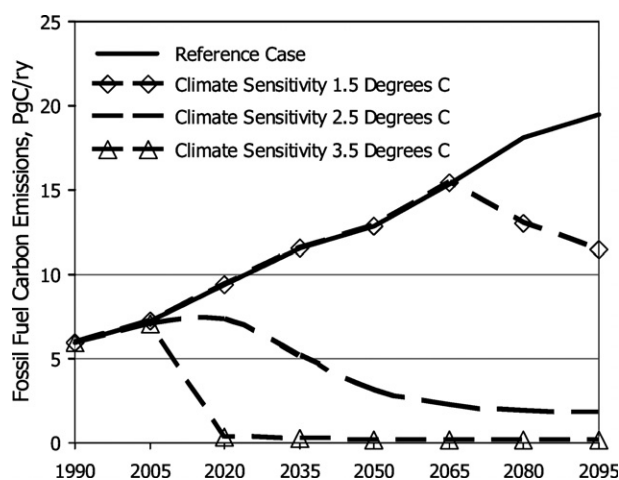
The increase in the atmospheric mixing ratio of CO<sub>2</sub> over the industrial period and the expected continued increase in CO<sub>2</sub> as a consequence of continued combustion of fossil fuels are confidently believed to have increased downwelling thermal infrared radiation from the atmosphere to Earth's surface. There is strong theoretical reason to believe that this radiative forcing would result in an increase in GMST, and this expected increase in GMST is borne out by observation. The Intergovernmental Panel on Climate Change in its 2007 Assessment Report<sup>1</sup> concluded that warming of Earth's climate system is unequivocal, that it is extremely unlikely that global climate change of the past fifty years can be explained without external forcing and very likely not due to known natural causes alone, and that continued greenhouse gas emissions at or above current rates would cause further warming and induce many changes in the global climate system during the 21st century that would very likely be larger than those observed during the 20th century.

The examination presented here of the uncertainty in Earth's climate sensitivity is in no way meant to call into question these main conclusions of the IPCC assessment report. Rather, the emphasis here has been on the uncertainty in the quantitative relation between forcing and climate system response, specifically, the change of global mean surface temperature per change in the emitted longwave flux or absorbed shortwave flux that control Earth's surface temperature and the energy balance of Earth's climate system: the magnitude of this uncertainty, the reasons that this uncertainty is so great at present, approaches to reduce this uncertainty, and consequences of this uncertainty. The IPCC Working Group characterized the magnitude of this uncertainty as the range of the estimate in temperature response for doubled CO<sub>2</sub>  $\Delta T_{2\times}$ , 2.0 to 4.5 K, with 66% probability of the actual value being within this range. Here it is argued that perhaps a more useful way of characterizing the uncertainty in climate sensitivity is as a multiplicative uncertainty, the ratio of the high end of the range to the low end, because such a multiplicative uncertainty immediately attaches to quantities that are proportional to the sensitivity. The multiplicative uncertainty that corresponds to the IPCC 66% likelihood range is thus a factor of 2.25; the arguments presented here suggest that the multiplicative uncertainty may actually be somewhat greater. Whatever the value of this uncertainty range, and uncertainties of highly uncertain quantities are inevitably difficult to characterize with great precision, it immediately attaches to the maximum incremental amount of CO<sub>2</sub> that can be introduced into the atmosphere subject to a constraint on increasing GMST or equivalently, on the amount of fossil fuel that can be burned,

without control of emissions by sequestration. There is thus a like uncertainty in the amount of this globally shared energy resource that is imposed by the uncertainty in climate sensitivity.

While the consequences of climate change involve many attributes of climate, not just global mean surface temperature, in considerations of strategies to limit climate change, these other consequences have generally been assumed to scale with, or expressed in terms of, increases in GMST, under the assumption that these further consequences become increasingly severe the greater the increase in GMST.<sup>2,70,71</sup> Such considerations have impelled development of strategies to limit the increase of GMST to some specified, mutually agreed upon value. It is thus clear that any strategy to achieve such a goal strongly depends on Earth's climate sensitivity, as setting a target level of CO<sub>2</sub> (or CO<sub>2</sub> equivalent, a target that takes into account the forcings of GHGs other than CO<sub>2</sub>, expressed as a mixing ratio of CO<sub>2</sub>) explicitly or implicitly requires an assumption of the value of Earth's climate sensitivity. The strong dependence of future temperature change on climate sensitivity was recently stressed by Hare and Meinshausen,<sup>72</sup> who noted on the basis of probabilistic assessments of temperature change the strong influence of climate sensitivity on increase in GMST for a given emissions pathway, concluding that climate sensitivity is "of quite fundamental significance for policy in general and specifically in relation to the question of long term warming commitments." The IPCC Working Group III similarly concludes in its 2007 report that "climate sensitivity is a key uncertainty for mitigation scenarios that aim to meet specific temperature levels," adding that "the timing and level of mitigation to reach a given temperature stabilization level is earlier and more stringent if climate sensitivity is high than if it is low" (ref. 4, p. 67).

The dependence of strategies for stabilizing Earth's temperature on climate sensitivity was explicitly examined by Edmonds and Smith,<sup>73</sup> who applied an integrated assessment model that takes into account population growth, economic growth, energy requirements, energy availability, alternative energy sources, and approaches to limitation of CO<sub>2</sub> emissions across various economic sectors globally to examine the means and costs of achieving various degrees of temperature stabilization for assumed values of climate sensitivity. The study posited a target maximum increase of GMST of 2 K above its preindustrial value and examined the approaches to and costs of stabilizing the increase of GMST to that level for three assumed values of climate sensitivity,  $\Delta T_{2\times} = 1.5, 2.5,$  and  $3.5$  K, that are within the range of current estimates. Not surprisingly, it was found that the assumed sensitivity plays a major role in determining the trajectory of maximum future emissions that would be required to stabilize the increase in GMST to 2 K above preindustrial, as shown in Fig. 16, which compares the several trajectories to that of a reference case based on assumed population increase and economic growth. For  $\Delta T_{2\times} = 1.5$  K it was found that it was not necessary to begin substantial reductions in emissions until after 2050. Such an extended time period would allow for phase-out of existing combustion facilities and introduction of new controlled emissions facilities in an economically effective manner. In contrast for  $\Delta T_{2\times} = 2.5$  K, stabilization of temperature required an early imposition of stringent reductions in CO<sub>2</sub> emissions, with the peak in emissions as early as 2020 and with reduction by the end of the century to below half of current emissions.



**Fig. 16** Carbon emissions pathways needed to achieve an increase in GMST prior to its preindustrial value not to exceed 2 K for three different climate sensitivity values, expressed as equilibrium increase in GMST for doubled atmospheric CO<sub>2</sub> mixing ratio  $\Delta T_{2\times}$ . Modified from ref. 73.

Edmonds and Smith also found that if  $\Delta T_{2\times}$  is 3.5 K or higher, it may be impossible to limit the increase in GMST to the 2 K target. These findings have economic implications as well. Whereas for climate sensitivity  $\Delta T_{2\times} = 1.5$  K, the cost of stabilization was viewed as rather trivial, for  $\Delta T_{2\times} = 2.5$  K, the discounted integrated cost of stabilization to 2 K increase in GMST was estimated as approximately 18 trillion US dollars (constant 1990 dollars); comparison of this figure to global annual GDP of \$30 trillion in 2002 gives a sense of the magnitude of effort that would be required to achieve this level of climate stabilization and the importance of climate sensitivity to estimating that cost and in turn as input to decision making.

Edmonds and Smith<sup>73</sup> explicitly addressed the important policy implications of the present uncertainty in climate sensitivity:

Policy decisions must be taken today in the context of profound uncertainty, a feature which highlights the usefulness of framing the problem in terms of risk management. From this perspective the presence of uncertainty is not a reason for inaction, but rather shapes the nature of near-term actions and recommends policies that provide flexibility in future actions. The uncertainty virtually guarantees that today's decisions will eventually be deemed inappropriate, but it is impossible to determine before the fact whether their inadequacy will be in too aggressively preserving other socially desirable resources at the expense of climate or climate at the expense of other socially desirable resources.

These considerations suggest that decreasing uncertainty in climate sensitivity would be of great economic and social value by leading to reduction in inappropriate decision making—decision making that errs either on the side of too little caution or too much caution—that would otherwise occur because of uncertainty in climate sensitivity. Based on the monetary costs of efforts to stabilize GMST to a 2 K rise for the different climate sensitivities it would appear that the value of early

knowledge of climate sensitivity could itself be in the trillions of dollars.

There are several key reasons for the present large uncertainty in climate sensitivity. The first, and perhaps most intrinsic, is the small fractional changes in temperature and in radiative fluxes that are of concern. An increase of 2 K in GMST, which is typical of values that have been suggested as characterizing the onset of dangerous climate change, represents an increase in GMST of less than one percent of its present value of 288 K. The entire increase over the industrial period in downwelling longwave flux from the atmosphere to the surface from the long lived greenhouse gases that is at the root of the present concern over climate change,  $2.6 \text{ W m}^{-2}$ , is likewise less than one percent of that total flux, about  $327 \text{ W m}^{-2}$ . A first-principles calculation, with a computer model of the climate system, of the effect of such a 1% change in flux to an accuracy of, say, 10 or 20% thus imposes an accuracy requirement of order 0.1 or 0.2%, respectively, with a similar requirement on the accuracy of evaluation of the model. Such an overall accuracy requirement makes quantifying climate sensitivity a truly tough scientific problem.

As discussed above, accurate determination of climate sensitivity is limited by other major hindrances. Much of the model-to-model difference in climate sensitivity (Fig. 10) arises from differences in treatment of clouds. Differences in treatment of clouds are also exhibited in the comparison of modeled albedo with observations (Fig. 7). Accurately representing clouds in models, even high resolution models, is inherently difficult as the condensed-phase water represents a small fraction of the total water substance in a given volume of air, being the difference between the actual water content and the equilibrium water content at the local temperature. Temperature is subject to variation as a consequence of vertical motions (adiabatic compression or expansion), latent heat uptake or release, and mixing; these processes are strongly coupled, as well, as latent heat exchange affects the buoyancy of a given air parcel, inducing vertical motions and mixing. Condensed water content is also controlled by precipitation, which is subtly dependent on cloud microphysical properties. Representation of clouds in current climate models is highly parameterized, in large part because of the large horizontal and vertical dimensions of grid cells in these models relative to the actual scales of the motions of the eddies that are responsible for cloud formation. It certainly seems that the accuracy of cloud parameterizations would benefit from much more detailed comparisons with observations. Recent work points to important new directions. One such direction is the so-called superparameterization approach of Randall *et al.*<sup>74</sup> in which a GCM grid cell would be divided into much smaller cells, say one tenth of the dimension of the primary cell, in each horizontal dimension, and the model equations integrated for these smaller cells, not for the entirety of the initial grid cell, but for a stripe across the cell, which would allow for communication between the smaller cells. The computational requirements for such an approach would be several orders of magnitude greater than for present GCMs, but advances in computational power may make such an approach practical. Recent work describing clouds at much higher resolution (100 m or less in the horizontal and vertical) has yielded a very realistic representation of deep clouds and mesoscale organization of convection in computations that are free of many of the parameterizations that are



required for GCMs.<sup>75</sup> While such scales are entirely impractical for global climate models, it may be possible to use studies on this scale to develop accurate parameterizations that can be employed in GCMs.

A further major hindrance to reducing uncertainty in climate sensitivity arises from uncertainty in climate forcing over the industrial period. Although some uncertainty arises from uncertainty in radiative forcing by CO<sub>2</sub> and other GHGs, the overwhelming contribution to this uncertainty comes from uncertainty in the radiative forcing by aerosols. This uncertainty limits both empirical inference of climate sensitivity from observed temperature change over the instrumental record and evaluation of the performance of climate models over this period. The lack of constraint on forcing over the period of instrumental record has led to the present situation in which the observed change in global mean temperature can be accurately reproduced by models whose sensitivities differ by more than a factor of 2, with the more sensitive models employing a lower forcing and *vice versa*. The ability of current models to represent twentieth century climate would be more accurately assessed as the envelope of change in GMST obtained by exercising each of the models over the uncertainty range of forcing.

Although both observational and modeling approaches have been brought to bear to determine aerosol forcing, these approaches have not converged, and in fact recent work, by identifying hitherto unrecognized contributions to aerosol burden and forcing, such as by secondary organic aerosols, has actually led to a situation in which the uncertainty in aerosol forcing has appeared to increase. Improving the representation of aerosol forcing in climate models would require substantial effort: measuring emissions of primary particles and precursor gases and developing emissions inventories; characterizing the rates of atmospheric reactions that produce condensable gases responsible for aerosol growth and the dependence of these rates on concentrations of precursor gases and other controlling variables; developing accurate treatments of aerosol physical evolution through condensation of gases and coagulation; and representing the interactions of aerosols with water vapor governing aerosol light scattering, absorption, and cloud drop nucleation. There is a strong coupling of aerosol forcing with cloud processes, as aerosol particles modify clouds and precipitation formation, affecting cloud albedo and persistence, and as clouds are central to removal of aerosol particles through precipitation and otherwise affect the composition and properties of aerosols through in cloud reactions; these couplings would need to be accurately represented in climate models. While substantially reducing uncertainty in aerosol forcing would require a great deal of effort,<sup>48</sup> this effort could lead to important payoff in two directions. First, representation of aerosol forcing in climate models would be much more greatly constrained than at present, permitting much more rigorous evaluation of the performance of climate models over the industrial period. Perhaps just as important, a tightly constrained aerosol forcing would permit much more confident empirical inference of climate sensitivity directly from measured temperature change and known forcing over the period of instrumental temperature record.<sup>25,76</sup>

Determination of aerosol forcing is essential also to permit informed decision making about greenhouse gas policy that takes into account the extent to which the positive (warming)

forcing due to increases in greenhouse gases has been offset by negative (cooling) forcing by anthropogenic aerosols. Knowledge of the extent of offset is very much at the heart of this decision making. Because of the short lifetime of tropospheric aerosols, about a week, compared to the long atmospheric residence times of excess CO<sub>2</sub> and of other long-lived GHGs, whatever fraction of the forcing by the GHGs that is being offset by aerosols, it is a week's worth of aerosols—ast week's emissions—that is offsetting that fraction of decades worth of GHG emissions. As has been shown in model studies (*e.g.*, ref. 9), an abrupt decrease in aerosol forcing, as might occur in conjunction with a major decrease in emissions of CO<sub>2</sub> would have the effect of abruptly increasing total (positive) forcing exacerbating, perhaps greatly, the climate change that the decrease in CO<sub>2</sub> emissions was intended to forestall. Quantification of aerosol forcing is also essential to interpretation of climate change under conditions of rapidly increasing emissions, such as has occurred in the past several years in China. To the extent that increased emissions of sulfur dioxide (the precursor of sulfate aerosol) scale with the increase in CO<sub>2</sub> emissions, then a new source, such as a new power plant, will initially exert a net cooling effect because the sulfate aerosol in a given pulse of emissions exerts its cooling influence immediately, whereas the CO<sub>2</sub> in the same pulse of emissions exerts its warming influence over decades. In the long run the warming influence of the CO<sub>2</sub> dominates, but at short times, perhaps a decade or more, the cooling influence of the sulfate aerosol dominates.<sup>77,78</sup>

The difficulties in determining climate sensitivity together with the renewed recognition<sup>37</sup> that errors in modeled feedback are amplified as the feedback strength approaches unity, have led to the suggestion by Allen and Frame,<sup>79</sup> perhaps facetious, that the quest for determining climate sensitivity be called off. Allen and Frame go on to state that “once the world has warmed by 4 °C, conditions will be so different from anything we can observe today... that it is inherently hard to say when the warming will stop.” Rather than determine climate sensitivity, they suggest stabilizing CO<sub>2</sub> at 450 ppm and letting our descendants find out whether this was the appropriate target and make necessary corrections.

Several arguments may be raised in opposition to this suggestion. First, Allen and Frame confuse the sensitivity, expressed as  $\Delta T_{2\times}$  with the actual equilibrium increase temperature that would result from a sustained doubled mixing ratio of CO<sub>2</sub>.  $\Delta T_{2\times}$ , as defined, is not an actual temperature change; rather it is Earth's climate sensitivity, a local derivative of GMST with forcing, defined at some climate state of interest, expressed in units of temperature change. Further, although it is not unlikely that sensitivity would change with GMST because of changes in the climate state, such a dependence should not be a reason for abandoning the sensitivity concept; rather it should be an impetus to determine that dependence. More intrinsically Allen and Frame suggest a target CO<sub>2</sub> mixing ratio be selected based on current understanding, for example 450 ppm, and that our descendants revise the target—upwards or downwards as needed—once they observe how much the climate has changed. How did Allen and Frame come up with such a target mixing ratio? It is based on their perception of the climate sensitivity, the amount of increase in GMST that would be acceptable, and the

ability of society to meet such a target. Developing a strategy to limit the increase in GMST to a target level, and even more so, developing an efficient strategy, requires knowledge of Earth's climate sensitivity.

This paper has not examined means of reducing emissions of greenhouse gases. Broadly speaking there are four approaches to this: reduction in energy use (from decrease in services and from enhanced end-use efficiency); use of non-carbon sources of energy; carbon capture and storage; and reduction in the emissions of GHGs other than CO<sub>2</sub> and of absorbing (soot) aerosols. Within these categories there is much opportunity for reducing emissions at costs which range from quite low (or even negative, that is, net reduction in costs) to substantial, as compared to the present situation in which costs of externalities are not borne by the emitter. It certainly seems that reduction in energy use and use of alternative sources of energy will become increasingly attractive for the explicit purpose of reducing CO<sub>2</sub> emissions in addition being driven by the increasing cost of carbon-based fuels. Prospects for these several approaches are examined and overviews of research in these areas are provided in ref. 3,80–83. An approach that is now receiving some attention<sup>84,85</sup> is large-scale removal of CO<sub>2</sub> from combustion streams or even from ambient air by the use of sorbents, which would need to be followed by long-term storage, as by conversion of silicate mineral to carbonate. Arguments for the technical and economic feasibility of such an approach have been presented by Lackner *et al.*,<sup>86</sup> and the removal of CO<sub>2</sub> from air has been demonstrated at the pilot level.<sup>87</sup> Such an approach as well, should it prove feasible, would afford the opportunity of rapidly reversing global warming by geoengineering the amount of CO<sub>2</sub> in the atmosphere as well as permitting continued reliance on fossil fuels to meet the world's energy needs.

The present paper has focused largely on forcing by CO<sub>2</sub>, but forcings by other gases, and by aerosols, also need to be considered in developing strategies to limit the increase in GMST, especially as limiting emissions by these other forcing agents might lessen constraints on CO<sub>2</sub> emissions and be more readily achievable. Here, attention is called to the suggestion of limiting emissions of absorbing aerosols, principally black carbon (soot),<sup>88,89</sup> for which atmospheric concentrations and forcing could, in principle, be rapidly reduced by stringent reductions of emissions because of the short atmospheric residence time of about a week. Clearly, reduction in emissions of methane and nitrous oxide would also contribute to reduction in warming forcing. In this context, attention would have to be paid to the possibility of inadvertent increases in emissions of N<sub>2</sub>O associated with enhanced use of nitrogen fertilizers as might result from cultivation of corn (maize) for ethanol production.<sup>90</sup> Nonetheless, the focus here on CO<sub>2</sub> would seem appropriate given its intrinsic relationship to energy production and the resultant inevitable connection between energy production and climate change.

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## References

- Intergovernmental Panel on Climate Change, *Climate Change 2007–The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the IPCC*, ed. S. Solomon, D. Qin, M. Manning, Z. Chen, M. C. Marquis, K. Averyt, *et al.*, Cambridge University Press, Cambridge, UK, World Meteorological Organization, Geneva, 2007, <http://www.ipcc.ch/ipccreports/ar4-wg1.htm>.
- Intergovernmental Panel on Climate Change, *Climate Change 2007–Impacts, Adaptation and Vulnerability, Contribution of Working Group II to the Fourth Assessment Report of the IPCC*, ed. M. L. Parry, O. F. Canziani, J. P. Palutikof, P. J. van der Linden and C. E. Hanson, World Meteorological Organization, Cambridge University Press, Cambridge, UK, World Meteorological Organization, Geneva, 2007, <http://www.ipcc.ch/ipccreports/ar4-wg2.htm>.
- Intergovernmental Panel on Climate Change, *Climate Change 2007–Mitigation of Climate Change, Contribution of Working Group III to the Fourth Assessment Report of the IPCC*, ed. B. Metz, O. R. Davidson, P. R. Bosch, R. Dave and L. A. Meyer, Cambridge University Press, Cambridge, UK, World Meteorological Organization, Geneva, 2007, <http://www.ipcc.ch/ipccreports/ar4-wg2.htm>.
- Intergovernmental Panel on Climate Change, *Climate Change 2007–Synthesis Report*, World Meteorological Organization, Geneva, 2007, [http://www.ipcc.ch/pdf/assessment-report/ar4/syr/ar4\\_syr.pdf](http://www.ipcc.ch/pdf/assessment-report/ar4/syr/ar4_syr.pdf).
- J. E. Hansen and S. Lebedeff, Global trends of measured surface air temperature, *J. Geophys. Res.*, 1987, **92**, 13345–13372.
- E. N. Lorenz, Deterministic nonperiodic flow, *J. Atmos. Sci.*, 1963, **20**, 130–141.
- D. Rind, The consequences of not knowing low- and high-latitude climate sensitivity, *Bull. Am. Meteorol. Soc.*, 2008, **89**, 855–864.
- S. J. Cox, W.-C. Wang and S. E. Schwartz, Climate response to radiative forcings by aerosols and greenhouse gases, *Geophys. Res. Lett.*, 1995, **22**, 2509–2512.
- G. P. Brasseur and E. Roeckner, Impact of improved air quality on the future evolution of climate, *Geophys. Res. Lett.*, 2005, **32**, L23704.
- M. Engardt and H. Rodhe, A comparison between patterns of temperature trends and sulfate aerosol pollution, *Geophys. Res. Lett.*, 1993, **20**, 117–120.
- Intergovernmental Panel on Climate Change, *Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the IPCC*, ed. J. T. Houghton, Y. Ding, D. J. Griggs, M. Noguer, P. van der Linden, X. Dai *et al.*, Cambridge Univ. Press, Cambridge, UK, 2001, [http://www.grida.no/climate/ipcc\\_tar/wg1/index.htm](http://www.grida.no/climate/ipcc_tar/wg1/index.htm).
- S. Arrhenius, On the influence of carbonic acid in the air upon the temperature of the ground, *Phil. Mag.*, 1896, **41**(251), 237–276.
- J. G. Charney, A. Arakawa, D. J. Baker, B. Bolin, R. E. Dickinson, R. M. Goody *et al.*, *Carbon Dioxide and Climate: A Scientific Assessment*, National Academy of Sciences, Washington DC, 1979.
- G. C. Hegerl, T. Crowley, W. T. Hyde and D. Frame, Constraints on climate sensitivity from temperature reconstructions of the past seven centuries, *Nature*, 2006, **440**, 1029–1032.
- United Nations, *United Nations Framework Convention on Climate Change*, 1992, <http://www.unfccc.int/resource/docs/convkp/conveng.pdf>.
- Council of the European Union, *Community Strategy on Climate Change–Council Conclusions*. 1939th Council Meeting Environment, Document Nr. 8518/96, Luxembourg, 1996, [http://ue.eu.int/ueDocs/cms\\_Data/docs/pressData/en/envir/011a0006.htm](http://ue.eu.int/ueDocs/cms_Data/docs/pressData/en/envir/011a0006.htm).
- B. C. O'Neill and M. Oppenheimer, Dangerous climate impacts and the Kyoto protocol, *Science*, 2002, **296**, 1971–1972.
- J. Hansen, Defusing the global warming time bomb, *Sci. Am.*, 2004, **290**(3), 68–77.
- International Scientific Steering Committee, *Avoiding Dangerous Climate Change: International Symposium on the Stabilization of Greenhouse Gas Concentrations*, Hadley Centre, Met Office, Exeter, UK, 2005, [http://www.stabilisation2005.com/Steering\\_Committee\\_Report.pdf](http://www.stabilisation2005.com/Steering_Committee_Report.pdf).
- S. E. Schwartz, The Whitehouse Effect–Shortwave radiative forcing of climate by anthropogenic aerosols: An overview, *J. Aerosol. Sci.*, 1996, **27**, 359–382.

- 21 V. Ramanathan, The role of earth radiation budget studies in climate and general circulation research, *J. Geophys. Res.*, 1987, **92**, 4075–4095.
- 22 R. Kandel and M. Viollier, Planetary radiation budgets, *Space Sci. Rev.*, 2005, **120**, 1–26.
- 23 R. W. Wood, Note on the theory of the greenhouse, *Phil. Mag.*, 1909, **17**, 319–320.
- 24 C. D. Keeling and T. P. Whorf, Atmospheric CO<sub>2</sub> records from sites in the SIO air sampling network, in *Trends: A Compendium of Data on Global Change*, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, TN, 2005, <http://cdiac.esd.ornl.gov/trends/CO2/sio-mlo.htm>.
- 25 J. M. Gregory, R. J. Stouffer, S. C. B. Raper, P. A. Stott and N. A. Rayner, An Observationally based estimate of the climate sensitivity, *J. Climate*, 2002, **15**, 3117–3121.
- 26 C. K. Folland, N. A. Rayner, S. J. Brown, T. M. Smith, S. S. P. Shen, D. E. Parker and et al., Global temperature change and its uncertainties since 1861, *Geophys. Res. Lett.*, 2001, **28**, 2621–2624.
- 27 G. A. Meehl, W. M. Washington, W. D. Collins, J. M. Arblaster, A. Hu, E. Lawrence and et al., How much more global warming and sea level rise?, *Science*, 2005, **307**, 1769–1772.
- 28 T. M. L. Wigley, The climate change commitment, *Science*, 2005, **307**, 1766–1770.
- 29 P. Friedlingstein and S. Solomon, Contributions of past and present human generations to committed warming caused by carbon dioxide, *Proc. Nat. Acad. Sci. U. S. A.*, 2005, **102**, 10832–10836.
- 30 J. Hansen, L. Nazarenko, R. Ruedy, M. Sato, J. Willis and A. D. Genio et al., Earth's energy imbalance: Confirmation and implications, *Science*, 2005, **308**, 1431–1435.
- 31 H. N. Pollack, S. J. Hunter and J. R. Johnson, Heat flow from the earth's interior: Analysis of the global data set, *Rev. Geophys.*, 1993, **31**, 267–280.
- 32 C. Jaupart, S. Labrosse and J. C. Mareschal, Temperatures, Heat and Energy in the Mantle of the Earth, in *Treatise on Geophysics*, ed. D. Bercovici, Mantle Dynamics, American Geophysical Union, Washington DC, 2007, vol. 7, pp. 253–303.
- 33 BP, *Statistical Review of World Energy*, 2007, [http://www.bp.com/liveassets/bp\\_internet/globalbp/globalbp\\_uk\\_english/reports\\_and\\_publications/statistical\\_energy\\_review\\_2007/STAGING/local\\_assets/downloads/pdf/statistical\\_review\\_of\\_world\\_energy\\_full\\_report\\_2007.pdf](http://www.bp.com/liveassets/bp_internet/globalbp/globalbp_uk_english/reports_and_publications/statistical_energy_review_2007/STAGING/local_assets/downloads/pdf/statistical_review_of_world_energy_full_report_2007.pdf).
- 34 J. Hansen, A. Lacis, D. Rind, G. Russell, P. Stone, I. Fung et al., Climate Sensitivity: Analysis of Feedback Mechanisms, in *Climate Processes and Climate Sensitivity*, AGU Geophysical Monograph 29, Maurice Ewing Vol. 5, ed. J. E. Hansen and T. Takahashi, American Geophysical Union, Washington DC, 1984, pp. 130–163.
- 35 R. D. Cess, G. L. Potter, J. P. Blanchet, G. J. Boer, S. J. Ghan and J. T. Kiehl et al., Interpretation of cloud climate feedback is produced by 14 atmospheric general circulation models, *Science*, 1989, **245**, 513–516.
- 36 R. S. Lindzen and C. Giannitsis, On the climatic implications of volcanic cooling, *J. Geophys. Res.*, 1998, **103**, 5929–5941.
- 37 G. H. Roe and M. B. Baker, Why is climate sensitivity so unpredictable?, *Science*, 2007, **318**, 629–632.
- 38 F. A. M. Bender, H. Rodhe, R. J. Charlson, A. M. L. Ekman and N. Loeb, 22 views of the global albedo - Comparison between 20 GCMs and two satellites, *Tellus*, 2006, **58**, 320–330.
- 39 M. J. Webb, C. A. Senior, D. M. H. Sexton, W. J. Ingram, K. D. Williams and M. A. Ringer et al., On the contribution of local feedback mechanisms to the range of climate sensitivity in two GCM ensembles, *Climate Dyn.*, 2006, **27**, 17–38.
- 40 W. D. Collins, V. Ramaswamy, M. D. Schwarzkopf, Y. Sun, R. W. Portmann and Q. Fu et al., Radiative forcing by well-mixed greenhouse gases: Estimates from climate models in the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4), *J. Geophys. Res.*, 2006, **111**, D14317.
- 41 P. Brohan, J. J. Kennedy, I. Harris, S. F. B. Tett and P. D. Jones, Uncertainty estimates in regional and global observed temperature changes: A new dataset from 1850, *J. Geophys. Res.*, 2006, **111**, D12106.
- 42 S. E. Schwartz, R. J. Charlson and H. Rodhe, Quantifying climate change - Too rosy a picture?, *Nature Reports - Climate Change*, 2007, **1**, 23–24.
- 43 J. T. Kiehl, Twentieth century climate model response and climate sensitivity, *Geophys. Res. Lett.*, 2007, **34**, L22710.
- 44 J. M. Haywood and O. Boucher, Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review, *Rev. Geophys.*, 2000, **38**, 513–543.
- 45 B. N. Holben, D. Tanré, A. Smirnov, T. F. Eck, I. Slutsker and N. Abuhassan et al., An emerging ground-based aerosol climatology: Aerosol optical depth from AERONET, *J. Geophys. Res.*, 2001, **106**, 12067–12097.
- 46 Y. J. Kaufman, D. Tanré and O. Boucher, A satellite view of aerosols in the climate system, *Nature*, 2002, **419**, 215–223.
- 47 H. Yu, Y. J. Kaufman, M. Chin, G. Feingold, L. A. Remer and T. L. Anderson et al., A review of measurement-based assessments of the aerosol direct radiative effect and forcing, *Atmos. Chem. Phys.*, 2006, **6**, 613–666.
- 48 S. J. Ghan and S. E. Schwartz, Aerosol properties and processes: A path from field and laboratory measurements to global climate models, *Bull. Am. Meteorol. Soc.*, 2007, **88**, 1059–1083.
- 49 C. Textor, M. Schulz, S. Guibert, S. Kinne, Y. Balkanski and S. Bauer et al., Analysis and quantification of the diversities of aerosol life cycles within AeroCom, *Atmos. Chem. Phys.*, 2006, **6**, 1777–1813.
- 50 T. S. Bates, T. L. Anderson, T. Baynard, T. Bond, O. Boucher and G. Carmichael et al., Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and North Indian Oceans: Estimates based on in-situ chemical and optical measurements and chemical transport modeling, *Atmos. Chem. Phys.*, 2006, **6**, 1657–1732.
- 51 C. L. Heald, D. J. Jacob, R. J. Park, L. M. Russell, B. J. Huebert and J. H. Seinfeld et al., A large organic aerosol source in the free troposphere missing from current models, *Geophys. Res. Lett.*, 2005, **32**, L18809.
- 52 J. A. De Gouw, A. M. Middlebrook, C. Warneke, P. D. Goldan, W. C. Kuster and J. M. Roberts et al., Budget of organic carbon in a polluted atmosphere: Results from the New England Air Quality Study in 2002, *J. Geophys. Res.*, 2005, **110**, D16305.
- 53 R. Volkamer, J. L. Jimenez, F. S. Martini, K. Dzepina, Q. Zhang and D. Salcedo et al., Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected, *Geophys. Res. Lett.*, 2006, **33**, L17811.
- 54 L. I. Kleinman, S. R. Springston, P. H. Daum, Y. N. Lee, L. J. Nunnermacker and G. I. Senum et al., The time evolution of aerosol composition over the Mexico City plateau, *Atmos. Chem. Phys.*, 2008, **8**, 1559–1575.
- 55 Q. Zhang, J. L. Jimenez, M. R. Canagaratna, J. D. Allan, H. Coe and I. Ulbrich et al., Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced northern hemisphere midlatitudes, *Geophys. Res. Lett.*, 2007, **34**, L13801.
- 56 J. H. Kroll, N. L. Ng, S. M. Murphy, R. C. Flagan and J. H. Seinfeld, Secondary organic aerosol formation from isoprene photooxidation, *Environ. Sci. Technol.*, 2006, **40**, 1869–1877.
- 57 D. K. Henze and J. H. Seinfeld, Global secondary organic aerosol from isoprene oxidation, *Geophys. Res. Lett.*, 2006, **33**, L09812.
- 58 J. N. Smith, M. J. Dunn, T. M. VanReken, K. Iida, M. R. Stolzenburg and P. H. McMurry et al., Chemical composition of atmospheric nanoparticles formed from nucleation in Tecamac, Mexico: Evidence for an important role for organic species in nanoparticle growth, *Geophys. Res. Lett.*, 2008, **35**, L04808.
- 59 T. L. Anderson, R. J. Charlson, S. E. Schwartz, R. Knutti, O. Boucher and H. Rodhe et al., Climate forcing by aerosols—A hazy picture, *Science*, 2003, **300**, 1103–1104.
- 60 H. Rodhe, R. J. Charlson and T. L. Anderson, Avoiding circular logic in climate modeling, *Climatic Change*, 2000, **44**, 419–422.
- 61 S. E. Schwartz, Heat capacity, time constant, and sensitivity of Earth's climate system, *J. Geophys. Res.*, 2007, **112**, D24S05.
- 62 S. E. Schwartz, Reply to Comments by G. Foster et al., R. Knutti et al., and N. Scafetta on “Heat capacity, time constant, and sensitivity of Earth's climate system”, *J. Geophys. Res.*, 2008, **113**, p. D15105.
- 63 S. Levitus, J. Antonov and T. Boyer, Warming of the world ocean, 1955–2003, *Geophys. Res. Lett.*, 2005, **32**, L02604.
- 64 J. Hansen, R. Ruedy, M. Sato and R. Reynolds, Global surface air temperature in 1995: Return to pre-Pinatubo level, *Geophys. Res. Lett.*, 1996, **23**, 1665–1668.
- 65 A. Einstein, Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen, *Ann. Phys.*, 1905, **17**, 549–560.

- 66 C. E. Leith, Climate response and fluctuation dissipation, *J. Atmos. Sci.*, 1975, **32**, 2022–2026.
- 67 N. Scafetta, Comment on “Heat capacity, time constant, and sensitivity of Earth’s climate system” by S. E. Schwartz, *J. Geophys. Res.*, 2008, **113**, p. D15104.
- 68 G. Foster, J. D. Annan, G. A. Schmidt and M. E. Mann, Comment on “Heat capacity, time constant, and sensitivity of Earth’s climate system” by S. E. Schwartz, *J. Geophys. Res.*, 2008, **113**, p. D15102.
- 69 R. Knutti, S. Krähenmann, D. J. Frame and M. R. Allen, Comment on “Heat capacity, time constant, and sensitivity of Earth’s climate system” by S. E. Schwartz, *J. Geophys. Res.*, 2008, **113**, p. DD15103.
- 70 W. Nordhaus and J. G. Boyer, *Roll the DICE Again: Economic Models of Global Warming*, MIT Press, Cambridge, MA, 1999, <http://www.econ.yale.edu/~nordhaus/homepage/web%20table%20of%20contents%20102599.htm>.
- 71 N. Stern, S. Peters, V. Bakhshi, A. Bowen, C. Cameron, S. Catovsky et al., *Stern Review: The Economics of Climate Change*, HM Treasury, Cambridge University Press, London, 2006, [http://www.hm-treasury.gov.uk/independent\\_reviews/stern\\_review\\_economics\\_climate\\_change/stern\\_review\\_report.cfm](http://www.hm-treasury.gov.uk/independent_reviews/stern_review_economics_climate_change/stern_review_report.cfm).
- 72 B. Hare and M. Meinshausen, How much warming are we committed to and how much can be avoided?, *Climatic Change*, 2006, **75**, 111–149.
- 73 J. Edmonds and S. Smith, The Technology of Two Degrees, in *Avoiding Dangerous Climate Change*, ed. H. J. Schellenhuber, W. Cramer, N. Nakicenovic, T. Wigley and G. Yohe, Cambridge University Press, 2006, pp. 385–392.
- 74 D. A. Randall, M. Khairoutdinov, A. Arakawa and W. Grabowski, Breaking the cloud-parameterization deadlock, *Bull. Am. Meteorol. Soc.*, 2003, **84**, 1547–1564.
- 75 M. F. Khairoutdinov and D. A. Randall, High-resolution simulation of shallow-to-deep convection transition over land, *J. Atmos. Sci.*, 2006, **63**, 3421–3436.
- 76 S. E. Schwartz, Uncertainty requirements in radiative forcing of climate change, *J. Air Waste Manage. Assoc.*, 2004, **54**, 1351–1559.
- 77 S. E. Schwartz, Does fossilfuel combustion lead to global warming?, *Energy Int. J.*, 1993, **18**, 1229–1248.
- 78 K. P. Shine, J. S. Fuglestedt, K. Hailemariam and N. Stuber, Alternatives to the global warming potential for comparing climate impacts of emissions of greenhouse gases, *Climate Change*, 2005, **68**, 281–302.
- 79 M. R. Allen and D. J. Frame, Call off the quest, *Science*, 2007, **318**, 582–583.
- 80 U.S. Climate Change Technology Program, *Strategic Plan*, U.S. Department of Energy, Washington, 2006, <http://www.climatechange.gov/stratplan/final/CCTP-StratPlan-Sep-2006.pdf>.
- 81 Royal Society of Chemistry, Chemical Science Priorities for Sustainable Energy Solutions, 2005, [http://www.rsc.org/images/ChemicalSciencePrioritiesSustainableEnergySolutions\\_tcm18-12642.pdf](http://www.rsc.org/images/ChemicalSciencePrioritiesSustainableEnergySolutions_tcm18-12642.pdf).
- 82 J. Creyts, A. Derkach, S. Nyquist, K. Ostrowski and J. Stephenson, *Reducing U.S. Greenhouse Gas Emissions: How Much at What Cost? Executive Report; U.S. Greenhouse Gas Abatement Mapping Initiative*, McKinsey & Company Analysis, Washington, DC, 2007, <http://mckinsey.com/clientervice/ccsi/greenhousegas.asp>.
- 83 M. C. MacCracken, Prospects for future climate change and the reasons for early action, *J. Air Waste Manage. Assoc.*, 2008, **58**, 735–786.
- 84 W. S. Broecker, CO<sub>2</sub> Arithmetic, *Science*, 2007, **315**, 1371.
- 85 R. Lal, Sequestration of atmospheric CO<sub>2</sub> in global carbon pools, *Energy Environ. Sci.*, 2008, **1**, 86–100.
- 86 K. S. Lackner, P. Grimes and H. J. Ziock, Capturing Carbon Dioxide From Air, in *First National Conference on Carbon Sequestration*, 2001, [http://www.netl.doe.gov/publications/proceedings/01/carbon\\_seq/carbon\\_seq01.html](http://www.netl.doe.gov/publications/proceedings/01/carbon_seq/carbon_seq01.html).
- 87 J. K. Stolaroff, D. W. Keith and G. V. Lowry, Carbon dioxide capture from atmospheric air using sodium hydroxide spray, *Environ. Sci. Technol.*, 2008, **42**, 2728–2735.
- 88 M. Sato, J. Hansen, D. Koch, A. Lacis, R. Ruedy, O. Dubovik and et al., Global atmospheric black carbon inferred from AERONET, *Proc. Natl. Acad. Sci. U. S. A.*, 2003, **100**, 6319–6324.
- 89 V. Ramanathan and G. Carmichael, Global and regional climate changes due to black carbon, *Nature Geoscience*, 2008, **1**, 221–337.
- 90 P. J. Crutzen, A. R. Mosier, K. A. Smith and W. Winiwarter, N<sub>2</sub>O release from agro-biofuel production negates global warming reduction by replacing fossil fuels, *Atmos. Chem. Phys.*, 2008, **8**, 389–395.