ATMOSPHERIC SCIENCE

Climate Forcing by Aerosols—a Hazy Picture

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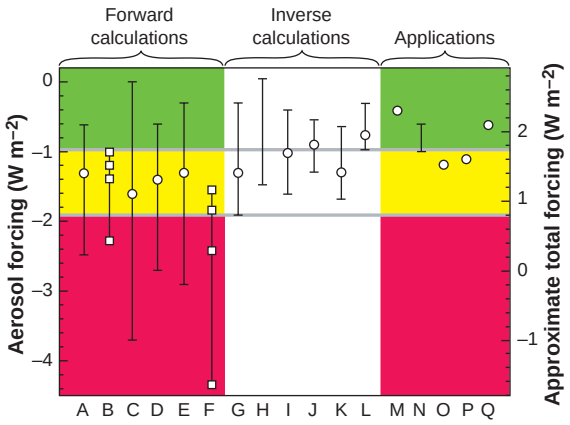
The global average surface temperature has risen by 0.6 K since the late 19th century. Ocean heat content has increased, and other climate indices also point to a warming world. Many studies have attributed this warming largely to top-of-atmosphere radiative forcing—a change in planetary heat balance between incoming solar radiation and outgoing infrared radiation—by anthropogenic greenhouse gases (GHGs) (*1*, *2*).

Such attribution studies compare temperature observations to climate model simulations forced by various industrial-era agents. Among these agents, GHGs have well-constrained positive forcings (creating a warming influence) (*3*). In contrast, the mostly negative forcings (cooling) associated with anthropogenic aerosols are highly uncertain (*3*, *4*).



Uncertainties in aerosol forcings. Global-mean anthropogenic aerosol forcing over the industrial era (left axis) as estimated by forward (A to F) and inverse (G to L) calculations and as used in applications (M to Q) (*20*). Circles with error bars are central values and 95% confidence limits. Bare error bars are stated range. Squares represent specific forcing calculations using alternative formulations within the same study. Right axis: Total forcing over the industrial era using the approximation that nonaerosol forcings are 2.7 W m^{-2} (*3*, *4*).

Different forcings have different spatio-temporal patterns; however, model studies indicate that climate sensitivity (the ratio of global mean equilibrium temperature response to global mean forcing) is approximately equal for almost all of the major forcing agents (*3*). Thus, total forcing (the global mean sum of all industrial-era forcings) is a widely used diagnostic parameter.

Here we argue that the magnitude and uncertainty of aerosol forcing may affect the magnitude and uncertainty of total forcing to a degree that has not been adequately considered in climate studies to date. Inferences about the causes of surface warming over the industrial period and about climate sensitivity may therefore be in error.

Anthropogenic aerosol forcings arise from multiple aerosol components and various forcing mechanisms. The sum of these forcings has been calculated by two independent methods. First, forward calculations are based on knowledge of the pertinent aerosol physics and chemistry. Second, inverse calculations infer aerosol forcing from the total forcing required to match climate model simulations with observed temperature changes.

Inverse calculations are based on the premise that the observed warming is caused by a positive total forcing over the industrial era (rather than by natural variability and/or unrecognized forcings). They constrain aerosol forcing to around –1 W m^{-2}, with uncertainties that extend no farther than –1 to –1.9 W m^{-2}, depending on the study (see the figure). Aerosol forcing determined by the forward calculations is considerably greater, centered around –1.5 W m^{-2}, with an uncertainty range that extends beyond –3 W m^{-2}. The larger magnitude aerosol forcings from the forward calculations greatly exceed the largest values allowed by the inverse calculations (see colored bands in the figure).

The substantial region of inconsistency shown in the figure (the red and, depending on the study, yellow bands) implies either that the large-magnitude aerosol forcings from the forward calculations are erroneously high or, alternatively, that the limits on aerosol-forcing magnitude inferred from the inverse calculations are erroneously low. We caution against simply assuming the former. The forward calculations are based on a substantial body of aerosol and cloud measurements, observation-based parameterizations of aerosol-cloud interactions, and well-understood physics of radiative transfer.

The inverse calculations are also based on sound physical principles. However, to the extent that climate models rely on the results of inverse calculations, the possibility of circular reasoning arises (*5*)—that is, using the temperature record to derive a key input to climate models that are then tested against the temperature record. Rather than rely exclusively on one ap-

Reflection of sunlight by aerosols. The southeast coast of China and the island of Taiwan viewed toward the southwest from the Space Shuttle at an altitude of 278 km above Okinawa, Japan. An aerosol plume (between arrows) is carried by northwest winds from China a distance of more than 600 km over the ocean; small clouds are embedded in the plume. Albedo enhancement is evident over the ocean, and indirect effects on clouds are possible.

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proach or the other, it is prudent to acknowledge the current inconsistency and seek to understand and resolve it.

Unfortunately, virtually all climate model studies that have included anthropogenic aerosol forcing as a driver of climate change (diagnosis, attribution, and projection studies; denoted "applications" in the figure) have used only aerosol forcing values that are consistent with the inverse approach. If such studies were conducted with the larger range of aerosol forcings determined from the forward calculations, the results would differ greatly.

The forward calculations raise the possibility that total forcing from preindustrial times to the present (right axis in the figure) has been small or even negative. If this is correct, it would imply that climate sensitivity and/or natural variability (that is, variability not forced by anthropogenic emissions) is much larger than climate models currently indicate.

Although even the sign of the current total forcing is in question, the sign of the forcing by the middle of the 21st century will certainly be positive. The reason is that GHGs accumulate in the atmosphere, whereas aerosols do not. Even if the most negative value of aerosol forcing shown in the figure turns out to be correct, the current range of plausible emissions scenarios (*6*) indicates that GHG forcing will exceed aerosol forcing somewhere between 2030 and 2050. Thus, despite current uncertainties, forward calculations lead to the unambiguous conclusion that anthropogenic activity will inevitably result in a strong, positive forcing of Earth's climate system.

In addressing the critical question of how the climate system will respond to this positive forcing, researchers must seek to resolve the present disparity between forward and inverse calculations. Until this is achieved, the possibility that most of the warming to date is due to natural variability, as well as the possibility of high climate sensitivity, must be kept open.

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20. Forward calculations: (A) statistical sum by (*4*) of the radiative forcings given in (*3*); (B) sulfate-only forcings by (*7*); (C) (*8*); (D) (*9*); (E) (*10*); (F) indirect (that is, cloud-mediated) effects only by (*11*). Inverse calculations: (G) (*12*); (H) (*13*); (I) (*14*); (J) (*15*); (K) (*16*); (L): (*17*). Applications, anthropogenic aerosol forcing as used in (M) the detection/attribution study by (*2*); (N) detection/attribution review by (*1*); (O) climate projections by (*6*); (P) detection/attribution study by (*18*); (Q) forcing/response diagnostic study by (*19*).

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MATERIALS SCIENCE

Molecular Fuel Tanks

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Concern about finite petroleum reserves and climate change has reinvigorated interest in alternative fuel technologies. Among these, hydrogen-based fuel cells have attracted considerable attention for power generation. A hydrogen-based economy, however, hinges on systems that guarantee safe transport and storage of the hydrogen fuel.

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On page 1127 of this issue, Rosi *et al.* (*1*) report an important advance toward safe hydrogen storage. They describe the adsorption of hydrogen by porous metal-organic frameworks (MOFs). The storage capacities of the materials are not yet sufficiently high for practical applications, but the ability to modify the organic components of the frameworks offers a unique opportunity for increasing the hydrogen uptake. Furthermore, the authors show how the chemical and structural factors that govern hydrogen storage—including the adsorption sites—can be probed in these porous molecular materials.

Hydrogen has several advantages as a fuel. Its energy content is about three times that of liquid hydrocarbons. When reacted with oxygen, either by combustion or in a fuel cell configuration, the sole by-product is water—a particularly attractive feature for transport (*2*) in congested urban areas where pollution is a concern.

Hydrogen is plentiful but is bound up primarily in water and to a lesser extent in hydrocarbons. Economical and environmentally clean production of hydrogen from these sources is crucial to hydrogen-based power generation. Equally important are safe transport, storage, and delivery. The substantial safety concerns associated with hydrogen storage in high-pressure containers have prompted a search for materials capable of sequestering hydrogen at low pressures and ambient temperatures.

Many metal alloys can store hydrogen in the form of metal hydrides such as $LaNi_5$. However, the large mass of this alloy precludes capacities much larger than 2% by weight (the benchmark set by the U.S. Department of Energy is 6.5% by weight). Other, lighter weight metal alloys exhibit higher capacities, but hydride formation is often too slow or irreversible.

In the quest for lightweight hydrogen-storage materials, investigators also have examined high-surface-area carbon materials, recently including carbon single-walled nanotubes (SWNTs). The amount of hydrogen loading reported for SWNTs differs substantially among research groups (*3*), apparently because of complications in attaining compositional and structural uniformity in these materials (*4*).

The MOFs of Rosi *et al.* (*1*) circumvent this problem because they are prepared under mild conditions from solution as well-defined single crystals. Single-crystal x-ray diffraction reveals a structure consisting of $[OZn_4]^{6+}$ building blocks assembled into a highly uniform cubic lattice by organic connectors (see the figure) (*5*). Solvent molecules captured by the framework pores during crystallization can be removed while retaining the framework; this structural stability is unusual in crystalline molecular materials. The solvent-free crystals contain substantial interior surface area (>2000 m^2/g) that is accessible to small molecules.

The group has previously reported nitrogen and methane adsorption by these framework materials (*6, 7*). Now they demonstrate hydrogen uptake of 0.5% by weight in the parent compound (MOF-5).

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