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ATMOSPHERIC SCIENCE

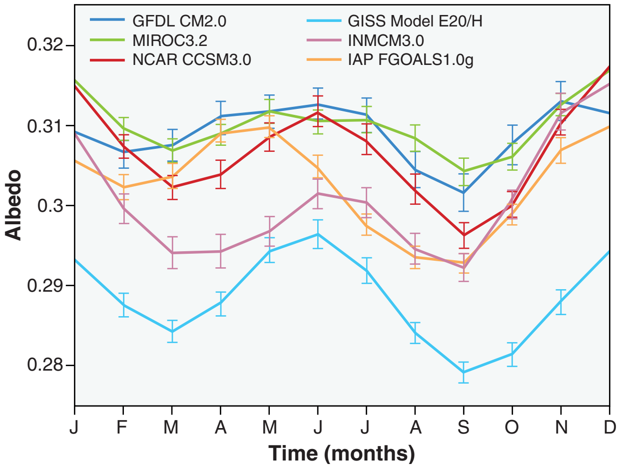
In Search of Balance

Robert J. Charlson, Francisco P. J. Valero, John H. Seinfeld

The climate of Earth and its global mean surface temperature are the consequence of a balance between the amount of solar radiation absorbed by Earth's surface and atmosphere and the amount of outgoing longwave radiation emitted by the system. The former is governed by the albedo (reflectivity) of the system, whereas the latter depends strongly on the atmospheric content of gases and particles (such as clouds and dust). Although the theory of absorption of infrared radiation by gases in the atmosphere (*1*) is well accepted and embodied in climate models, the observational and theoretical treatments of albedo, aerosols, and clouds are still under development. One brevium (*2*) and two reports (*3*, *4*) in this issue report estimates of Earth's albedo and of solar radiation reaching the surface, but the uncertainties remain large.

The buildup of CO_2 (*5*), CH_4, and other greenhouse gases during the past century has led to an increased absorption of infrared radiation in the atmosphere (enhanced greenhouse effect) and a consequent warming ("positive forcing") of the climate. But human-made changes in aerosols and clouds can cause enhanced albedo and hence cooling ("negative forcing"), and they may already have offset a substantial part of the enhanced greenhouse effect. Present trends suggest that by 2050, the magnitude of the enhanced greenhouse effect will be so large that the net anthropogenic forcing will be unequivocally positive and substantial in magnitude (*6*).

Changes in energy balance affect a host of climatic factors, such as temperature, sea level, meteorological patterns, and precipitation. To understand and quantify these effects, the enhanced greenhouse effect and all other forcings must be known accurately. To complicate matters further, the enhanced greenhouse effect is suspected of causing changes in clouds and hence albedo, resulting in feedbacks on both incoming and outgoing radiation (*7*).



Apparent agreement. Monthly mean annual cycle and standard deviation (vertical bars) of albedo from six models (*12*, *15*). These and other models are used by the Intergovernmental Panel on Climate Change (IPCC) for preindustrial control simulations.

Increased albedo could counteract the enhanced greenhouse effect on a global scale. However, the spatial and temporal characteristics of aerosols, clouds, and greenhouse gases differ widely. Clouds change rapidly, and atmospheric residence times for aerosols are short relative to those for the key greenhouse gases (which remain in the atmosphere for centuries). Albedo therefore changes rapidly, whereas the enhanced greenhouse effect simply increases as a result of the slow accumulation of greenhouse gases. Local and regional changes in energy balance would occur even if the albedo change could offset the enhanced greenhouse effect globally. Light-absorbing aerosols further complicate the picture by cooling Earth's surface, heating the atmosphere, and making clouds more absorbing; they may even reduce cloud cover, thereby decreasing albedo further.

These considerations underscore the importance of understanding the natural and anthropogenic changes in Earth's albedo and the need for sustained, direct, and simultaneous observations of albedo with all methods that are currently available. Albedo changes may be as important as changes in greenhouse gases for determining changes in global climate.

Many methods have been used to estimate albedo, which cannot be measured directly. These methods differ in their scattering geometries, calibration accuracy, and in spectral, space, and time coverage. The different modes of observation include measurements of earthshine reflected from the Moon (*8*, *9*), broadband radiometer data from low orbits around Earth [Wielicki *et al.* on page 825 (*2*)], geostationary cloud-cover observations (*10*), deep space radiometry (*11*), and surface radiometry [Pinker *et al.* on page 850 (*3*), Wild *et al.* on page 847 (*4*)]. All these methods require a theoretical model for relating the measured parameters to albedo, and they all rely on different assumptions. It is critical to compare the results from different approaches to test the consistency among them.

The scientific community has recognized this essential need for years, but major impediments have developed. For example, the broadband data collected by the ERBS (Earth Radiation Budget Satellite) between 2000 and 2004 are not being analyzed for budgetary reasons. The DSCOVR (Deep Space Climate Observatory) satellite has been built but has since fallen victim to the delayed space shuttle program and is now in storage awaiting a launch opportunity. The CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) and CloudSat satellites have been built and have scheduled launches, but recent budget cuts imposed on the Earth sciences in NASA will severely constrain the analysis and interpretation of the data. Inasmuch as

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LARGE INCONSISTENCIES

Climatic observations and forcings	Equivalent change in albedo × 10^3
Enhanced greenhouse effect during industrial era (2.4 ± 0.2 W/m^2) (*6*)	−7 ± 0.6
Anthropogenic aerosol forcing during industrial era (*6*)	+4 ± 4
Albedo change estimated from earthshine data (2000 to 2004) (*2, 8, 9*)	+16
Albedo change estimated from low-orbit satellite data (2000 to 2004) (*2*)	−6
Change in irradiance at Earth's surface measured with satellites (1983 to 2001) (*3*)	−8
Change in irradiance at Earth's surface measured at the surface (1985 to 2000) [Fig. 1 in (*4*)]	−13
Change in irradiance at Earth's surface measured at the surface (1950 to 1990) [Fig. 1 in (*4*)]	+20

the primary objectives of these three satellites include studies of the effects of aerosols and clouds on albedo, what seemed to be real progress could be delayed or thwarted.

Several global climate models appear to calculate nearly the same albedo (see the figure); however, clouds are treated very differently in these models, the seasonal cycles that are prominent in the figure are not apparent in data from the CERES (Clouds and the Earth's Radiant Energy System) experiment or from earthshine data (*2*, *8*, *9*, *12*), and the global amount of condensed water varies among the models by as much as a factor of 5. Hence, little certainty can be gained from models alone.

To date, the results from different measurement and modeling approaches are inconsistent among themselves and with each other. The magnitudes of the inconsistencies exhibited by both measurements and models of albedo changes and effects are as large as, or larger than, the entire enhanced greenhouse gas effect when compared in terms of the albedo change equivalent of climate forcing (see the table). In fact, the albedo change that is the equivalent of the enhanced greenhouse effect is barely detectable by the available methods for measuring albedo.

To quantify all changes in energy balance, and in view of the discrepancies in magnitude and even sign (see the table and the figure), it will be necessary to develop a strategy to strengthen research efforts on albedo-related quantities, including modeling and analysis of the data from the yet-to-be-launched satellites. To help achieve a balance of effort, care must be exercised in the use of potentially misleading terms like "global warming" (*13*) and "global dimming" (*14*). Their use may constitute an obstacle in reaching an understanding of the issues driving the fundamental scientific questions of Earth's energy balance, albedo, greenhouse effect, and interactions of solar and infrared radiation with aerosols and clouds.

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13. Global warming formally means an increase in the mean temperature at Earth's surface, but may seem to imply (incorrectly) that the whole Earth will warm more or less uniformly.
14. Global dimming formally means a decrease in "global radiation" (the sum of direct plus diffuse solar radiation measured at a point on Earth's surface), but might seem to imply that the Sun's radiation has dimmed or that the effect is global in extent.
15. GFDL: Geophysical Fluid Dynamics Laboratory, USA. NCAR: National Center for Atmospheric Research, USA. GISS: Goddard Institute for Space Studies, USA. INMCM: Institute for Numerical Mathematics, Russia. IAP: Institute of Atmospheric Physics, China. Miroc is a medium-resolution model run by the Center for Climate System Research (University of Tokyo), the National Institute for Environmental Studies, and the Frontier Research Center for Global Change of the Japan Agency for Marine-Earth Science and Technology.
16. We thank the international modeling groups for providing their data for analysis, the Program for Climate Model Diagnosis and Intercomparison for collecting and archiving the model data, the JSC/CLIVAR Working Group on Coupled Modelling and their Coupled Model Intercomparison Project and Climate Simulation Panel for organizing the model data analysis, and the IPCC Working Group I Technical Support Unit for technical support. The IPCC Data Archive at Lawrence Livermore National Laboratory is supported by the Office of Science, U.S. Department of Energy.

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GEOCHEMISTRY

The Paradox of Mantle Redox

Catherine McCammon

Redox reactions (those involving reduction or oxidation) occur in many everyday processes, from photosynthesis and metabolism to fuel combustion and household cleaning. They also play a critical role in many geological systems. Processes on Earth's surface are intimately linked to the oxidation state of the mantle through the geochemical cycles of elements such as carbon, sulfur, oxygen, and hydrogen. Recent studies have advanced our understanding of the oxidation state of the mantle, elucidating the redox relations within Earth and their consequences for global processes.

The term "oxidation state" has caused some confusion in the geological literature, because it has two different meanings in the context of mantle properties. First, it is used to indicate the valence state of elements, for example, divalent iron (Fe^{2+}) and trivalent iron (Fe^{3+}). Second, it is used to indicate the chemical potential of oxygen, more commonly referred to as oxygen fugacity. High oxygen fugacity means oxidizing conditions, whereas low oxygen fugacity implies reducing conditions.

In everyday experience, these two definitions of oxidation state are almost always coupled: Oxidizing conditions favor the formation of Fe^{3+} (for example, rust on a car), whereas reducing conditions favor the formation of Fe^{2+} or even metallic iron (Fe^0). However, paradoxical behaviors can arise when solids are present, because crystal structures impose additional constraints: Some minerals incorporate almost no Fe^{3+} even under oxidizing conditions, whereas others incorporate Fe^{3+} even under reducing conditions. A classic example is iron oxide, Fe_xO, which always contains a measurable amount of Fe^{3+} in its crystal structure, even under reducing conditions where metallic iron is stable.

Studies of mantle rocks show that the oxygen fugacity of the upper mantle is relatively high (*1*), even though the abundance of oxidized iron (Fe^{3+}) is low (*2*) (see the figure). How can we reconcile these apparently contradictory observations? The answer lies in the unfavorable energetics of defect incorporation in olivine, the most abundant mineral in the upper mantle. This property leads to an almost negligible Fe^{3+} concentration in olivine even under relatively oxidizing conditions (*3*). Fe^{3+} is readily incorporated into the minerals spinel and garnet, but because they are at least 1/10th as abundant as olivine, their presence causes only a small increase in Fe^{3+}

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