ner. In one example, two chiral catalysts were used to induce conjugate addition reactions of cyanide (*7*). Only one stereocenter is created in that process, but substantial enhancement in enantioselectivity was observed through the "matching" of the two chiral catalysts. In another example, researchers used two different chiral secondary amine catalysts in the same reaction to effect a sequential enantioselective conjugate addition and then trapping of the intermediate enolate (*8*). Through ingenious reaction design, the first catalyst was induced to fall off after the initial step, and the second catalyst entered and controlled the diastereoselectivity of the second stereocenter formation. As such, both stereocenters of the product can be controlled, and thus all four of the possible stereoisomers could be accessed through proper choice of the two chiral catalysts. That work represented an extraordinary illustration of a well-established strategy in asymmetric synthesis: chiral catalyst-controlled diastereoselectivity in a reaction of an enantioenriched substrate.

Krautwald *et al*. also use two different chiral catalysts to control the formation of two stereocenters. However, their approach is fundamentally different from previous efforts in that it relies on the independent activation of two distinct reacting partners in a single reaction (see the figure, panel B). The goal is not for one catalyst to overcome or complement the effect of the other, but rather for the two catalysts to induce stereoselectivity independently yet simultaneously.

The concept is illustrated in the α-allylation of branched aldehydes, a carbon-carbon bond-forming reaction that generates two contiguous stereocenters (see the figure, panel C). Relying on previously established catalytic reactivity principles, the aldehyde is activated by an amine catalyst, and the allylic electrophile is activated by an iridium catalyst. The authors show in an unambiguous set of experiments that each catalyst controls the stereoselectivity of one of the substrates but has essentially no effect on the stereocenter derived from the other reacting partner. Then, in a stunning set of results, they show that two chiral catalysts working together achieve the desired effect of nearly perfect and independent control of each center. Selective access to every possible stereoisomer can thus be achieved in a single transformation from the same set of substrates simply by choice of a distinct catalyst combination.

In principle, this has the potential to emerge as a powerful new strategy for reaction design, applicable to a wide range of important reactions. Many of the most powerful transformations in organic chemistry involve bond formation between two potentially prochiral reaction partners and thus can introduce multiple stereocenters in one operation. Notable examples include the Diels-Alder reaction, the aldol reaction, and cyclopropanation reactions. Effective enantioselective catalytic variants are known for each of these classes of reactions but in all cases rely on the use of a single chiral catalyst. The question now is whether dual chiral catalysis of the type uncovered by Krautwald *et al*. might be applied to these and other broadly useful reactions in organic chemistry, thereby making it possible to access every stereochemical permutation of the products of interest by the same method, varying only the stereochemistry of the catalysts.

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3. Enantiomers are stereoisomers, i.e., isomers that have the same bond connectivity but different spatial relationships, that differ in being nonsuperimposable mirror-image partners (such as a matched pair of left and right shoes). In organic compounds, this occurs when there is a single stereochemical element, most often a stereogenic carbon center bearing four different substituents. Diastereomers are stereoisomers that are not enantiomers (such as a left foot in a left shoe versus a left foot in a right shoe), and these occur in structures that have multiple stereogenic carbon centers.
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CLIMATE CHANGE

What Are Climate Models Missing?

A better representation of the coupling between atmospheric water and circulation is necessary to reduce imprecision in climate model projections.

Bjorn Stevens[1] and Sandrine Bony[2]

Fifty years ago, Joseph Smagorinsky published a landmark paper (*1*) describing numerical experiments using the primitive equations (a set of fluid equations that describe global atmospheric flows). In so doing, he introduced what later became known as a General Circulation Model (GCM). GCMs have come to provide a compelling framework for coupling the atmospheric circulation to a great variety of processes. Although early GCMs could only consider a small subset of these processes, it was widely appreciated that a more comprehensive treatment was necessary to adequately represent the drivers of the circulation. But how comprehensive this treatment must be was unclear and, as Smagorinsky realized (*2*), could only be determined through numerical experimentation. These types of experiments have since shown that an adequate description of basic processes like cloud formation, moist convection, and mixing is what climate models miss most.

From GCMs to Earth System Models

Smagorinsky's GCM was designed around the premise that studies of the general circulation required a model capable of resolving the heat transport from the equator to the poles. Its formulation was the next logical step in a program of hierarchical model development best known for its pioneering contributions to numerical weather prediction (*3*). The work paved the way for fundamental studies of the atmospheric general circulation, and hence Earth's climate.

Over the past half century, many of these studies have focused on the types of numerical experiments anticipated by Smagorinsky. Beginning with basic processes like moist convection and cloud formation, which have long been appreciated as central to the energetics of the troposphere, a long succession of processes and couplings have been added to primitive-equation descriptions of the atmospheric general circulation. In so doing, GCMs have gradually morphed into Global Climate Models, and with the more recent incorporation of models of the biosphere and the associated cycles of important chemical nutrients, Earth System Models (*4*, *5*).

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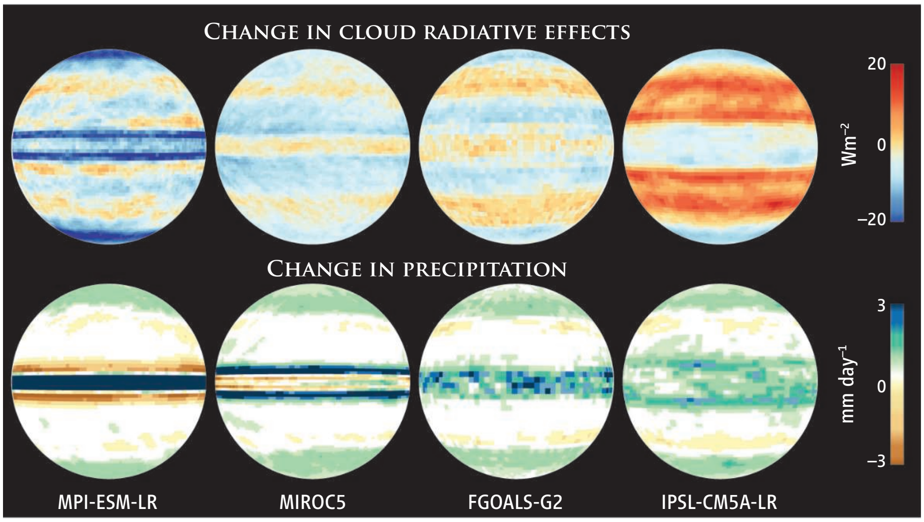

Wide variation. The response patterns of clouds and precipitation to warming vary dramatically depending on the climate model, even in the simplest model configuration. Shown are changes in the radiative effects of clouds and in precipitation accompanying a uniform warming (4°C) predicted by four models from Phase 5 of the Coupled Model Intercomparison Project (CMIP5) for a water planet with prescribed surface temperatures.

Key Uncertainties

The increase in complexity has greatly expanded the scope of questions to which GCMs can be applied (*5*). Yet, it has had relatively little impact on key uncertainties that emerged in early studies with less comprehensive models (*6*). These uncertainties include the equilibrium climate sensitivity (that is, the global warming associated with a doubling of atmospheric carbon dioxide), arctic amplification of temperature changes, and regional precipitation responses. Rather than reducing biases stemming from an inadequate representation of basic processes, additional complexity has multiplied the ways in which these biases introduce uncertainties in climate simulations (*7*, *8*).

For instance, a poor understanding of what controls the distribution of tropical precipitation over land, and hence vegetation dynamics, limits attempts to understand the carbon cycle (*9*). Similarly, uncertainties in arctic amplification of warming hinder predictions of permafrost melting and resultant changes in soil biogeochemistry.

Although the drive to complexity has not reduced key uncertainties, it has addressed Smagorinsky's question (*2*) as to what level of process detail is necessary to understand the general circulation. There is now ample evidence that an inadequate representation of clouds and moist convection, or more generally the coupling between atmospheric water and circulation, is the main limitation in current representations of the climate system.

That this limitation constitutes a major roadblock to progress in climate science can be illustrated by simple numerical experiments. In idealized simulations of a waterworld that neglect complex interactions among land surface, cryosphere, biosphere, and aerosol and chemical processes (see the figure), the key uncertainties associated with the response of clouds and precipitation to global warming are as large as they are in comprehensive Earth System Models (*10*).

Differences among the simulations in the figure are especially evident in the tropics, where the sign of cloud changes and the spatial structure of the precipitation response differ fundamentally between models. This diversity of responses arises because, at low latitudes, the coupling between water and circulation is disproportionately dependent on the representation of unresolved processes, such as moist convection and cloud formation (*11*, *12*). The mid-latitudes show more robust responses because much of the energy transport is carried by baroclinic eddies; these, too, are fundamentally coupled to water, but they are much better described and resolved by modern GCMs, as foreseen by Smagorinsky (*1*).

The uncertain interplay between water and circulation that underlies differences in the response of the climate system to warming (see the figure) can be expressed in terms of more specific questions. For instance, how do marine boundary-layer clouds depend on their environment? Or how do atmospheric circulations couple to moist convection through surface and radiative fluxes? The first question ends up being key to explaining the intermodel spread in climate sensitivity (*13*, *14*), the second to the pattern of the regional response to warming. Differences in regional responses also influence ocean circulations, and hence how oceans take up heat, as well as patterns of precipitation, and hence how the land biosphere takes up carbon.

Back to Basics

A deeper understanding and better representation of the coupling between water and circulation, rather than a more expansive representation of the Earth System, is thus necessary to reduce the uncertainty in estimates of the climate sensitivity and to guide adaptation to climate change at the regional level. This knowledge should help focus efforts and lead to progress in reducing the imprecision of climate models in the next 50 years. Here, Numerical Weather Prediction (NWP) provides a good example. By focusing on key limitations in the model initialization, spatial resolution, and the representation of key parameterized processes, NWP has improved forecast skill substantially over the past 30 years (*15*).

It is time to draw lessons from the era of experimentation that Smagorinsky launched half a century ago, and focus climate modeling efforts on advancing understanding and improving the numerical representations of how clouds, moist convection, and heating couple to the general circulation.

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