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1 The Concept of Model

1.1 Introduction

This book describes the foundations of mathematical models for atmospheric chemistry. Atmospheric chemistry is the science that focuses on understanding the factors controlling the chemical composition of the Earth's atmosphere. Atmospheric chemistry investigates not only chemical processes but also the dynamical processes that drive atmospheric transport, the radiative processes that drive photochemistry and climate forcing, the evolution of aerosol particles and their interactions with clouds, and the exchange with surface reservoirs, including biogeochemical cycling. It is a highly interdisciplinary science.

Atmospheric chemistry is a young and rapidly growing science, motivated by the societal need to understand and predict human perturbations to atmospheric composition. These perturbations have increased greatly over the past century due to population growth, industrialization, and energy demand. They are responsible for a range of environmental problems including degradation of air quality, damage to ecosystems, depletion of stratospheric ozone, and climate change. Quantifying the link between human activities and their atmospheric effects is essential to the development of sound environmental policy.

The three pillars of atmospheric chemistry research are laboratory studies, atmospheric measurements, and models. Laboratory studies uncover and quantify the fundamental chemical processes expected to proceed in the atmosphere. Atmospheric measurements probe the actual system in all of its complexity. Models simulate atmospheric composition using mathematical expressions of the driving physical and chemical processes as informed by the laboratory studies. They can be tested with atmospheric measurements to evaluate and improve current knowledge, and they can be used to make future projections for various scenarios. Models represent a quantitative statement of our current knowledge of atmospheric composition. As such, they are fundamental tools for environmental policy.

Atmospheric chemistry modeling has seen rapid improvement over the past decades, driven by computing resources, improved observations, and demand from policymakers. Thirty years ago, models were so simplified in their treatments of chemistry and transport that they represented little more than conceptual exercises. Today, state-of-science *chemical transport models* provide realistic descriptions of the 3-D transport and chemical evolution of the atmosphere. Although uncertainties remain large, these models are used extensively to interpret atmospheric observations and to make projections for the future. The state of the science is advancing rapidly, and atmospheric chemists 30 years from now may well scoff at the crude nature of present-day models. Nevertheless, we are now at a point where models can provide a credible, process-based mathematical representation of the atmosphere to serve the needs of science and policy. It is with this perspective of a mature yet evolving state of science that this book endeavors to describe the concepts and algorithms that provide the foundations of atmospheric chemistry models.

This chapter is intended to introduce the reader to the notion and utility of models, and to provide a broad historical perspective on the development of atmospheric chemistry models. It starts with general definitions and properties of mathematical models. It then covers the genesis and evolution of meteorological models, climate models, and finally atmospheric chemistry models, leading to the current state of science. It describes conceptually different types of atmospheric chemistry models and the value of these models as part of atmospheric observing systems. It finishes with a brief overview of the computational hardware that has played a crucial role in the progress of atmospheric modeling.

1.2 What is a Model?

A model is a simplified representation of a complex system that enables inference of the behavior of that system. The *Webster New Collegiate Dictionary* defines a model as a description or analogy used to help visualize something that cannot be directly observed, or as a system of postulates, data, and inferences presented as a mathematical description of an entity or state of affairs. The *Larousse Dictionary* defines a model as a formalized structure used to account for an ensemble of phenomena between which certain relations exist. Models are abstractions of reality, and are often associated with the concept of metaphor (Lakoff and Johnson, 1980). Humans constantly create models of the world around them. They observe, analyze, isolate key information, identify variables, establish the relationships between them, and anticipate how these variables will evolve in various scenarios.

One can distinguish between cognitive, mathematical, statistical, and laboratory models (Müller and von Storch, 2004). Cognitive models convey ideas and test simple hypotheses without pretending to simulate reality. For example, the Daisyworld model proposed by Lovelock (1989) illustrates the stability of climate through the insolation–vegetation–albedo feedback. This model calculates the changes in the geographical extent of imaginary white and black daisies covering a hypothetical planet in response to changes in the incoming solar energy. It shows that the biosphere can act as a planetary thermostat. Such apparently fanciful models can powerfully illustrate concepts. More formal mathematical models attempt to represent the complex intricacies of real-world systems, and describe the behavior of observed quantities on the basis of known physical, chemical, and biological laws expressed through mathematical equations. They can be tested by comparison to observations and provide predictions of events yet to be experienced. Examples are meteorological models used to perform daily weather forecasts. Statistical models describe the behavior of variables in terms of their observed statistical relationships with other variables, and use these relationships to interpolate or extrapolate

Figure 1.1 Prussian naturalist and explorer Alexander von Humboldt (a) and French mathematician and astronomer Pierre-Simon, Marquis de Laplace (b).

behavior. They are empirical in nature, as opposed to the physically based mathematical models. Laboratory models are physical replicas of a system, at a reduced or enlarged geometric scale, used to perform controlled experiments. They mimic the response of the real system to an applied perturbation, and results can be extrapolated to the actual system through appropriate scaling laws.

In his 1846 book *Kosmos*, German scientist Alexander von Humboldt (1769–1859, see Figure 1.1) states that the structure of the universe can be reduced to a problem of mechanics, and reinforces the view presented in 1825 by Pierre-Simon Laplace (1749–1827, see Figure 1.1). In the introduction of his *Essai Philosophique sur les Probabilités* (Philosophical Essay on Probabilities), Laplace explains that the present state of the Universe should be viewed as the consequence of its past state and the cause of the state that will follow. Once the state of a system is known and the dynamical laws affecting this system are established, all past and future states of the system can be rigorously determined. This concept, which applies to many aspects of the natural sciences, is extremely powerful because it gives humanity the tools to monitor, understand, and predict the evolution of the Universe.

Although von Humboldt does not refer explicitly to the concept of model, he attempts to describe the functioning of the world by isolating different causes, combining them in known ways, and asking whether they reinforce or neutralize each other. He states that, "by suppressing details that distract, and by considering only large masses, one rationalizes what cannot be understood through our senses." This effectively defines models as idealizations of complex systems designed to achieve understanding. Models isolate the system from its environment, simplify the relationships between variables, and make assumptions to neglect certain internal variables and external influences (Walliser, 2002). They are not fully objective tools because they emphasize the essential or focal aspects of a system as conceived by their authors. They are not universal because they include assumptions and simplifications that may be acceptable for some specific applications but not others. Indeed, the success of a model is largely the product of the skills and imagination of the authors.

During the twentieth century, models started to become central tools for addressing scientific questions and predicting the evolution of phenomena such as economic cycles, population growth, and climate change. They are extensively used today in many disciplines and for many practical applications of societal benefit, weather forecasting being a classic example. As computing power increases and knowledge grows, models are becoming increasingly elaborate and can unify different elements of a complex system to describe their interactions. In the case of Earth science, this is symbolized by the vision of a "virtual Earth" model to describe the evolution of the planet, accounting for the interactions between the atmosphere, ocean, land, biosphere, cryosphere, lithosphere, and coupling this natural system to human influences. Humans in this "virtual Earth" would not be regarded as external factors but as actors through whom environmental feedbacks operate.

1.3 Mathematical Models

Mathematical models strip the complexity of a system by identifying the essential driving variables and describing the evolution of these variables with equations based on physical laws or empirical knowledge. They provide a quantitative statement of our knowledge of the system that can be compared to observations. Models of natural systems are often expressed as mathematical applications of the known laws that govern these systems. As stated by Gershenfeld (1999), mathematical models can be rather general or more specific, they can be guided by *first principles* (physical laws) or by empirical information, they can be analytic or numerical, deterministic or stochastic, continuous or discrete, quantitative or qualitative. Choosing the best model for a particular problem is part of a modeler's skill.

Digital computers in the 1950s ushered in the modern era for mathematical models by enabling rapid numerical computation. Computing power has since been doubling steadily every two years ("Moore's law") and the scope and complexity of models has grown in concert. This has required in turn a strong effort to continuously improve the physical underpinnings and input information for the models. Otherwise we have "garbage in, garbage out." Sophisticated models enabled by high-performance computing can extract information from a system that is too complex to be fully understood or quantifiable by human examination. By combining a large amount of information, these models point to system behavior that may not have been anticipated from simple considerations. From this point of view, models generate knowledge. In several fields of science and technology, computer simulations have become a leading knowledge producer. In fact, this approach, which does not belong either to the theoretical nor to the observational domains, is regarded as a new form of scientific practice, a "third way" in scientific methodology complementing theoretical reasoning and experimental methods (Kaufmann and Smarr, 1993).

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For a model to be useful it must show some success at reproducing past observations and predicting future observations. By definition, a model will always have some error that reflects the assumptions and approximations involved in its development. The question is not whether a model has error, but whether the error is small enough for the model to be useful. As the saying goes, “all models are wrong, but some are useful.” A crucial task is to quantify the error statistics of the model, which can be done through error propagation analyses and/or comparison with observations. The choice of observational data sets and statistics to compare to the model is an important part of the modeler’s skill, as is the interpretation of the resulting comparisons. Discrepancies with observations may be deemed acceptable, and used to compile model error statistics, but they may also point to important flaws in the founding assumptions or implementation of the model. The modeler must be able to recognize the latter as it holds the key to advancing knowledge. Some dose of humility is needed because the observations cannot sample all the possible realizations of a complex system. As a result, the error statistics of the model can never be characterized fully.

Many mathematical models are based on differential equations that describe the evolution in space and time of the variables of interest. These are often conservation equations, generalizing Newton’s second law that the acceleration of an object is proportional to the force applied to that object. Atmospheric chemistry models are based on the *continuity equation* that describes mass conservation for chemical species. Consider an ensemble of chemical species ($i = 1, \ldots n$) with mole fractions (commonly called *mixing ratios*) assembled in a vector $\mathbf{C} = (C_1, \ldots C_n)^T$. The continuity equation for species i in a fixed (*Eulerian*) frame of reference is given by

$$\frac{\partial C_i}{\partial t} = -\mathbf{v}\bullet\nabla C_i + P_i(\mathbf{C}) - L_i(\mathbf{C}) \quad (i = 1, \ldots n) \tag{1.1}$$

Here, $\mathbf{v}$ is the 3-D wind vector, and P_i and L_i are total production and loss rates for species i that may include contributions from chemical reactions (coupling to other species), emissions, and deposition. The local change in mixing ratio with time ($\partial C_i/\partial t$) is expressed as the sum of transport in minus transport out (flux divergence term $\mathbf{v}\bullet\nabla C_i$) and net local production ($P_i - L_i$). Similar conservation equations are found in other branches of science. For example, replacing C_i with momentum yields the Navier–Stokes equation that forms the basis for models of fluid dynamics.

A system is said to be *deterministic* if it is uniquely and entirely predictable once initial conditions are specified. It is *stochastic* if randomness is present so that only probabilities can be predicted. Systems obeying the laws of classical mechanics are generally deterministic. The two-body problem (e.g., a satellite orbiting a planet or a planet orbiting the Sun), described by Newton’s laws and universal gravitation, is a simple example of a deterministic system. An analytic solution of the associated differential equations can be derived with no random element. All trajectories derived with different initial conditions converge toward the same subspace called an *attractor*. By contrast, when trajectories starting from slightly different initial conditions diverge from each other at a sufficiently fast rate, the system is said to be *chaotic*. Meteorological models are a classic example. They are deterministic but exhibit chaotic behavior due to nonlinearity of the Navier–Stokes equation. This chaotic behavior is called *turbulence*. Chaotic systems evolve in a manner that is exceedingly dependent on the precise choice of initial conditions. Since initial conditions in a complex system such as the weather can never be exactly defined, the model results are effectively stochastic and multiple simulations (*ensembles*) need to be conducted to obtain model output statistics.

1.4 Meteorological Models

The basic ideas that led to the development of meteorological forecast models were formulated about a century ago. American meteorologist Cleveland Abbe (1838–1916) first proposed a mathematical approach in a 1901 paper entitled "The physical basis of long-range weather forecasting." A few years later, in 1904, in a paper entitled "Das Problem von der Wettervorhersage betrachtet vom Standpunkte der Mechanik und der Physik" (The problem of weather prediction from the standpoint of mechanics and physics), Norwegian meteorologist Vilhelm Bjerknes (1862–1951) argued that weather forecasting should be based on the well-established laws of physics and should therefore be regarded as a deterministic problem (see Figure 1.2). He wrote:

> If it is true, as every scientist believes, that subsequent atmospheric states develop from the preceding ones according to physical law, then it is apparent that the necessary and sufficient conditions for the rational solution of forecasting problems are the following:
>
> 1. A sufficiently accurate knowledge of the state of the atmosphere at the initial time;
> 2. A sufficiently accurate knowledge of the laws according to which one state of the atmosphere develops from another.

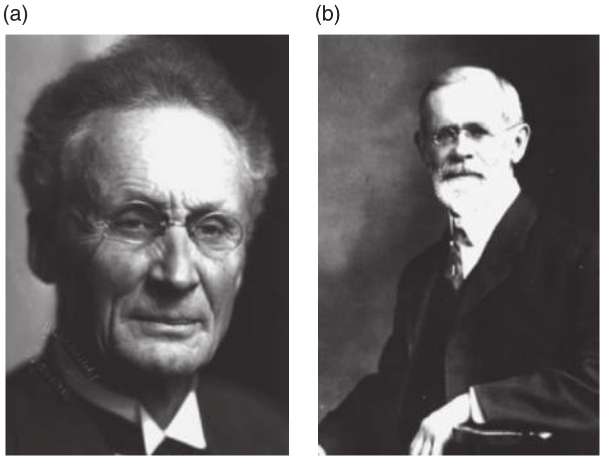

Figure 1.2 Norwegian meteorologist Vilhelm Bjerknes (a), and American meteorologist Cleveland Abbe (b). Source: Wikimedia Commons.

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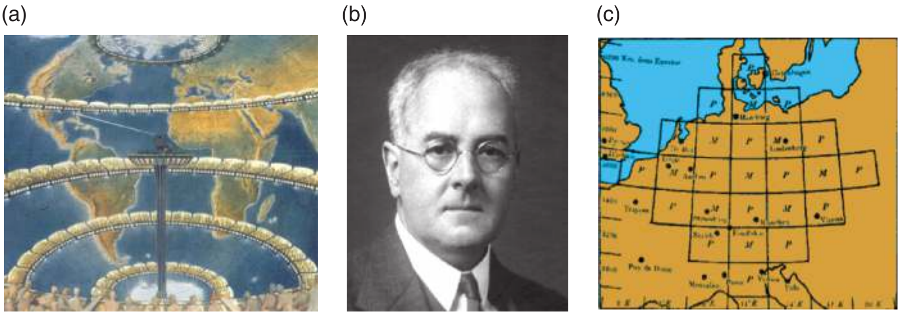

Figure 1.3 British meteorologist Lewis Fry Richardson (b), the map grid he used to make his numerical weather forecast (c), and an artist's view of a theater hall (a) imagined by Richardson to become a "forecast factory." Panel (a) reproduced with permission from "Le guide des cités" by François Schuiten and Benoît Peeters, © Copyright Casterman.

Bjerknes reiterated his concept in a 1914 paper entitled "Die Meteorologie als exakte Wissenschaft" (Meteorology as an exact science). He used the medical terms "diagnostics" and "prognostics" to describe the two steps shown. He suggested that the evolution of seven meteorological variables (pressure, temperature, the three wind components, air density, and water vapor content) could be predicted from the seven equations expressing the conservation of air mass and water vapor mass (continuity equations), the conservation of energy (thermodynamic equation, which relates the temperature of air to heating and cooling processes), as well as Newton's law of motion (three components of the Navier–Stokes equation), and the ideal gas law (which relates pressure to air density and temperature). Bjerknes realized that these equations could not be solved analytically, and instead introduced graphical methods to be used for operational weather forecasts.

During World War I, Lewis Fry Richardson (1881–1951; see Figure 1.3), who was attached to the French Army as an ambulance driver, attempted during his free time to create a numerical weather forecast model using Bjerknes' principles. He used a numerical algorithm to integrate by hand a simplified form of the meteorological equations, but the results were not satisfying. The failure of his method was later attributed to insufficient knowledge of the initial weather conditions, and to instabilities in the numerical algorithm resulting from an excessively long time step of six hours. Richardson noted that the number of arithmetic operations needed to solve the meteorological equations numerically was so high that it would be impossible for a single operator to advance the computation faster than the weather advances. He proposed then to divide the geographic area for which prediction was to be performed into several spatial domains, and to assemble for each of these domains a team of people who would perform computations in parallel with the other teams, and, when needed, communicate their information between teams. His fantasy led him to propose the construction of a "forecast factory" in a large theater hall (Figure 1.3), where a large number of teams would perform coordinated computations. This construction was a precursor vision of modern massively parallel supercomputers. The methodology used by Richardson to solve numerically the

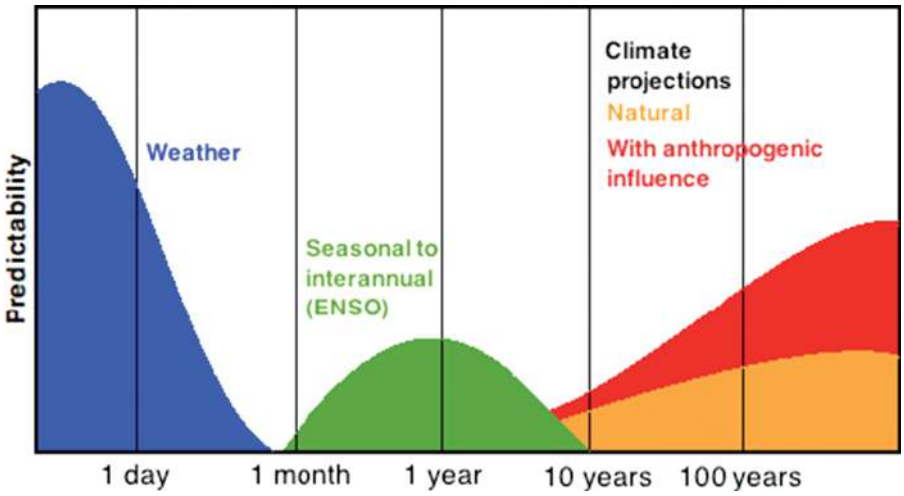

Figure 1.4 Qualitative representation of the predictability of weather, seasonal to interannual variability (El Nino – Southern Oscillation) and climate (natural variations and anthropogenic influences). Adapted from US Dept. of Energy, 2008.

meteorological equations was published in 1922 in the landmark book *Weather Prediction by Numerical Process*.

The first computer model of the atmosphere was developed in the early 1950s by John von Neumann (1903–1957) and Jule Charney (1917–1981), using the Electronic Numerical Integrator and Computer (ENIAC). The computation took place at about the same pace as the real evolution of the weather, and so results were not useful for weather forecasting. However, the model showed success in reproducing the large-scale features of atmospheric flow. Another major success of early models was the first simulation of cyclogenesis (cyclone formation) in 1956 by Norman Phillips at the Massachusetts Institute of Technology (MIT). Today, with powerful computers, meteorological models provide weather predictions with a high degree of success over a few days and some success up to ten days. Beyond this limit, chaos takes over and the accuracy of the prediction decreases drastically (Figure 1.4). As shown by Edward Lorenz (1917–2008), lack of forecasting predictability beyond two weeks is an unavoidable consequence of imperfect knowledge of the initial state and exponential growth of model instabilities with time (Lorenz, 1963, 1982). Increasing computer power will not relax this limitation. Lorenz's finding clouded the optimistic view of forecasting presented earlier by Bjerknes. Predictions on longer timescales are still of great value but must be viewed as stochastic, simulating (with a proper ensemble) the statistics of weather rather than any prediction of specific realization at a given time. The statistics of weather define the *climate*, and such long-range statistical weather prediction is called *climate modeling*,

Meteorological models include a so-called dynamical core that solves Bjerknes' seven equations at a spatial and temporal resolution often determined by available computing power. Smaller-scale turbulent features are represented through somewhat empirical parameterizations. Progress in meteorological models over the past decades has resulted from better characterization of the initial state, improvements in the formulation of physical processes, more effective numerical algorithms, and higher resolution enabled by increases in computer power. Today, atmospheric models may be used as *assimilation* tools, to help integrate observational data into a coherent theoretical framework; as *diagnostic* tools, to assist in the interpretation of observations and in the identification of important atmospheric processes; and as *prognostic* tools, to project the future evolution of the atmosphere on timescales of weather or climate.

Data assimilation plays a central role in weather forecasting because it helps to better define the initial state for the forecasts. Observations alone cannot define that state because they are not continuous and are affected by measurement errors. The meteorological model provides a continuous description of the initial state, but with model errors. Data assimilation blends the information from the model state with the information from the observations, weighted by their respective errors, to achieve an improved definition of the state. Early approaches simply nudged the model toward the observations by adding a non-physical term to the meteorological equations, relaxing the difference between model and observations. Optimal estimation algorithms based on Bayes' theorem were developed in the 1960s and provide a sounder foundation for data assimilation. They define a most likely state through minimization of an error-weighted least-squares cost function including information from the model state and from observations. Current operational forecast models use advanced methods to assimilate observations of a range of meteorological variables collected from diverse platforms and at different times. Four-dimensional *variational* data assimilation (4DVAR) methods ingest all observations within a time window to numerically optimize the 3-D state at the initial time of that window.

1.5 Climate Models

The climate represents the long-term statistics of weather, involving not only the atmosphere but also the surface compartments of the Earth system (atmosphere, oceans, land, cryosphere). It is a particularly complex system to investigate and to model. The evolution of key variables in the different compartments can be described by partial differential equations that represent fundamental physical laws. Solution of the equations involves spatial scales from millimeters (below which turbulence dissipates) to global, and temporal scales from milliseconds to centuries or longer. The finer scales need to be parameterized in order to focus on the evolution of the larger scales. Because of the previously described chaos in the solution to the equations of motion, climate model simulations are effectively stochastic. *Ensembles* of climate simulations conducted over the same time horizon but with slightly modified initial conditions provide statistics of model results that attempt to reproduce observed climate statistics.

The first climate models can be traced back to the French mathematician Joseph Fourier (1768–1830, see Figure 1.5), who investigated the processes that have maintained the mean Earth's temperature at a relatively constant value during its

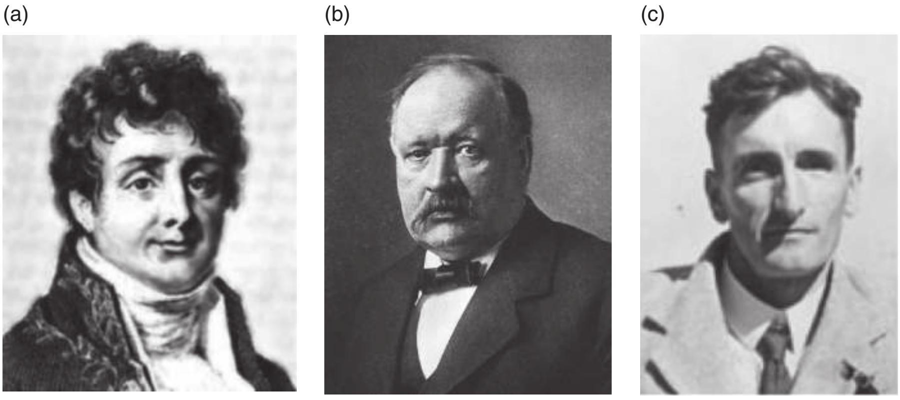

Figure 1.5 French mathematician and physicist Jean Baptiste Joseph Fourier (a), Swedish chemist Svante August Arrhenius (b), and British scientist Guy Stewart Callendar (c). Source of panel (c): G. S. Callendar Archive, University of East Anglia.

history. In 1896, the Swedish scientist Svante Arrhenius (1859–1927; see Figure 1.5) made the first estimate of the changes in surface temperature to be expected from an increase in the atmospheric concentration of CO_2. He did so by using measurements of infrared radiation emitted by the full Moon at different viewing angles to deduce the sensitivity of absorption to the CO_2 amount along the optical path, and then using the result in an energy balance equation for the Earth.

In 1938, Guy S. Callendar (1898–1964; see Figure 1.5) used a simple radiative balance model to conclude that a doubling in atmospheric CO_2 would warm the Earth surface by 2 °C on average, with considerably more warming at the poles. In the following decades, more detailed calculations were performed by 1-D (vertical) radiative–convective models allowing for vertical transport of heat as well as absorption and emission of radiation. Increasing computing power in the 1950s and 1960s paved the way for 3-D atmospheric climate models, called *general circulation models* (GCMs) for their focus on describing the general circulation of the atmosphere. Early GCMs were developed by Norman Phillips at MIT, Joseph Smagorinsky and Syukuro Manabe at the Geophysical Fluid Dynamics Laboratory (GFDL) in Princeton, Yale Mintz and Akio Arakawa at the University of California at Los Angeles (UCLA), and Warren Washington and Akira Kasahara at the National Center for Atmospheric Research (NCAR).

Climate models today have become extremely complex and account for coupling between the atmosphere, the ocean, the land, and the cryosphere. The Intergovernmental Panel on Climate Change (IPCC) uses these models to inform decision-makers about the climate implications of different scenarios of future economic development. Several state-of-science climate models worldwide contribute to the IPCC assessments, and yield a range of climate responses to a given perturbation. Attempts to identify a "best" model tend to be futile because each model has its strengths and weaknesses, and ability to reproduce present-day climate is not necessarily a gauge of how well the model can predict future climate. The IPCC uses instead the range of climate responses from the different models for a given