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Interactive Chemistry and Climate Models in Global Change Studies

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To inform processes of policy development and implementation, climate change research needs to focus on improving the prediction of those variables that are most relevant to economic, social, and environmental effects. In turn, the greenhouse gas and atmospheric aerosol assumptions underlying climate analysis need to be related to the economic, technological, and political forces that drive emissions, and to the results of international agreements and mitigation. Further, assessments of possible societal and ecosystem impacts, and analysis of mitigation strategies, need to be based on realistic evaluation of the uncertainties of climate science.

This report is one of a series intended to communicate research results and improve public understanding of climate issues, thereby contributing to informed debate about the climate issue, the uncertainties, and the economic and social implications of policy alternatives. Titles in the Report Series to date are listed on the inside back cover.

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Chien Wang and Ronald G. Prinn[†]

Abstract

Continually increasing atmospheric concentrations of radiatively important chemical species such as CO_2, CH_4, N_2O, tropospheric O_3, and certain halocarbons most likely will cause future climate changes, which could in turn impact chemical reaction rates and thus lifetimes of many important chemical species. Complicated interactions between climate dynamics and atmospheric chemistry strongly suggest that a fully interactive, comprehensive chemistry-climate modeling system is needed to study the issue. This article reviews recent work in the new and challenging field of interactive chemistry-climate modeling, describing major efforts in model development and summarizing in detail applications of and results from these models.

Contents

1. INTRODUCTION

Since the Industrial Revolution, human activity has greatly increased emissions of such chemicals as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and halocarbons, and changed the chemical composition of Earth's atmosphere [1, 2]. Increased concentrations of these trace gases can absorb and reradiate more outgoing long-wave radiation of the Earth-atmosphere system and thus change the radiative budget of the system, causing radiative forcing of climate change. Studies have shown that CO_2, CH_4, tropospheric ozone (O_3) (in the lower troposphere, O_3 is mainly formed by anthropogenically emitted nitrogen oxides, or NO_x), chlorofluorocarbons (CFCs), and N_2O cause positive radiative forcing and, potentially, atmospheric warming [3, 4]. In contrast, aerosols (tiny particles in the atmosphere that are either formed by reactions in the atmosphere or emitted from the surface) can produce either negative forcing by reflecting incoming solar radiation [*e.g.*, 5, 6] or positive forcing by absorbing the solar radiation [*e.g.*, 7–9].

Burning of fossil fuels and changes in land use (*e.g.*, deforestation) are examples of major anthropogenic sources of atmospheric CO_2. In addition, the chemical reaction of carbon monoxide (CO) with the hydroxyl radical (OH) forms a certain amount of CO_2. The atmospheric budget of CO_2 is thus determined by these sources in combination with two major sinks: oceanic uptake [10] and biospheric uptake [11].

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Anthropogenic production of methane is attributable mainly to fossil fuel burning, rice paddies, human and animal wastes, biomass burning, and changes in land use. CH_4 is a reactive species in the troposphere, where its major sink is reaction with the OH radical, which subsequently leads to formation of O_3 (in NO_X-rich environment) and formaldehyde (CH_2O), and ultimately to CO and thus CO_2 [12–14].

N_2O is primarily emitted from soil and water with a relatively small contribution from anthropogenic emissions. This species is destroyed predominantly in the stratosphere due to photolysis and reaction with $O(^1D)$ [2, 15]. For two short-lived but radiatively important species, tropospheric O_3 and aerosols, anthropogenic emissions of nitrogen and sulfur compounds are the primary precursors.

Economic development analyses indicate that anthropogenic emissions and hence the atmospheric concentrations of several long-lived chemicals most likely will continue to increase [16-18]. Therefore, the greenhouse effect produced by these chemicals in the atmosphere will increase. Many natural processes such as ocean circulation and clouds can also influence climate, however. By how much the enhanced greenhouse effect, when combined with other feedback processes, will warm the climate remains a challenging question. Changes in water vapor concentrations due to climate change, anthropogenic emissions of CH_4, N_2O, CO, NO, and non-methane hydrocarbons (NMHCs), and changes in the flux of solar ultraviolet radiation entering the troposphere can all affect chemical reaction rates and OH radical concentrations. In turn, changes in OH affect the lifetimes of many chemical species, and hence the strength of the enhanced greenhouse effect. To predict climate, a modeling system is needed that incorporates not only climate dynamics but also the chemistry of the atmosphere and ocean as well as the biogeochemistry of ecosystems, in order to include important feedbacks among these subsystems. Because climate change is a serious societal issue, in order to assess the impact of anthropogenic activities on climate change including proposed policy adjustments, the modeling system should also allow for policy-related simulations.

A modeling system suitable for this type of analysis is called an Integrated Assessment model (IA model), which usually includes submodels of coupled atmospheric, land, and oceanic dynamics, atmospheric chemistry, natural and anthropogenic emissions, ecosystem dynamics, and policy evaluation, as well as socioeconomic impact analysis [19]. However, traditional IA models consist of highly parameterized submodels. Specifically, the climate, ocean, and atmospheric chemistry components in IA models are often highly simplified formulae that seek to mimic the behavior of very complicated systems. Quite often, these formulae are too simple to simulate even fundamental feedbacks existing in the atmosphere-ocean system. In addition, without spatial representation of the atmosphere and ocean, transport of trace chemicals and water cannot be realistically handled.

Climate dynamics and atmospheric chemistry are traditionally considered separate research fields. Climate models containing coupled atmospheric and oceanic circulation submodels are usually employed in the study of climate evolution using exogenous assumptions about atmospheric chemical composition [*e.g.*, 20–26], while the spatial distributions of chemical species in the atmosphere as a result of transport and chemical reactions are studied using chemical transport models (CTMs) [*e.g.*, 27–41]. Chemical transport models use dynamic data such as wind speeds, temperature, and water vapor concentration from either climate models or observations to calculate the evolution of chemical species. However, modeled results cannot incorporate feedbacks to the climate model. This type of procedure is generally called “off-line” modeling. The temporal resolution of input dynamic data in CTMs is often too coarse to correctly represent important physical processes such as radiation, convection, and clouds, unfortunately. To overcome this problem, some CTMs have their own physical modules that duplicate the corresponding components in the “source model,” *i.e.*, the climate model. Great amounts of input and output during model integration and/or the use of analyzed observed transport data further limit the use of CTMs to short simulations of the current atmosphere.

Obviously, a model including both climate dynamics and atmospheric chemistry based on physical and chemical theories rather than on oversimplified formulae would greatly improve predictions of climate change in IA models. On-line data communication between the climate and chemistry models provides the opportunity for studies of interactions between climate dynamics and atmospheric chemistry. However, great uncertainties exist in both climatic and chemical processes owing to our limited knowledge about these systems. In order to quantify the magnitude of these uncertainties, the interactive chemistry-climate model must be computationally efficient to allow repeated integrations over a 100-year time scale.

This paper reviews the most recent developments in interactive chemistry-climate models and summarizes important results from these models. As an example, we use the Massachusetts Institute of Technology (MIT) coupled chemistry-climate model [42], which is central to the MIT IA model called the Integrated Global System Model (IGSM) [43]. In the following section, we describe the models. Results then are discussed in two categories: diagnostic and prognostic simulations. Perspectives on the future of this research field are presented in the last section.

2. INTERACTIVE CHEMISTRY AND CLIMATE MODELS

A fully interactive chemistry-climate model comprises two closely integrated submodels: a climate model that simulates the dynamics and thermodynamics of the atmosphere and ocean, and an atmospheric chemistry model that predicts emissions, transport, and chemical conversions of targeted trace gases. An ideal coupling of these two models would allow them to communicate with one another as frequently as demanded by the physics of the processes.

The climate submodel is central to any interactive model. It should be able to simulate atmospheric and oceanic circulations as well as their interactions in reasonable detail, and predict radiative fluxes (both long- and short-wave), cloud coverage, precipitation rates, and water vapor content in the atmosphere. Obviously, three-dimensional general circulation models (GCMs) are the best choice but they do have tremendous computational demands which limits their ability to explore uncertainty and differing emission scenarios. In developing the MIT interactive model, we wanted to address uncertainty and multiple scenarios. Therefore, we have used a two-dimensional (*i.e.*, latitude and altitude) land-ocean-resolving statistical-dynamical model [44] as the climate submodel. This model runs 20 times faster than equivalent three-dimensional models having similar latitudinal and vertical resolution. While two-dimensional, this model resolves the ocean and land separately at each latitude, and reproduces many characteristics of the current zonally averaged observed climate. Responses of this model to varied climate forcings resemble those of three-dimensional GCMs. This model simulates current climate reasonably well, and mimics the much of behavior of its parent three-dimensional GCM, developed at the Goddard Institute for Space Studies (GISS) [20]. Also, the model is flexible because through various parameter choices in the model it can simulate quite closely the gross behavior of most other GCMs. This flexibility is central to our ability to address uncertainty in predictions.

Fundamental issues for the chemistry submodel in an interactive chemistry-climate model system are similar to those of non-interactive CTMs. First, its advection scheme, the numerical method used to calculate transport of chemical properties by the winds, must be positive-definite and accurate. The most popular schemes include the monotone mass conservative scheme [*e.g.*, 45–51] (the scheme used in the MIT model was developed based on [47], [49–51]) and the semi-Lagrangian scheme [52–55]. The monotone scheme appears to be superior in maintaining mass conservation, and is computationally inexpensive [55]. Another issue in developing a chemistry model is that the ordinary differential equations used to formulate chemical reactions are stiff equations and thus require a complicated solver [56, 57]. However, even with a fast solver, the computational requirement is great, so quite often researchers must reduce the size of the equation matrix using various assumptions to save computer time [42, 58].

The most interesting—while challenging—aspect of developing an interactive chemistry-climate model is the coupling of the climate and chemistry submodels. The major coupling must be implemented for the following processes: advection and eddy diffusion of trace chemicals, cloud and precipitation scavenging of chemicals, and radiative forcing by greenhouse gases and aerosols. In the MIT interactive chemistry-climate model, the transport and mixing of trace chemicals are calculated at 20-minute time intervals, when the climate submodel updates its predicted wind fields.

Clouds and precipitation can effectively scavenge some important chemical species, such as sulfur dioxide (SO_2), sulfuric acid gas (H_2SO_4), nitric acid gas (HNO_3), hydrogen peroxide (H_2O_2), and CH_2O. The first two species determine the formation and concentration of sulfate aerosols and the latter two are important species in the tropospheric chemical cycle. On the other hand, cloud coverage significantly alters incoming solar radiation, and hence photochemical processes. In the MIT model, the cloud and precipitation scavenging of chemical species are calculated at each time step when the climate model performs its cloud calculation. The rates at which soluble chemicals are taken up into the cloud droplets and precipitating droplets are derived using relevant equilibrium and aqueous-phase reactions. In addition, the solar radiance derived from the climate model, after including the cloud effects, is used to calculate photochemical reaction rates.

The evolving spatial distributions of greenhouse gases and aerosols predicted by the chemistry model need to be incorporated into the radiation module of the climate model in order to interpret their climatic effects. In the MIT model, concentrations of CO_2, CH_4, N_2O, two CFCs ($CFCl_3$ and CF_2Cl_2), tropospheric O_3, *HFCs*, *PFCs*, SF_6, and sulfate aerosols predicted by the chemistry submodel are used by the climate submodel in a 4-hour time step for calculating the influences of these chemical species on radiative fluxes. As a result, variations in atmospheric chemical composition, which themselves are influenced by modeled climate changes, can almost instantly impact climate prediction. Directly coupling the chemistry and climate submodels, therefore, provides us an opportunity to investigate the two-way interactions between climate dynamics and atmospheric chemistry. A direct coupling of the chemistry and climate models through the radiation module requires a multidisciplinary effort, and often demands revision of a pre-existing complicated radiation code. As a result, this approach is rare.

Table 1 lists several selected interactive chemistry-climate models reported in recent years. Their applications in real research are discussed in the next two chapters. Despite their computational expense, three-dimensional GCMs have been used to develop interactive models. Two models have been developed from the European Center Hamburg Model (ECHAM) version 4, a general circulation model of the Max-Planck Institute (MPI) in Germany. One, the "ECHAM-SULFUR" model [59] in Table 1, was developed primarily for studies of sulfate aerosol-climate interactions. The model has a rather simplified gaseous sulfur-sulfate chemistry as well as equilibrium and aqueous reactions related to $S(IV)$ and $S(VI)$. Important tropospheric chemical properties such as OH free radical concentrations and the aqueous concentrations of H_2O_2 are prescribed. The transport is handled by a semi-Lagrangian scheme. The radiation package in the GCM does not use predicted chemical concentrations to calculate radiative fluxes. In order to estimate radiative forcing by aerosols, an off-line radiative-transfer model is used.

Another interactive model built on the ECHAM GCM, ECHAM-OZONE in Table 1, has a far more complicated chemistry submodel than ECHAM-SULFUR, and was designed mainly to study tropospheric O_3 [60, 61]. The chemistry submodel calculates the transport of seven individual species or grouped species. Chemical reactions of $CO\text{-}CH_4\text{-}O_3\text{-}HO_X\text{-}NO_X$ [*e.g.*, 32] are formulated with a forward-Eulerian solver [58]. Because modeled distributions of CH_4 (the lifetime of which is about nine years) are difficult to evaluate in a few years' integration, the prescribed surface concentrations of CH_4 are used. As with ECHAM-SULFUR, this model calculates the transport of chemicals by modeled winds at each dynamical time step of the GCM.

Table 1. Selected interactive chemistry-climate models

Model	Type of Chemistry Model	Climate Model	Horizontal Resolution Lati./Long.	Prognostic Chemicals/ Reactions	Chemistry/ Radiation Coupling[1]	Reference
ECHAM-SULFUR	3D Eulerian	ECHAM4	5.6° × 5.6°	3/7[2]	No	[59]
ECHAM-OZONE	3D Eulerian	ECHAM4	5.6° × 5.6° or 3.75° × 3.75°	7/~29[3]	No	[60, 61]
LLNL	3D Lagrangian	CCM1	4.5° × 7.5°[4]	3/3[5]	No	[62, 33]
MIT	2D Eulerian	GISS/MIT	7.8°	18/54	Yes	[42]
NCAR/CNRS	2D Eulerian	[64]	5°	60/130	No	[63]
IMAGE	0D Box	UD/EB	NA[7]	22/~29[6]	No	[65]

[1] Listings indicate whether predicted concentrations of chemical species are used directly in radiation calculations for the climate model.

[2] Concentrations of H_2O_2 [69] as well as OH, NO_2, and O_3 [60] are prescribed based on the respective studies cited here.

[3] Odd-nitrogen species ($NO + NO_2 + NO_3 + 2N_2O_5 + HNO_4$) are grouped into one species in transport; reactions are formulated based on [28]; surface concentrations of CH_4 are prescribed.

[4] Size of the Eulerian mesh for wind fields and for counting air parcels.

[5] OH concentrations are prescribed based on a two-dimensional model [see 33]; aqueous conversion of S(IV) to S(VI) by H_2O_2 is assumed to have an e-folding lifetime of 30 hours at 40°N at the surface in summer.

[6] The chemical reaction rates are calculated by taking the global averages of local rates; chemical sinks of N_2O and halocarbons are calculated using prescribed destruction frequencies.

[7] NA = Not Applicable

The direct coupling of the chemistry model and climate model through the GCM radiation module was not implemented.

Chuang *et al*. [62] reported a study using the interactive version of the Lawrence Livermore National Laboratory's three-dimensional model [33]. The chemistry submodel, GRANTOUR, is a Lagrangian parcel model. When calculating the concentrations of chemical species, it launches as many as 50,000 air parcels from random locations, then derives the concentrations at each grid-point in an Eulerian grid mesh by averaging parcels occupying the same grid. Specified OH concentrations based on a two-dimensional model [see 33] are used to derive chemical conversion rates. Aqueous reactions of $S(IV)$ and $S(VI)$ are parameterized based on assumed lifetimes. The climate submodel (first generation of the National Center for Atmospheric Research's Community Climate Model, or NCAR CCM1) and the chemistry submodel communicate with one another every twelve hours, with the CCM1 providing averaged wind fields to the chemistry submodel. A separate radiation module is used off-line to analyze the radiative forcing of aerosols.

Compared with these three-dimensional models, the MIT interactive chemistry-climate model, though two-dimensional, is much more comprehensive in its treatment of processes [42]. When driven with calculated or estimated trace gas emissions from both anthropogenic and natural sources, it predicts as functions of time, latitude, and altitude the zonally averaged concentrations of the major chemically and radiatively important trace species in the atmosphere as well as climate variables. As described above, the chemistry and climate submodels in this system are fully interactive. Specifically, the transport of eighteen chemical species is driven by dynamical variables predicted by the climate submodel and the calculations of 54 gaseous and aqueous phase reactions are based on the temperatures, radiative fluxes, and precipitation rates computed in the climate model. Predicted mixing ratios of CO_2, CH_4, N_2O, two CFCs ($CFCl_3$ and CF_2Cl_2), tropospheric O_3, $HFCs$, $PFCs$, SF_6, and sulfate aerosols are then used to calculate the radiative forcing in the climate submodel.

The most comprehensive chemical reaction package appears in another two-dimensional model developed by the National Center for Atmospheric Research (NCAR) and the Centre National de la Recherche Scientifique (CNRS) [63]. It includes 130 chemical reactions and employs 60 chemical species as prognostic variables. The climate submodel [64] is relatively simple, especially in formulating surface conditions, ocean circulation, and clouds (prescribed). The transports of chemical species are calculated excluding convection. This model was designed primarily to study the middle atmosphere and therefore has a very high vertical resolution and vertical extent (from 0 to 85 km above the surface, with 1-km resolution).

The so-called "box" model or zero-dimensional model is a much simpler approach compared with these multi-dimensional models. As an example of this type of model, the climate predictions of the IMAGE model [65] are calculated by an upwelling diffusion-energy balance (UD/EB) model [66–68]. The UD/EB model separates the Earth-atmosphere system into several boxes: land, ocean, and a few atmospheric boxes. Energy is allowed to exchange between boxes. The future temperature change is linearly proportional to the net energy budget, which relates to total radiative forcing change, mixed-layer net energy flux, and climate sensitivity defined as the relative response to doubled CO_2 forcing. Simulation of atmospheric chemistry is handled in the atmospheric boxes, based on box-averaged reaction rates (CH_4-CO-OH) or box-averaged loss rates (N_2O and halocarbons) and is driven by chemical emissions predictions.

3. DIAGNOSTIC STUDIES USING CHEMISTRY-CLIMATE MODELS

A common diagnostic study using interactive chemistry-climate models involves investigation of chemical interactions among chemicals and derivation of individual chemical budgets. A typical approach is to supply the model with either preindustrial or present-day emissions, along with same initial conditions for the climate model (*e.g.*, land and ocean surface temperatures) and carry out runs to simulate the atmospheric chemical composition of the corresponding era. Integration periods are usually one to two years, and are therefore relatively inexpensive.

A good example of this type of simulation is the study of the distribution and radiative forcing of tropospheric O_3. Roelofs and Lelieveld [60] calculated the tropospheric O_3 budget of the current atmosphere using the ECHAM-OZONE model with prescribed surface CH_4 concentrations. Their work in agreement with earlier work suggested that the main production of ozone in tropical and sub-tropical regions come from reaction of NO and HO_2, which produces NO_2 and thus O_3. The total net production of ozone derived using their model is significantly lower than that using other CTMs, according to a published World Meteorological Organization (WMO) reports [70]. Comparing their model's results for preindustrial times against runs representing the present day, they concluded [61] that: (*a*) the seasonal variations and latitudinal gradients of tropospheric O_3 over continents in the Northern Hemisphere (NH) are relatively weak in the preindustrial runs, (*b*) photochemical production of ozone from anthropogenically emitted precursors contributes about 30% to present-day tropospheric O_3, and (*c*) present-day biomass burning in subtropical regions in the Southern Hemisphere (SH) significantly increases ozone in the middle and high latitudes of the SH. Their calculations suggested a global-mean radiative forcing of $+0.42\ \mathrm{W{\cdot}m^{-2}}$, with $+0.33\ \mathrm{W{\cdot}m^{-2}}$ in the SH and $+0.51\ \mathrm{W{\cdot}m^{-2}}$ in the NH due to anthropogenic perturbations in tropospheric O_3. This result is qualitatively consistent with that obtained by Hauglustaine *et al*. [63]. According to the latter study, which used a two-dimensional model, the globally averaged radiative forcing due to anthropogenic perturbations in tropospheric O_3 is about $+0.55\ \mathrm{W{\cdot}m^{-2}}$, while the maximum value in the NH is about $+0.88\ \mathrm{W{\cdot}m^{-2}}$. Their results also suggest that tropospheric O_3 in the midlatitudes of the NH have increased 80 to 120% since the Industrial Revolution.

The distribution and radiative forcing of sulfate aerosols have also been studied using interactive chemistry-climate models. Feichter *et al*. [59] studied the global sulfur cycle using the ECHAM-SULFUR model. About two-thirds of the atmospheric sulfate loading derived from their model comes from the aqueous-phase oxidation of $S(IV)$, primarily (59%) by H_2O_2. The same model was also used to link stratospheric sulfate production with the tropospheric transport of carbonyl sulfide (OCS) and SO_2 [71]. The result suggests that SO_2 transported from the troposphere is the most important precursor for the stratospheric sulfate aerosol layer, in agreement with an earlier study using a CTM [72].

The radiative forcing induced by tropospheric aerosols has been categorized as direct and indirect forcing [73], referring respectively to the fact that aerosols can directly reflect incoming solar radiation and also cause changes of cloud properties and thus alter cloud contributions to the radiative budget. The calculated direct forcing caused by sulfate aerosols is about $-0.4\ \mathrm{W{\cdot}m^{-2}}$ in one study [62] and ranges from -0.81 to $-0.55\ \mathrm{W{\cdot}m^{-2}}$ in another study [9]. Indirect forcing could range from –0.6 to –1.6 W·m^{-2} based on an assumption that a considerable amount (65 to 85%) of the total sulfate loading is attributable to the evaporation of liquid particles [62]. These results are in general agreement with the earlier IPPC estimation [4]. Another aerosol-related issue which has recently attracted much attention is the warming potential of certain types of carbonaceous aerosols. Including estimated emissions from both biomass burning and fossil-fuel use, calculations have suggested that the total forcing of carbonaceous aerosols emitted from fossil fuel burning sources is about +0.16 to +0.20 W m^{-2}, due to the black carbon components of these aerosols [9]. These calculations also assume carbonaceous aerosols from biomass burning to be mainly less-absorbing organic carbon aerosols, yielding a negative forcing in the range from –0.23 to –0.16 W m^{-2}.

4. TRANSIENT PROJECTIONS USING CHEMISTRY-CLIMATE MODELS

Another application of interactive chemistry-climate models is to help understand transient phenomena. Some current interactive models are computationally efficient enough to allow uncertainty studies through multiple transient integrations over long time periods. Others are not.

One simple transient integration is a “scenario study.” Evolution of the atmospheric concentrations of chemical species is derived based on prescribed scenarios of emissions that do or do not take into account atmospheric chemical processes. The model runs are repeated using various scenarios to predict climate. In [74], simple interactive models were used to provide this type of information and give a wide range of predicted temperature changes under various emission scenarios.

More complicated and illuminating analyses have been conducted using the MIT interactive model [*e.g.*, 42, 43, 75]. To illustrate a range of plausible predictions, we carried out a series of numerical experiments. In each of these experiments one of three plausible assumptions was made regarding each of the following three processes:

1) emissions due to human activity,

2) aerosol radiative forcing intensity (direct, indirect) and the rate of oceanic uptake of CO_2 and heat,

3) sensitivity of the climate submodel to doubling CO_2.

The three plausible assumptions are identified using the labels: H for the “high” warming case, R for the “reference” case, and L for the “low” warming case—all labels being determined specifically by the effect of the assumption on the surface temperature predicted in the year 2100. Each run is then designated by listing the labels for the three processes in the order given above. For example, the “HHH” run consists of high anthropogenic emissions, low aerosol radiative forcing intensity with slow oceanic uptake of CO_2 and heat, and high sensitivity of the climate submodel to doubled CO_2—a combination leading to the highest temperature change in the year 2100. **Tables 2** and **3** list the integrated emissions of various chemical gases and the experimental design for each run, respectively.

Table 2. Integrated chemical emissions between 1977 and 2100 for the high (H), reference (R), and low (L) emission predictions

	$CFCl_3$ (Tg)	**CF_2Cl_2 (Tg)**	**N_2O (PgN)**	**NO_x (PgN)**	**CO_2 (PgC)**	**CO (PgC)**	**CH_4 (Pg)**	**SO_2 (PgS)**
H	5.27	7.52	1.98	8.55	1912.28	98.27	110.89	13.94
R	5.27	7.52	1.94	8.40	1654.83	97.05	105.73	13.39
L	5.27	7.52	1.91	8.26	1368.73	95.73	100.21	12.86

Table 3. Designs of numerical experiments

Test	**Emissions**	**Aerosol Forcing/Ocean Uptake**[1]		**Climate Submodel Sensitivity**[2]
HHH	high	low	slow	high
HRR	high	reference	reference	reference
HLL	high	high	fast	low
RHH	reference	low	slow	high
RRR	reference	reference	reference	reference
RLL	reference	high	fast	low
LHH	low	low	slow	high
LRR	low	reference	reference	reference
LLL	low	high	fast	low

[1] Calculated optical depths of anthropogenic sulfate aerosols including direct and indirect effects are multiplied by 2 for the high and 0.5 for the low aerosol forcing setting; and ocean vertical diffusion coefficients for heat and CO_2 are five times larger or smaller than their reference value for fast or slow ocean uptake, respectively.

[2] Climate submodel sensitivity is defined as the difference in global average surface temperature between doubled CO_2 and current-day equilibrium climates. The value is 3.5, 2.5, and 2.0°C for the high, reference, and low setting respectively.

As a result of increasing emissions, the MIT model predicts that concentrations of long-lived species such as CO_2, CH_4, and N_2O will increase significantly through to the year 2100. In the "reference" (RRR) run, the global mean mole fraction in 2100 is predicted to be 745 ppm for CO_2, 410 ppb for N_2O, and 4.4 ppm for CH_4. Due to these major increases in greenhouse gas concentrations, the global mean surface temperature under these conditions increases by 2.5°C from its 1990 value by the end of the 21st Century [42]. The different emission scenarios, aerosol forcings, oceanic uptakes, and climate sensitivities in these runs produce markedly different results in terms of climate change predictions [43, 76]. For example, projected increases in surface temperature from the current level to the year 2100 range from 1.34°C for the LLL run to 5.06°C for the HHH run. Associated relative changes in global-average water vapor concentration for these same scenarios during the same period range from 6.7% to 33.4% (**Table 4**). These results qualitatively demonstrate the significant range of uncertainty of climate model predictions.

Table 4. Relative changes of selected variables

Test Set	Test	R(OH)	R(Ts)	R(CO)	R(CH_4)	R(LCH_4)	R(H_2O)
A	HHH	-20.7	37.4	102.9	180.5	123.9	33.4
	RHH	-19.3	31.3	89.2	152.8	105.5	27.4
	LHH	-16.5	27.5	73.2	125.7	89.6	23.6
B	HRR	-28.5	20.0	124.7	206.5	119.7	14.9
	RRR	-24.9	19.2	103.6	173.2	105.6	15.3
	LRR	-23.0	16.2	87.1	142.7	88.6	12.0
C	HLL	-31.1	12.2	133.0	220.3	120.3	7.6
	RLL	-29.1	10.5	114.5	185.3	103.5	6.7
	LLL	-25.7	10.0	93.5	152.2	89.9	6.7
D	HHH	-20.7	37.4	102.9	180.5	123.9	33.4
	HRR	-28.5	20.0	124.7	206.5	119.7	14.9
	HLL	-31.1	12.2	133.0	220.3	120.3	7.6
E	RHH	-19.3	31.3	89.2	152.8	105.5	27.4
	RRR	-24.9	19.2	103.6	173.2	105.6	15.3
	RLL	-29.1	10.5	114.5	185.3	103.5	6.7
F	LHH	-16.5	27.5	73.2	125.7	89.6	23.6
	LRR	-23.0	16.2	87.1	142.7	88.6	12.0
	LLL	-25.7	10.0	93.5	152.2	89.9	6.7

Here, the relative change of a given variable x is defined as $R(x) = (x_{2100} - x_{1977})/x_{1977} \times 100\%$. Here x_{1977} and x_{2100} represents the value of x in the year 1977 and in the year 2100, respectively. OH represents the tropospheric average concentration of OH (in 10^5 radicals/cm^3); Ts is the global-mean surface temperature; CO and CH_4 are the tropospheric mole fractions of CO (ppb) and CH_4 (ppm), respectively; LCH_4 is the annual photochemical loss rate of CH_4 (Tg/year); and H_2O is the tropospheric average concentration of water vapor in g/kg.

The different temperatures projected could in turn accelerate or retard the various chemical reaction rates. Also, the different water-vapor concentrations projected could significantly affect the concentration of *OH* radicals, and thus the chemical lifetimes of many species, including CH_4, *CO*, and SO_2. Results from numerical experiments using various emission predictions with the same chemistry-climate model assumptions enable calculation of the impact of emissions on future trends in tropospheric concentrations of reactive chemicals such as CH_4 and *CO* in relation to *OH* (Table 4, Groups A, B, and C). Higher emissions for the various gases (including CH_4 and *CO*) lead to lower tropospheric *OH* concentrations for the same climate model settings. For example, in runs using the "reference" climate model settings, the predicted tropospheric-mean *OH* concentrations at the end of the runs (*i.e.*, December 2100) for the three emissions scenarios are 7.9, 8.2, and 8.3×10^5 radicals/cm^3, respectively. Since runs are started from the same level, these numbers represent 28.5%, 24.9%, and 23.0% relative decreases, respectively, in the *OH* radical concentration from its 1977 level. Note that higher emissions lead to a warmer climate and higher water vapor concentrations, which, along with higher NO_X and hence O_3 concentrations, favor an increase in *OH* concentration. This partially offsets the *OH* depletion due to the rising CH_4 and *CO* levels. The contribution of the climate change to this offset is quantified by comparing a "no-forcing" run with the corresponding "forced" run to show that the increased water vapor concentration associated with climate warming offsets about 14 to 17% of the emission-induced reductions in *OH* concentration.

Comparing results from sensitivity tests using the same emissions scenario but different chemistry-climate model settings, we find that a warm climate leads to less decrease in OH concentrations due to the partial offsetting effect of increased atmospheric water vapor (Table 4, Groups D, E, and F). The difference between the high and low cases is about 6%—approximately twice the difference seen in the above emission sensitivity runs. This result indicates that changes in various chemistry-climate model parameters modify the rate of decrease in tropospheric OH even more than do changes in emissions scenarios, over the parameter ranges under investigation. The relationships among emissions, chemistry, and climate that are revealed by these sensitivity runs are apparently very complicated, strongly suggesting that further research in this field is needed.

5. FUTURE PERSPECTIVES

Current research with fully coupled chemistry-climate models indicates complicated interactions between climate change and atmospheric chemistry, and highlights their importance to accurate climate prediction. However, the drawbacks of current models are obvious: three-dimensional models are not computationally efficient enough to allow modelers to fully evaluate the models' sensitivity to assumptions and to conduct uncertainty studies. At the same time, more computationally efficient models suitable for long-term multiple integrations are inherently less realistic. In order to calculate transport of chemical species, the representations of circulation, clouds, and precipitation patterns in current climate models need to be improved. We presently know very little quantitatively about the climatic effects of aerosols, especially the indirect effect or cloud-aerosol interactions. Unless improvements are made, inadequate interpretations of the aqueous phase chemistry and cloud processes in global models, which often contradict more detailed process models [*e.g.*, 77–79], will continue to add to the uncertainties in these models.

Great uncertainties exist in the CO_2 budgets of current models, primarily due to our limited knowledge of biospheric, and to a lesser extent, oceanic sinks of CO_2. To deal with biospheric CO_2 uptake, an ecosystem model [11, 80, 81] run interactively with the chemistry-climate model is essential. Process studies are also needed to improve various aspects of current models including hydrological processes and agricultural activities. Coupling a three-dimensional ocean GCM (OGCM) with an atmospheric GCM (AGCM) is difficult but is now a common practice. Oceanic chemistry and biology involved in the cycling of CO_2 and other species will also hopefully be incorporated more realistically into future chemistry-climate models.

Because of computational limitations, the chemistry models used in interactive studies currently are still too simple to include accurately some important reactions, such as those involving NMHCs that influence O_3 production in the troposphere. Another challenge that modelers face is the accurate simulation of the chemistry of NO_X, a central player in tropospheric chemistry. The typical resolution of current models is larger than 200 km. The lifetime of NO_X (~1 day) is therefore too short to allow modelers to consider its related chemistry as a "grid-scale" processes. Ongoing research at MIT suggests that an efficient parameterization of urban pollution chemistry including NO_X-NMHC reactions, for global models could overcome this problem [82].

It is clear that computers are not fast enough currently to allow us to achieve many of the improvements discussed above. Fortunately, we expect continuing increases in computer speed, allowing us to develop better three-dimensional interactive chemistry-climate models in the next three to five years.

Finally and most importantly, observational networks with better spatial coverage are needed to evaluate models. Over the past two decades, observational networks for long-lived species have been established, providing valuable data sets for global chemistry model development [83–85]. However, observations of CO and O_3, both of which have lifetimes of about 2 to 4 months in the troposphere and therefore strong horizontal gradients, are still rare [86]. Recent use of and ongoing plans for remote sensing platforms to measure CO offer some hope for filling this gap.

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