Atmospheric chemistry–climate feedbacks

Frank Raes,1,2 Hong Liao,3 Wei-Ting Chen,2,4 and John H. Seinfeld2

Received 29 September 2009; revised 2 February 2010; accepted 24 February 2010; published 26 June 2010.

We extend the theory of climate feedbacks to include atmospheric chemistry. A change in temperature caused by a radiative forcing will include, in general, a contribution from the chemical change that is fed back into the climate system; likewise, the change in atmospheric burdens caused by a chemical forcing will include a contribution from the associated climate change that is fed back into the chemical system. The theory includes two feedback gains, Gche and Gcli. Gche is defined as the ratio of the change in equilibrium global mean temperature owing to long-lived greenhouse gas radiative forcing, under full climate-chemistry coupling, to that in the absence of coupling. Gcli is defined as the ratio of the change in equilibrium mean aerosol or gas-phase burdens owing to chemical forcing under full coupling, to that in the absence of coupling. We employ a climate-atmospheric chemistry model based on the Goddard Institute for Space Studies (GISS) GCM II’, including tropospheric gas-phase chemistry, sulfate, nitrate, ammonium, black carbon, and organic carbon. While the model describes many essential couplings between climate and atmospheric chemistry, not all couplings are accounted for, such as indirect aerosol forcing and the role of natural dust and sea salt aerosols. Guided by the feedback theory, we perform perturbation experiments to quantify Gche and Gcli.

We find that Gche for surface air temperature is essentially equal to 1.00 on a planetary scale. Regionally, Gche is estimated to be 0.80–1.30. The gains are small compared to those of the physical feedbacks in the climate system (e.g., water vapor, and cloud feedbacks). These values for Gche are robust for the specific model used, but may change when using more comprehensive climate-atmospheric chemistry models. Our perturbation experiments do not allow one to obtain robust values for Gcli. Globally averaged, the values range from 0.99 to 1.28, depending on the chemical species, while, in areas of high pollution, Gcli can be up to 1.15 for ozone, and as large as 1.40 for total aerosol. These preliminary values indicate a significant role of climate feedbacks in the atmospheric chemistry system.


1. Introduction

Attributing and predicting climate change requires linking perturbations in the Earth’s radiation balance to changes in temperature and other climate variables. Radiative perturbations, also called radiative “forcings” under certain conditions, can be caused by many factors [Forster et al., 2007]. Here, we are concerned with changes in the concentrations of long-lived and short-lived greenhouse gases and aerosols. Their role in radiative forcing has been recently reviewed by Isaksen et al. [2009]. The response of the global mean temperature to radiative forcings is embodied in the climate sensitivity [Knutti and Hegerl, 2008]. It is composed of changes in the blackbody radiation of the Earth and physical feedbacks involving water vapor, clouds and ice/snow effects [Hansen et al., 1984; Bony et al., 2006, equation (24)]. Initially, climate analyses were performed using general circulation models (GCM) including only the radiative effects of long-lived greenhouse gases (LLGHGs). Later, the effects of short-lived greenhouse gases (SLGHGs; e.g., ozone) and aerosols were included, based on prescribed fields or on fields calculated from fixed emissions. While the latter studies quantified the radiative forcings by SLGHGs and aerosols (dashed arrow in Figure 1), they initially did not consider the effects of a changing climate on the emissions and other formation and removal processes of SLGHGs and aerosols (dotted arrow in Figure 1).
processes. The review by Isaksen et al. [2009], dealing with climate-chemistry interactions, largely divides the work into, on one hand, the impacts of changes in atmospheric composition on climate, and, on the other hand, impacts of climate change on atmospheric composition. Many of the studies reviewed note the existence of feedbacks but, in fact, refer only to a one-way coupling between climate and atmospheric chemistry, or vice versa. Two recent papers more systematically address the feedback effects resulting from two-way coupling. Liao et al. [2009] approach the climate-atmospheric chemistry system by studying the production, transformation, and removal of tropospheric ozone and aerosols fully coupled to the evolving climate. GCM simulations with and without full coupling show significant differences in the predicted levels of equilibrium global mean temperature and global and regional levels of ozone and aerosols. Their study did not include aerosol-cloud interactions: the aerosol indirect effect. Unger et al. [2009] looked, in particular, at the effect of coupling of ozone and aerosols with cloud microphysical processes. They also found significant effects of full coupling on levels of air pollutants.

The above mentioned review and studies, while giving an exhaustive overview of processes that couple climate with atmospheric chemistry, indicate that atmospheric chemistry research would benefit from a consistent framework for discussing, quantifying and comparing feedbacks in the climate-atmospheric chemistry system. We propose a framework based on Figure 1. Although we deal essentially with one feedback loop within the fully coupled system, we can, and will, throughout this paper, maintain two points of view. The first is that of the climatologist, who is interested in how the presence of chemically active species in the atmosphere leads to feedbacks in the climate system, in the same way as, for example, water vapor leads to feedbacks and enhances climate sensitivity. As such we will speak about the atmospheric chemistry feedback in the climate system. The second view is that of the atmospheric chemist, who is interested how climate leads to feedbacks in the atmospheric chemistry system that affect the relationship between emissions and burdens of chemical components, or, generally speaking, the atmospheric chemistry sensitivity. We will speak about the climate feedback in the atmospheric chemistry system. Together these constitute the atmospheric chemistry–climate feedbacks referred to in the title of this paper.

The working of feedbacks in the coupled climate-atmospheric chemistry system can be illustrated with a concrete example. Consider a scenario in which both climate and atmospheric chemistry are at steady state and global SO2 emissions were to be suddenly reduced. Reduction in SO2 emissions leads immediately to a reduction in the formation of airborne sulfate aerosol. A global reduction in sulfate aerosol leads to a decrease in the negative radiative forcing associated with sulfate aerosol. As the Earth warms in response to the aerosol perturbation, the hydrological cycle adjusts such that the removal of sulfate aerosol by precipitation is, say, increased. At this point the feedback loop is closed. The additional decrease in sulfate aerosol, beyond that resulting from the original SO2 reduction, further warms the system, and so on, until a new steady state is achieved. The eventual new steady state results from the two-way coupling between climate and atmospheric chemistry. The atmospheric chemistry sensitivity, linking the SO2 emission reduction to a change in sulfate burden, affords a way coupling between climate and atmospheric chemistry.
as well as the climate sensitivity, linking the sulfate forcing to a temperature change, are each changed by the feedback within the fully coupled system. The question is: by how much?

[7] Hence, at a general level, we seek to evaluate the extent to which the presence of chemically active species in the atmosphere leads to feedbacks in the climate system. At the same time, we will evaluate how climate processes lead to feedbacks in the atmospheric chemistry system. At a more practical level, we want know what degree of sophistication is needed to perform integrated climate change and air pollution analysis. Ideally, the best tool is a fully coupled climate-atmospheric chemistry model, with comprehensive treatment of all important climate and chemical processes. However, even the most comprehensive current coupled models require the maximum computing capacity available in, for example, a typical research or meteorological center. They are therefore not practical for evaluating multiple scenarios needed in the analysis of combined climate change and air pollution mitigation options. Before making the case that fully coupled models are indeed necessary, it is of interest to evaluate the importance of coupling, i.e., to quantify the strength of the atmospheric chemistry-climate feedbacks.

[8] In this work, we use Figure 1 as a framework to study more systematically the full coupling between climate and atmospheric chemistry and the resulting feedbacks. In doing so, we use the traditional analysis of climate sensitivity and feedbacks [e.g., Hansen et al., 1984; Bony et al., 2006; Schwartz, 2007; Roe and Baker, 2007; Roe, 2009] and extend it to include atmospheric chemistry sensitivity and feedbacks. In section 2, we describe this framework theoretically, and in section 3, guided by the framework, we perform a number of perturbation experiments with a fully coupled climate-atmospheric chemistry GCM. This allows us to study the effect of full coupling on both global climate and levels of air pollutants. In section 4, we focus on the climate system, quantify the atmospheric chemistry feedback and compare it with the known physical feedbacks due to changing water vapor, lapse rate, albedo, and clouds.

2. Coupling the Climate and Atmospheric Chemistry Systems: Theoretical Framework

[9] In order to illustrate the coupling between climate and atmospheric chemistry, it is sufficient to describe the coupled system in terms of the Earth’s global mean temperature, \( T \), and the concentration of a single generic chemical component, \( C \).

[10] The basic dynamic equation for \( T \) follows from the overall planetary energy balance,

\[
\frac{dT}{dt} = \frac{S_0}{4} (1 - A(T, C)) - \varepsilon(T, C) \sigma T^4,
\]

where \( c \) is the effective heat capacity of the atmosphere-ocean system, and \( cdT \) is the change in the heat content of the system arising from an imbalance between incoming and outgoing radiation. \( S_0 \) is the solar constant and \( \sigma \) the Stefan-Boltzmann constant. \( A(T, C) \) is the planetary albedo, determined partly by cloudiness, aerosols, and the presence of snow and ice, each of which depends in a complex manner on \( T \) and \( C \). \( \varepsilon(T, C) \) is the planetary longwave emissivity, which depends on the level of LLGHGs in the atmosphere, including water vapor and SLGHGs, such as tropospheric ozone, which themselves depend on \( T \) and \( C \).

[11] The dynamic equation for \( C \) follows from the global material balance of a species,

\[
\frac{dC}{dt} = E(T, C) - D(T, C) + RX(T, C).
\]

\( E(T, C) \) and \( D(T, C) \) are the emission and physical removal rates, respectively, of the chemical component. The latter includes dry and wet deposition rates, which are dependent on the concentration of the species and on climatic conditions. Emissions of one species could, in principle, depend on the concentration of other chemical components. \( RX(T, C) \) describes the net rate of chemical production of the species.

[12] At this stage we have introduced two ways to describe the coupling between climate and atmospheric chemistry: the set of coupled differential equations (1) and (2), for \( T \) and \( C \), respectively, and the "box diagram" in Figure 1, with its sensitivity parameters \( \lambda \) and coupling factors \( c \). It is useful to show how these two descriptions relate to one another, as it will help us in the discussion of feedbacks.

2.1. Perturbation Analysis

[13] We can express equations (1) and (2) in terms of radiative and chemical imbalances as follows,

\[
\frac{dT}{dt} = R_0(T, C) = \text{the radiative imbalance (W m}^{-2})
\]

\[
\frac{dC}{dt} = R_1(T, C) = \text{the chemical imbalance (\mu g m}^{-3} s}^{-1}).
\]

Starting from a steady state, \( R_0(T^{ss}, C^{ss}) = 0 \) \( (i = 0,1) \), and applying sustained perturbations, \( \Delta R_{0f} \) and \( \Delta R_{1f} \) \( (i.e., \) radiative and chemical forcings, respectively), the climate-atmospheric chemistry system will act to restore radiative and chemical equilibrium. It will relax to a new steady state at which incoming and outgoing energy fluxes and sources and sinks of chemical species are again in balance: \( R_i(T^{ss} + \Delta T^{ss}, C^{ss} + \Delta C^{ss}) = 0 \) \( (i = 0,1) \). At any moment during the relaxation, the change of the imbalances can be written as follows:

\[
\delta R_i = \left. \frac{\partial R_i}{\partial T} \right|_C \delta T + \left. \frac{\partial R_i}{\partial C} \right|_T \delta C \quad i = 0, 1.
\]

Integrating between the start of the perturbation (when \( R_i = \Delta R_{0f} \)) and reaching the new steady state (when \( R_i = 0 \)) yields

\[
\int_{\Delta R_{0f}}^0 \delta R_0 = \left. \int_{T^{ss}}^{T^{ss} + \Delta T^{ss}} \frac{\partial R_0}{\partial T} \right|_C \delta T + \left. \int_{C^{ss}}^{C^{ss} + \Delta C^{ss}} \frac{\partial R_1}{\partial C} \right|_T \delta C \quad i = 0, 1.
\]

Assuming linear behavior in the vicinity of the steady states, it follows that

\[
\Delta R_{0f} = - \left. \frac{\partial R_0}{\partial T} \right|_C \Delta T^{ss} - \left. \frac{\partial R_1}{\partial C} \right|_T \Delta C^{ss} \quad i = 0, 1
\]
or, in matrix form,

$$
\begin{bmatrix}
\Delta R_{0f} \\
\Delta R_{1f}
\end{bmatrix} = - J \begin{bmatrix}
\frac{\partial R_0}{\partial T} & \frac{\partial R_0}{\partial C} \\
\frac{\partial R_1}{\partial T} & \frac{\partial R_1}{\partial C}
\end{bmatrix} \begin{bmatrix}
\Delta T^{ss} \\
\Delta C^{ss}
\end{bmatrix}.
$$

\[ J, \text{ the Jacobian of the coupled system described by equations (1) and (2), contains the complete sensitivity information. The result as expressed in equation (7) is a standard result from that forcing, plus a contribution from the chemical change that also results and that is fed back into the climate system. Hence,}

$$
\Delta T^{ss} = \lambda_{dl}(\Delta R_{0f} + c_{10}\Delta C^{ss}),
$$

where \( \lambda_{dl} \) is the sensitivity parameter of the climate system, and \( c_{10} \) is the factor that represents one-way coupling of chemical change with climate. Similarly, for the change in the concentration of a chemical compound, caused by a chemical forcing \( \Delta R_{1f} \), we can write

$$
\Delta C^{ss} = \lambda_{che}(\Delta R_{1f} + c_{10}\Delta T^{ss}),
$$

where \( \lambda_{che} \) is the sensitivity parameter of the atmospheric chemistry system, and \( c_{10} \) is the factor that represents one-way coupling of change on atmospheric chemistry.

\[ \text{Comparing (8a) and (8b) for } \Delta R_{0f} \text{ and } \Delta R_{1f}, \text{ and writing in matrix notation yields}

$$
\begin{bmatrix}
\Delta R_{0f} \\
\Delta R_{1f}
\end{bmatrix} = \begin{bmatrix}
\lambda_{dl} & - c_{10} \\
-c_{10} & \lambda_{che}
\end{bmatrix} \begin{bmatrix}
\Delta T^{ss} \\
\Delta C^{ss}
\end{bmatrix}.
$$

\[ \text{Comparing (7) with (9) gives an immediate interpretation of the elements in the Jacobian, of which we will make use later.}

\[ \text{Inverting equation (9) yields}

$$
\begin{bmatrix}
\Delta T^{ss} \\
\Delta C^{ss}
\end{bmatrix} = \frac{1}{1 - \lambda_{dl}\lambda_{che}c_{10}} \begin{bmatrix}
\lambda_{che} & - c_{10} \\
-c_{10} & 1
\end{bmatrix} \begin{bmatrix}
\Delta R_{0f} \\
\Delta R_{1f}
\end{bmatrix}.
$$

\[ \text{2.2. Feedback Analysis}

\[ \text{[14] Figure 1 is a “box diagram,” used in traditional feedback analysis. It shows (see Appendix A) that the change in temperature, caused by a radiative forcing \( \Delta R_{0f} \), will be a result from that forcing, plus a contribution from the chemical change that also results and that is fed back into the climate system.}

\[ \text{Hence,}

$$
\Delta T^{ss} = \lambda_{dl}(\Delta R_{0f} + c_{10}\Delta C^{ss}),
$$

where \( \lambda_{dl} \) is the sensitivity parameter of the climate system, and \( c_{10} \) is the factor that represents one-way coupling of chemical change with climate. Similarly, for the change in the concentration of a chemical compound, caused by a chemical forcing \( \Delta R_{1f} \), we can write

$$
\Delta C^{ss} = \lambda_{che}(\Delta R_{1f} + c_{10}\Delta T^{ss}),
$$

where \( \lambda_{che} \) is the sensitivity parameter of the atmospheric chemistry system, and \( c_{10} \) is the factor that represents one-way coupling of change on atmospheric chemistry.

\[ \text{[15] Solving (8a) and (8b) for } \Delta R_{0f} \text{ and } \Delta R_{1f}, \text{ and writing in matrix notation yields}

$$
\begin{bmatrix}
\Delta R_{0f} \\
\Delta R_{1f}
\end{bmatrix} = \begin{bmatrix}
\lambda_{dl} & - c_{10} \\
-c_{10} & \lambda_{che}
\end{bmatrix} \begin{bmatrix}
\Delta T^{ss} \\
\Delta C^{ss}
\end{bmatrix}.
$$

\[ \text{Comparing (7) with (9) gives an immediate interpretation of the elements in the Jacobian, of which we will make use later.}

\[ \text{Inverting equation (9) yields}

$$
\begin{bmatrix}
\Delta T^{ss} \\
\Delta C^{ss}
\end{bmatrix} = \frac{1}{1 - \lambda_{dl}\lambda_{che}c_{10}} \begin{bmatrix}
\lambda_{che} & - c_{10} \\
-c_{10} & 1
\end{bmatrix} \begin{bmatrix}
\Delta R_{0f} \\
\Delta R_{1f}
\end{bmatrix}.
$$

\[ \text{2.3. Assessing the Atmospheric Chemistry Feedback in the Climate System}

\[ \text{[17] From equation (10) it follows that}

$$
\Delta T^{ss} = \frac{\lambda_{dl}}{1 - \lambda_{che}c_{10}} (\Delta R_{0f} + \lambda_{che}c_{10}\Delta R_{1f}).
$$

\[ \text{This shows that in the fully, i.e., two-way, coupled climate-atmospheric chemistry system, the global mean temperature is, in principle, sensitive to both radiative and chemical forcings. The effects of both forcings are additive, but that holds only as long as it is justified to assume linear behavior (see equations (6) and (8)), which must be checked subsequently.}

\[ \text{[18] If there is no full coupling between climate and chemistry (} c_{10} = 0 \text{ or } c_{10} = 0 \text{ or } c_{10} = c_{10} = 0 \text{), equation (11) becomes}

$$
\Delta T^{ss} = \lambda_{dl}(\Delta R_{0f} + \lambda_{che}c_{10}\Delta R_{1f}).
$$

\[ \text{Comparing (11) and (12) shows how, because of full coupling with atmospheric chemistry, the climate sensitivity parameter changes,}

$$
\lambda = \frac{\lambda_{dl}}{1 - \lambda_{che}c_{10}}.
$$

\[ \text{where } \lambda \text{ is the climate sensitivity parameter of the coupled system. According to the feedback analysis (see equation (A4)), } \lambda_{che}c_{10} \text{ has the significance of a feedback parameter, and can be termed the atmospheric chemistry feedback parameter, } \lambda_{che}.

\[ \text{c_{che} = \lambda_{che}c_{10}.}

\[ \text{In fact, if we are interested only in radiative perturbations (} \Delta R_{1f} = 0 \text{), but still want to describe the feedback due to atmospheric chemistry, Figure 1 can be simplified into Figure 2.}

\[ \text{[19] Following equation (A5), the gain } G_{che} \text{ of the system, i.e., the ratio of output of the coupled system to the output of the uncoupled system, can be calculated as the ratio of equations (11) and (12),}

$$
G_{che} = \frac{\Delta T^{ss}}{\Delta T^{ss}_{an}} = \frac{1}{1 - \lambda_{che}c_{10}}.
$$

\[ \text{If } c_{che} < 0, \text{ then } G_{che} < 1 \text{ and atmospheric chemistry will dampen the climate sensitivity; if } c_{che} > 0, \text{ then } G_{che} > 1 \text{ and it will amplify the climate sensitivity.}

\[ \text{[20] As shown in equation (14), the atmospheric chemistry feedback is composed of three factors, and using the comparison of the matrices in (7) and (9), it can be written more explicitly as}

$$
c_{che} = \lambda_{che}c_{10}.
$$

\[ \text{Each of the three factors can determine the sign and the magnitude of the atmospheric chemistry feedback. What this means can be illustrated by considering a simple system, consisting of nonreactive black carbon (BC) aerosols only. In this case, equation (2) becomes}

$$
\frac{dC}{dt} = E - \beta(T)C = R(T).
$$

\[ \text{where } C \text{ is the global burden of BC, } E \text{ its global emissions rate, and } \beta(T) \text{ its globally average removal rate. The latter is taken to be proportional to the precipitation rate } P, \text{ which is known to be dependent on the global mean temperature } T: \beta(T) = \alpha P(T).
The first and third factors on the LHS of equation (16) can be obtained by differentiating equation (17). Hence

\[ c_{che} = \frac{\partial P}{\partial T} \frac{\partial R_0}{\partial C} \bigg|_C = -hr. \]  

(18)

The first factor on the RHS of equation (18) is the hydrological sensitivity, \( h \), defined as the fractional change in global mean precipitation per unit of increase of global mean temperature. The second factor can be called the radiative forcing sensitivity, \( r \), defined as the change in global radiative forcing per fractional change in the burden of the forcing agent. The latter can be calculated from the radiative forcing of BC between pre-industrial and present day, which is 0.60 W m\(^{-2}\) in the model used here, and from the consideration that this forcing is caused by about 90% of the present burden of BC [Chung and Seinfeld, 2005]. Hence \( r = 0.60/0.90 = 0.66 \) W m\(^{-2}\). The sign of \( h \) depends critically on how the increase in global mean temperature occurs. If it is due to an increase in LLGHG, it is about 0.02/K; if it is the result of an increase in LLGHG and aerosols combined, it can be −0.02/K, depending on the aerosol mixture and assumed scenario for the aerosol increase [Feichter et al., 2004]. Remembering that we are assessing the atmospheric chemistry feedback that possibly affects the warming initially caused by a change in LLGHGs, we take \( h = 0.02/K \), and hence, \( c_{che} = -0.013 \) W m\(^{-3}\)K. The negative sign means that the presence of BC aerosols in the climate system results in a damping of the climate sensitivity; an increase in \( T \) will lead to an increase in precipitation and therefore increased removal of BC, and since BC is a warming agent, its removal will lead to a cooling, and a damping of the initial warming. The value of \( c_{che} \) in this example is, however, quite small and, as we will see in section 4, essentially negligible compared to the values of the physical feedback parameters in the climate system. This is despite the fact that BC aerosols exert a strong positive radiative forcing. However, in the overall feedback loop, the contribution of radiative forcing is diminished by the low hydrological sensitivity. In other words, the fact that a chemical component exerts a strong radiative forcing is clearly a necessary, but not a sufficient, condition for it to lead to a nonnegligible feedback in the climate system.

The simple calculation above for black carbon cannot be easily repeated for the mixture of reactive chemical components in the atmosphere, the burdens of which are interdependent in a complex manner and can lead, for example, to either a positive or negative hydrological sensitivity. Instead, we will perform perturbation experiments with a fully coupled climate-atmospheric chemistry model, as described in section 3. These experiments will lead to an overall gain, \( G_{che} \), which describes the feedback produced by all interacting chemical components in the climate system. From \( G_{che} \) the chemical feedback parameter, \( c_{che} \), can be obtained, using equation (15), assuming that \( l_{cli} \) is known.

### 2.4. Assessing the Climate Feedback in the Atmospheric Chemistry System

From equation (10) it also follows that

\[ \Delta C^{cli} = \frac{\lambda_{cli}}{1 - \lambda_{cli} c_{che}} (\Delta R_{1f} + \lambda_{cli} c_{che} \Delta R_{0f}). \]  

(19)

Further developing this equation, in the same way as we did for \( \Delta T^{ss} \) above, is of limited use, because it is difficult to select a generic chemical component from the
Table 1a. Climate Variables, Considered in the Present Study, That Influence Emissions, Chemical Reactions, Transport, and Deposition of Gas-Phase Species and Aerosols

<table>
<thead>
<tr>
<th>Climate Variables</th>
<th>Atmospheric Chemistry Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature, precipitation</td>
<td>NOx emissions (soil)</td>
</tr>
<tr>
<td>Frequency of convective events</td>
<td>NOX emissions (lightning)</td>
</tr>
<tr>
<td>Temperature, solar radiation</td>
<td>biogenic hydrocarbon emissions</td>
</tr>
<tr>
<td>Surface wind, temperature</td>
<td>dimethyl sulfide (DMS) emissions</td>
</tr>
<tr>
<td>Wind</td>
<td>sea salt emissions</td>
</tr>
<tr>
<td>Wind, precipitation</td>
<td>mineral dust emissions</td>
</tr>
<tr>
<td>Temperature, water vapor</td>
<td>chemical reaction rates</td>
</tr>
<tr>
<td>Temperature, relative humidity</td>
<td>aerosol-gas-phase equilibrium</td>
</tr>
<tr>
<td>Cloud amount, cloud water content</td>
<td>in-cloud aerosol formation</td>
</tr>
<tr>
<td>Winds, atmospheric stability</td>
<td>transport of chemical species</td>
</tr>
<tr>
<td>Atmospheric stability, clouds,</td>
<td>deposition (wet and dry)</td>
</tr>
<tr>
<td>precipitation</td>
<td></td>
</tr>
</tbody>
</table>

many that are present in the atmosphere. A vector/matrix analysis could be carried out to describe the effect of climate feedback on the chemical components, but it is not necessary to present that here.

[24] Perturbation experiments with the full climate-atmospheric chemistry model will yield, for each of the chemical components $i$, a gain,

$$ G_{cl,i} = \Delta C_{i}^{gas} / \Delta C_{i}^{sun}, $$

(20)

where $\Delta C_{i}^{gas}$ is the change in the concentration of component $i$ due to a forcing in the fully coupled climate-chemistry system, and $\Delta C_{i}^{sun}$ is the corresponding change in the uncoupled system. These gains will give insight into the importance of climate as a feedback within the atmospheric chemistry system.

3. Quantifying Atmospheric Chemistry and Climate Feedbacks

[25] Given the complexity of the coupled climate-atmospheric chemistry system, one cannot estimate a priori in many cases even the sign of the atmospheric chemistry and climate feedbacks. This can only be done through simulations with a fully coupled climate-atmospheric chemistry model. While many processes in such a model are known to be nonlinear, the structure of the linear feedback theory is used here as a guide for a series of perturbation experiments that are required to compare the magnitude and sign of the various feedbacks within a consistent conceptual framework. In particular, the experiments allow the quantification of the gains, $G_{chem}$ and $G_{cl,i}$, and their related feedback parameters.

[26] It should be clear that the values for the gains and feedback parameters are descriptors of the particular climate-atmospheric chemistry used in the experiments, in the same way as the climate sensitivity is a descriptor of a particular climate model. The robustness of the values for the gains and feedback factors obtained from the perturbation experiments will need to be discussed, especially when deviations from the linear theory do occur.

3.1. Model Description: CACTUS Unified Model

[27] The Unified Model developed in the National Aeronautics and Space Administration (NASA) project, Chemistry, Aerosols, and Climate: Tropospheric Unified Simulation (CACTUS), simulates the fully coupled interactions of chemistry, aerosol, and climate based on the 4° latitude by 5° longitude, nine-layer Goddard Institute for Space Studies (GISS) GCM II’, as described in detail in previous studies [Liao et al., 2003, 2004; Liao and Seinfeld, 2005; Liao et al., 2006, 2009].

[28] The model includes a detailed simulation of tropospheric O$_3$-NO$_x$-hydrocarbon chemistry, as well as sulfate, nitrate, ammonium, black carbon (BC), primary organic aerosols (POA), secondary organic aerosols (SOA), sea salt, and mineral dust. Two-way coupling between aerosols and gas-phase chemistry provides consistent chemical fields for aerosol dynamics and aerosol mass, for heterogeneous processes and calculations of gas-phase photolysis rates. Additional information on the treatment of atmospheric chemistry is given in Appendix B. Table 1a list the climate variables in the model that influence emissions, chemical reactions, transport, and deposition of gas-phase species and aerosols, whereas Table 1b lists the atmospheric chemistry variables in the model that influence climate. Hence, in the fully coupled mode, the climate responds to radiative perturbations associated with the varying concentrations of LLGHGs, aerosols and O$_3$. Only the direct radiative effect of aerosols, through scattering and absorption of radiation, is considered here. The aerosol indirect effect is not considered; clouds do, however, respond to changes in climate variables driven by LLGHGs, tropospheric ozone, and direct aerosol forcing. Consideration of the aerosol indirect radiative effect in climate-chemistry feedbacks should be the subject of future work. For aerosol and gas-phase species, the removal and transport processes, the thermodynamic partitioning, and reaction rates of the temperature-sensitive reactions are calculated simultaneously based on relevant climate variables such as temperature, precipitation, and wind fields. Coupling between climate and the chemistry fields can be turned on and off, depending on the need of the experiments.

[29] In the GCM the atmosphere is coupled to a “Q-flux” ocean [Hansen et al., 1984], in which the Monthly horizontal heat transport fluxes are held constant as in work by Mickley et al. [2004], while changes in the sea surface temperature and sea ice are calculated based on energy exchange with the atmosphere, ocean heat transport, and the ocean mixed layer heat capacity [Hansen et al., 1984]. In the atmosphere nine vertical layers in a $\sigma$-coordinate system extend from the surface to 10 mbar. The dynamical time step in the GCM is 1 h, while the chemistry subroutines are called every 4 h. The GISS GCM-II’ has been used extensively to probe the climate response to perturbations in LLGHG concentrations, solar luminosity, and tropospheric O$_3$ and aerosol burdens [e.g., Grenfell et al., 2001; Rind et al., 2006].

Table 1b. Atmospheric Chemistry Variables, Considered in the Present Study, That Influence Climate

<table>
<thead>
<tr>
<th>Atmospheric Chemistry Variable</th>
<th>Climate Processes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Concentration of tropospheric ozone</td>
<td>radiative effect</td>
</tr>
<tr>
<td>Concentration of all aerosol species, except sea salt and mineral dust</td>
<td>direct radiative effect only (assuming internal mixing of aerosol components)</td>
</tr>
</tbody>
</table>
3.2. Perturbation Experiments: Setup

Aa A1: 2000 aerosol and gas
N2O (ppbv) 316 447
Ethane (Tg C yr⁻¹) 32 109.7

B1: 2000 aerosol and gas
BC (Tg C yr⁻¹) 6.7 28.1
Acetone (Tg C yr⁻¹) 30.1 60.5
SO2 (Tg S yr⁻¹) 71.3 62.6

Ethane (Tg C yr⁻¹) 8.6 100.1
Propane (Tg C yr⁻¹) 4.0 11.0
≥C4 alkanes (Tg C yr⁻¹) 32 109.7
≥C3 alkenes (Tg C yr⁻¹) 22 41

Acetone (Tg C yr⁻¹) 9 9
SO2 (Tg S yr⁻¹) 46.9 102.6
NH3 (Tg N yr⁻¹) 2.0 6.7

Table 3. Description of the Model Experiments and Their Outcome

<table>
<thead>
<tr>
<th>Experiments</th>
<th>Outcome</th>
<th>Gains</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aa A1: 2000 aerosol and gas-phase concentrations kept fixed; 2000 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to GHG radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>A1: 2000 aerosol and gas-phase concentrations kept fixed; 2100 GHG concentrations</td>
<td>ΔCspi: change in equilibrium global air pollutant concentrations owing to a chemical forcing, without allowing atmospheric chemistry to affect climate (uncoupled)</td>
<td>Gclcli = ΔCspi</td>
</tr>
<tr>
<td>B1: 2000 aerosol and gas-phase precursor emissions; 2000 GHG concentrations</td>
<td>ΔTspi and ΔCspi: changes in equilibrium global mean temperature and in levels of air pollutants, owing to GHG radiative forcing, allowing for full climate-chemistry coupling</td>
<td>Gclcli = ΔCspi</td>
</tr>
<tr>
<td>B1: 2000 aerosol and gas-phase precursor emissions; 2100 GHG concentrations at year 2100 level</td>
<td>ΔCspi: change in equilibrium global air pollutant concentrations owing to a chemical forcing, without allowing atmospheric chemistry to affect climate (uncoupled)</td>
<td>Gclcli = ΔCspi</td>
</tr>
<tr>
<td>C1: 2000 aerosol and gas-phase precursor emissions; 2000 climate kept fixed</td>
<td>ΔTssr and ΔCssr: changes in equilibrium global mean temperature and in levels of air pollutants owing to the chemical forcing, allowing for full climate-chemistry coupling</td>
<td>Comparing, e.g., ΔTssr + ΔCssr allows quantifying the linearity of the system</td>
</tr>
<tr>
<td>C: 2100 aerosol and gas-phase precursor emissions; 2000 climate kept fixed</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>D1: 2000 aerosol and gas-phase precursor emissions; 2000 climate kept fixed</td>
<td>ΔTssr and ΔCssr: changes in equilibrium global mean temperature and in levels of air pollutants owing to a radiative forcing, allowing for full climate-chemistry coupling</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>D2: 2100 aerosol and gas-phase precursor emissions; 2000 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>E1: 2000 aerosol and gas phase precursor emissions; 2000 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>E2: 2100 aerosol and gas phase precursor emissions; 2100 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>F1: 2100 aerosol and gas phase precursor emissions; 2000 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>F2: 2100 aerosol and gas phase precursor emissions; 2100 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>G1: 2100 aerosol and gas phase precursor emissions; 2000 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
<tr>
<td>G2: 2100 aerosol and gas phase precursor emissions; 2100 GHG concentrations</td>
<td>ΔTssr: change in equilibrium global mean temperature owing to a radiative forcing, without allowing climate to affect air pollutant concentrations (uncoupled)</td>
<td>Gche = ΔTssr</td>
</tr>
</tbody>
</table>

B: Reference year for atmospheric chemical composition: 2100. Note: G2 = E2.
3.3. Perturbation Experiments: Results

Results of the perturbation experiments are summarized in Table 4.

3.3.1. Effect of Atmospheric Chemistry Feedback on Climate (Temperature and Precipitation) Sensitivity

In experiments A and B, the radiative forcing at the top of the atmosphere, as a result of perturbing the concentrations of LLGHGs, is 6.63 W m\(^{-2}\). The steady state temperature and precipitation increase by 5.32 K and 0.336 mm d\(^{-1}\) in experiment A, and by 5.28 K and 0.342 mm d\(^{-1}\) in experiment B. As a result of the latter increases, the burdens of chemical species also undergo significant changes in experiment B, for example, -12% for ozone, -48% for nitrate aerosols, -10% for black carbon and -14% for total aerosols (i.e., the sum of all individual aerosol species). With a positive hydrological sensitivity, a decrease in scattering aerosols amplifies the climate sensitivity, while a decrease in ozone and black carbon dampens it (see illustrative example above). Apparently, the latter effect dominates, as the gain \(G_{che}\) for temperature, derived from experiments A and B is 5.28/5.32 = 0.99, i.e., a small damping of the climate (= temperature) sensitivity. The gain \(G_{che}\) for precipitation, 0.342/0.336 = 1.02, indicates a small amplification of the precipitation sensitivity. The latter can be explained by the fact that the initial reduction in aerosol burdens owing to an increase in precipitation allows for more radiation reaching the surface, more evaporation, and a further increase in precipitation. The strength of these feedbacks appears to be dependent on the chemical state of the atmosphere; this is shown by experiments F and G, in which the chemical composition of the atmosphere is that of 2100. The temperature and precipitation sensitivities now increase by factors 1.02 and 1.05, respectively. The values of these gains are very small, indicating that atmospheric chemistry, as treated here, does not lead to important feedbacks in the climate system at the planetary scale. We will discuss this further in section 4.

So far we have calculated the gain \(G_{che}\) from the globally averaged values of surface temperature and precipitation. We can also use values in each surface grid point of the model and study the geographical distribution of \(G_{che}\). This is done in Figure 3 for the gain for temperature. It shows that, locally, this gain can be significant: as low as 0.8 and as high as 1.3. As can be expected, \(G_{che}\) depends on the

\[ D_2, \text{ and } F_1 \] were performed to complete the present study. All experiments were run for a sufficiently long time, ranging from 35 to 80 years, to establish a steady state in climate and atmospheric chemistry. Each of the pairs allows one to calculate a particular forcing and its effect on climate and chemical burdens. This is done with climate and chemistry uncoupled (pairs A, C and F), and with climate and chemistry fully coupled (pairs B, D, E and G). By combining results from the coupled and uncoupled runs, values for \(G_{che}\) and \(G_{cli,i}\) are obtained (see Table 3). For the A–E runs summarized in Table 3, the reference case, which is subsequently perturbed, has 2000 GHG levels and chemical composition. For the F and G runs in Table 3 the reference case has 2000 GHG levels but a 2100 chemical composition. In this way the effect of the atmospheric chemical state on the atmospheric chemistry feedback can be evaluated.

3.3. Perturbation Experiments: Results

Results of the perturbation experiments are summarized in Table 4.

3.3.1. Effect of Atmospheric Chemistry Feedback on Climate (Temperature and Precipitation) Sensitivity

In experiments A and B, the radiative forcing at the top of the atmosphere, as a result of perturbing the concentrations of LLGHGs, is 6.63 W m\(^{-2}\). The steady state temperature and precipitation increase by 5.32 K and 0.336 mm d\(^{-1}\) in experiment A, and by 5.28 K and 0.342 mm d\(^{-1}\) in experiment B. As a result of the latter increases, the burdens of chemical species also undergo significant changes in experiment B, for example, -12% for ozone, -48% for nitrate aerosols, -10% for black carbon and -14% for total aerosols (i.e., the sum of all individual aerosol species). With a positive hydrological sensitivity, a decrease in scattering aerosols amplifies the climate sensitivity, while a decrease in ozone and black carbon dampens it (see illustrative example above). Apparently, the latter effect dominates, as the gain \(G_{che}\) for temperature, derived from experiments A and B is 5.28/5.32 = 0.99, i.e., a small damping of the climate (= temperature) sensitivity. The gain \(G_{che}\) for precipitation, 0.342/0.336 = 1.02, indicates a small amplification of the precipitation sensitivity. The latter can be explained by the fact that the initial reduction in aerosol burdens owing to an increase in precipitation allows for more radiation reaching the surface, more evaporation, and a further increase in precipitation. The strength of these feedbacks appears to be dependent on the chemical state of the atmosphere; this is shown by experiments F and G, in which the chemical composition of the atmosphere is that of 2100. The temperature and precipitation sensitivities now increase by factors 1.02 and 1.05, respectively. The values of these gains are very small, indicating that atmospheric chemistry, as treated here, does not lead to important feedbacks in the climate system at the planetary scale. We will discuss this further in section 4.

So far we have calculated the gain \(G_{che}\) from the globally averaged values of surface temperature and precipitation. We can also use values in each surface grid point of the model and study the geographical distribution of \(G_{che}\). This is done in Figure 3 for the gain for temperature. It shows that, locally, this gain can be significant: as low as 0.8 and as high as 1.3. As can be expected, \(G_{che}\) depends on the
The gain increases in NO an increase of 400% in nitrate burdens, reflecting the large burdens of the chemical components, for example, up to precursors. This generally leads to large changes in the imposed by changing emissions of air pollutants and their feedback, operating in the high latitudes. by the physical feedbacks, such as, e.g., the snow/ice atmospheres, and that climate sensitivity is rather governed chemistry feedback is not necessarily related to polluted chemistry has an amplifying effect in the 2100 atmosphere. Their geographical pattern does not correlate with those of the air pollution fields nor with that of warming. This shows that the atmospheric chemistry feedback is not necessarily related to polluted atmospheres, and that climate sensitivity is rather governed by the physical feedbacks, such as, e.g., the snow/ice feedback, operating in the high latitudes.

In experiments C and D a chemical forcing is imposed by changing emissions of air pollutants and their precursors. This generally leads to large changes in the burdens of the chemical components, for example, up to an increase of 400% in nitrate burdens, reflecting the large increases in NOx and NH3 emissions in the A2 scenario (see Table 2). These changes in the burdens also result in a top of the atmosphere radiative forcing. This forcing is calculated from experiment C, where climate is kept fixed, and amounts to 0.95 W m\(^{-2}\); 0.72 W m\(^{-2}\) due to the change in ozone burden plus 0.23 W m\(^{-2}\) due to the change in aerosol burden. This forcing is effective in experiment D where it leads to an increase of temperature of 0.48 K and a decrease of precipitation, −0.019 mm d\(^{-1}\). The warming is again explained by the dominance of ozone and BC, whereas the drying is explained by a reduction of incoming radiation, hence a reduction in evaporation at the surface due to the increased aerosol burden.

Experiment E evaluates the effects of the radiative and chemical forcings combined. In a purely linear system, these effects on climate are expected to be equal to the sum of the effects of the individual forcings, obtained in experiments B and D (see equation (11)). The Lin column in Table 4 shows that the temperature increase owing to increasing LLGHGs and air pollutants simultaneously is within 3% of the sum of the temperature increases obtained by increasing LLGHGs and air pollutants separately. The same conclusion holds for precipitation change. A similar conclusion was reached by Kirkevag et al. [2008] and Kloster et al. [2010]. Feichter et al. [2004], on the other hand, found significant deviations from additivity. We will refrain from making numerical comparisons with these or other studies, because the air pollutants considered as well as their interactions with the climate system are different. To explain the differences would require a detailed intercomparison study, which is beyond the scope of the present study.

The fact that additivity holds in the present experiments is an indication that the imposed perturbations do not lead the modeled temperature and precipitation out of the linear range. The changes in temperature and precipitation, at most 5.93 K and 0.35 mm d\(^{-1}\), are indeed small compared to the unperturbed values of 287.4 K and 3.19 mm d\(^{-1}\), respectively. In addition, the values for \(G_{\text{che}}\) are obtained by radiative forcing experiments alone, hence we are not concerned about the issues existing with chemical forcings mentioned earlier. We conclude that the values for \(G_{\text{che}}\) are robust, but valid only for the particular climate-atmospheric chemistry model used.

### 3.3.2. Effects of Climate Feedback on Atmospheric Chemistry Sensitivity

Changing the emissions of air pollutants and their precursors in experiment C leads to a change in the steady state global burdens. As pointed out earlier, these changes are large, for example, up to +59% in ozone burdens, +382% in nitrate aerosol, +134% in BC and +87% in total aerosol. The same change of emissions in experiment D, in which full coupling between atmospheric chemistry and climate occurs, leads to different changes: +59%, +399%, +144% and +91% for ozone, nitrate aerosol, BC and total aerosol burdens, respectively. These changes show that climate does feed back on the atmospheric chemistry sensitivity, or, more simply, on the relationship between emissions and burdens.

In the case of ozone, the global gain \(G_{\text{cli}}\) is 0.99, but with local values that are significant and up to 1.15, especially in the Northern Hemisphere, and down to 0.8 in some areas over the remote oceans (see Figure 4a). The positive feedback values can be explained by the positive effect of chemical forcing on temperature (+0.48 K), which over the continents of the Northern Hemisphere has a positive impact on ozone production, by, for example, more active NO\(_x\) chemistry and increased biogenic ozone precursors. Over the oceans the higher water vapor, resulting from the slight warming, dominates and contributes to ozone destruction.

For aerosols, the climate feedback is generally positive; \(G_{\text{cli}} > 1\). As pointed out by Liao et al. [2009], this is
underpredictions compared to the results of applying these forcings separately (experiments B and D), can lead to large overprediction or underprediction of climate feedbacks. This can be explained by the fact that in experiment E, sulfate aerosol chemistry is occurring at a temperature that is 5.93 K higher than the unperturbed run, compared to only 0.48 K higher in experiment D. At this higher temperature, global ozone destruction through higher water vapor concentrations seems to be more important than global ozone production by more active NOx chemistry and higher biogenic emissions. In the case of nitrate and ammonium, the overpredictions are 101% and 74%, respectively. This can again be explained by the much larger temperature increase in experiment E and the temperature dependence of the ammonium/nitrate-ammonia/nitric acid equilibrium, which leads to less ammonium nitrate in experiment E.

The fact that additivity does not hold in these experiments is an indication that the imposed perturbations do lead to modeled chemical burdens that are out of the linear range. In addition, we need to recognize the problem that the calculated gains are dependent on the particular IPCC A2 scenario from which the perturbations were derived. The values for the gains $G_{cli}$ in Table 4 and Figure 4 should be considered as preliminary and give an indication of the strength of climate feedback in the atmospheric chemistry of individual species. In future experiments, it will be worthwhile to perturb systematically the emissions of individual chemical compounds or groups of components, for example, emitted by a single economic sector, to come to robust values of $G_{cli}$. 

### 4. Comparison of Atmospheric Chemistry Feedback With Known Physical Feedbacks in the Climate System

The atmospheric chemistry feedback parameter, $c_{che} = \lambda_{che} c_{ch0} + c_{che}$, derived in section 2.3, equation (14), can be compared directly with the parameters of the physical feedbacks. This can be understood by making the physical feedbacks explicit in the analysis. Figure 2a can be transformed into Figure 2b, in which $\lambda_{cli}$ is the sensitivity parameter of a new reference system, with respect to which the physical feedbacks, $c_{00}$, as well as the atmospheric chemistry feedback, $c_{che}$, can be assessed. The climate sensitivity parameter, $\lambda_{cli}$, appearing in Figures 1 and 2a, can thus be written as

$$\lambda_{cli} = \frac{\lambda_0}{1 - \lambda_0 c_{ch0}}$$

Introducing this in the expression for the climate sensitivity parameter of the coupled climate-atmospheric chemistry system, $\lambda$ (see equation (13)), yields

$$\lambda = \frac{\lambda_{cli}}{1 - \lambda_{cli} c_{che}} = \frac{\lambda_0}{1 - \lambda_0 (c_{ch0} + c_{che})}$$

Equation (22) shows how the physical and chemical feedback parameters are additive and can be quantitatively compared.
[45] The physical significance of $\lambda_0$ and $c_{00}$ is straightforward. Comparison of (7) and (9) shows that

$$\lambda_{0l}^{-1} = -\frac{\partial R_0}{\partial T} \bigg|_c .$$

Differentiating equation (1) with respect to $T$ yields

$$\lambda_{cl}^{-1} = 4\sigma T^3 + \frac{S_0}{4} \frac{\partial A}{\partial T} \bigg|_c + \sigma T^{\alpha_s} \frac{\partial E}{\partial T} \bigg|_c ,$$

and from a comparison with equation (21) we can write

$$\lambda_0^{-1} = 4\sigma T^3$$

and

$$c_{00} = \frac{S_0}{4} \frac{\partial A}{\partial T} \bigg|_c - \sigma T^{\alpha_s} \frac{\partial E}{\partial T} \bigg|_c .$$

Equation (25) shows that the new reference system for a joint analysis of physical and chemical feedbacks is the Earth acting as a blackbody, for which $\lambda_0 = 0.31$ K/W m$^{-2}$ [e.g., Soden and Held, 2006]. Equation (26) shows that $c_{00}$ can be decomposed into various contributions. The first term on the RHS of (26) describes feedbacks related to changes in the planetary albedo with temperature, such as cloud and snow/ice feedbacks. The second term describes feedbacks related to changes in the emissivity with temperature, such as water vapor and atmospheric lapse rate feedback. The feedback parameters of each of these processes are continuously being refined by the climate modeling community. Most probable values are given by Randall et al. [2007] (see Table 5), and their summation leads to a value for $c_{00}$ of 1.91 W m$^{-2}$K. (Note that with the latter value for $c_{00}$ and $\lambda_0 = 0.31$ K/W m$^{-2}$, it follows from (21) that $\lambda_{cl} = 0.78$ K/W m$^{-2}$. That is practically equal to the climate sensitivity of the particular climate model used in our study, GISS GCM II', $\lambda_{cl} = 0.80$ K/W m$^{-2}$.)

[46] Finally, using equation (15), the atmospheric chemistry feedback parameter $c_{che}$ can be calculated from the values for $G_{che}$ obtained in the perturbation analyses and $\lambda_{cl} = 0.80$ K/W m$^{-2}$. This results in a value of $-0.013$ W m$^{-2}$K under the present-day chemical composition of the atmosphere, and 0.025 W m$^{-2}$K under a 2100 chemical composition. The gains $G_{che}$ obtained in the perturbation analyses are those with respect to a reference system including the physical feedbacks. In order to calculate the gains with respect to the Earth as a blackbody, and be able to compare them with the gains of the physical feedbacks, we can use the values just obtained for $c_{che}$ and plug them in equation (15) using $\lambda_0 = 0.31$ K/W m$^{-2}$, instead of $\lambda_{cl} = 0.80$ K/W m$^{-2}$. The result, shown in Table 5, is a gain of 0.996 under present-day chemical composition and 1.008 under 2100 chemical composition. Hence, a change in atmospheric chemical composition according to the A2 scenario might turn the atmospheric chemistry feedback from slightly negative to slightly positive.

[47] Table 5 shows how the values for the atmospheric chemistry feedback parameter and the corresponding gain are quite small compared to the parameters and gains of the known physical feedbacks: for example, atmospheric chemistry gains of 0.996 and 1.008, compared to gains of 2.26, 0.80, 1.28 and 1.09, for the water vapor, lapse rate, cloud and surface albedo feedback, respectively. While atmospheric chemistry is found to lead only to a minor feedback in the globally averaged climate system, chemical feedbacks on climate can be important regionally, owing to the unique features of ozone and aerosols in the climate system.

### Table 5. Climate Feedback Parameters and Gains

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$c_i$ (W m$^{-2}$K)</th>
<th>$G_i = \frac{1}{\lambda_i}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water vapor</td>
<td>1.80 ± 0.18</td>
<td>2.27</td>
<td>Randall et al. [2007]</td>
</tr>
<tr>
<td>Lapse rate</td>
<td>-0.84 ± 0.26</td>
<td>0.80</td>
<td>Randall et al. [2007]</td>
</tr>
<tr>
<td>Clouds</td>
<td>0.69 ± 0.38</td>
<td>1.28</td>
<td>Randall et al. [2007]</td>
</tr>
<tr>
<td>Surface albedo (2000)</td>
<td>0.26 ± 0.08</td>
<td>1.09</td>
<td>Randall et al. [2007]</td>
</tr>
<tr>
<td>Atmospheric chemistry (2000)</td>
<td>-0.013</td>
<td>1.00</td>
<td>this work</td>
</tr>
<tr>
<td>Atmospheric chemistry (2100)</td>
<td>0.025</td>
<td>1.01</td>
<td>this work</td>
</tr>
</tbody>
</table>

5. Summary and Conclusions

[48] We have developed a framework to analyze and quantify atmospheric chemistry-climate feedbacks. We do so by extending the familiar feedback analysis used in climate studies to include atmospheric chemistry.

[49] Feedbacks exist only when there is a full, i.e., two-way, coupling between climate and atmospheric chemistry; i.e., when a feedback loop is created (see Figure 1). Atmospheric chemistry creates a feedback in the climate system, and by the same token, climate creates a feedback in the atmospheric chemistry system.

[50] The analysis of the coupled climate-atmospheric chemistry system shows how each of these feedbacks consists of at least three factors. For instance, the atmospheric chemistry feedback in the climate system is determined by: (1) the change in chemical imbalance, per unit of temperature change ($c_{10}$ in Figure 1); (2) the atmospheric chemistry sensitivity, $\lambda_{che}$; and (3) the radiative imbalance per unit of chemical burden ($c_{01}$). Hence, the fact that a change in the global burden of a chemical compound leads to a large radiative imbalance (forcing) is a necessary, but not a sufficient, condition for a strong feedback of atmospheric chemistry on climate. For aerosols in particular, the hydrological sensitivity, i.e., the fractional change in precipitation per degree K warming, seems to play an important role in determining both the magnitude and sign of the atmospheric chemistry feedback. In the case of a positive hydrological sensitivity, absorbing aerosols like black carbon will dampen the climate (temperature) sensitivity, while scattering species like sulfate and nitrate aerosols will amplify the climate sensitivity. In the case of a negative hydrological sensitivity, the roles of absorbing and scattering aerosols will reverse.

[51] We use the framework to define a number of perturbation experiments with the CACTUS model, which simulates many, but not all, of the interactions of chemistry,
aerosol, and climate within the GISS GCM II’ climate model. Radiative perturbations allow one to quantify the effect of atmospheric chemistry on climate sensitivity and to calculate an atmospheric chemistry feedback parameter that can be directly compared with the corresponding parameters of known physical feedbacks: the water vapor, cloud, lapse rate and surface albedo feedbacks. Chemical perturbations allow one to quantify the effect of climate on atmospheric chemistry sensitivity. The following conclusions can be drawn from these experiments.

Within the context of our particular climate-atmospheric chemistry model (CACTUS), atmospheric chemistry has only a small effect on the climate sensitivity on a planetary scale. In the presence of the physical feedbacks, the climate sensitivity would be reduced by a factor of 0.99 and enhanced by a factor of 1.02, in an atmosphere with a 2000 and 2100, respectively, chemical composition. Locally, however, damping can be by as much as a factor 0.80 and amplification by as much as a factor 1.30. These values seem to be robust descriptors of the CACTUS model.

Mainly because of the way the chemical perturbations were prescribed, it has not been possible to assess in a robust way the effect of climate on atmospheric chemistry sensitivity, for example, on the relationship between emissions and atmospheric burdens. The values obtained by experiments are indicative only of a significant climate feedback in the atmospheric chemistry system. The CACTUS model does not include all possible processes that could contribute to creating feedbacks. First and foremost, it does not consider the indirect radiative effects of aerosols through their effect on clouds and precipitation. It also does not consider the climate effect of changes in levels of sea salt and mineral dust through direct and indirect radiative effects, and we have not considered the longer-term climate feedback associated with methane perturbations. More comprehensive climate-atmospheric chemistry models will lead to different gains and feedback factors. Second, the results regarding climate feedbacks in the atmospheric chemistry system are expected to be dependent on the particular scenario used in performing the perturbations experiments in our study. Systematic perturbations by small changes in the emissions of one species or one group of species at a time are in order.

We stress once again that the quantitative results of our study should be viewed as preliminary for two reasons. The CACTUS model does not include all possible processes that could contribute to creating feedbacks. First and foremost, it does not consider the indirect radiative effects of aerosols through their effect on clouds and precipitation. It also does not consider the climate effect of changes in levels of sea salt and mineral dust through direct and indirect radiative effects, and we have not considered the longer-term climate feedback associated with methane perturbations. More comprehensive climate-atmospheric chemistry models will lead to different gains and feedback factors. Second, the results regarding climate feedbacks in the atmospheric chemistry system are expected to be dependent on the particular scenario used in performing the perturbations experiments in our study. Systematic perturbations by small changes in the emissions of one species or one group of species at a time are in order.

Appendix A: Block Diagram Analysis of Climate Feedbacks

We follow the notation and much of the terminology of Roe [2009]. Consider, as a reference, the open loop system in Figure A1a. The system behaves in such a way
that a sustained perturbation of the input, $\Delta R_f$, leads to a change in the output $\Delta T_{0s}^{ss}$. The equilibrium sensitivity parameter $\lambda_0$ of this system is defined by

$$\Delta T_{0s}^{ss} \equiv \lambda_0 \Delta R_f.$$  \hfill (A1)

We now consider the presence of a number of feedbacks as depicted in Figure A1b. [57] The system relaxes to a new steady state at which

$$\Delta T_{0s}^{ss} = \lambda_0 (\Delta R_f + \sum_i c_i \Delta T_{0s}^{ci}).$$  \hfill (A2)

The coefficients $c_i$ are called the feedback parameters. The customary and inherent assumption is that feedbacks are linearly proportional to the system output, and that they are independent of one another. Solving (A2) for $\Delta T_{0s}^{ss}$ gives

$$\Delta T_{0s}^{ss} = \frac{\lambda_0}{1 - \lambda_0 \sum_i c_i} \Delta R_f.$$  \hfill (A3)

Comparing equations (A1) and (A3) shows how, because of the feedback coupling, the sensitivity parameter has changed

$$\lambda_0 \xrightarrow{\text{feedback coupling}} \lambda = \frac{\lambda_0}{1 - \lambda_0 \sum_i c_i} \text{ or } \lambda_0^{-1} \rightarrow \lambda^{-1} = \lambda_0^{-1} - \sum_i c_i.$$  \hfill (A4)

The second form of the equation highlights the additivity of the feedback parameters. In climate studies, $\lambda_0^{-1}$ is often also called a feedback parameter. [58] Another descriptor that follows from the feedback analysis is the gain $G$ of the system, defined as the ratio of the output in the closed loop system, $\Delta T_{0s}^{ss}$, to the output in the reference system, $\Delta T_{0s}^{ref}$. By dividing (A3) by (A1), we obtain

$$G = \frac{\Delta T_{0s}^{ss}}{\Delta T_{0s}^{ref}} = \frac{\lambda_0}{1 - \lambda_0 \sum_i c_i}.$$  \hfill (A5)

The gain $G$ results from the effect of all feedbacks acting in concert. The gain $G_i$ for an individual feedback process is

$$G_i = \frac{1}{1 - \lambda_0 c_i}.$$  \hfill (A6)

Appendix B: Treatment of Atmospheric Chemistry in the CACTUS Unified Model

[59] The gas-phase atmospheric chemical mechanism includes 225 chemical species and 346 reactions for simulating gas-phase species and aerosols. The partitioning of ammonia and nitrate between gas and aerosol phases is determined by the online thermodynamic equilibrium model ISORROPIA [Nenes et al., 1998], and the formation of secondary organic aerosol from monoterprenes is based on equilibrium partitioning and experimentally determined yield factors [Chung and Seinfeld, 2002]. Note that the simulation of secondary organic aerosol does not include isoprene or more recent laboratory yields. Heterogeneous reactions considered in the model include those of $\text{N}_2\text{O}_5$, $\text{NO}_2$, and $\text{HO}_2$ on wet aerosols, the uptake coefficients are taken to depend on atmospheric temperature and relative humidity, as described by Liao and Seinfeld [2005]. Upper boundary layer conditions for $\text{O}_3$ and $\text{NO}_x$ are applied at the tropopause (about 150 hPa) to represent transport across the tropopause, as described by Wang et al. [1998].

[60] The direct radiative effect of $\text{O}_3$ as well as that of internally mixed aerosols including sulfate, nitrate, ammonium, black carbon, and organic carbon is fed back into the GISS GCM-II’. Aerosol optical properties (extinction cross section, single-scattering albedo, and asymmetry factor) are calculated by Mie theory with a look-up table based on wavelength-dependent refractive indices and aerosol size distributions. Assumptions and parameters used for the calculations of aerosol optical properties are given by Liao et al. [2004]. Water uptake by sulfate/nitrate/ammonium aerosols is determined by the aerosol thermodynamic equilibrium module, ISORROPIA [Nenes et al., 1998]. Water uptake by organic carbon aerosol follows the treatment of Chung and Seinfeld [2002]. The refractive index of internally mixed aerosols is calculated by volume-weighting of the refractive index of each aerosol species and water. It should be noted that the actual (unknown) mixing state of aerosols in the atmosphere depends on the aging and coagulation of aerosol particles, and to some extent internal and external mixtures coexist. In our work the refractive index of the internal mixture is calculated by volume weighting as in most previous studies, but such a linear combination may represent an oversimplification. Present-day global optical depths and single-scattering albedos predicted by this model have been evaluated by comparison with measurements from Liao et al. [2004].

[61] Biomass burning emissions are, in part, anthropogenic and, in part, natural. We assume in this study that biomass burning emissions remain unchanged in 2000 and 2100; the effect of climate change on the occurrence and intensity of wildfires is therefore not considered. The seasonal and geographical distributions of BC and POA emissions in year 2100 are obtained by scaling year 2000 monthly values, grid by grid, using projected changes in IPCC SRES A2 CO emissions. Climate-sensitive natural emissions, including lightning $\text{NO}_x$, $\text{NO}_x$ from soil, biogenic hydrocarbons, sea salt, desert dust and dimethyl sulfide (DMS), are calculated as described by Liao et al. [2006]. Ozone and all aerosols, except sea salt and desert dust, affect the radiative balance in the model. Global and annual GHG concentrations and species emissions, for the years 2000 and 2100 are given in Table 2.

[62] Acknowledgments. This work was supported by the U.S. Environmental Protection Agency STAR Program (grant RD-83337001). Hong Liao acknowledges support from the National Science Foundation of China (grant 40775083). Frank Raes acknowledges support by the JRC Director General Sabbatical Programme.

References


Chen, W. T., H. Liao, and J. H. Seinfeld (2007), Future climate impacts of direct radiative forcing of anthropogenic aerosols, tropospheric ozone,


