

MODELING CHEMICAL CONSTITUENTS OF THE ATMOSPHERE

Interactions between the atmosphere, ocean, ice, land surface, and the marine and terrestrial biosphere control the global climate system. These components are coupled by the exchange of momentum, radiative energy, and trace constituents' mass. Various processes drive this exchange and require different computational methods to model it.

Changes in the solar cycle, shifts in terrestrial vegetation, variations in sea ice coverage, and volcanic eruptions have always led to climate variability, as can be seen from proxy data such as ice cores, tree rings, or lake sediments. In the past century, a growing suite of anthropogenic perturbations has been added to the mix and has begun to significantly change the atmosphere's state. Trace constituents, such as greenhouse gases and aerosols, can alter the radiative balance and thus exert a warming or cooling influence throughout the atmospheric column. Agricultural and industrial activities as well as land-use practices have also significantly changed the atmospheric composition.

Consequently, between 1860 and 1990, the atmospheric volume mixing ratio of carbon dioxide (CO₂) increased from 280 to 350 ppmv, methane (CH₄) rose from 0.7 to 1.7 ppmv, and nitrous oxide (N₂O) rose from 0.28 to 0.31 ppmv. Ozone has nearly doubled in the troposphere yet decreased in the lower stratosphere due to anthropogenic halocarbon emissions. The sources and

sinks of the canonical greenhouse gases CO₂, CH₄, and N₂O are closely linked to nutrient cycles and physical uptake in the ocean and the terrestrial biosphere. Other greenhouse gases (for example, ozone) and aerosols are formed in the atmosphere from precursor species. Chemical reactions and microphysical and transport processes determine their concentrations.

In light of these complex interactions, we need comprehensive Earth system models to assess the climate system's current state. Such models also help us reliably predict future changes in temperature and precipitation and their impact on human welfare and the economy. Researchers recently took an important step toward this goal of creating such models by enhancing global general circulation models (GCMs) to simulate the atmospheric transport and chemical reactions of gaseous and particulate constituents. They've implemented other simulation codes that do not compute the meteorological fields (for example, wind speed, temperature, or humidity) but read them from input files. Either a GCM (such as the ECHAM-5 model) can provide this data, or researchers can obtain it as archived data from a weather center (such as the European Center for Medium-Range Weather Forecast).

The atmosphere's gaseous and particulate constituents play an important role in climate change through their ability to affect radiative

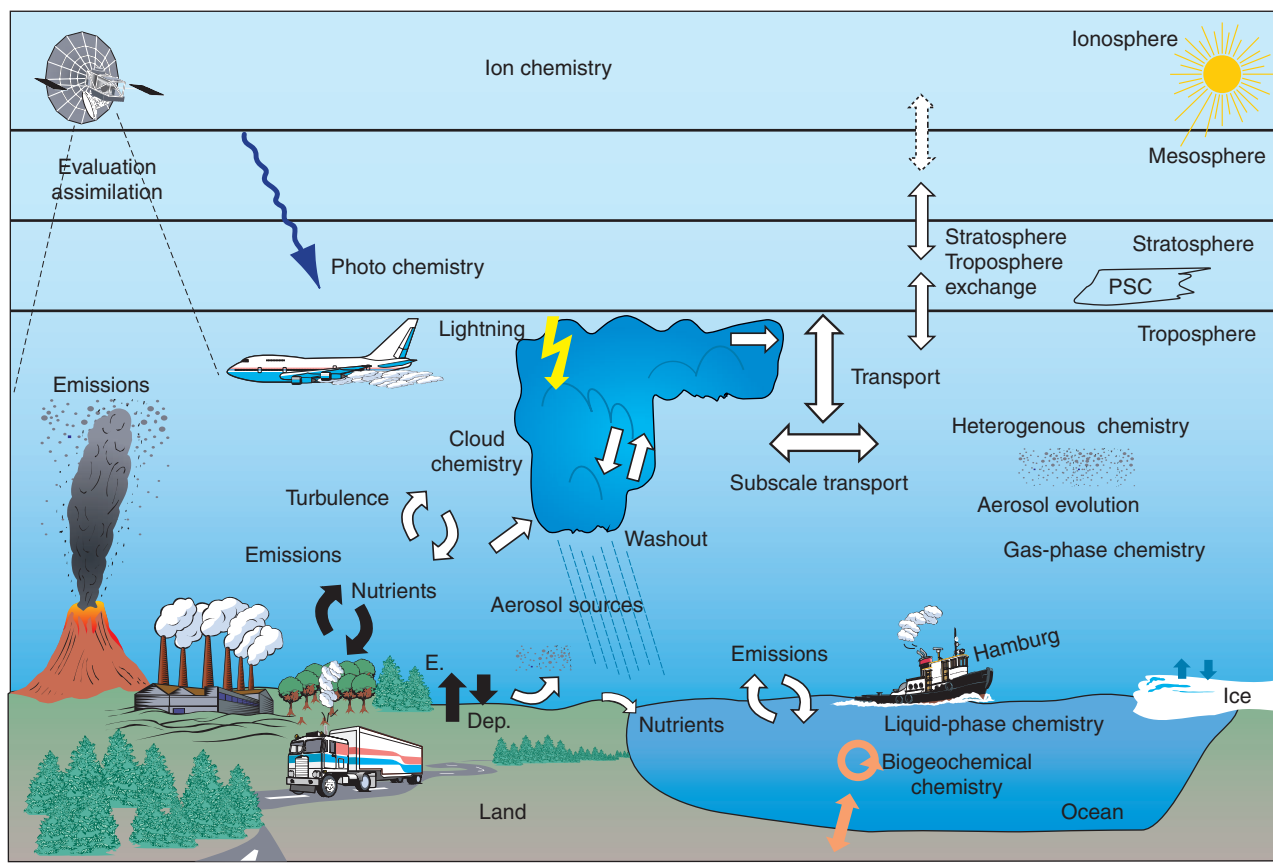


Figure 1. The processes affecting the concentration of trace gases and aerosols in the atmosphere, and how they interact with the climate system.

fluxes and the atmosphere's "oxidation power"—that is, its ability to clean itself from pollutants. Chemistry and aerosol models are not only complex due to the manifold processes and interactions they represent (see Figure 1), they are also demanding in terms of the computer power they require for scientific experiments. For a given species i at location x , the change in mixing-ratio μ with time is expressed by

$$\begin{aligned} \frac{\partial \mu_{i,x}}{\partial t} = & \left(\frac{\partial \mu}{\partial t} \right)_{emissions} \\ & + \left(\frac{\partial \mu}{\partial t} \right)_{loss} + \left(\frac{\partial \mu}{\partial t} \right)_{transport} \\ & + \left(\frac{\partial \mu}{\partial t} \right)_{photochemistry} + \left(\frac{\partial \mu}{\partial t} \right)_{heterogeneouschem} \end{aligned}$$

Here, we describe key processes controlling the atmosphere's compositions and state-of-the-art approaches to solving the chemical equations and aerosol microphysical processes. We start by describing chemistry models that parameterize the emission of gaseous and particulate constituents.

Emissions

Terrestrial and marine biological processes, biomass burning, volcanic eruptions, and weathering constitute natural sources of numerous trace constituents. Anthropogenic activities, such as energy production, industrial production, traffic, and waste disposal, have also led to emissions that are noticeable on a global scale. Furthermore, emissions from wildfires and anthropogenic phytomass burning play an important role in the tropospheric chemistry. For some species, the emissions from fossil fuel combustion rival or exceed natural emission sources; other compounds are present in the atmosphere only because of human activities. Chemical constituents released in the atmosphere are chemically transformed, deposited at the Earth's surface, and removed by precipitation. Removal by rain and dry deposition at the ground level control the atmospheric residence time of gases and aerosol particles and contribute to the acidification and eutrophication of ecosystems.

To derive global estimates of trace gas and aerosol emissions, researchers collect large amounts of data on energy consumption, fuel-use efficiency, activity patterns, and emission factors. They then

process the data in a database system to create annual or monthly gridded data sets, which they can then use as input for atmospheric models. For example, the Global Emissions Inventory Activity provides emission data about key species from anthropogenic and biogenic sources valid for 1985 and 1990. Other organizations have derived data sets for past¹ and future scenarios.²

Although gridded data sets are easy to use, they do not consider the variability of emissions that meteorological parameters or human behavior can cause. For example, temperature, insolation, humidity, and precipitation influence the production of volatile species and their subsequent emission through plant physiological and soil microbial processes. Recently, several research groups began using explicit parameterizations for such emissions, thereby building on the expertise formed from carbon cycle modeling. Other areas in which the variability of emissions is important include the burning of vegetation due to forest clearing, agricultural waste disposal, and the release of dust and sea salt aerosol (which strongly depends on wind speed). Researchers are currently developing parameterizations for these processes.

Validating emission inventories and source parameterizations is another important issue. Only recently have satellite observations of tropospheric trace gases and aerosols become available. Researchers are using these observations in inverse modeling studies to estimate errors in emission estimates. Other efforts involve carefully evaluating model simulations with field observations and analyzing measured and predicted trace gas correlations.

Loss processes

Loss processes remove material from the atmosphere. For many reactive gases, the primary atmospheric loss process is reaction with the hydroxyl radical (OH) or photolytic dissociation. Other species are removed from the atmosphere primarily through deposition on aerosol and land surfaces, uptake by oceans and lakes, and uptake in cloud droplets and subsequent precipitation.

The dry deposition of gases and aerosol particles from the atmosphere to a receptor surface is governed by their concentration near ground and by the turbulent transport processes in the boundary layer. It is also governed by the chemical and physical nature of the depositing species and the surface's ability to capture or absorb gases and particles. For particles, the transport from the free atmosphere to the receptor surface is similar to gas

transport, but transport processes through the laminar layer differ considerably. The receptor surface's microstructure and the particles' size distribution and density largely control these processes. Brownian diffusion is predominant for particles with radii less than $0.1 \mu\text{m}$, impaction for particles in the size range 0.1 to $1.0 \mu\text{m}$, and gravitational settling (sedimentation) for supermicron particles.

Wet deposition (also called wet scavenging) is the process that attaches and dissolves gases or particles in cloud droplets and precipitation, thereby depositing them at the Earth's surface. Wet scavenging is a complex phenomenon acting on a wide spectrum of spatial scales from the microphysical to the storm scale. We distinguish between two processes for removing gases or particles from the atmosphere:

- *In-cloud scavenging* results from incorporating species into cloud droplets. Precipitation finally removes these droplets and, with them, the aerosol particles from the atmosphere (in-cloud scavenging + removal = rainout).
- *Below-cloud scavenging* occurs when raindrops or snowflakes collect particles or absorb gases below clouds (washout). Cloud droplets or rain drops can absorb gaseous species to the extent that these are soluble in water. If sufficient time is available to reach equilibrium, Henry's law gives the concentration in the water droplets.

The dissolution of reactive gases thus proceeds in two steps: the substance is hydrated, then the hydrate forms ions. Molecular diffusion inside and outside of the liquid water drop determines the flux of material through the transport resistance at the gas-liquid interface and the rate of hydrolysis and ion formation. Gas scavenging by ice clouds occurs predominantly if the temperature is close to the freezing point and if a pseudoliquid layer covers the ice particle. Because global GCMs do not yet predict the size-spectrum of cloud droplets—only cloud water amount—all these processes are highly parameterized.

Aerosol particles serve as cloud condensation nuclei and are scavenged from the atmosphere when the cloud precipitates. Because drops form predominantly on larger particles, which contain most of the aerosol mass, about 80 to 99 percent of the aerosol mass is scavenged. However, 10 to 90 percent of the particle number is scavenged, because small particles can be left unactivated and can remain present as an interstitial aerosol. Essentially all particles with radii greater than $0.2 \mu\text{m}$ undergo nucleation scavenging.

Transport

At a given location and time, the atmospheric chemical composition is mostly determined by transporting the substance or its precursor into or out of the area. The atmosphere possesses a large spectrum of motions from planetary waves and synoptic scale disturbances (approximately 1,000 km or 3 to 7 days), to mesoscale processes (such as orographic flow), to turbulent exchange (down to the mm scale). The importance of a particular transport process for a specific species depends strongly on its atmospheric residence time. On larger spatial scales, the winds transport species with long lifetimes far from the source region. Pollutants predominantly released in the northern hemisphere continents move across entire continents and contribute by interhemispheric transport to the southern hemisphere's pollution. Subgrid-scale processes, such as turbulent exchange and vertical transport in clouds, dilute quite efficiently polluted boundary-layer air by mixing with clean free tropospheric air masses. The degree of vertical mixing controls the dry deposition at the ground and the transit time until a parcel enters a cloud. GCMs calculate the large-scale transport of atmospheric constituents (advection) explicitly and parameterize subgrid-scale vertical transport by turbulent exchange and in convective clouds. Horizontal diffusion of trace constituents is mostly neglected.

The advection equation for the trace constituents is $\partial q/\partial t + \mathbf{v} \cdot \nabla q = 0$, where q represents a "mixing ratio-like" quantity, and \mathbf{v} is the vector wind. The numerical method used to solve this equation should fulfill several constraints:

- Accuracy
- Monotonicity (that is, not introducing new extrema)
- Positive definiteness (not generating negative values)
- Mass conservation

Furthermore, the method should be local (the solutions at a given point should not be influenced by processes far from that point) and transportive (the information should propagate primarily downwind). An introduction to solving the advection equation appears elsewhere.^{3,4}

In light of the computational demands of 3D chemistry transport simulations, the advection scheme must also be computationally efficient. This becomes important when using an approximately equiangular mesh in spherical geometry. In this geometry, the spacing between longitudes

becomes increasingly small as you approach the poles; thus, very small time steps are required to maintain stability due to the Courant-Fredrichs-Lewy condition (pole problem). One method with a much less stringent time-stepping restriction is the semi-Lagrangian technique (SLT).⁵ However, this method is not mass conserving.

In recent years, researchers have developed other methods, based on the advection equation's flux form. These are inherently mass conserving and allow for an SLT-like time step.^{6,7} However, some of them are not strictly monotonic, and others do not provide the required accuracy in certain situations.

Another issue in present-day models is the inconsistency between the prescribed surface pressure and the surface pressure as the model implicitly predicts it. This inconsistency manifests itself by violating the conservation of the tracer mass.⁸ It arises if the method used to predict the meteorological tendencies is inconsistent with the advection scheme used to transport the chemical species. This is the case when an offline model reads in data from a spectral GCM—for example, a model that computes the meteorological data in spectral space, which is the most common GCM. Researchers are developing new models with the goal of performing all the computations in grid point space, using a special grid covering the sphere to determine the dynamical variables and the species in a consistent way. Thus, these models will not have inconsistency problems—if successfully implemented. For now, researchers have suggested several methods for removing this inconsistency. The general idea is to adjust the wind fields so that the surface pressure change that the advection scheme implies is consistent with the surface pressure tendency from the input fields.

Gas-phase chemistry

The atmosphere is an oxidative medium, but the temperature is too low for reactions with molecular oxygen to play an important role. Chemical reactions in the atmosphere are thus controlled by the availability of radical species (that is, molecules with unpaired electrons). The radicals themselves are generated by photodissociation of trace gases in the ultraviolet, and they can be propagated in subsequent reaction chains (for example, degradation of hydrocarbons). OH is the most important oxidizing agent in the troposphere. It is generated by photodissociation of ozone and subsequent reaction of excited atomic oxygen with water vapor. The oxidation of trace gases takes place either as

Mozart-2: A Global 3D Chemical Transport Model

The Model of Ozone and Related Chemical Tracers, version 2, is an offline global chemical transport model designed to simulate the distribution of tropospheric ozone and its precursors (such as nitrogen oxides (NO_x) and carbon monoxide (CO)).^{1,2} Researchers at the Atmospheric Chemistry Division of NCAR, the NOAA Geophysical Fluid Dynamics Laboratory, and the Max Planck Institute for Meteorology have jointly developed Mozart-2. They are releasing it as a community model, which will be available through NCAR and MPI-Met Web sites (<http://acd.ucar.edu/models/MOZART> and www.mpimet.mpg.de/en/extra/models/mozart). It predicts the concentrations of 63 chemical species from the surface up to the lower stratosphere and includes 167 chemical and photochemical reactions.

At each time step, the chemical species are updated by a sequence of operators, which we can symbolically write as

$$q_i(t_{n+1}) = C \cdot K \cdot D \cdot A q_i(t_n),$$

where A represents advection, D represents diffusion (including dry deposition and surface emissions), K represents convection, and C represents the chemistry including rainout and wash-out processes. q_i is the mixing ratio of species i at time step t_n .

The system of chemical reaction equations is solved via a fully implicit Euler backward method with a Newton-Raphson iteration, which converges quickly if the initial guess is sufficiently close to the solution:

$$(\mathbf{q}_{n+1} - \mathbf{q}_n) / \Delta t - P(\mathbf{q}_{n+1}, t_{n+1}) + L(\mathbf{q}_{n+1}, t_{n+1}) = 0,$$

which we can write as $G(\mathbf{q}_{n+1}) = 0$. A system of nonlinear equations in \mathbf{q}_{n+1} is solved iteratively by

$$\mathbf{q}_{n+1}^{m+1} = \mathbf{q}_{n+1}^m + \Delta \mathbf{q}_{n+1}^m,$$

where m denotes the iteration and $\Delta \mathbf{q}_{n+1}^m$ is determined by the equation

$$J(\mathbf{q}_{n+1}^m) \Delta \mathbf{q}_{n+1}^m = -G(\mathbf{q}_{n+1}^m). \quad (A)$$

\mathbf{q} is the vector of all species and J is the Jacobian ($J_{ij} = \partial G_i / \partial \mathbf{q}_{n+1,j}$). P and L denote the chemical production and loss rates, respectively. Because J is a sparse matrix (a certain species reacts only with few other species), we can use computationally efficient techniques based on the decomposition of J into a lower triangular matrix L and an upper triangular matrix U to obtain the solution vector $\Delta \mathbf{q}_{n+1}^m$ of equation A. Contrary to explicit methods, the implicit Euler backward method is stable for relatively long time steps in the case of a stiff system of differential equations.

Mozart-2 is designed to run efficiently on almost any computer architecture, from Linux laptops to workstations to vector supercomputers. The Mozart package includes a preprocessor written in Fortran 90 in addition to the model source code. This preprocessor takes parameters as the horizontal and vertical resolution, chemical species and reactions, output variables, and machine-specific information as input and it then generates various Fortran-90 source files for a run. The preprocessor generates the chemical solver routines, which carry the highest computational load of all subroutines, in a highly optimized version for the desired architecture (cache- or vector-based machines).

The Mozart code is parallelized using a hybrid approach. The first level of parallelization uses the message passing interface and a 1-dimensional domain decomposition, where the number of latitudes is decomposed onto the number of MPI tasks. In addition, an OpenMP-based parallelization is implemented. Depending on whether the code runs on a cache-based or vector architecture, the longitudes are split among the OpenMP threads (resulting in rather short data segments for cache-based machines) or the latitudes are split among the threads (so that complete latitudes are processed per iteration for vector machines). The code can run in either pure MPI mode, pure OpenMP mode, or with OpenMP on top of MPI (hybrid mode).

The standard resolution used at MPI-Met is currently 1.9° × 1.9° (192 × 96 grid boxes) with 31 vertical levels extending up to a pressure of 10 hPa, which approximately corresponds

abstraction of a hydrogen atom, or by addition of the hydroxy radical after breaking of a double bond. Some radicals are also generated at night—for example, by ozonolysis of volatile organic compounds such as isoprene or terpenes. Peroxy radicals play a key role in tropospheric ozone formation. In the presence of suitable amounts of nitrogen oxides, they react with nitric oxide (NO) to form nitrogen dioxide (NO₂), which is then photodissociated. This process releases an unbound oxygen atom, which quickly reacts with molecular oxygen to form ozone. Under low NO_x

(= sum of nitric oxide (NO) and nitrogen dioxide (NO₂)) conditions, peroxy radicals react primarily with themselves, leading to a decrease in the ozone concentration.

In the stratosphere, where water vapor concentrations are much lower, but UV intensity is greater, the key players are oxygen and halogen atoms, halogen radicals, and NO. Ozone formation in the stratosphere occurs directly through the photodissociation of molecular oxygen and the subsequent reaction of two oxygen atoms with two oxygen molecules.^{9,10}

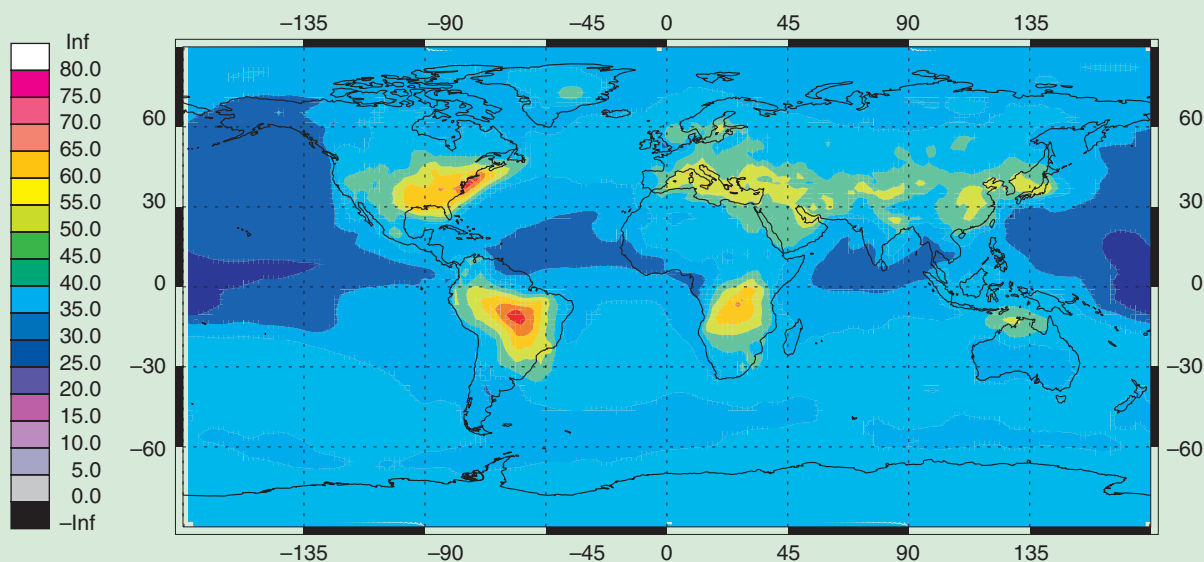


Figure A. The calculated monthly mean ozone surface-mixing ratio for August 1997 (units: parts per billion by volume). ECMWF meteorological analyses have driven Mozart (T63, 31 vertical levels). The figure shows distinct ozone maxima in regions affected by vegetation fires and in industrialized regions. High ozone values over the north Atlantic demonstrate long-range transport from the polluted regions in the eastern US and Canada.

to an altitude of 30 km and with a time step of 15 minutes. We prepare the required input files containing the meteorological fields at time intervals of six hours by extracting the corresponding data from analyses in the data archive of the European Centre for Medium-Range Weather Forecast.

Our main compute server at MPI-Met is a NEC SX-6 located at the German Climate Computing Center (DKRZ). The single CPU performance of Mozart-2 in this resolution is currently about 2 Gflops (25 percent of the peak performance). For long-term integrations, we use the model in hybrid mode on two nodes, each made up of eight CPUs (two MPI tasks, eight OpenMP threads per task). The time-to-solution for one year is in this case about 17 hours.

The memory requirements are 2.6 Gbytes to 10.7 Gbytes, depending on the number of CPUs and the parallelization approach. The disk storage requirement is about 1.3 Terabytes per decade. Figure A shows a typical species concentration field obtained from the Mozart-2 model.

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Atmospheric chemical reaction rates usually depend on temperature and sometimes pressure as well. Researchers have performed considerable work in laboratories to determine the rate constants for key atmospheric reactions, and they've compiled the results in comprehensive evaluations.^{11,12} Yet, because of experimental difficulties, several reaction rates (in particular, of radical species or soluble compounds) remain unknown or uncertain. Photodissociation reactions are governed by the available UV intensity (the actinic flux), the molecule's absorption cross sec-

tion, and the quantum yield determining the dissociation reaction's efficiency. All these parameters are wavelength dependent (which requires computing at least eight additional bands in the radiative transfer calculations in the GCM). The absorption cross section and quantum yield might also depend on temperature and pressure.

An atmospheric chemistry model computes the concentration changes of trace gases at each time step by solving a set of ordinary differential equations. The equations describe the molecule's production and loss rates and the reaction

stoichiometry. The following lists the major terms for tropospheric ozone as an example (the square brackets denote concentration):

$$\frac{\partial[\text{O}_3]}{\partial t} = k_1 \cdot [\text{NO}] \cdot [\text{HO}_2] + k_2 \cdot [\text{NO}] \cdot [\text{CH}_3\text{O}_2] - j_{\text{O}^1\text{D}} \cdot [\text{O}_3] - k_3 \cdot [\text{O}_3] \cdot [\text{HO}_2].$$

Given the typical set of about 50 to 60 species, which react in about 150 to 200 reactions, these equations form a considerably sized sparse matrix that must be solved for each model grid box. Due to the different timescales for atmospheric reactions (spanning several orders of magnitude), the system is stiff, and special solvers had to be developed to treat the matrix numerically.

Techniques that global modelers often use are quasi-steady-state approximation,¹³ the Euler backward iterative method (see the “Mozart-2: A Global 3D Chemical Transport Model” sidebar), and a sparse-matrix, vectorized version of Gear’s method.^{14,15}

Heterogenous chemistry and aerosol microphysics

Atmospheric chemical processes are not limited to the gas phase—they also occur on the surface of solid particles and in liquid particles, such as aerosols and cloud droplets. Studies of the stratospheric ozone hole have shown the importance of heterogenous reactions on aerosol surfaces. Reactions on the sea salt of organic halogens might also play an important role in the marine boundary layer. Condensable matter condenses on mineral dust and other particles. Clouds receive trace gases from the boundary layer, vertical winds redistribute the gases, and the cloud then transforms the gases through gas and aqueous-phase chemistry. Clouds also control the formation of aerosols and their removal by scavenging. For example, oxidating SO₂ to sulfate in cloud droplets is much more efficient than in the gas phase. Generally, reaction pathways and rates differ considerably from those in cloud-free air. Moreover, clouds also affect photochemistry by enhancing the actinic fluxes above the cloud and by reducing it below the cloud compared to clear-sky conditions.

Liquid or solid aerosol particles are injected into the atmosphere from natural and anthropogenic sources (primary particles) or are formed in the atmosphere from condensable gases (sec-

ondary particles). The size of particles, which undergo long-range transport, ranges from a cluster with few molecules to 10 μm. Processes controlling the temporal evolution of atmospheric aerosols are homogenous and heterogenous nucleation, condensation, coagulation, and removal by dry deposition and precipitation. Homogenous nucleation is the condensation to small droplets in supersaturated vapor; heterogenous nucleation of a single species is the nucleation on a foreign substance or surface. In polluted air, condensable gases will more likely condense on preexisting particles rather than form new particles.

Coagulation is the main sink for small particles (less than 0.1 μm), which undergo diffusion and collide and stick to one another, shifting a particle population’s size distribution to larger radii. Aerosol particles affect the climate in two ways:


- Particles scatter and absorb solar radiation and cool the Earth’s surface
- Particles act as cloud condensation and ice nuclei and change the physical and optical properties of clouds

To describe the aerosol population, we must predict the particle mass per volume, solving the same processes as in the case of gaseous constituents as well as the intensive aerosol properties (that is, particle number concentration, size distribution, and size-resolved chemical composition). Numerical modeling of these properties must consider the continuous modification by processes such as coagulation, condensational growth, evaporation, and nucleation. Particle population in climate models is represented either by

- Subdividing the spectrum of particle sizes into several bins, where each bin is characterized by the upper- and lower-limit particle diameter
- Several modes, each described by a lognormal distribution

Recently, higher-order moments of the particle distribution have also been used to represent particle population.¹⁶

To solve the equations that describe the aerosol dynamics, we apply an operator-splitting technique to treat each process separately. We calculate the condensation and evaporation as a transport process of vapor on or from aerosol surfaces by solving an advection equation.^{9,10}

Until now, a model's affordable horizontal and vertical resolution, which is a function of the available computing power and time interval that the simulation must cover, has constrained the explicit representation of physical processes. Parameterizations of processes—such as turbulent and cloud transport of chemical species, cloud formation, precipitation and wet removal, aerosol microphysics, and radiative transfer—rely on detailed observational and high-resolution modeling studies. Due to the sparseness of observational data, the atmosphere's chemical composition is not as well known as the distribution of dynamical and thermodynamical parameters. This situation is beginning to improve as global distributions of tropospheric trace gases and aerosols derived from satellite observations become available (for example GOME, Schiavacy, and Mopitt). 

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