Chapter 11

CLOUD DYNAMICS AND CHEMISTRY

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ABSTRACT

Chapter 11 describes the cloud module that is currently incorporated into CMAQ. This module simulates the physical and chemical processes of clouds that are important in air quality simulations. Clouds affect pollutant concentrations by vertical-convective mixing, scavenging, aqueous chemistry, and removal by wet deposition. The CMAQ cloud module includes parameterizations for sub-grid convective precipitating and non-precipitating clouds and grid-scale resolved clouds. Cloud effects on both gas-phase species and aerosols are simulated by the cloud module.

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11.0   CLOUD DYNAMICS AND CHEMISTRY

11.1   Background

Clouds play an important role in boundary layer meteorology and air quality. Convective clouds transport pollutants vertically, allowing an exchange of air between the boundary layer and the free troposphere. Cloud droplets formed by heterogeneous nucleation on aerosols grow into rain droplets through condensation, collision, and coalescence. Clouds and precipitation scavenge pollutants from the air. Once inside the cloud or rain water, some compounds dissociate into ions and/or react with one another through aqueous chemistry (i.e., cloud chemistry is an important process in the oxidation of sulfur dioxide to sulfate). Another important role for clouds is the removal of pollutants trapped in rain water and its deposition onto the ground. Clouds can also affect gas-phase chemistry by attenuating solar radiation below the cloud base which has a significant impact on the photolysis reactions.

The Models-3/CMAQ cloud model incorporates many of these cloud processes. The model includes parameterizations for several types of clouds, including sub-grid convective clouds (precipitating and non-precipitating) and grid-scale resolved clouds. It includes an aqueous chemistry model for sulfur, and includes a simple mechanism for scavenging.

11.2   Model Description

The cloud model can be divided into two main components, including the sub-grid cloud model (*subcld*) and the resolved cloud model (*rescld*). For large horizontal grid resolutions, the grid size will be larger than the size of a typical convective cloud, requiring a parameterization for these sub-grid clouds. The sub-grid cloud scheme simulates convective precipitating and non-precipitating clouds. The second component of the cloud model considers clouds which occupy the entire grid cell and have been "resolved" by the meteorological model. The rate of change in pollutant concentrations ($\overline{m}_i$) due to cloud processes is given by:

$$\left.\frac{\partial \overline{m}_i}{\partial t}\right|_{cld} = \left.\frac{\partial \overline{m}_i}{\partial t}\right|_{subcld} + \left.\frac{\partial \overline{m}_i}{\partial t}\right|_{rescld} \tag{11-1}$$

The terms on the right-hand side of Equation 11-1 are solved separately at different times. The influence of sub-grid clouds are instituted once an hour while the resolved clouds impact concentrations every synchronization timestep. Each subcomponent of the cloud model is described in detail below.

11.2.1 Subgrid Convective Cloud Scheme

$$\left.\frac{\partial \overline{m}_i}{\partial t}\right|_{subcld} \sim f\left(mixing, scav, aqchem, wetdep\right) \tag{11-2}$$

The current sub-grid cloud scheme in CMAQ was derived from the diagnostic cloud model in RADM version 2.6 (Dennis et al., 1993; Walcek and Taylor, 1986; Chang, et al., 1987; Chang, et al., 1990). Seaman (1998) noted that most convective parameterizations are based on the assumption that "the area of an updraft is small compared to the area of the grid cell" and most parameterizations can be used at grid resolutions a small as 12 km (Wang and Seaman, 1997). In CMAQ, subgrid clouds are considered only for horizontal grid resolutions on the order of 12 km or more. Seaman (1998) also pointed to a study by Weisman et al. (1997) that showed that explicit cloud models can resolve convection for resolution finer than 5 km. Within CMAQ, for resolutions of 4 km or less, vertical convection is assumed to be resolved at the grid level; therefore, the resolved cloud model will be the only cloud scheme used at small grid scales.

The effects of sub-grid clouds on grid-averaged concentrations are parameterized by modeling the mixing, scavenging, aqueous chemistry, and wet deposition of a "representative cloud" within the grid cell. For all sub-grid clouds, a 1-hour lifetime (τ_{cld}) has been assumed. Sub-grid clouds can be either precipitating or non-precipitating, and the non-precipitating subgrid clouds are further categorized as pure fair weather (PFW) clouds and non-precipitating clouds coexisting (CNP) with precipitating clouds.

The subgrid cloud model determines if precipitating or nonprecipitating clouds exist over each grid cell. Precipitating clouds are simulated when the meteorological preprocessor (currently the Mesoscale Model version 5 or MM5, Grell et al., 1994) indicates precipitation from its convective cloud model. The CMAQ implementation differs from the RADM in that only the convective precipitation amounts from MM5 are used to drive the subgrid precipitating cloud. RADM used the total precipitation (convective and nonconvective precipitation) to drive the subgrid cloud model. In CMAQ, the nonconvective precipitation is used in the resolved cloud model. Nonprecipitating clouds are modeled if the moisture and temperature profiles support the development of a cloud (Dennis et al., 1993). Nonprecipitating clouds are modeled only when the relative humidity of the source level is above 70% and the calculated cloud base is below 1500 m for PFW clouds or 3000 m for CNP clouds. For both precipitating and nonprecipitating cloud types, the geometry of the cloud (base, top, and spatial extent) are determined next. The cloud base is calculated by lifting a parcel of air from the cloud source level (the level between the surface and 650 mb with the highest equivalent potential temperature) to the lifting condensation level (LCL). The cloud top calculation depends upon the cloud type and atmospheric stability. For precipitating clouds in unstable conditions, the cloud top is found by following the moist adiabatic lapse rate from the cloud base up to the level where it becomes cooler than the surrounding environment. For precipitating clouds under stable conditions, the cloud top is set as the first layer above the cloud base in which the relative humidity falls below 65%, but limited to less than the 600 mb height. For nonprecipitating clouds, further restrictions are placed on the cloud top. The cloud top calculation for nonprecipitating clouds uses the same relative humidity criterion as the precipitating clouds, but the cloud top is allowed to extend up to 500 mb for CNP clouds and only to 1500 m for PFW clouds. If the atmosphere is unstable, the nonprecipitating cloud top may be reduced in height if the parcel method calculated a lower cloud top. The fractional cloud coverage calculations depend on cloud type and have been

described thoroughly by Dennis et al. (1993). For precipitating clouds, the model uses a parameterization similar to approach of Kuo (1974). The fractional coverage parameterization for the nonprecipitating clouds is based on relative humidity.

The convective cloud simulated by the sub-grid cloud model is considered to be composed of air transported vertically-from below the cloud, entrained from above the cloud (for precipitating clouds), and entrained from the sides of the cloud. Concentrations of pollutants for each layer of the cloud are calculated by:

$$\overline{m}_i^{cld}(z) = f_{ent}\left[(1 - f_{side})\overline{m}_i^{down} + f_{side}\,\overline{m}_i(z)\right] + (1 - f_{ent})\overline{m}_i^{up} \quad (11\text{-}3)$$

where f_{side} is the fraction of entraining air originating from the side of the cloud. For nonprecipitating clouds, no entrainment of air from above the cloud is allowed and therefore $f_{side}{=}1$. The entrainment, f_{ent}, is calculated by iteratively solving conservation and thermodynamic equations (Dennis et al., 1993; Chang et al, 1990,1987; and Walcek and Taylor, 1986). The terms $\overline{m}_i^{up}$ and $\overline{m}_i^{down}$ represent the above and below cloud concentrations, respectively. Once the cloud volume has been determined, vertically-averaged cloud temperature, pressure, liquid water content, total water content, and pollutant concentrations, are computed with liquid water content (W_c) as the weighting function (gives the most weight to the layers with the highest liquid water content). Thus, the average pollutant concentrations within the cloud are calculated by:

$$\overline{m}_i^{cld} = \frac{\displaystyle\int_{z_{cbase}}^{z_{ctop}} \overline{m}_i^{cld}(z)W_c(z)dz}{\displaystyle\int_{z_{cbase}}^{z_{ctop}} W_c(z)dz} \quad (11\text{-}4)$$

With the averages over the cloud volume, the processes of scavenging, aqueous chemistry, and wet deposition are considered. The final step in cloud mixing is the reapportioning of mass back into individual layers. This is accomplished using cloud fractional coverage, initial in-cloud concentrations, final in-cloud concentration, and the initial vertical concentration profile. For precipitating clouds, the average pollutant concentration for the grid cell within the cloud layers is computed by:

$$\overline{m}_i(z,t_0 + \tau_{cld}) = \overline{m}_i^{cld}(z,t_0)\left[\frac{\overline{m}_i^{cld}(t_0 + \tau_{cld})}{\overline{m}_i^{cld}(t_0)}\right]cfrac + \overline{m}_i(z,t_0)\left[1 - cfrac\right] \quad (11\text{-}5)$$

where *cfrac* is the fractional cloud coverage. There are variations on this equation for below cloud, above cloud, and for nonprecipitating clouds.

11.2.1.1   Scavenging and Wet Deposition

Pollutant scavenging is calculated by two methods, depending upon whether the pollutant participates in the cloud water chemistry and on the liquid water content. (1) For those pollutants that are absorbed into the cloud water and participate in the cloud chemistry (and provided that the liquid water content is > 0.01 g/m^3), the amount of scavenging depends on Henry's law constants, dissociation constants, and cloud water pH. (2) For pollutants which do not participate in aqueous chemistry (or for all water-soluble pollutants when the liquid water content is below 0.01 g/m^3), the model uses the Henry's Law equilibrium equation to calculate ending concentration and deposition amounts. The rate of change for in-cloud concentrations (m_i^{cld}) for each pollutant (i) following the cloud timescale (τ_{cld}) is given by:

$$\left.\frac{\partial \overline{m}_i^{cld}}{\partial t}\right|_{scav} = \overline{m}_i^{cld}\left(\frac{e^{-\alpha_i \tau_{cld}} - 1}{\tau_{cld}}\right) \tag{11-6}$$

where α_i is the scavenging coefficient for the pollutant. For subgrid convective clouds, τ_{cld} is 1 hour and for grid resolved clouds it is equal to the CMAQ's synchronization timestep. For gases, the scavenging coefficient is given by:

$$\alpha_i = \frac{1}{\tau_{washout}\left(1 + \dfrac{TWF}{H_i}\right)} \tag{11-7}$$

where H_i is the Henry's Law coefficient for the pollutant, *TWF* is the total water fraction given by:

$$TWF = \frac{\rho_{H_2O}}{\overline{W}_T RT} \tag{11-8}$$

where ρ_{H2O} is the density of water, $\overline{W}_T$ is the mean total water content (kg/m^3), R is the Universal gas constant, and T is the in-cloud air temperature (K). The washout time, $\tau_{washout}$ represents the amount of time required to remove all of the water from the cloud volume at the specified precipitation rate (P_r),
and is given by:

$$\tau_{washout} = \frac{\overline{W}_T \Delta z_{cld}}{\rho_{H_2O} P_r} \tag{11-9}$$

Here, Δz_{cld} is the cloud thickness.

11-4

The accumulation mode and coarse mode aerosols are assumed to be completely absorbed by the cloud and rain water. Therefore, the scavenging coefficients for these two aerosol modes are simply a function of the washout time:

$$\alpha_i = \frac{1}{\tau_{washout}}$$ (11-10)

The Aitken mode aerosols are treated as interstitial aerosol and are slowly absorbed into the cloud/rain water. This process is discussed in detail in the aerosol chapter (Chapter 10).

The wet deposition algorithms in CMAQ were taken from the RADM (Chang et al., 1987). In the current implementation, deposition is accumulated over 1-hour increments before being written to the output file. The wet deposition amount of chemical species i ($wdep_i$) depends upon the precipitation rate (P_r) and the cloud water concentration (m_i^{cld}):

$$wdep_i = \int_0^{\tau_{cld}} \overline{m}_i^{cld} P_r \, dt$$ (11-11)

Deposition amounts are accumulated for each of the modeled species, but the user specified which species are written to the output file. This is handled in the Program Control Processor (see Chapter 15).

11.2.1.2   Aqueous Chemistry

The aqueous chemistry model evolved from the original RADM model (Chang et al., 1987; and Walcek and Taylor, 1986). The model considers the absorption of chemical compounds into the cloud water; the amount that gas-phase species absorb into the cloud water depends on thermodynamic equilibrium, while accumulation-mode aerosols are considered to have been the nucleation particles for cloud droplet formation and are 100% absorbed into the cloud water. Then the model calculates the dissociation of compounds into ions, oxidation of S(IV) to S(VI), and wet deposition. The species that participate in the aqueous chemistry are given in Table 11-1. This version of the aqueous chemistry model differs from Walcek's scheme in that it tracks contributions from gases and aerosols separately. It also considers the scavenging of interstitial aerosols, and it allows for variable-length cloud time scales.

Table 11-1. List of Species Considered in the Aqueous Chemistry Model

Gases	Aerosols
SO_2	$SO_4^=$ (Aitken & accumulation)
HNO_3	NH_4^+ (Aitken & accumulation)
N_2O_5	NO_3^- (Aitken, accumulation, & coarse)
CO_2	Organics (Aitken & accumulation)
NH_3	Primary (Aitken, accumulation, & coarse)
H_2O_2	$CaCO_3$
O_3	$MgCO_3$
formic acid	NaCl
methyl hydrogen peroxide	Fe^{3+}
peroxy acetic acid	Mn^{2+}
H_2SO_4	KCl
	Number (Aitken, accumulation, & coarse)

11.2.2 Resolved Cloud Scheme

At any grid resolution, clouds may be resolved by the MM5, which could include stratus, cumulus, or cirrus type clouds. The resolved clouds have been simulated by the MM5 to cover the entire grid cell. No additional cloud dynamics are considered in CMAQ for this cloud type since any convection and/or mixing would have been resolved and considered in the vertical wind fields provided by MM5. A resolved cloud horizontally covers the entire grid cell and vertically extends over the whole depth of the layer, thus $\overline{m_i}^{cld}$ and $\overline{m_i}$ are equivalent in resolved clouds. These clouds are activated in MM5 when the humidity is high enough for water vapor to condense, and then MM5 computes cloud and rain water amounts according to any of several microphysical submodels. Using the total of the condensed cloud water and rain water reported by MM5, the CMAQ resolved cloud model then considers scavenging, aqueous chemistry, and wet deposition. The average liquid water content $\overline{W}_c$ in a model layer (z) for the resolved cloud is given by:

$$\overline{W_C}(z) = \left[Q_C(z) + Q_R(z) \right] \rho(z) \qquad (11\text{-}12)$$

where $Q_C(z)$ is the cloud water mixing ratio (kg/kg), $Q_R(z)$ is the rain water mixing ratio (kg/kg), $\rho(z)$ is the air density (kg/m^3). Precipitation amounts for resolved cloud layers, $P_r(z)$, are derived

using the MM5 non-convective precipitation amounts (R_n), apportioned into individual model layers with the vertical profile of condensed liquid water as follows:

$$P_r(z) = R_n \left[\frac{\overline{W_c}(z)}{\int \overline{W_c}(z)dz} \right] \tag{11-13}$$

Once quantities for precipitation rate, liquid water content, etc. have been calculated, then the scavenging, aqueous chemistry, and wet deposition are solved using the same procedures as in the subgrid clouds.

$$\left. \frac{\partial \overline{m}_i}{\partial t} \right|_{rescld} = \left. \frac{\partial \overline{m}_i}{\partial t} \right|_{scav} + \left. \frac{\partial \overline{m}_i}{\partial t} \right|_{aqchem} \tag{11-14}$$

Several assumptions have been made in the current implementation of the resolved cloud model. (1) The lifetime of the resolved cloud computations varies based on the synchronization timestep of CMAQ. (2) Following the method of operator splitting, the effect of the resolved clouds on pollutant concentrations occurs at the end of the cloud lifetime, thus no exchange between layers is permitted during the cloud life-cycle. (3) The pollutants, cloud water, and rain water are uniformly distributed within the grid cell. Because of the separation of MM5 from CMAQ, we do not have the information to do precipitation fluxes. Even if a complete cloud precipitation model was developed within CMAQ, there is no guarantee that it would be consistent with what was done in MM5.

11.3   Conclusions

One of the concepts for Models-3 was that multiple modules may exist for each physical process of the air quality model. The implementation described here is the first module available for modeling cloud physics and chemistry. Other subgrid cloud models (i.e., the Kain-Fritsch (1990, 1993) and Betts-Miller (1986)) are under consideration and may be included as optional modules for CMAQ. In addition, a more detailed resolved cloud model is under development which will include a microphysical submodel for following the evolution of the cloud (i.e., cloud droplet formation, growth of rain droplets, and descent through model layers to the ground). It will also consider resolved cloud lifetimes which extend beyond the CMAQ synchronization timestep, thus maintaining the partition between gas and aqueous-phase pollutants during the gas-phase chemistry calculations. The current implementation of the cloud model in CMAQ will be evaluated using available datasets and will be used as a reference for evaluating future cloud modules for CMAQ.

11.4 References

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