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A GENERALIZED MULTIDIMENSIONAL MODEL FOR PRECIPITATION SCAVENGING AND ATMOSPHERIC CHEMISTRY

JEREMY M. HALES
Battelle, Pacific-Northwest Laboratory, PO Box 999, Richland, WA 99352, U.S.A.

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Abstract—This paper describes a new, general-purpose computer code for modeling air pollutants, both in the gas phase and in conjunction with cloud and precipitation systems. The code is based on Eulerian representations of conservation equations for chemical species, energy, and the physical media (e.g. air, cloud water, rain water, ice, . . .) in which the chemical species reside. Because energy and moisture conservation equations are included, the code is capable of simulating cloud and storm formation, and can deal directly with the attachment, wet-chemistry and deposition processes associated with precipitating systems.

The code has been structured to allow considerable flexibility in its use. One-, two- or three-dimensional simulations can be performed, and selection of modeled chemical species, physical media, physicochemical interaction mechanisms, spatial/temporal domain and grid spacing is at the option of the user. A simple example simulation, corresponding to the scavenging of sulfur and nitrogen oxides in a frontal storm system, is presented to illustrate the code's use.

Key word index: Model, chemistry, scavenging, Eulerian, multidimensional, numerical, storm, cloud, precipitation.

NOMENCLATURE

A	Coefficient in general conservation Equation (10), l^2/t
B	Coefficient in general conservation Equation (10), l/t
c	Concentration, moles/total volume
C_p	Specific heat at constant pressure, (fl)/(moles T)
C_v	Specific heat at constant volume, (fl)/(moles T)
C_z	Coefficient in general conservation Equation (10), l/t or T/t
D	Substantial-derivative operator
g	Acceleration of gravity, l/t^2
g_c	International gravitation constant, $(ml)/(ft)$
G	Coefficient in general conservation Equation (10), l/t or T/t
i, j, k	Unit vectors in i, j and k directions, respectively
K	Component eddy diffusivity, l^2/t
K	Eddy diffusivity tensor, l^2/t
L	Collective symbol for latent heating terms excluding those associated with cloud-water condensation, T/t
M_{air}	Molecular weight of air
p	Pressure, f/l^2
r	Mixing ratio, moles/mole air
r^*	Physical or chemical reaction rate, moles/$(l^3\,t)$
R	Gas-law constant, (lf)/(mole T)
R_{Am}	Source-sink term, moles/$(l^3\,t)$
t	Time, t
T	Temperature, T
U, V, W	Velocity components, l/t
v	Velocity vector, l/t
w_{Am}	Vertical fall velocity of pollutant with respect to air motion, l/t
x, y, z	Cartesian distance components, l
γ_d	Dry adiabatic lapse rate, T/l
Γ	Collective latent heating term, $(fl)/t$
Δ	Difference operator
ζ	Dependent variable in general Equation (15)
θ	Potential temperature, T
λ	Latent heat associated with phase transformation, (fl)/mole
ξ	Dummy independent variable
χ_1	Collective variable in Equation (6), dimensionless
∂	Partial derivative operator
∇	Gradient operator, $1/l$
$\nabla\cdot$	Divergence operator, $1/l$.

Subscripts

A	Species index
c	Denotes "cloud" or "condensation"
d	Denotes "dry" or "deposition"
f	Denotes "freezing"
H	Denotes "heat"
m	Medium index
s	Denotes "saturated"
v	Denotes "water vapor"
0	Denotes "initial".

INTRODUCTION

During the past several years a number of computer codes have been prepared for simulating the mechanistic behavior of cloud chemistry and precipitation scavenging. These have been significantly different from conventional air-pollution simulation codes in the sense that they treat simultaneous heat and mass transfer, and internal chemistry of the cloud-precipitation system explicitly; thus the phase transitions associated with cloud-physics processes can be blended with the pollutant-capture and reaction steps in a

mathematically specific and physically realistic fashion. This advancement offers the opportunity for more refined simulation of natural-removal processes; in addition, it provides a highly convenient forum for direct incorporation of specific physicochemical mechanisms.

Earlier examples of such codes have been presented by Hegg *et al.* (1984), Molencamp (1983), Hales (1981*, 1982), and Easter and Hales (1982, 1984*). These particular codes have been of low dimensionality (one- or two-dimensional). More recently, however, Tremblay and Leighton (1986) have presented an excellent description of a complete three-dimensional model for reactive-scavenging simulations of a convective storm. The starred references among these citations are user's manuals, which are generally available for application by the scientific community.

The goal of this paper is to present a new, general-purpose code, with the intention of providing a central reference point for later scientific articles describing code applications. This new code, which is intended ultimately for application by a large number of users, will be eventually documented in a user's manual similar to its predecessors (Hales, 1981; Easter and Hales, 1984). The code has considerable flexibility, in the sense that it can be applied in three-, two- or one-dimensional mode, and can be modified rapidly to simulate a diversity of chemical and/or cloud-microphysics schemes at the option of the user. It approximates solutions to the coupled conservation equations for energy and pollutant mass in a manner similar to its predecessors, and thus allows the natural simulation of simultaneous cloud-physics, pollution-attachment, and chemical-conversion processes. Like its older relatives this new code is kinematic, in the sense that it does not contain a momentum balance. As such, it depends on an outside source to supply the wind fields necessary for its operation.

The following section of this paper summarizes the governing equations and boundary conditions applied within the code. This is followed in turn by a discussion of the numerical methods used for approximating solutions to these equations, and the general architecture of the computational framework. Finally, a summary of the code's operating features is given, indicating the procedure for implementing the system for solution of specific problems, and some example simulations are described. Formal derivations of the governing equations are presented in the Appendix.

GOVERNING EQUATIONS AND BOUNDARY CONDITIONS

Material balances

The code is able to accommodate as many chemical species as desired by the user, as computer resources permit. Chemical species are partitioned between a group of physical 'media', in which the species may reside. The number and classification of media are determined by the user; for example, a particular simulation may classify the gas phase as one medium and condensed water as a second. A more elaborate simulation may partition the condensed water into separate 'media' of cloud-water, ice and rain. Still more complex simulations may subdivide water groups into additional classifications, based on hydrometeor size and/or morphology. Regardless of the choice of media, all species-medium pairs are treated as dependent variables in individual material balances of the simulation. Derivation of these balances is based on the form (cf. Bird *et al.*, 1960)

$$\frac{\partial c_{\mathrm{Am}}}{\partial t} = -\nabla\cdot(c_{\mathrm{Am}}\mathbf{v}_{\mathrm{Am}}) + R_{\mathrm{Am}} \tag{1}$$

where c_{Am} is the concentration of chemical constituent A in medium m, and $\mathbf{v}_{\mathrm{Am}}$ is its velocity vector in Cartesian coordinates. R_{Am} is a source-sink term which accounts for the contributions of physicochemical processes to the overall balance.

Combining Equation (1) with the continuity equation for air, time-smoothing and applying gradient-transport theory in the conventional manner, and expressing the dependent variables as mixing ratios rather than concentrations, yields the form

$$\frac{\mathrm{D}r_{\mathrm{Am}}}{\mathrm{D}t} = -\frac{1}{c}\frac{\partial}{\partial z}(cr_{\mathrm{Am}}w_{\mathrm{Am}}) + \frac{1}{c}\nabla\cdot(c\mathbf{K}_{\mathrm{m}}\cdot\nabla r_{\mathrm{Am}}) + \frac{R_{\mathrm{Am}}}{c} \tag{2}$$

where

$\frac{\mathrm{D}}{\mathrm{D}t}$ is the conventional substantial-derivative operator, $\frac{\partial}{\partial t} + \mathbf{v}\cdot\nabla$

r_{Am} is the time-smoothed pollutant mixing ratio (molar basis),

$\mathbf{v}$ is the time-smoothed wind-velocity vector,

c is the molar concentration of air,

w_{Am} is the vertical fall velocity of m-bound pollutant A (in stagnant air), and

$\mathbf{K}_{\mathrm{m}}$ is the associated diffusivity tensor.

Energy balance

The energy balance employed by the code is based on the form (cf. Bird *et al.*, 1960)

$$cC_{\mathrm{v}}\frac{\mathrm{D}T}{\mathrm{D}t} = p\nabla\cdot\mathbf{v} + \Gamma \tag{3}$$

where

C_{v} is the specific heat of air at constant volume (molar basis),

T is the absolute temperature,

p is the local pressure, and

Γ accounts for energy input arising from latent-heating effects.

As shown in the Appendix this may be combined with