NUMERICAL MODELING OF CUMULUS CLOUD CHEMISTRY —PART I. MODEL DEVELOPMENT *

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Received October 9, 1995; revised January 19, 1996

ABSTRACT

A one-dimensional cumulus cloud chemistry model (1CCCM) is developed to simulate cloud physical processes and chemical processes during the evolution of a convective cloud. The cloud physical submodel includes a detailed microphysical parameterized scheme of 20 processes. The chemistry submodel is composed of three parts: gas phase chemistry, aqueous phase chemistry and scavenging of soluble gases. The gas phase reaction mechanism contains 85 reactions among 45 species including 13 organics. The aqueous phase reaction mechanism contains 54 reactions among 40 species and 12 ion equilibria. Mass of 19 gases is transported between the gas phase and the aqueous phase. With this model, studies may be made to analyze the interactions among processes during lifetime of a cumulus cloud.

Key words: cloud model, numerical method, aqueous chemistry, gas scavenging

I. INTRODUCTION

It is well recognized that clouds play an important role in the redistribution and transformation of chemical species and the developing of acid deposition. A number of models have been formulated to study the chemical processes taking place in clouds. Most of them focused on cumulus cloud chemistry (Tremblay and Leighton 1986; Niewiadomski 1989; Walcek and Taylor 1986; Lee 1986), because cumulus clouds contain a mechanism of intensive vertical transports as well as provide sites for aqueous chemical reactions with large liquid water content within clouds.

Most of these cumulus models focused on acidification and their basic chemical processes are quite similar. Solubility equilibrium is generally assumed between gas phase and aqueous phase in cloud droplets (and rain droplets) for chemical species such as SO_2, HNO_3, NH_3 and CO_2. In aqueous phase, oxidation of sulfur dioxide by O_3 and H_2O_2 is considered. Formic acid has been identified as an important contributor to the acidification of precipitation at remote sites (Norton 1985; Keene and Galloway 1986; Jacob 1986). Chamedies and Davis (1983) and Chamedies (1984) pointed out that clouds are a major global source of HCOOH in the troposphere. So it is great need to introduce organic chemistry reactions into cloud chemistry model.

It is recognized that the gas phase chemistry and the aqueous chemistry are closely coupled and some mesoscale acid deposition models include the gas phase mechanism

* This work was supported by the National Natural Science Foundation of China.

(Carmichael 1986; Chang et al. 1987). Usually the role of gas phase chemistry is neglected in cumulus cloud chemistry model because of the short lifetime of cumulus clouds. Due to the intensive vertical transport, cumulus clouds can change the vertical distribution of chemical species and thereby influence the chemical process of the troposphere, especially the photochemical process. In view of the formation of acidification, O_3, H_2O_2 and free radicals such as OH and HO_2 formed through photochemical reactions in the gas phase are scavenged by cloud water and take part in the aqueous phase oxidation of S (IV). In the gas phase, OH radical is the major species oxidizing SO_2 and NO_2 into sulfuric and nitric acid, respectively. For the same reason, the inclusion of gas phase chemistry in the cumulus cloud chemistry models is also necessary.

More recently, oxidation capacity of the troposphere is investigated by studying the interaction of chemical processes and cloud processes (Lelieveld and Crutzen 1990; Chamedies and Davis 1982; Chamedies 1984). Thus the gas phase chemistry included in cloud chemistry model must not only be able to describe the formation of acidification, but also to predict the concentrations of free radicals, O_3, H_2O_2 and other oxidation products.

Our objective is to develop a cumulus cloud chemistry model as a tool with wide applications to study interaction among cloud processes and chemical processes during lifetime of a cumulus cloud, which can be used to study the roles of cumulus clouds in recycle of atmospheric species, the impact of clouds on oxidizing capacity of the troposphere and acid deposition. In this paper, a description of one-dimensional cumulus cloud chemistry model (1CCCM) is presented. Table 1 shows the components of 1CCCM. A companion paper will present model results of analysis of ozone chemistry during a cumulus cloud.

Table 1. List of Gas Phase Species

No.	Species	No.	Species	No.	Species	No.	Species	No.	Species
1	O_2	2	O_3	3	$O^1(D)$	4	$O_3(P)$	5	OH
6	HO_2	7	H_2O	8	H_2O_2	9	NO	10	NO_2
11	NO_3	12	N_2O_5	13	HNO_2	14	HNO_3	15	HNO_4
16	NH_3	17	CO	18	CO_2	19	HCl	20	SO_2
21	H_2SO_4	22	CH_4	23	ALK	24	ETHE	25	OLE
26	HCHO	27	RCHO	28	TOLU	29	XYLE	30	ROOH
31	HCOOH	32	PAA	33	PAN	34	NPHE	35	MO_2
36	RO_2	37	RCO_2	38	CHO_2	39	GLYX	40	MGLY
41	DIAL	42	MHO_2	43	CRES	44	BZO	45	BZNO

II. MODEL DESCRIPTION

1. *Cumulus Cloud Submodel*

The cumulus cloud submodel adopted here is a modified version of the one-dimensional, time-dependent cumulus cloud parameterized model developed by Hu and He (1987).

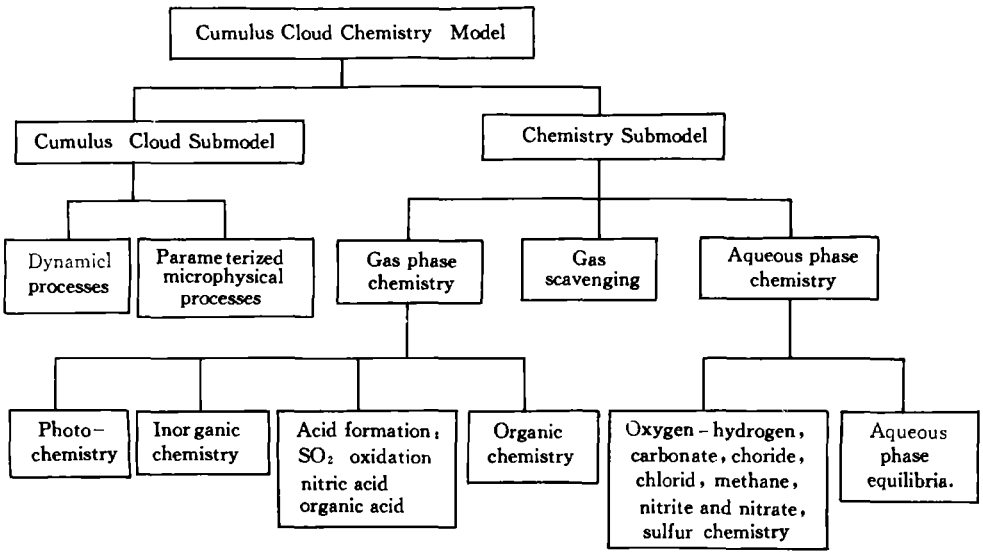

Fig. 1. Components of 1CCCM.

The water substance is grouped into five categories: water vapor, cloud water, rainwater, ice particles and graupels. A detailed parameterized scheme which consists of 20 processes is applied to deal with the microphysical processes between these five water categories. A detailed description of the governing equations can be found in the paper of Hu and He (1987).

2. *Chemical Submodel*

The chemical submodel is executed simultaneously with the cumulus cloud submodel at each step using the output of the cloud physical model. It is a flexible submodel which can describe the chemical processes in the tropospheric atmosphere, including gas phase chemistry and aqueous chemistry, as well as the gases scavenging. In order to make the model have a broad applications, some necessary mechanisms are considered such as photochemistry reactions in the troposphere, free radicals reactions and the various pathways of formation of acidification. With minor modification, the codes of chemistry submodel are designed to be easily adopted by other models such as mutil-dimensional cloud models and long-range transport models.

The gas phase chemical reaction mechanism used here is based on the work of Lurmann and Alan (1986) and Zhang (1990). Using 85 reactions among 45 species, it simulates concentrations and conversing rates of sulfur oxides, nitrogen oxides, OH radical, hydrogen peroxide (H_2O_2), organic peroxides, ozone and other species. Of these reactions those involving free radicals form the largest part of the mechanism. The gas phase chemistry in 1CCCM is not only used to describe the formation of acidification but also to predict the concentrations of the oxidants generated by the photochemistry processes (such as H_2O_2, HO_2, OH etc.). It includes the gas phase acid production pathways of sulfate,

nitrate and organic acids. Sulfate (H_2SO_4) is the final products of the gas phase oxidation of SO_2 by OH. The lists of gas phase species and reaction formulations are persented in Table 2 and Table 3.

Table 2. Gas Phase Chemistry Mechanism

No.	Reactions	No.	Reactions
G1	$NO_2 + h\nu \rightarrow NO + O_3$	G2	$NO_3 + h\nu \rightarrow 0.15NO + 0.85NO_2 + 0.85O_3$
G3	$O_3 + h\nu \rightarrow O^1(D)$	G4	$HNO_2 + h\nu \rightarrow NO + OH$
G5	$H_2O_2 + h\nu \rightarrow 2OH$	G6	$HCOH + h\nu \rightarrow 2HO_2 + CO$
G7	$HCHO + h\nu \rightarrow CO$	G8	$RCHO + h\nu \rightarrow CO + HO_2 + RO_2$
G9	$GLYX + h\nu \rightarrow 0.13HCHO + 1.87CO$	G10	$MGLY + h\nu \rightarrow RCO_3 + HO_2 + CO$
G11	$DIAL + h\nu \rightarrow HO_2 + CO + RCO_3$	G12	$CH_3OOH + h\nu \rightarrow HO_2 + OH$
G13	$PAA + h\nu \rightarrow MO_2 + OH$	G14	$NO_2 + NO_3 \rightarrow N_2O_5$
G15	$NO + OH \rightarrow HNO_2$	G16	$NO_2 + OH \rightarrow HNO_3$
G17	$SO_2 + OH \rightarrow H_2SO_4 + HO_2$	G18	$N_2O_5 \rightarrow NO_2 + NO_3$
G19	$CO + OH \rightarrow HO_2$	G20	$NO_2 + HO_2 \rightarrow HNO_4$
G21	$HNO_4 \rightarrow NO_2 + HO_2$	G22	$HO_2 + HO_2 \rightarrow H_2O_2$
G23	$NO + O_3 \rightarrow NO_2$	G24	$NO_2 + O_3 \rightarrow NO_3$
G25	$NO + NO_3 \rightarrow 2NO_2$	G26	$NO + NO \rightarrow 2NO_2$
G27	$N_2O_5 + H_2O \rightarrow 2HNO_3$	G28	$NO_2 + NO_3 \rightarrow NO + NO_2$
G29	$O^1(D) + H_2O \rightarrow 2OH$	G30	$O^1(D) \rightarrow O_3$
G31	$NO_2 \rightarrow HNO_2 - 1.0NO_2 + HNO_3$	G32	$HNO_3 + OH \rightarrow NO_3$
G33	$O_3 + OH \rightarrow HO_2$	G34	$NO + HO_2 \rightarrow NO_2 + OH$
G35	$HNO_4 + OH \rightarrow NO_2$	G36	$O_3 + HO_2 \rightarrow OH$
G37	$NO_3 + HO_2 \rightarrow HNO_3$	G38	$H_2O_2 + OH \rightarrow HO_2$
G39	$ALK + OH \rightarrow 0.19HCHO + 0.48RCHO + RO_2$	G40	$ETHE + OH \rightarrow RO_2 + 1.56HCHO + 0.22RCHO$
G41	$ETHE + O_3 \rightarrow HCHO + 0.12HO_2 + 0.42CO + 0.4CHO_2$	G42	$OLE + OH \rightarrow HCHO + RO_2 + RCHO$
G43	$OLE + O_3 \rightarrow 0.64HCHO + 0.17HO_2 + 0.5RCHO + 0.29CO + 0.06OH + 0.135MO_2 + 0.4CHO_2$	G44	$RO_2 + NO \rightarrow NO_2 + HO_2$
G45	$RO_2 + HO_2 \rightarrow RCHO + RCHO$	G46	$RO_2 + RO_2 \rightarrow HO_2$
G47	$HCHO + OH \rightarrow HO_2 + CO$	G48	$HCHO + HO_2 \rightarrow MHO_2$
G49	$RCHO + OH \rightarrow RCO_3$	G50	$RCO_3 + NO \rightarrow NO_2 + HCHO + RO_2$

(to be continued)

No.	Reactions	No.	Reactions
G51	$RCO_3 + HO_2 \rightarrow HCHO + PAA$	G52	$RCO_3 + RO_2 \rightarrow HO_2 + HCHO$
G53	$RCO_3 + RCO_3 \rightarrow 2HO_2 + 2HCHO$	G54	$RCO_3 + NO_2 \rightarrow PAN$
G55	$PAN \rightarrow NO_2 + RCO_3$	G56	$GLYX + OH \rightarrow 0.63HO_2 + 1.26CO + 0.37RCO_3$
G57	$MGLY + OH \rightarrow RCO_3 + CO$	G58	$TOLU + OH \rightarrow 0.16CRES + 0.16HO_2 + 0.4DIAL + 0.84RCO_2 + 0.144MGLY + 0.144GLYX$
G59	$DIAL + OH \rightarrow RCO_3$	G60	$XYLE + OH \rightarrow 0.17CRES + 0.17HO_2 + 0.83RO_2 + 0.65DIAL + 0.316MGLY + 0.095GLYX$
G61	$CRES + OH \rightarrow 0.2MGLY + RO_2$	G62	$CRES + NO_3 \rightarrow HNO_3 + BZO$
G63	$BZO + NO_2 \rightarrow NPHE$	G64	$NPHE + NO_3 \rightarrow HNO_3 + BZN_2$
G65	$BZN_2 + NO_2 \rightarrow$	G66	$BZN_2 + HO_2 \rightarrow NPHE$
G67	$BZN_2 \rightarrow NPHE$	G68	$CHO_2 + SO_2 \rightarrow H_2SO_4 + HCHO$
G69	$CH_4 + OH \rightarrow MO_2$	G70	$MO_2 + HO_2 \rightarrow HCHO + CH_3OOH$
G71	$MO_2 + NO \rightarrow HCHO + NO_2 + HO_2$	G72	$MO_2 + NO_2 \rightarrow 1.4HCHO + 0.8HO_2$
G73	$CHO_2 + H_2O \rightarrow HCOOH$	G74	$MHO_2 + NO \rightarrow HCOOH$
G75	$MHO_2 + MHO_2 \rightarrow 2HCOOH + 2HO_2$	G76	$MHO_2 + HO_2 \rightarrow HCOOH$
G77	$HCOOH + OH \rightarrow HO_2$	G78	$HCHO + NO_3 \rightarrow HNO_3 + HO_2 + CO$
G79	$RCHO + NO_3 \rightarrow HNO_3 + RCO_3$	G80	$GLYX + NO_3 \rightarrow HNO_3 + 0.63HO_2 + 1.26CO + 0.37RCO_3$
G81	$MGLY + NO_3 \rightarrow HNO_3 + RCO_3 + CO$	G82	$DIAL + NO_3 \rightarrow HNO_3 + RCO_3$
G83	$CHO_3 + NO \rightarrow HCHO + NO_2$	G84	$PAA + OH \rightarrow RCO_3$
G85	$PAN + OH \rightarrow CHO_2 + CO + NO_2$		

The aqueous chemistry mechanism used in this model is similar to the work of Zhang (1990) which consists of 40 species and 54 reactions listed in Table 4 and Table 5. The oxidants (O_3, H_2O_2, HO_2, OH, NO_3) generated in the gas phase are rapidly scavenged by water droplets and incorporated into the aqueous phase where they play an important role in aqueous phase. It is recognized that the NO_3 radicals can be an important oxidant of S(IV) (via reaction AQ48 in Table 4) under night time conditions in rural environment (Chamedies 1986; Cho et al. 1989). The inclusion of NO_3 radical reactions enlarges the application of this chemical submodel.

Table 3. List of Aqueous Phase

No.	Species	No.	Species	No.	Species	No.	Species	No.	Species
1	OH	2	HO_2	3	H_2O_2	4	O_3	5	SO_2
6	CO_2	7	NO	8	NO_2	9	NO_3	10	HNO_2
11	HNO_3	12	Cl	13	HCl	14	NH_3	15	MO_2
16	HCHO	17	HCOOH	18	ROOH	19	H_2COH_2	20	PAA
21	PAN	22	H^+	23	OH^-	24	O_2^-	25	HSO_3^-
26	SO_3^{2-}	27	SO_4^-	28	SO_4^{2-}	29	SO_5^-	20	HSO_5^-
31	$HMSA^-$	32	NO_2^-	33	NO_3^-	34	HCO_3^-	35	CO_3^{2-}
36	NH_4^+	37	Cl^-	38	Cl_2^-	39	$ClOH^-$	40	$HCOO^-$

Table 4. Aqueous Phase Chemistry Mechanism

No.	Reactions	No.	Reactions
AQ1	$H_2O_2+h\nu\rightarrow 2OH$	AQ2	$OH + HO_2\rightarrow$
AQ3	$OH + O_2^-\rightarrow OH^-$	AQ4	$OH + H_2O_2\rightarrow HO_2$
AQ5	$HO_2+ HO_2\rightarrow H_2O_2$	AQ6	$HO_2+ O_2^-\rightarrow H_2O_2+OH^-$
AQ7	$O_2^- + O_3\rightarrow OH + OH^-$	AQ8	$HCO_3^- + OH\rightarrow CO_3^-$
AQ9	$HCO_3^- + O_2^-\rightarrow H_2O_2+CO_3^- + H^+$	AQ10	$CO_3^- + O_2^-\rightarrow HCO_3^- + OH^-$
AQ11	$CO_3^- + H_2O_2\rightarrow HO_2+ HCO_3^-$	AQ12	$Cl^-+OH\rightarrow ClOH^-$
AQ13	$ClOH^-\rightarrow Cl^-+OH$	AQ14	$ClOH^- + H^+\rightarrow Cl$
AQ15	$Cl\rightarrow ClOH^- + H^+$	AQ16	$HO_2+Cl_2^-\rightarrow 2Cl^- + H^+$
AQ17	$O_2^- + Cl_2^-\rightarrow 2 Cl^-$	AQ18	$H_2O_2+ Cl_2^-\rightarrow 2 Cl^- + HO_2+ H^+$
AQ19	$H_2O_2+Cl\rightarrow Cl^- + HO_2+H^+$	AQ20	$NO_3^-+h\nu\rightarrow OH +OH^- + NO_2$
AQ21	$NO_3+HO_2\rightarrow NO_3^-+H^+$	AQ22	$NO_3+O_2^-\rightarrow NO_3^-$
AQ23	$NO_3+Cl^-\rightarrow NO_3^- + Cl$	AQ25	$H_2COH_2+OH\rightarrow HCOOH + HO_2$
AQ26	$HCOOH +OH\rightarrow CO_2+ HO_2$	AQ27	$HCOO^- + OH\rightarrow CO_2+ HO_2+OH^-$
AQ28	$HCOO^- + CO_3^-\rightarrow CO_2+ HCO_3^- +HO_2 + OH^-$	AQ29	$HCOO^- + Cl_2^-\rightarrow CO_2+ HO_2+ 2Cl^-$
AQ30	$H_2O_2+SO_2\rightarrow 2 H^+ + SO_4^{2-}$	AQ31	$O_3+ HSO_3^-\rightarrow H^+ +SO_4^{2-}$
AQ32	$O_3+ SO_3^{2-}\rightarrow SO_4^{2-}$	AQ33	$SO_3^{2-} + OH\rightarrow SO_5^- + OH^-$
AQ34	$HSO_3^- +OH\rightarrow SO_5^-$	AQ35	$SO_5^- + HSO_3^-\rightarrow HSO_5^- + SO_5^-$

(to be continued)

No.	Reactions	No.	Reactions
AQ36	$SO_5^- + SO_3^{2-} \rightarrow HSO_5^-$	AQ38	$SO_5^- + SO_5^- \rightarrow 2SO_4^-$
AQ39	$HSO_5^- + HSO_3^- + H^+ \rightarrow 2SO_4^{2-} + 3H^+$	AQ40	$HSO_5^- + OH \rightarrow SO_5^-$
AQ41	$SO_4^- + HSO_3^- \rightarrow SO_4^{2-} + H^+ + SO_5^-$	AQ42	$SO_4^- + HO_2 \rightarrow SO_4^{2-} + H^+$
AQ43	$SO_4^- + O_2^- \rightarrow SO_4^{2-}$	AQ44	$SO_4^- + H_2O_2 \rightarrow SO_4^{2-} + H^+ + HO_2$
AQ45	$SO_4^- + Cl^- \rightarrow SO_4^{2-} + Cl$	AQ46	$HO_2 + HSO_3^- \rightarrow SO_4^{2-} + H^+ + OH$
AQ47	$O_2^- + SO_3^{2-} \rightarrow SO_4^{2-} + OH + OH^-$	AQ48	$HSO_3^- + NO_3 \rightarrow NO_3^- + 2H^+ + SO_4^{2-} + SO_4^- + HSO_3^-$
AQ49	$HCHO + HSO_3^- \rightarrow HMSA^-$	AQ50	$HCHO + SO_3^{2-} \rightarrow HMSA^- + OH^-$
AQ51	$HMSA^- + OH \rightarrow SO_5^- + HCHO$	AQ52	$HSO_3^- + Cl_2^- \rightarrow SO_5^- + 2Cl + H^+$
AQ53	$Cl_2^- \rightarrow Cl^- + Cl$	AQ54	$Cl^- + Cl \rightarrow Cl_2^-$

Table 5. Aqueous Phase Eqilibria

No.	Reactions	No.	Reactions
E1	$H_2O \Leftrightarrow H^+ + OH^-$	E2	$SO_2 \Leftrightarrow H^+ + HSO_3^-$
E3	$HSO_3^- \Leftrightarrow H^+ + SO_3^{2-}$	E4	$HNO_3 \Leftrightarrow H^+ + NO_3^-$
E5	$CO_2 \Leftrightarrow H^+ + HCO_3^-$	E6	$HCO_3 \Leftrightarrow H^+ + CO_3^{2-}$
E7	$NH_3 \Leftrightarrow NH_4^+ + OH^-$	E8	$HO_2 \Leftrightarrow H^+ + O_2^-$
E9	$HNO_2 \Leftrightarrow H^+ + NO_2^-$	E10	$HCHO \Leftrightarrow H_2C(OH)_2$
E11	$HCOOH \Leftrightarrow H^+ + HCOO^-$	E12	$HCl\ H^+ + Cl^-$

A total of 12 ion equilibirum equations are taken into account in this model. The reactions are listed in Table 5. The concentrations of ions are calculated under the electrical-neutrality constraint and acid-alkali balance constraint.

Proper representation of mass transferring of gas species from air into liquid drops is an important component of cloud chemistry model. Dissolution rates in cloud water and rainwater of 19 kinds of gas species are similar to Qin and Chamedies (1986). Table 6 lists 19 gaseous species. The rate of scavenging of species I from the gas phase into cloud water (rainwater) is given by

$$\frac{dG(I)}{dt} = -4\pi D_i N_c R_c f_1 f_2 \left(G(I) - \frac{C(I)}{P^* H(I)} \right), \quad (1)$$

where $G(I)$ is the mixing ratio of species I in gas phase; $C(I)$ the mixing ratio of species I in cloud water (or rainwater); D_i molecular diffusion coefficient of the species I; P atmosphere pressure; $H(I)$ efficient solubility constant of the species I; N_c number concentration of cloud droplets (or raindrops); R_c average radius of cloud droplets (or raindrops); f_1 corrective coefficient of molecular boundary layer for cloud droplets; f_2 ventilation

factor, corrective coefficient for convective mass-transport for raindrops.

Table 6. List of Gas-Aqueous Phase Equilibria

No.	Gas phase Aqueous phase	No.	Gas phase Aqueous phase
1	(G) SO_2 ⇔ (AQ) SO_2	2	(G) H_2O_2 ⇔ (AQ) H_2O_2
3	(G) HNO_3 ⇔ (AQ) HNO_3	4	(G) HNO_2 ⇔ (AQ) HNO_2
5	(G) O_3 ⇔ (AQ) O_3	6	(G) NO_2 ⇔ (AQ) NO_2
7	(G) NO ⇔ (AQ) NO	8	(G) MO_2 ⇔ (AQ) MO_2
9	(G) CO_2 ⇔ (AQ) CO_2	10	(G) NH_3 ⇔ (AQ) NH_3
11	(G) PAN ⇔ (AQ) PAN	12	(G) HCHO ⇔ (AQ) HCHO
13	(G) HCOOH ⇔ (AQ) HCOOH	14	(G) HCL ⇔ (AQ) HCL
15	(G) ROOH ⇔ (AQ) ROOH	16	(G) PAA ⇔ (AQ) PAA
17	(G) NO_3 ⇔ (AQ) NO_3	18	(G) OH ⇔ (AQ) OH
19	(G) HO_2 ⇔ (AQ) HO_2		

III. NUMERICAL SCHEME

The numerical scheme of cloud physical submodel can be found in papers of Hu and He (1987). Here we just give a description of the numerical scheme of the chemistry submodel.

A time-splitting method is used to integrate the governing equations of the cloud chemistry submodel. The chemical reaction equations are solved by two-step method in short time step (0. 2 s) and 50-time iteration. This method is a modification from QSSA method described in detail by Hesstvedt et al. (1978). The variations of the concentration of species i can be written in the following form

$$\frac{dC_i^n}{dt} = P_i^n - D_i^n C_i^n, \tag{2}$$

where P_i^n and $D_i^n C_i^n$ are production rate and depletion rate of species C_i^n in unit (ppbv s^{-1}), respectively, which vary in space and with time. They also depend on the concentrations of the other species at the grid point and the time under consideration. Thus Eq. (2) is in general nonlinear, although it looks linear. An attempt to choose automatically the optimal algorithm for each equation is carried out by checking the value of factor *Tau*, which equals to the ratio of time step and the character time of species i.

Let $Tau = \Delta t / \tau$, and $\tau_i = 1/D_i^n$. The criterion of solving Eq. (2) is

$$\begin{aligned}
&\text{if } Tau < 0.01,\ C_i^{n+1} = C_i^n + (P_i^n - D_i^n C_i^n)cdt,\\
&\text{if } Tau > 10,\ C_i^{n+1} = \frac{P_i^{n+1}}{D_i^{n+1}},\\
&\text{if } 0.01 \leqslant Tau \leqslant 10,\ C_i^{n+1} = \frac{P_i^n}{D_i^n} + \left(C_i^n - \frac{P_i^n}{D_i^n}\right)\exp(-Tau),
\end{aligned} \tag{3}$$

where cdt is the chemical time step of 0. 2 s. $n+1$ and n represent at time $n+1$ and time n.

The value of *Tau* is surveyed throughout the calculation and an iterative procedure is devised to improve the accuracy of the solution. The two-step method is that the calculation of C_i^{n+1} is divided into two steps. In the first step we calculate concentrations of C_i^* with input concentrations of C_i^n and the time step is a half of the chemical time step (0.1 s), then obtain P_i^* and D_i^* with these concentrations (C_i^*). In the second step we calculate the concentrations of species C_i^{n+1} with the values of P_i^*, D_i^* and C_i^*, and the time step is 0.2 s.

With 1CCCM, ozone chemistry processes during a single convective event are analyzed in the succeeding paper (Part II).

IV. CONCLUDING REMARKS

A one-dimensional comprehensive cumulus cloud chemistry model (1CCCM) is developed to simulate the physical and chemical processes during cumulus processes.

The cumulus cloud submodel includes a detailed microphysical scheme in 20 processes. It predicts the variables of updraft velocity, temperature, bulk mass mixing ratios of water vapor, cloud water, rainwater, ice particles and graupels, as well as the bulk number concentrations of rainwater, ice particles, and graupels. The chemical submodel is composed of gas phase chemistry, aqueous phase chemistry, and gases scavenging. The gas phase reaction mechanism contains 85 reactions among 46 species including 13 organics. The aqueous phase reaction mechanism contains 54 reactions among 40 species and 12 ion equilibra. Mass of 19 gases is transported between the gas phase and the aqueous phase. The two-step method is used to integrate the chemical equations.

The 1CCCM is designed as a powerful tool and it has a broad applications. With this model, the simulation gives a picture of the cumulus cloud chemical processes. Studies may be made to analyze the interactions among processes taking place during a convection event. Formation of acidification, problems of tropospheric oxidizing capacity, and roles of clouds in tropospheric chemistry can be evaluated through numerical experiments. Ozone chemistry processes during a single cumulus cloud process are analyzed in the succeeding paper (Part II).

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