

Acid Rain Model Development Considering Altitudinal Precipitation Rate

고도별 강우율 변화를 고려한 산성비 모델의 개발

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國 文 抄 錄

개발된 산성비 모델에서는 다종의 대기오염물 제거로 인한 강우내의 화학성분과 산성도의 고도별 및 시간별 변화를 예측하기 위하여, 강우시 구름내와 밑에서 일어나는 대기 오염물의 강우내로의 물질전달 현상과 화학반응 현상을 동시에 고려하였다. 또한, 강우 산성도의 형성에 중요한 영향을 미치는 구름내에서의 복잡한 동력학적 특성을 고려하기 위하여 강우율의 높이별 변화를 단순화하여 해석하였다.

개발모델을 이용하여 1985년과 1987년 가을의 서울시 산성비에 대하여, 산성비 모델을 이용하여 추정된 pH값과 실측 pH값 사이의 상관관계는 0.57을 나타내었고, 효과적이고 정량적인 관리를 위하여 대기중의 이산화황 농도, 구름층의 두께, 지상에서의 강우율 및 황산이온의 형성에 중요한 역할을 하는 산화제들의 강우 산성도에 미치는 영향을 민감도 분석을 통해서 나타내었다.

INTRODUCTION

The chemical composition of raindrops is influenced by the removals of atmospheric gaseous pollutants and of aerosols, which are occurring both in-cloud and below-cloud. From the importance of this wet removal processes, much research efforts have been devoted to relate the atmospheric concentration of gaseous pollutants with the

rain acidity. However, those relationships are not yet fully understood due to the complex mechanisms in natural system such as the dynamic processes of cloud, the gas absorption, and the aqueous-phase chemical reactions.

Liljestrand *et al.* (1979) have proposed a equilibrium model assuming the equilibria between atmospheric pollutants and raindrops. Durham *et al.* (1984) and Kumar (1985) estimated the rain

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acidity considering the gaseous mass transfer and aqueous-phase chemical reactions below cloud. However, they have not treated the mechanisms within cloud. To quantify the removal efficiency of atmospheric pollutants within cloud, Fisher (1982) and Scott (1982) considered dynamic characteristics of cloud and simple chemical kinetics rather than the removal of various atmospheric pollutants.

In this study, coupled processes of mass transfer and chemical kinetics both in-cloud and below-cloud are considered, and the complex dynamic processes within cloud are represented by the variation of precipitation rate for the simplicity. Therefore, the objective of this study is to develop an acid rain model, in which the chemical composition of raindrops is a function of the atmospheric concentration of various pollutants and the precipitation rate. The rain acidity can be estimated from the simultaneous processes; absorption of various gaseous pollutants and chemical reactions in aqueous phase.

In addition, the sensitivity analyses for the rain acidity are carried out to investigate the effects of atmospheric pollutants, precipitation rate, and various oxidants for sulfate.

MODEL DESCRIPTION

As shown in Figure 1, a model domain consists of two layers to analyze the acidification processes of raindrops. Thus, entire region is divided into two regions of in-cloud and below-cloud. The altitudinal variations of updraft velocity and liquid water contents in each sub-region are utilized to obtain the characteristic size and fallspeed of raindrops. Then the chemical composition and acidity of raindrops are estimated in each sub-region from the solution of mass transfer equations that include the coupled processes of

gas absorption and chemical kinetics in aqueous phase. In the sections followed, the steps for the analysis of governing equations are evaluated.

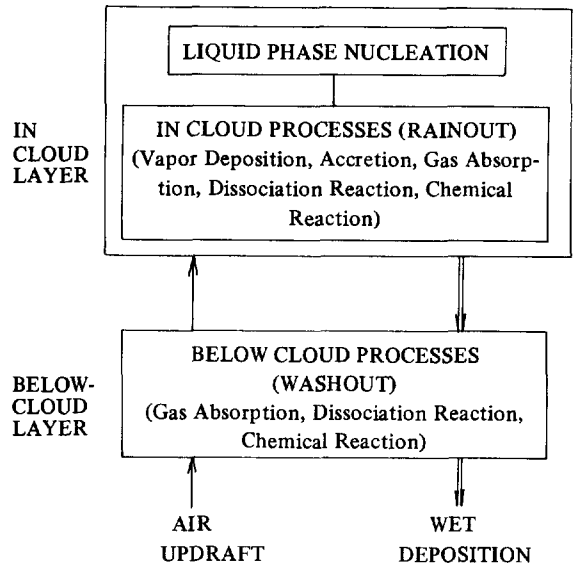


Fig. 1 Two layers for acid rain development

Altitudinal Variation of Precipitation Rate

The precipitation rate at each height has been specified to simplify the complex processes within cloud during precipitation and to analyze gas absorption and chemical kinetics occurring simultaneously in gas and aqueous phases. The characteristic values for raindrops are obtained by using the relationships of the followings (Scott, 1982).

$$M = 0.074 J^{0.89} \quad (1a)$$

$$V = 3.75 J^{0.11} \quad (1b)$$

$$V = 130 d^{0.5} \quad (1c)$$

where, M is precipitation mass concentration (g/m^3), J is precipitation rate (mm/hr), V is fallspeed of raindrop (m/s), and d is the diameter

of raindrop (m).

To estimate altitudinal variation of precipitation rate, the formulas proposed by Scott (1982) are employed. That is, the initial precipitation embryos are assumed to be the ice crystal, and they become raindrops at the ground level through the growth by vapor deposition and accretion of liquid cloud droplets at each level.

Some knowledge of the vertical profiles of cloud water concentration and air updraft velocity are required to obtain precipitation rate as a function of height. In this study, they are assumed to have a parabolic profiles, which is known to be the most representative precipitation condition in a steady state (Scott, 1982; and Fisher, 1982). Thus, the altitudinal variation of precipitation rate is expressed as the following equation from Scott's study (1982).

$$J(z)^{0.22} = J(b)^{0.22} - 3.10 \times 10^{-4} \bar{m}(z - z_b) \quad (2)$$

where, $\bar{m}(g/m^3)$ is the average cloud water concentration between the ground and height z , $J(b)$ is the precipitation rate at the cloud base (mm/hr), and $z_b(m)$ is the height of the cloud base.

As the equation (2) is applicable for the rain at height z where the temperature is higher than $0^\circ C$, the zone is called as the raingrowth zone by Scott (1982). The equation representing the precipitation rate as a function of height with temperature lower than $0^\circ C$ is different from equation (2). However, since precipitation droplets are small and are advected from the top of cloud, they do not contribute to the wet deposition in that region. Therefore, the removal of gaseous pollutants by them is neglected in the analyses of acidification processes for the simplicity.

The precipitation rate below cloud is assumed

to be identical with the value at cloud base, because the effects of vapor deposition and evaporation on the precipitation rate are small and are regarded to be canceled out each other (Hong *et al.*, 1988).

In Figure 2, the solution of equation (2) for surface precipitation rate of 2.5mm/hr is illustrated. As shown in Figure 2, the total cloud depth is 8km, the height from surface to cloud base is 1km, and the height from surface with temperature higher than $0^\circ C$ is assumed to be 6km, of which values are similar to that proposed by Scott (1982).

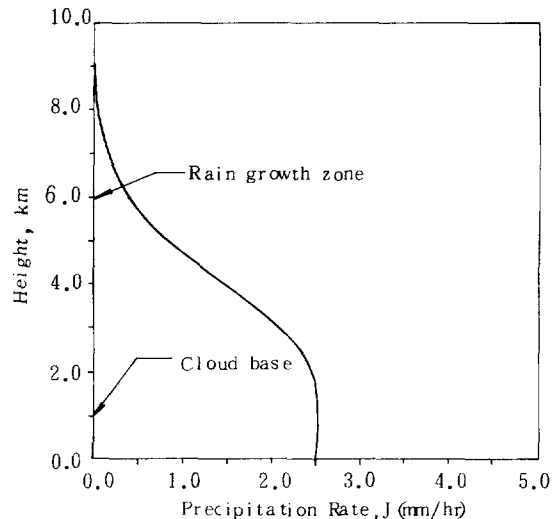


Fig. 2 Precipitation rate as a function of height

To investigate the acidification process of raindrops through gas absorption and aqueous-phase chemical kinetics, the empirical relationships between characteristic variables for raindrop are used from the equations of (1a), (1b), and (1c).

Physical Aspects of a Model

Basic assumption in this work are the followings: First, complete mixing in the gas-phase at

each interval during a rain event with a constant dispersion coefficient is assumed. Second, it is known that the shear on the surface of a falling drop creates a significant velocity gradient and due to the rapid mixing inside the drop. Thus, a liquid-phase resistance to mass transfer (Bird, 1969) is assumed to be negligible. The gas-phase mass transfer coefficient, k_g , for the drop in motion can be calculated from the Frössling equation (Kumar, 1985).

Then, the mass balance equation for the pollutant in gas-phase, of which concentration $P(z, t)$ is taking into account of the mass loss transferred into the raindrops of the entire size spectrum is

$$\begin{aligned} & \frac{\partial P_i(z, t)}{\partial t} + \frac{\partial}{\partial z} (W \cdot P_i) \\ & = K \cdot \frac{\partial^2 P_i(z, t)}{\partial z^2} - \int 4\pi r^2 \cdot n(r) k_g \\ & \cdot [P_i(z, t) - \frac{c_i(z, t)}{H}] dr \end{aligned} \quad (3)$$

where, $c(z, t)$ is the aqueous-phase concentration in raindrop of radius r_p , $n(r)$ is the size distribution function, and $n(r)dr$ represents the number of drops between radius r and $r+dr$ per unit volume of air. And K is the effective dispersion coefficient which represents a sum of turbulent diffusion and molecular diffusion. Also, $W(z)$ is the updraft velocity of air parcel at an arbitrary height and H is the Henry's law constant.

The equation (3) is simplified because the calculation considering the entire size spectrum of raindrops is very cumbersome. Thus, it is assumed that the uniform size of the raindrop is r_p , and the number of drops per unit volume of air (N_p) corresponds to $\frac{3}{4} \cdot \pi r_p^3 \cdot n(r)$. The value of r_p can be obtained using equation (1c). Thus, the equation for species mass balance in gas-phase can be simplified as

$$\begin{aligned} & \frac{\partial P_i(z, t)}{\partial t} + \frac{\partial}{\partial z} (W \cdot P_i) \\ & = K \cdot \frac{\partial^2 P_i(z, t)}{\partial z^2} - 4\pi r_p^2 \cdot N_p \cdot k_g \cdot [P_i(z, t) - \frac{c_i(z, t)}{H}] \end{aligned} \quad (4)$$

The governing mass balance equation for solute in a raindrop of which aqueous concentration is $c(z, t)$ becomes

$$\begin{aligned} & \frac{\partial c_i(z, t)}{\partial t} + \frac{\partial}{\partial z} (V \cdot C_i) \\ & = \frac{3k_g}{r_p RT} [P_i(z, t) - \frac{c_i(z, t)}{H}] \\ & + R(c_1, c_2, \dots, c_n); \quad i = 1, 2, \dots, n \end{aligned} \quad (5)$$

where, R is the universal gas constant, T is the absolute temperature, and $R(c_1, c_2, \dots, c_n)$ represents the chemical reaction of each aqueous-phase species.

To solve the above differential equations, the IMSL (International Mathematics and Statistics Library) which contains the variable time step and error tolerance method is used.

Chemical Aspects of a Model

The most important gases which influence the composition and acidity of precipitation are known as HNO_3 , SO_2 , NH_3 , O_3 , and H_2O_2 . The SO_2 is absorbed into raindrops, undergoes dissociation, and is oxidized to produce sulfate. HNO_3 directly contributes to the rain acidity through the absorption and the dissociation. NH_3 is highly basic and serves to neutralize some of the rain acidity. O_3 and H_2O_2 are considered to be the most important oxidizing agents for S(IV) in the aqueous-phase.

For these pollutant species, the concept of effective Henry's law constant (H^*) indicating the total solubility of pollutant species including the dissociated forms is used in this study. (Seinfeld,

1986) That is, the effective Henry's law constants depending on the raindrop's acidity and the solubilities of HNO_3 , H_2O_2 , and SO_2 decrease as the concentration of hydrogen ion $[\text{H}^+]$ increases. Ammonia solubility on the other hand increases as $[\text{H}^+]$ increases.

In table 1, the forward (k_f) and reverse (k_r) chemical reactions selected for determining the acidification of falling raindrops are listed. Equations (6)-(12) are the reversible reactions for the CO_2 - H_2O - HNO_3 - NH_3 - H_2O system.

Equations (13)-(14) are the reactions for sulfate production by ozone oxidation. The rate constant value of the O_3 /S(IV) reaction is a pH-dependent, and decreases with decreasing pH. The phenomenon of pH dependence of the rate constant coupled with the decreasing solubility of S(IV) with decreasing pH causes the value of overall reaction rate to be decreased strongly with decreasing pH. Because of this strong pH dependence, the reaction tends to be "self-limiting" since it produces $[\text{H}^+]$ as it proceeds (Schwartz, 1984).

Equation (15) is the irreversible S(IV) oxidation by H_2O_2 . In this reaction, the increase of the rate constant with decreasing pH essentially compensates the phenomenon of decreased solubility of SO_2 with decreasing pH. Thus, it gives an overall reaction rate that is nearly independent of pH. Because of this pH independence, the reaction is not "self-limiting" and can proceed at a high rate even at low pHs.

Schwartz (1984) has concluded that the rate of aqueous-phase oxidation of NO_x is expected to be of only minor importance in the atmosphere and contributes negligibly to nitrate level in precipitation. On the other hand, though gaseous HNO_3 is presented as a trace pollutant, it is highly soluble in water. Thus, scavenging of HNO_3 is a major source of nitrate in rainwater. Here, the nitrate aerosols serving as the cloud condensation

Table 1. Chemical reactions in raindrops (Durham, 1984)

Major Reactions	Kinetic Constants
$\text{H}_2\text{O} \rightleftharpoons \text{H}^+ + \text{OH}^-$	$k_f = 1.3 \times 10^{-3}$, $k_r = 1.3 \times 10^{11}$ (6)
$\text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{HCO}_3^- + \text{H}^+$	$k_f = 2.4 \times 10^{-2}$, $k_r = 4.9 \times 10^4$ (7)
$\text{HCO}_3^- \rightleftharpoons \text{CO}_2 + \text{OH}^-$	$k_f = 1.0 \times 10^{-4}$, $k_r = 1.4 \times 10^4$ (8)
$\text{SO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{H}^+ + \text{HSO}_3^-$	$k_f = 3.4 \times 10^6$, $k_r = 2.0 \times 10^4$ (9)
$\text{HSO}_3^- \rightleftharpoons \text{H}^+ + \text{SO}_3^{2-}$	$k_f = 1.0 \times 10^4$, $k_r = 1.0 \times 10^{11}$ (10)
$\text{HNO}_3 \rightleftharpoons \text{H}^+ + \text{NO}_3^-$	$k_f = 2.2 \times 10^4$, $k_r = 1.0 \times 10^4$ (11)
$\text{NH}_3 \rightleftharpoons \text{NH}_4^+ + \text{OH}^-$	$k_f = 6.0 \times 10^4$, $k_r = 3.4 \times 10^{10}$ (12)
$\text{O}_3 + \text{HSO}_3^- \rightarrow \text{SO}_4^{2-}$	$k = 2.2 \times 10^9$ (13)
$\text{O}_3 + \text{SO}_3^{2-} \rightarrow \text{SO}_4^{2-}$	$k = 3.1 \times 10^5$ (14)
$\text{HSO}_3^- + \text{H}_2\text{O}_2 + \text{H}^+ \rightarrow 2\text{H}^+ + \text{SO}_4^{2-}$	$k = 5.2 \times 10^1$ (15)

nuclei are not considered in this work.

In addition to the above pollutant species considered in this model, the species such as Ca^{2+} , Mg^{2+} , K^+ and Cl^- exist in raindrops. Thus, it is needed that $[\text{Ca}^{2+}]$, $[\text{Mg}^{2+}]$, $[\text{K}^+]$, and $[\text{Cl}^-]$ should be considered in the evaluation of rain acidity to satisfy the charge balance of rainwater. From the report of Shin *et al.* (1986) who have measured the concentration of ionic species in rain samples collected in Seoul, Korea, the average value of $2[\text{Ca}^{2+}] + 2[\text{Mg}^{2+}] + [\text{K}^+] - [\text{Cl}^-]$ is calculated, and then the rain acidity by using the charge balance of rainwater is recomputed. The effective value of BNC (base neutralizing capacity) in Seoul is found to be $28 \mu\text{eq}/\ell$.

$$2[\text{Ca}^{2+}] + 2[\text{Mg}^{2+}] + [\text{K}^+] - [\text{Cl}^-] = 28 \mu\text{eq}/\ell \quad (16)$$

MODEL TEST

To test the numerical error of the developed model with which the composition and acidity of raindrops can be estimated, the values of the ion concentrations in raindrops evaluated by dividing the entire height into 10 segments are compared

with the value from the division into 2 segments. As the rain event continues, the difference for each values are as follows; 0.2–0.9%, 0.2–7.9%, 0.8–13%, and 5.9–19% for $[H^+]$, $[SO_4^{2-}]$, $[NO_3^-]$, and $[NH_4^+]$ respectively. It is considered that these differences are caused by the time difference needed for the mass transfer and numerical dispersion. In this model, the entire height is divided into 10 segments of complete mixing to minimize the numerical dispersion.

Also, the total mass balance is checked for each species. The initial total quantities of each species are compared with the sum of each quantity distributed both in gas and aqueous phases at any time. For example, the total $[HNO_3]$ is distributed between gas and aqueous phases as the following.

$$[NHO_3]_{total} = \frac{p(t)}{RT} + [HNO_3] \cdot L + [NO_3^-] \cdot L \quad (\text{unit} = \text{moles/l}) \quad (17)$$

where, L is the liquid water content of the atmosphere (m^3/m^3). The errors in total mass balance after 30 minutes rain event are 8.3% and 11% for HNO_3 and NH_3 , respectively.

VALIDATION OF MODEL

Figure 3 shows the correlation of the pH values from observed rain acidity in the field and from the estimated acidity based on this model. The observed ambient SO_2 concentration and rain acidity data of 1985 and 1987 during fall seasons are obtained from Korea Environmental Administration and Seoul Metropolitan Government Institute of Health and Environment. To calibrate the model, the O_3 and H_2O_2 concentrations are varied from 30ppb to 80ppb and from 1.5 to 2.5 ppb, respectively, and the correlation coefficient values are checked in the process. Correlation coefficient (r) thus obtained is 0.57 in the case of 40ppb

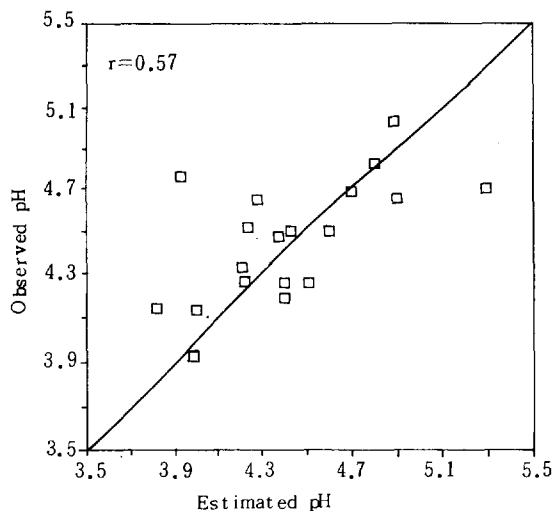


Fig. 3 Comparison of observed and estimated pH values

$[O_3]$ and 1.5ppb $[H_2O_2]$.

Here, the average BNC in Seoul area at equation (16) is considered to satisfy the charge balance in rainwaters for the estimation of rain acidity.

RESULT AND DISCUSSION

Simulations are performed to quantify the variation of concentrations of various pollutants which are dependent on time and altitude. For the simulations, it is assumed that the height from the ground to cloud top (H) is 8km, the rain growth zone is 5km, and the height of cloud base above the ground is 1km. Effective dispersion coefficient (K) is assumed to be the $100cm^2/sec$. Table 2 shows the rainfall parameters at each level obtained by equation during rainfall with surface precipitation rate (J) of 5mm/hr.

The initial concentrations of gaseous species are 20ppb for SO_2 , 1.5ppb for HNO_3 , 1ppb for NH_3 , 50ppb for O_3 , and 2ppb for H_2O_2 . In addition, it is assumed that the initial raindrop pH is 5.6, which is equilibrated with carbonate system ($CO_2=320ppm$).

Table 2. Rainfall parameters at each layer

	Layer (Km)				
	0 - 1	1 - 2	2 - 3	3 - 4	4 - 5
J(mm/hr)	5.0	4.56	3.62	2.43	1.37
r_p (mm)	0.592	0.581	0.552	0.506	0.446
N_p (/m ³)	355	348	330	301	264
V (m/s)	4.47	4.43	4.32	4.13	3.88

Temporal Concentration Variation of Various Species in Raindrops at Ground Level

Shin *et al.* (1986) measured ionic species in rainwaters as a function of time, and reported that their concentration in rainwaters shows the exponential decrease as the gas-phase concentration is depleted. In addition, Ester *et al.* (1984) and Dana *et al.* (1984) proposed that the concentration of ionic species in rainwaters is slowly decreased because of the sequential removal of pollutants as time goes by.

Figure 4 shows the the sharp decreasing of $[SO_4^{2-}]$, $[NO_3^-]$, $[NH_4^+]$ and increasing of pH as the time is passed.

The decrease of $[SO_4^{2-}]$ is a result of the rapid decline in the gas-phase H_2O_2 concentration and a result of the slow decline in the gas-phase SO_2 and O_3 concentrations. The decrease of $[NO_3^-]$ and $[NH_4^+]$ is a direct consequence of high water solubility of HNO_3 and NH_3 which leads to a rapid reduction in gas-phase HNO_3 and NH_3 concentrations.

From this study, the reduction rates are 37%, 43%, and 1.2% for HNO_3 , NH_3 , and SO_2 respectively for 60 minute run. Because the effects of gas absorption into the cloud droplets are not considered in the model, the reduction rates are smaller than that of Hong *et al.* (1988)

Sensitivity Analyses on Rain Acidity

In this section, the sensitivities of the important parameters affecting the raindrop pH are investigated. In the model studied, the rain acidity is expressed as a function of gas-phase concentration of various species, precipitation rate, and cloud depth. The effects of these parameters on raindrop pH are depicted in Figure 5. In these analyses, it is assumed that the initial SO_2 concentration is 20ppb, the surface precipitation rate (J) is 2.5mm/hr, and the rain growth zone is 4km.

The rain acidity is increased the SO_2 concentration increases, as the surface precipitation rate (J) decreases due to large specific interfacial area, and as the rain growth zone (H_r) increases.

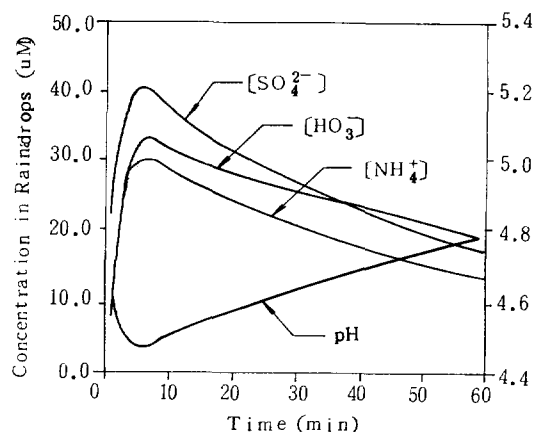


Fig. 4 Temporal concentration profiles of sulfate, nitrate, ammonium, and pH in raindrops at ground level.

Kumar (1986) compared the oxidation rates of S(IV) by O_3 and by H_2O_2 in raindrops, and proposed that the oxidation rate by O_3 decreases sharply with decreasing pH due to its “self-limiting” characteristics, while that by H_2O_2 remains roughly constant because the concentration of H_2O_2 is independent of the rain acidity. Consequently, he reported that S(IV) oxidation by H_2O_2 is likely to dominate at the pH below 4.7.

Figure 6 shows the effects of oxidants on rain acidity for the S(IV) oxidation when SO_2 concent-

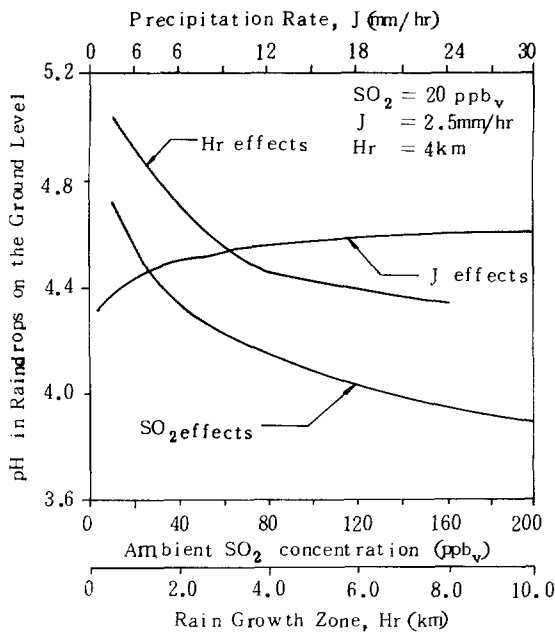


Fig. 5 Sensitivity analyses on the important parameters affecting the raindrop pH

ration is 15ppb and surface precipitation rate is 2.5mm/hr.

As shown in Figure 6, the pH at the ground level decrease sharply at first part as the concentration of H_2O_2 increases, and slowly decreases at

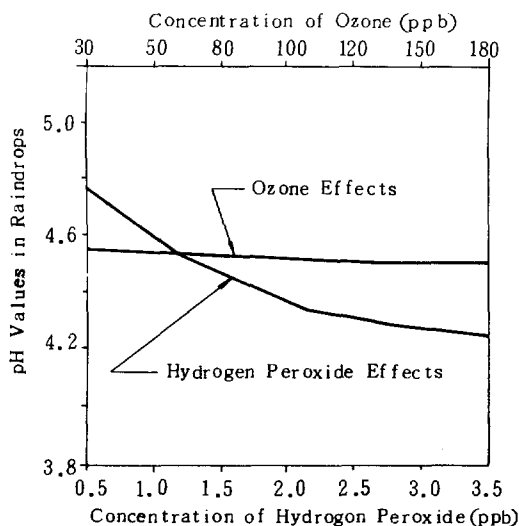


Fig. 6 Effect of oxidants on the rain acidity

later part because of the limited availability of species for the S(IV) oxidation. In case of S(IV) oxidation by O_3 , the pH value does not show significant difference as concentration of O_3 increases.

CONCLUSION

In this study, an acid rain model is developed from the solution of mass balance equations for the gas-phase depletion and aqueous-phase accumulation of various species to estimate the rain acidity. To simplify the complex dynamic processes within cloud, the altitudinal variation of precipitation rate is utilized to obtain the model parameters of the rainfall characteristics.

The results from the developed model which has correlation coefficient of 0.57 between estimated pH values and observed ones in Seoul area of Korea during fall seasons are as follows:

1. As the developed acid rain model is expressed as a function of various pollutant concentrations and precipitation rate, it can be effectively utilized for the management of acid rain problems in the condition of limited data, which is a common problem outside the developed countries.

2. It is revealed from the sensitivity analyses that the raindrop pH is decreased as the concentrations of SO_2 and H_2O_2 increase, as precipitation rate decreases, and as rain growth zone increases. In addition, the effect of ozone concentration on rain pH shows much smaller than that of hydrogen peroxide.

For the further research, it is necessary to consider the effect of aerosols and cloud water droplets on the rain acidity.

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