THE IMPACT OF RELATIVE HUMIDITY ON THE RADIATIVE PROPERTY AND RADIATIVE FORCING OF SULFATE AEROSOL*

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ABSTRACT

With the data of complex refractive index of sulfate aerosol, the radiative properties of the aerosol under 8 relative humidity conditions are calculated in this paper. By using the concentration distribution from two CTM models and LASG GOALS/AGCM, the radiative forcing due to hygroscopic sulfate aerosol is simulated. The results show that: (1) With the increase of relative humidity, the mass extinction coefficiency factor decreases in the shortwave spectrum; single scattering albedo keeps unchanged except for a little increase in longwave spectrum, and asymmetry factor increases in whole spectrum. (2) Larger differences occur in radiative forcing simulated by using two CTM data, and the global mean forcing is -0.268 and -0.816 W/m^2, respectively. (3) When the impact of relative humidity on radiative property is taken into account, the distribution pattern of radiative forcing due to the wet particles is very similar to that of dry sulfate, but the forcing value decreases by 6%.

Key words: relative humidity; radiative property; radiative forcing

I. INTRODUCTION

Aerosols can exert an influence on the energy budget of atmosphere-earth system in both direct and indirect ways. Because aerosols can lead to a radiative forcing comparable in magnitude but of opposite to that of greenhouse gases, much attention has been paid to their climate effects in recent years (Houghton 1996). It should be noted that, the most of the studies focused on the radiative forcing under a dry atmosphere condition and did not deal with the impact of relative humidity (RH) on the radiative property of aerosols. However, the real atmosphere is not just an ideal "dry" one, meaning that the RH in the atmosphere is not always less than 50%. Therefore, it is very necessary to include the impact of RH on the radiative property and forcing of hygroscopic aerosols in the models.

Charlson et al. (1999) reviewed the extinction coefficients used in different studies in recent years and analyzed the impact of RH on the extinction coefficients. They found that there is a large difference in the values of extinction coefficient used by different studies. The normalized radiative forcings simulated by Haywood and Ramaswamy (1998), Penner et al. (1998) and Grant et al. (1999), are larger than those of other researchers by a

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factor of 2 or 3. These authors implied that once RH surpasses a critical value, the whole grid-box is covered by cloud, correspondingly, the radiative forcing will be enlarged due to the strong nonlinear RH effects. Chuang et al. (1997) made sensitivity experiments with and without cloud and got a radiative forcing less than those of the above studies. They suggested that the hygroscopic growth is obvious only when RH surpasses 99%. Generally, the use of the mean RH field will lead to smaller radiative forcing because without any consideration of the change of RH with time and the nonlinear effect of RH. If the dependence of aerosols on RH is improved, the normalized radiative forcing will increase (Kiehl and Briegleb 1993; Myhre et al. 1998). Haywood and Ramaswamy (1998) showed that the sulfate aerosol radiative forcing becomes large when the aerosol stays at the near surface, where the RH is larger and the hygroscopic growth is faster. Other sensitivity studies showed that the radiative forcing is a function of RH and is relatively insensitive to the chemical compositions of aerosol (Boucher and Anderson 1995; Nemesure et al. 1995). There is also a large difference of aerosol radiative forcing due to cloud effect on the global radiative forcing simulated by many researchers, the difference is about 4% (Haywood et al. 1997), 11% (Haywood and Ramaswamy 1998), 22% (Boucher and Anderson 1995) and 27% (Myhre et al. 1998).

Haywood et al. (1997) indicated that most studies use a single value of RH in each grid-box of GCMs to represent the properties over coarse ($>$ 10000 km^2) horizontal dimensions; no account is taken of the sub-grid scale in RH or of the effects of sub-grid scale cloud. These variations might have an important impact on the direct radiative forcing due to sulfate aerosol. Therefore, they believed that GCM calculations may considerably underestimate the direct radiative forcing. Ghan and Easter (1998), however, suggested that the direct radiative forcing due to sub-grid variations in relative humidity is overestimated by Haywood et al. (1997), and that treatment of sub-grid variations in RH may not be necessary for estimating direct radiative forcing. The property of solubility of aerosols is dominated by the aerosol activation and cloud condition, therefore, the estimation error of radiative forcing is, actually, not so large. If the contribution of those activated aerosol particles to radiative forcing is neglected, the direct radiative forcing from the cloud areas will be greatly reduced. Under some conditions, the calculated direct radiative forcing is less when the sub-grid variations in RH are correctly taken into account than that when a grid-box mean RH is used.

It is readily seen that, the impact of the RH on radiative property and radiative forcing of soluble sulfate is still a controversial problem and uncertainties still exist. In this paper, firstly, the impact of RH on radiative property of sulfate is accurately calculated, according to the original physical and chemical properties of aerosols; then the comparison simulation of radiative forcing due to sulfate was performed by GCM, and its input data come from the output of two CTMs.

II. THE DATA OF AEROSOL AND MODEL

The data used in this study include two parts: one is the concentration distribution in spatial and temporary dimensions, the other is the refractive index of aerosols within the whole solar spectrum (0.2—40 μm). The concentration data come from two sources: (1)

the output of the tropospheric chemical transport model GRANTOUR of LLNL (Lawrence Livemore National Laboratory) (Chuang et al. 1997) (C97 afterwards); and (2) the simulation results of the sulfur cycle model of MPI (Max Planck Institute) (Feichter et al. 1997) (F97 afterwards). The two models include the global monthly averaged natural sulfate and anthropogenic fossil fuel combustion sulfate concentration. The global sulfur emission amounts to 106.0 and 97.3 TgS per year, respectively, and of which 80% is attributed to anthropogenic emission.

The GCM model is LASG GOALS/AGCM (Global Ocean-Atmosphere-Land System) (Wu et al. 1997). It is a R15L9 model with a TOA (top of atmosphere) at 10 hPa. The vertical coordinate is η. The model includes all main physical processes and solar radiation diurnal cycle and an improved aerosol parameterization (Zhang 1999).

III. THE CHANGE OF THE RADIATIVE PROPERTY

Different from non-hygroscopic soot particles etc., sulfate particles are hygroscopic. Moreover, the real atmosphere is not so dry (RH not always less than 50%), and the global annually mean cloud fraction is about 60% (Charlson et al. 1991). Therefore, it is quite necessary to include the hygroscopic property of the sulfate aerosols in model.

With the increase of RH, the size of hygroscopic or dissoluble particles will enlarge. This enlargement, on the one hand, depends on the water vapor concentration in the atmosphere, on the other hand, depends also on the physical property and chemical composition of particles. When the particles interact with the air, they mix with water in the air. Sequentially, the size, size distribution, shape and chemical composition of the particles are changed with the RH. In this way, the refractive index and mode radii of the particles also change. It finally leads to the alteration of the radiative property of the particles. To most hygroscopic salt and salt mixture particles, the RH in which particles crystallize is usually less than 40%. Therefore, the lag phenomenon of the RH growth curve caused by the different dissolution and crystallization humidity is often neglected (Shaw and Rood 1990). In this paper, under the assumption of fixed standard deviation, the change of size distribution of the particles with RH is taken into account by altering the mode radii of the RH-dependable particles. The mode radii and densities under 8 RH conditions are listed in Table 1. It is not difficult to see that the radii of the particles have increased when RH reaches 50%. The RH of 0%, thereby, is a theoretical value, and is completely different from 30% in laboratory.

The refractive indices of dry particle is followed by Zhang and Shi (2000). Under different RH, the densities and refractive indices of the hygroscopic sulfate particles can be calculated according to the following equations:

$$\rho = \rho_{dry}\frac{r_{mdry}}{r_m^3} + \rho_{water}\frac{r_m^3 - r_{mdry}^3}{r_m^3}, \tag{1}$$

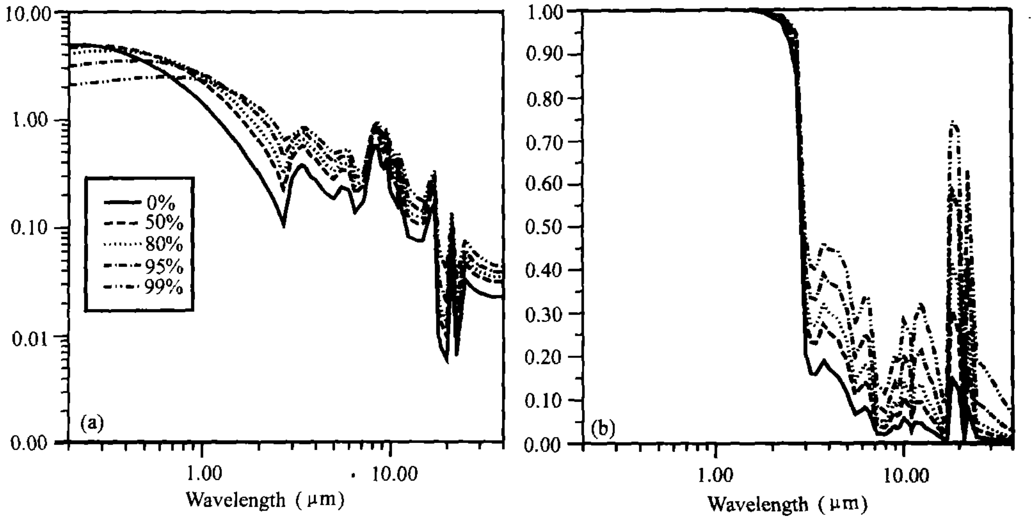
$$m^* = m^*_{water} + (m^*_{dry} - m^*_{water})\frac{r_{mdry}^3}{r_m^3}, \tag{2}$$

where ρ is the density of the particle, r_{mdry} is the mode radius of dry particle, r_m is the mode radius of wet particles, m^* is the refractive index of wet particle, and m^*_{water} is the refractive index of water from Liou (1992). The densities calculated under the different RH conditions are also shown in Table 1.

Table 1. The Change of Mode Radii and Densities of Sulfate Aerosol under 8 RH Classes

RH (%)	0	50	70	80	90	95	98	99
r_m (μm)	0.070	0.098	0.109	0.118	0.135	0.158	0.195	0.231
ρ (g/cm^3)	1.70	1.25	1.08	1.14	1.10	1.06	1.03	1.02

According to the above change of the density and the refractive index of the particles, the radiative properties under 8 RH classes are accurately calculated by using Mie algorithm (Wiscombe 1980). The size distribution of the particles is assumed to be log-normal distribution with mode radii in Table 1 and the same standard deviation of 2. The results are shown in Fig. 1 (For the sake of clarity, only the results under 5 RH classes are given in the figure).



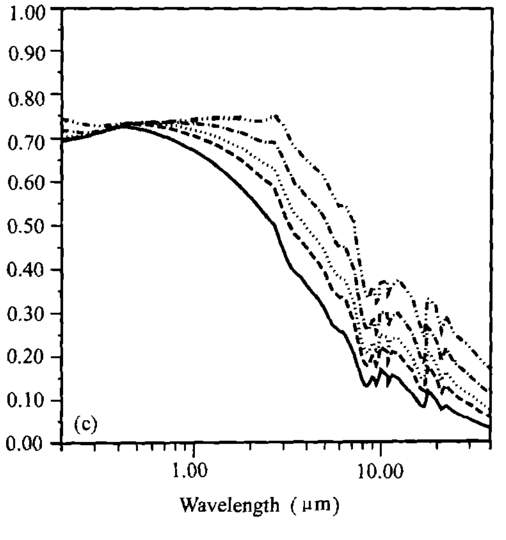


Fig. 1. The change of the mass extinction coefficiency factor (m^2/g) (a), single scattering albedo (b), and asymmetry factor (c) with wavelength under 5 RH conditions.

The mass extinction coefficiency factors decrease a little with increasing RH in the shortwave spectrum below 0. 7 μm, where the maximum scattering of the sulfate particles occurs (Fig. 1a). The largest decreasing value amounts to 2 m^2/g. It is easy to see that the maxima of the mass extinction coefficiency factors are inclined to the increasing wavelength. This also indicates that the scattering efficiency of the particles is the highest and the mass extinction coefficiency factor amounts to maximum when the radii of particles close to wavelength of solar radiation. The mass extinction coefficiency factors increase somewhat in the shortwave spectrum below 4 μm with the increasing of RH. If the interaction of aerosol particles with the water vapor in the atmosphere is taken into account, the mass of aerosol particles increases considerably. For RH of 99. 5%, the mass of the wet particles is a factor 30 — 40 higher than that of dry particles (Myhre et al. 1998). It can be induced that the increase of RH can influence the optical depth in two different ways through (1) modifying the mass extinction coefficiency factor by increasing the size of the particles; and (2) increasing the mass of the particles by enhancing the mixture of the particles with water vapor.

The single scattering albedo has almost no change in the shortwave less than 3 μm, but increases a little in the longwave spectrum (Fig. 1b). However, because of the low incoming solar radiation and low extinction efficiency in the longwave spectrum, there is minor influence of the aerosol on the solar radiation. It has been shown that the radiative forcing of aerosols in infrared spectrum only accounts for 3% of the total forcing (Haywood and Shine 1997).

The variation characteristics of the asymmetry factor are different from those of mass extinction coefficiency factor and single scattering albedo (Fig. 1c). There is a small change in shortwave spectrum. But it increases a little with the increase of RH in the whole spectrum. This implies that the backscattering of the particles decreases. The large particles can scatter the solar radiation into forward direction and lead to the decrease of the extinction efficiency of aerosol.

For the main sulfate compounds, such as H_2SO_4, $(NH_4)_2SO_4$, etc., some studies have shown that, under the typical RH condition in the troposphere (75% — 80%), the scattering energy per unit mass by these particles is twice as much as that of the corresponding dry particles (Charlson et al. 1991). According to the calculation in this work, however, the situation only occurs in some spectrum larger than 1 μm, if mass extinction coefficiency factor is concerned. But in ultraviolet spectrum, the extinction efficiency of the wet particles is less than that of the dry particles. The change of single scattering albedo and asymmetry factor is also very complicated. Obviously, the change of radiation property of the hygroscopic particles with RH can not be readily described by using a simple progressive relationship, which is the exact shortage of a box model. Therefore, it is very necessary to accurately recalculate the radiative property due to the hygroscopic characteristic of sulfate particles.

IV. THE GEOGRAPHICAL DISTRIBUTION AND VARIATION CHARACTERISTICS OF THE RADIATIVE FORCING

Five experiments are designed in this study, including three for C97 data (natural sulfate, total sulfate and the external mixture of total sulfate with soot), and two for F97 data (natural sulfate and total sulfate). All experiments are integrated for two years to simulate the seasonal variation and global mean characteristic of the radiative forcing due to these aerosols.

Since soot particles are non-hygroscopic, only their external mixture with sulfate particles is taken into account (Zhang 1999). The interaction process of the sulfate particles with water vapor and cloud in the atmosphere is also neglected in this study.

The seasonal distribution pattern and magnitude of the radiative forcing due to C97 natural sulfate is almost the same as those of the dry particles (not shown), but with a tiny difference. Although there is no clear variation in continent areas, a smaller decrease occurs in the North Pacific in summer.

Compared with the radiative forcing due to dry sulfate particles (Zhang and Shi 2001), the range of some isopleths of the radiative forcing due to wet particles decreases a little in Southern hemisphere in winter. A small difference occurs in the oceans and the

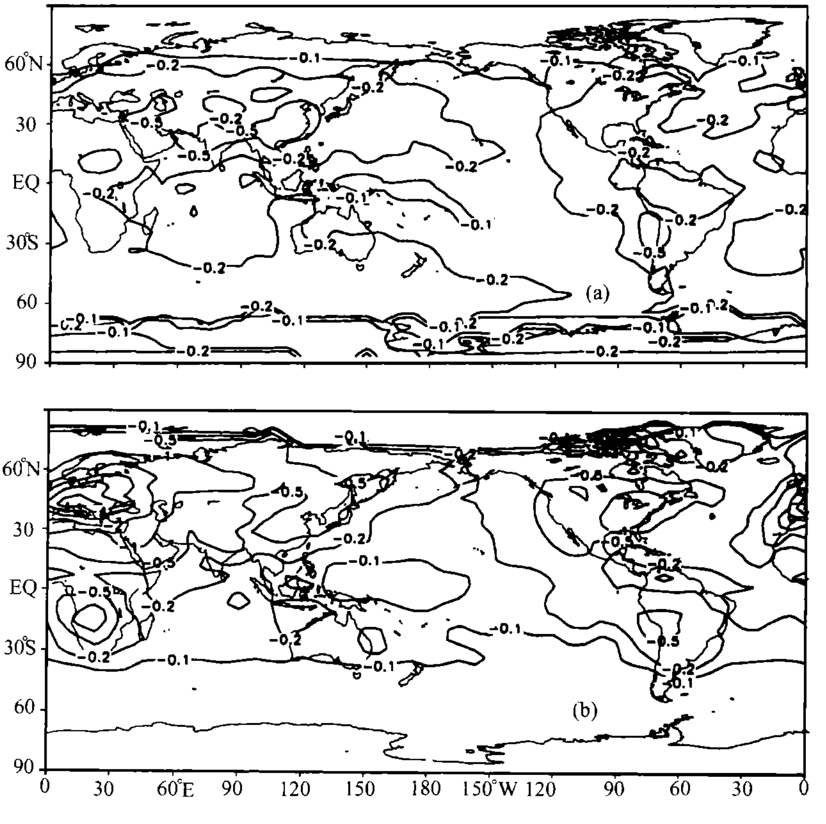


Fig. 2. C97 seasonal averaged radiative forcing due to total sulfate (W/m^2)
(a) winter; (b) summer.

intersection areas of the oceans and the lands in summer. However, the bulk trend of change is decreasing. It is obvious that the maximal centers are concentrated on the continents in both hemispheres, where the human activities are very serious. The largest forcing occurs in Western Europe area with the value of $-5\ W/m^2$ in summer.

The annual mean radiative forcing due to three experiments in C97 is given in Fig. 3. Comparing with that of dry particles, the forcing due to hygroscopic natural and total sulfate almost has no clear variation (Figs. 3a—3b). For the radiative forcing due to the external mixture of total sulfate and soot (Fig. 3c), it is obvious that the $-0.1\ W/m^2$ isopleth in equatorial Pacific Ocean area enlarges a little in winter but reduces in some extent in summer.

For the radiative forcing due to F97 natural sulfate and total sulfate (Fig. 4), the variation of distribution pattern of the annual mean radiative forcing in oceanic area is relatively obvious. But the variation in continent areas is very small. The largest value of the annual mean radiative forcing occurs in Eastern Asia with a value of $-3\ W/m^2$.

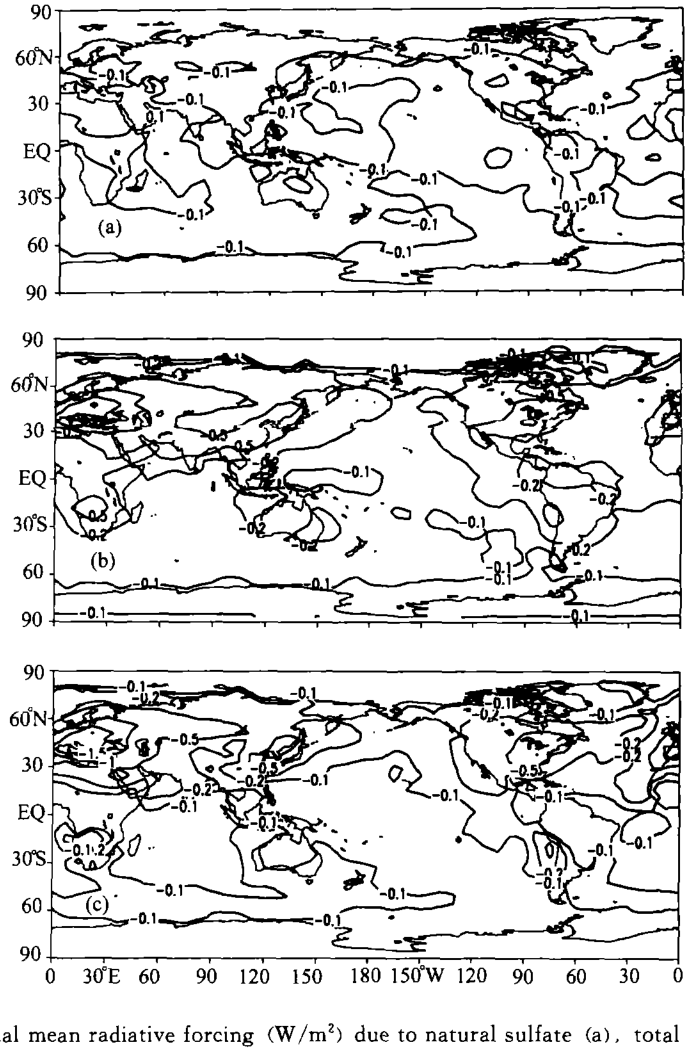


Fig. 3. C97 annual mean radiative forcing (W/m^2) due to natural sulfate (a), total sulfate (b), and the external mixture of total sulfate and soot (c).

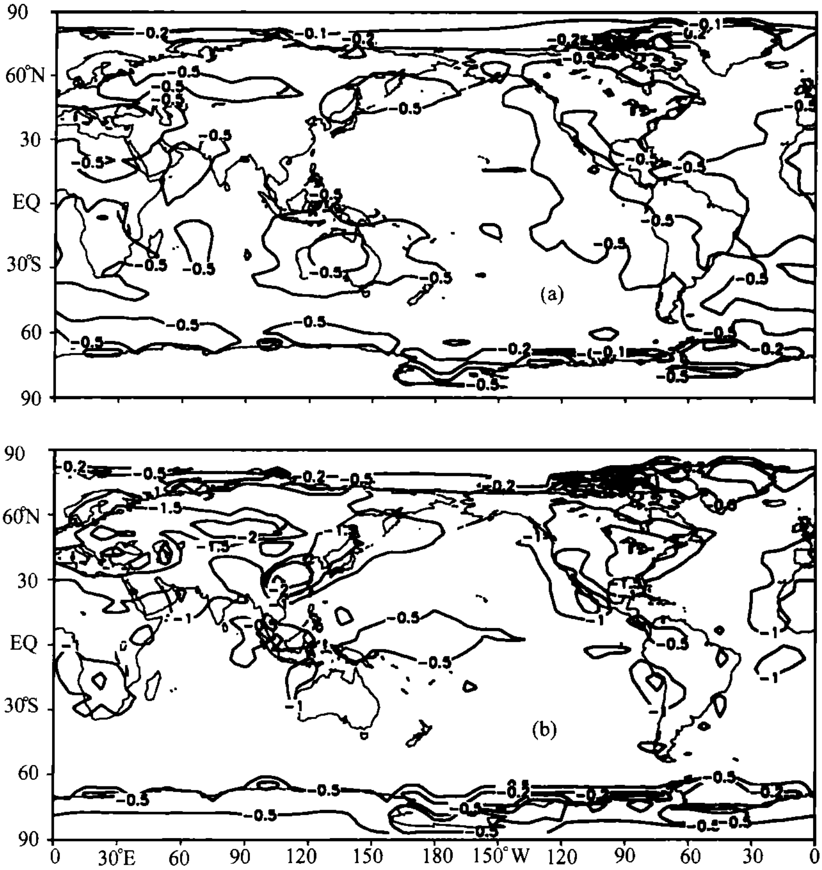


Fig. 4. F97 annual mean radiative forcing (W/m^2) due to (a) natural sulfate, and (b) total sulfate.

It is a little difficult to figure directly out the bulk change trend from the figures of the global monthly mean variation of the forcing (not shown). Therefore, the global annual mean radiative forcing of all experiment is also listed in Table 2. Compared with that of dry particles, it can be seen that the radiative forcing due to wet sulfate particles decreases a little to different extent. The maximal decrease rate is about 6% (F97 total sulfate experiment), and the minimal one is about 1% (C97 external mixture of total sulfate with soot).

Table 2. The Global Mean Radiative Forcing of All C97 and F97 Experiments (W/m^2)

Data	C97			F97	
Experiments	Natural sulfate	Total sulfate	External mixture	Natural sulfate	Total sulfate
Radiative Forcing	−0.106 (−0.11)	−0.268 (−0.28)	−0.174 (−0.18)	−0.47 (−0.50)	−0.816 (−0.87)

Note: The radiative forcing in the parentheses is from Zhang (1999) and Zhang and Shi (2001).

It should be emphasized here that our results are completely different from those of other studies. In the earliest simple estimation by a box model, Charlson et al. (1992) suggested that, the size of particles would increase with the increasing RH and larger particles can scatter more solar radiation. So they simply represented the impact of RH on radiative forcing, f (RH), by multiplying a factor of 1.7 to that of dry particles. Actually, only the impact of single scattering albedo and asymmetry factor were taken into account in the studies like this, so they magnified the influence of RH on the radiative forcing. Later, Haywood et al. (1997) simulated by using Hadler Centre GCM and showed that the radiative forcing due to wet particles is 1.6 times of that of the dry particles. They suggested that with the increase of RH, the mass extinction coefficiency factor of sulfate particles increases in the whole spectrum except for the ultra spectrum, and the asymmetry factor also has a trend of increase. Our calculation results in the impact of RH on radiative property are almost the same as that of Myhre et al. (1998). Their sensitivity experiment showed, however, that the factor f(RH), was reduced to 1.2 when the impact of RH was taken into account. In the meantime, they suggested that with the increase of RH the radiative forcing probably increases.

However, from Fig. 1, it can be seen that with the increase of RH, although the mass extinction coefficiency factor increases somewhat in most parts of the spectrum, it decreases in shortwave spectrum where the solar energy concentrates on. Meanwhile, the asymmetry factor increases in the whole spectrum, which means that the backscattering of the particles reduces. Under the condition of almost unchanged single scattering albedo, the radiative forcing caused by the change of these properties will reduce. Therefore, the results in this study show the decreasing trend of the radiative forcing due to the wet particles. Haywood et al. (1997) concluded that, with the increase of RH, the increasing impact of the asymmetry factor is much less than that of the mass extinction coefficiency factor. It should be noted that, as one of three basic radiative property factors, the asymmetry factor plays the equal important role in determining the radiative forcing due to aerosols. Even though the extinction coefficiency factor increases, the increase of the asymmetry factor can also lead to the reduction of the radiative forcing. Some our sensitivity experiments of a single air column model also indicated that, although the mass extinction coefficiency factor increases a little, it could not still cover the effect caused by the increasing asymmetry factor.

Nevertheless, there exist many uncertainties in simulating the impact of RH on the radiative forcing due to the hygroscopic aerosols. At present, it is still a controversy problem. When GCMs are used to simulate the impact of aerosols on climate, because of the limitation of CPU time, it is almost impossible to couple enough accurate radiative properties into the models. So it is inevitable that there are some biases between different results. Additionally, because the aerosol particle can act as CCN and has impact on the water vapor and cloud in atmosphere, it also influences the result a little. The interaction of aerosols and clouds (indirect effect) should be properly coupled into the models in the future.

V. RESULTS AND DISCUSSIONS

Because the radiative property and radiative forcing due to hygroscopic sulfate aerosols change with the increasing RH, therefore, the radiative properties of the sulfate aerosol under 8 RH conditions are first calculated in this study. Then these radiative properties are coupled into the GOALS/AGCM. Five experiments are designed and the change of the global radiative forcing due to sulfate aerosol is simulated finally.

Our results are completely different from those of other studies, namely, the global mean radiative forcing decreases a little, when the impact of RH is taken into account. It can be seen from Section III in this paper that, with the increase of RH, the mass extinction coefficiency factor increases somewhat in most regions of the spectrum, and decreases a little in shortwave spectrum; the change of single scattering albedo is not so obvious; in the meantime, the asymmetry factor increases a little, meaning that the backscattering ability of the particles reduces. In a word, the bulk effect of these changes is the reduction of the radiative forcing. It should be mentioned that, compared with other factors, the asymmetry factor also plays a very important role in estimating of the radiative forcing due to aerosols.

The simulation results show that the radiative forcing due to hygroscopic particles is insensitive to the impact of change of RH when the interaction of aerosols with cloud water is neglected. Comparied with the radiative forcing due to dry particles, the maximum reduction rate of the radiative forcing due to corresponding hygroscopic particles is about 6%. It can be referred, to some extent, that the box model exaggerated the effect of RH on the radiative forcing due to hygroscopic aerosol particles.

Some other studies also showed that when the optical depth of cloud is consideranbly larger than that of aerosols, the radiative forcing due to the aerosols is relatively smaller and the radiative forcing is not sensitive to the relative height of the cloud and aerosols (Haywood and Shine 1997; Liao and Seinfeld 1998). However, a competing effect that acts to weaken the radiative forcing is the co-existence of clouds and aerosols of high RH. Generally, cloudy area will produce a radiative forcing that is considerably less than in clear skies (Charlson et al. 1991; Boucher and Anderson 1995; Haywood et al. 1997); the magnitude of the decrease is dependent primarily on the optical depth of the cloud. Since clouds exist in regions of high RH, the strong radiative forcing at the high RH areas is reduced, thereby weakening the overall radiative forcing. The radiative forcing is reduced by over 50% at RH close to 100% but the reduction in the radiative forcing at RH of less than 90% is approximately 5%—20%. The radiative forcing weakens most in areas of high RH due to the co-existence of cloud and aerosols there. It can be seen from our results that, comparing with those of the oceanic areas, the cloud cover is relatively less in continent areas, so the change of radiative forcing in oceanic areas is relatively clear and also reasonable.

As implied by Haywood et al. (1997), because there are substantial spatial and temporal variations in RH, there are some problems in simulation of the radiative forcing due to aerosols by using GCMs with coarse resolution of more than several hundreds or a thousand kilometers. Further investigations of the variation from RH and cloud in global scale should be made later.

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