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NOTES AND CORRESPONDENCE

Using Sub-Grid Scale Method to Quantify Atmospheric Deposition of Sulfur in East China

Yan Zhang[1], Ti-Jian Wang [2, *], and Li-Min Chen[1]

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ABSTRACT

Atmospheric sulfur deposition in East China was studied using the Regional Acid Deposition Modeling System (RegADMS) with a horizontal resolution of 75 km. A one-year run was performed for the year 2000. As a result, the annually averaged V_d for SO_2 and SO_4^{2-} are 0.34 and 0.27 cm s^{-1}, respectively. The calculated V_d for SO_2 and SO_4^{2-} at Yingtan agreed well with the semi-measurements. And the modeled V_d over different land-use types were generally in agreement with those reported from the references. The sub-grid scale (25 × 25 km) method (SSM) was proven to better describe the dry deposition process and save computational time when estimating dry deposition velocity (V_d) than the coarse grid scale (75 × 75 km) method. The estimated total deposition of sulfur in East China is 2.22 × 10^6 ton yr^{-1}. Dry deposition accounts for almost 55% of the total deposition and more than 90% of sulfur was deposited on the soil-plant ecosystem. There was evident regional distribution of sulfur depositions in the provinces of the studied area, with most values more than 3 ~ 5 g m^{-2} yr^{-1}.

(Key words: Sub-Grid scale method, Sulfur, Dry deposition velocity, East China)

1. INTRODUCTION

In the atmosphere, gaseous SO_2, sulfate in aerosols and SO_4^{2-} ions in precipitation are the dominant sulfur-containing chemical species. They can be transported to the ground through

[1] Department of Environmental Science and Engineering, Fudan University, Shanghai, China

[2] Department of Atmospheric science, Nanjing University, Nanjing, China

* *Corresponding author address:* Prof. Ti-Jian Wang, Department of Atmospheric science, Nanjing University, Nanjing, China; E-mail: tjwang@nju.edu.cn

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both wet and dry deposition processes. In recent years, a good deal of research has been conducted on the regional deposition flux of SO_2 and sulfate aerosols because of their importance in ecosystems (Rodhe et al. 1995; Gorham 1998; Hu and Shen 1998). Wet deposition flux can be readily measured directly; however, field measurement of dry deposition flux is more difficult. Resistance models have proven to be a better way of calculating dry deposition velocity (V_d) (Pleim et al. 1984; Meyers et al. 1991; Harley et al. 1993; Padro 1996; Pratt et al. 1996; Kharchenko 1997; Wang 1998). Previous investigations have shown that V_d exhibits considerable variability (Stocker et al. 1995; Kruger and Tuovinen 1997; Wyers and Duyzer 1997; Xu and Carmichael 1998; Zhang and Brook 2001; Zhang et al. 2002) and is generally dependent upon meteorological conditions and surface characteristics in the regions under consideration.

Several kinds of techniques have been established to measure dry deposition velocity, in which, micrometeorological methods, including the gradient method, eddy correlation method and others, have been widely used. Also, many numerical models have been developed to estimate dry deposition velocity, such as the two-layer model, three-layer model and multi-layer model. Correspondingly, these V_d modules have been coupled in many long range transport models and regional acid deposition models (e.g., ADOM, RADM, EDACS, AURAMS, etc.) to estimate dry deposition flux of pollutants (Chang et al. 1987). In some of these models, the sub-grid scale method has been applied in dry deposition process modeling to improve the precision of V_d estimation over a large domain with surface ununiformity.

China has suffered a lot from excessive acid deposition regionally. South and South-west China have been recognized as regions exposed to heavy acid rain over long periods (Wang 1994; Huang et al. 1995; Zhou 1996; Seip et al. 1999). When compared with America and Europe, acid rain in China tends to be typically sulfate derived (Lin et al. 2000; Wang et al. 2000) with higher concentrations of SO_4^{2-} and lower concentrations of NO_3^-. Moreover, in the past, due to the rapid pace of industrial development and increased energy demand, consumption of sulfur-containing fuels has soared, resulting in large amounts of SO_2 emission in Eastern China. Although acid rain has been reported in this region (Mao and Li 1997; Li et al. 2000), few investigations of regional sulfur deposition in Eastern China have been conducted.

In this paper, the Regional Acid Deposition Modeling System (RegADMS) is applied to estimate SO_2 and SO_4^{2-} dry and wet deposition flux with an evaluation given on model improvements for the sub-grid scale method. The regional distribution of sulfur deposition in Eastern China is then presented.

2. METHODOLOGY

2.1 Regional Acid Deposition Modeling System (RegADMS)

RegADMS consists of a mesoscale meteorological model and an acid deposition model. The mesoscale model MM5 (Grell et al. 1994) developed at Pennsylvania State University and the National Center for Atmospheric Research was selected. MM5 has been widely used in meteorology and air pollution modeling on a regional scale. In this work, the model domain was designated as an area of 1800 × 1500 km, covering Eastern China and its surroundings, the center point of the domain is at 30°40'N, 116°24'E. A horizontal resolution of 75 km with sub-grid length of 25 km was selected and 10 altitude levels chosen. The air pressure at the top of the model space is 100 hPa. Blackadar's high resolution boundary layer scheme, time-dependent boundary condition and deep cumulus convection parameterization were selected for a one-year simulation. The output of MM5 was used to drive the acid deposition model.

The acid deposition model was developed by Wang et al. (1996, 2000); it is a three dimensional time-dependent Eulerian model and includes the processes of emission, transport, deposition and transformation of acidic compounds that can result in regional air pollution and acid deposition. The output of RegADMS gives atmospheric concentrations of SO_2 and sulfate (SO_4^{2-}) as well as total sulfur deposition.

2.1.1 Dry Deposition Module

In RegADMS, the dry deposition module is applied to compute dry deposition velocity by use of meteorological and land-use data. This module is based on the work of Walcek (1986), which uses a look-up table approach to give bulk surface resistance for uptaking gases.

The general formula for V_{dg} (V_d of gases) is:

$$V_{dg} = \frac{1}{R_a + R_b + R_c} \quad , \tag{1}$$

Where, R_a is the resistance caused by turbulence when gas is transported from a reference height to the ground; R_b is the resistance when the deposited gas passes through the quasi-laminar layer; R_c is the underlying surface resistance.

The formula for V_{da} (V_d of particles) is:

$$V_d = \frac{1}{R_a + R_b + V_g R_a R_b} + V_g \quad , \tag{2}$$

Where, R_a and R_b are the same as in Eq. 1, and V_g is the gravitational settling velocity, which can be calculated by the Stokes formula. R_a and R_b in Eqs. 1 and 2 can be calculated according to Wesely and Hicks (1977) and Wesely (1985) Considering the vertical distribution of the canopy and ground system, R_c consists of several components (Wesely 1989; Walmsley and Wesely 1996). It can be calculated as:

$$R_c = \frac{1}{\frac{1}{r_s + r_m} + \frac{1}{r_{lu}} + \frac{1}{r_{dc} + r_{cl}} + \frac{1}{r_{ac} + r_{gs}}} \quad , \tag{3}$$

Where, r_s, r_m and r_{lu} relate to the properties of high vegetation, r_s represents the surface bulk resistance component for stomata, r_m for the leaf mensophyll resistance, r_{lu} for the cuticles of living leaves in vegetation or the outer surfaces of the upper canopy; r_{dc} and r_{cl} relate to the lower canopy, r_{dc} for the gas-phase transfer resistance brought by buoyant convection in canopies, r_{cl} for leaves, twigs, barks, and other exposed surfaces in the lower canopy; and r_{ac} and r_{gs} relate to the ground, r_{ac} for the transfer that depends only on canopy height and density and r_{gs} for the soil, leaf litter, etc. at the ground surface.

2.1.2 Parameterization of Deposition Flux

Total deposition flux F_t is the sum of dry and wet deposition fluxes:

$$F_t(Z) = F_d(Z) + F_w(Z) \quad . \tag{4}$$

The dry fluxes $F_d(Z)$ are obtained from the vertical concentrations of pollutants, as well as their dry deposition velocities, which can be written as:

$$F_d(z) = V_{dg}(z) \times C_g(z) + V_{da}(z) \times C_a(z) \quad , \tag{5}$$

Where, z stand for the vertical direction, $C_g(z)$ and $C_a(z)$ are gas and aerosol concentrations at height level z. In this work, they mean the concentrations of SO_2 and SO_4^{2-}, respectively. $V_{dg}(z)$ and $V_{da}(z)$ are dry deposition velocities of SO_2 and SO_4^{2-} at the specific height level. The wet deposition flux (F_w) is got from the wet-scavenging coefficient and concentration of pollutants (Wang et al. 1996).

2.2 Sub-Grid Scale Method for Dry Deposition Velocities Estimation

Generally, dry deposition velocities in the early regional models have been assumed to remain universally constant in a grid, regardless of temporal changes and surface difference. However, many measurements have shown considerable variability of V_d depending on the type of surface and local meteorological conditions. If more than one pattern of land surface is included in one grid, the overall dry deposition velocity derived from the single predominated surface pattern may not represent the real deposition process well; thus, horizontal grid length has to be chosen as fine as necessary to depict the spatial distribution of surface types and meteorological conditions. This method is very time consuming. A more detailed distribution of land-use types can be included in a grid using a sub-grid scale method, by which dry deposition velocity can be estimated more realistically.

Here, three different methods are listed to estimate overall dry deposition velocity.

Coarse grid scale method (CSM): 75 × 75 km meteorological data derived from MM5 serve as input to RegADMS to calculate 75 × 75 km dry deposition velocities.

Sub-grid scale method (SSM): first, 75 × 75 km meteorological data is calculated by MM5 to estimate 25 × 25 km meteorological data in sub-grids, then 25 × 25 km dry deposition velocities are determined.

Fine grid scale methods (FSM): 25 × 25 km meteorological data are directly derived from MM5 (that needs very long computing time); thus, 25 × 25 km dry deposition velocities are determined.

Observations have shown that wind speed is smaller over rougher surfaces and larger over smoother surfaces within an averaging area (a model grid here), so Walcek (1986) assumed that the product of wind speed and friction velocity is approximately constant among varying surfaces or land-use type within a coarse grid. And based on the hypothesis (Walcek 1986), the parameters corresponding to the SSM are as below:

$$uu_* = u_i u_{*i} = \text{const} \quad , \tag{8}$$

Where, u and u_* refer to grid-averaged wind speed and friction velocity (from MM5), while u_i and u_{*i} refer to the corresponding values over individual land types within the averaging area. When the sub scale method was applied in this work, each 75 × 75 km grid (the averaging area) was divided into nine 25 × 25 km grids (sub grid areas). In this way, wind speed and friction velocity can be determined for each land type within a grid square if this constant is known. The grid-average friction velocity can be determined from grid-averaged Z_0:

$$Z_0 = \exp\left[\sum f_i \ln z_{0i}\right] \quad , \tag{9}$$

Where, f_i is the area fraction of land-use apportioned within the grid cell and Z_{0i} is the surface roughness for each land-use type. Land-use information of 13 categories is available at 2 minutes (about 3.7 km) resolution and thus can be obtained for both the RegADMS grid area (75 km) and its sub grid area (25 km).

By using Eq. 9 to determine the average surface roughness together with Eq. 8, wind speed over different land-use types can be predicted as:

$$u_i = u\left\{\left[\log\left(\frac{z_r}{z_{0i}}\right) - \Phi\right] \Big/ \left[\log\left(\frac{z_r}{z_0}\right) - \Phi\right]\right\}^{\frac{1}{2}} \quad , \tag{10}$$

$$u_{*i} = 0.4u_i \Big/ \left[\log\left(\frac{z_r}{z_{0i}}\right) - \Phi\right] \quad , \tag{11}$$

Where, Z_r is the reference height and Φ is a non-dimension stability function. Aerodynamic as well as quasi-laminar layer resistances were estimated, and V_{di} (V_d in each land area in sub grid) was then computed over each land area before computing the area-average deposition velocity. Thus, the grid-average deposition velocity V_d can be written as:

$$V_d = \sum f_i \, V_{di} \quad . \tag{12}$$

Meteorological data in 75- and 25-km scale were obtained from MM5. They were then input into the acid deposition model. The land-use data were from USGS.

3. RESULT AND DISCUSSIONS

3.1 Comparison of Dry Deposition Velocity with Measurement

To verify the reliability of calculated V_d by FSM, field measurements were performed at Yingtan (YT), Jiangxi province from 1999 to 2000 (Wang et al. 2003b; Zhang et al. 2004). An observation tower was established in farmland, where automatic sensors were installed at 5, 3.5, 2.0, and 1.0 m above the ground to measure wind speed/direction, air temperature, air pressure, and humidity. Similar microclimatic observations have been conducted in forest at 13.5, 8.0, 3.0, and 2.0 m. The observed meteorological data were input into the dry deposition module (Eqs. 1 - 3) to estimate V_d, which are called the semi-observed values.

The monthly averaged dry deposition velocities of SO_2 and SO_4^{2-} by RegADMS with FSM were chosen from Grid YT site and compared to the semi-observed ones (averaged value of V_d over farmland and forest) at YT site. The results are illustrated in Fig. 1. It shows that the

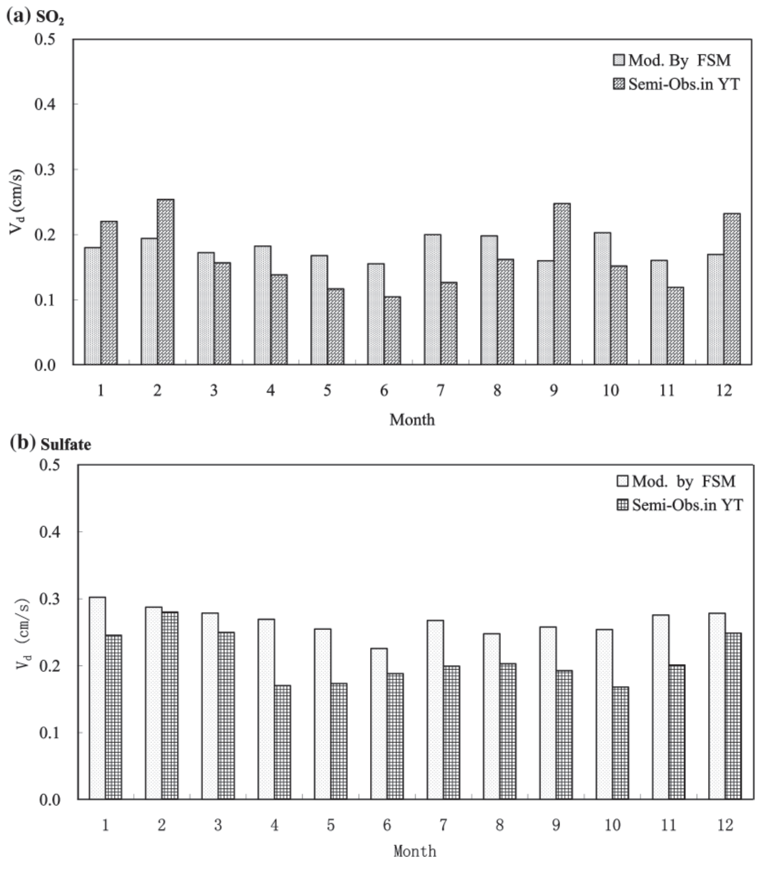

Fig. 1. Comparison of V_d for SO_2 (a) and Sulfate (b) modeled by FSM and the semi-observed V_d in Yingtan site.

simulated and observed V_d agreed well, except in summer. The semi-observed V_d may be influenced by seasonal variation due to crop planting and other land-use types near Yingtan station. However, the simulated V_d was a regionally averaged value for a single land-use type. This may cause some discrepancy between the two kinds of V_ds.

The annually averaged dry deposition velocities of SO_2 and SO_4^{2-} from FSM in different land-use types of Eastern China were also compared with other research results shown in Table 1 (Hicks and Liss 1976; Davies and Mitchell 1983; Sievering 1983; Wesely 1985; Hicks et al. 1989; Fan 1993; Erisman 1994; Granat and Richter 1995; Gallagher et al. 1997). For SO_2, V_d above water surface is 0.66 cm s^{-1}, which is larger than those above other land-use types. But it shows no significant difference in the V_d of SO_4^{2-}, since surface conditions have smaller impact on particles than on gases. The annual averaged V_d of SO_2 and SO_4^{2-} in the Eastern China region are 0.34 and 0.27 cm s^{-1}, respectively. V_ds of SO_2 and SO_4^{2-} above different land-use types by FSM were in good agreement with these measurements. These comparisons indicate that V_d estimated from RegADMS with FSM is reasonable.

In this comparison, FSM is considered the standard method because of its performance evaluation mentioned earlier. Dry deposition velocities obtained by CSM and SSM in all model grids were compared to evaluate the efficiency of SSM (Wang et al. 2006). The results for SO_2 and sulfate are shown in Table 2.

As shown in Table 2, the root-mean-square of error value is small because V_d is small in itself. But obviously, the value 3.44×10^{-3} cm s^{-1} from CSM is larger than that from SSM, which is 2.85×10^{-3} cm s^{-1}, and their ratio is near 1.21. SSM is effective (25-km sub-grid

Table 1. Dry deposition velocities of SO_2 and SO_4^{2-} by FSM (cm s^{-1}) and comparisons with the other research (cm s^{-1}).

Species	Surface	Review of Measured dry deposition velocities				V_d from FSM
		Method	Species	V_d (cm s^{-1})	Reference	Annual average
SO_2	Farmland	Gradient	SO_2	0.16 to 2.39	Fan (1993)	0.21 ± 0.007
	Water	/	SO_2	1.00 to 2.00	Hicks and Liss (1976)	0.66 ± 0.003
	Grass	Gradient	SO_2	0.04 to 3.40	Davies and Mitchell (1983)	0.36 ± 0.003
	Coni. forest	/	SO_2	0.10 to 0.30	Erisman (1994)	0.15 ± 0.002
	Tropical forest	/	SO_2	0.33	Granat and Richter (1995)	0.32 ± 0.004
	Average	/	/	/	/	0.34 ±0.004
Sulfate	Farmland	Eddy correlation	sulfate	0.09 to 0.7	Sievering (1983)	0.27 ± 0.005
	Coniferous forest	Eddy correlation	sulfate	0.02 to 1.80	Gallagher et al (1997)	0.30 ± 0.006
	Deciduous forest	/	sulfate	0.00 to 1.30	Hicks et al. (1989)	0.32 ± 0.007
	Grass	Gradient	sulfate	-0.33 to 0.57	Wesely (1985)	0.24 ± 0.009
	Average	/	/	/	/	0.27 ± 0.006

Table 2. Root-mean-square of error [a] between dry deposition velocities by CSM and SSM taking FSM as the standard, respectively (10^{-3} cm s^{-1}) and computational costs.

Methods \ Substance	SO_2	SO_4^{2-}	Averag e value	Computing Cost for pre-processors [c]	Computing Cost for dry deposition [d]
CSM[b]	4.38	2.49	3.44	T_1	T_2
SSM	3.63	2.08	2.85	T_1	$1.5T_2 \sim 2T_2$
Ratio of CSM to SSM	1.21	1.20	1.21	1.0	0.5 ~ 0.7
FSM	/	/	/	$6T_1$	$9T_2$

a: Root-mean-square of error for $\mathrm{CSM} = \left[\Sigma(\mathrm{V}_{\mathrm{di\ CSM}} - \mathrm{V}_{\mathrm{di\ FSM}})^2\right]^{1/2} \Big/ \text{the total number of grids}$; That is similar for SSM but for $V_{di\ CSM}$.

b: Considering the V_{di} in 25-km sub-grid equal the average value in corresponding coarse grid ($V_{di} = V_d$ in one coarse grid).

c: Computing cost here means total CPU time consumed in computing full-year meteorological conditions needed by CSM and SSM (20 × 24 grids) and FSM (60 × 72 grids) on a single-cpu PC platform, T_1 means about 100 ~ 150 hours of CPU time.

d: Computing cost here means total CPU time consumed in computing full-year dry deposition by CSM and SSM (20 × 24 grids) and FSM (60 × 72 grids) on a single-cpu PC platform, T_2 means about 4 ~ 6 hours of CPU time.

scale) in calculating V_d instead of CSM (75-km grid scale). More precise V_d will lead to further precise pollutant concentrations and deposition fluxes. The computational costs are also listed in Table 2. The differences in computing cost for SSM and FSM include two parts, which were cost by their corresponding pre-processors (meteorological conditions) based on different model grids and dry-deposition calculations using FSM and SSM. Combining pre-processor and dry deposition computations, SSM cost almost the same duration (about 106 ~ 165 CPU hours) in modeling a more precise result than CSM (about 104 ~ 156 CPU hours), and saved more than 500% of the computational cost compared to FSM. Moreover, when the sub-grid scale is divided into a smaller one such as 5 × 5 km, the computational cost saving is conceivably much more.

Furthermore, a typical coarse grid including different land-use types was chosen to better illustrate the variation of V_d due to the application of the sub-grid scale method (Wang et al. 2006). The grid is located in grid point (16, 2) of the domain and its land-use type is coniferous forest; sub-grid land-use types are coniferous forest (45%), deciduous forest (22%), tropical forest (12%), and water (11%). V_ds of SO_2 and SO_4^{2-} in CSM and SSM were compared and the result is shown in Fig. 2. With the transformation of land-use types, V_d of SO_4^{2-} from SSM decreases because the turbulence is weak against the deposition process above water, which brings great turbulent resistance. But for SO_2 the surface resistances above water and tropical forest are both much smaller and as it dominates compared with the turbulent resistance added

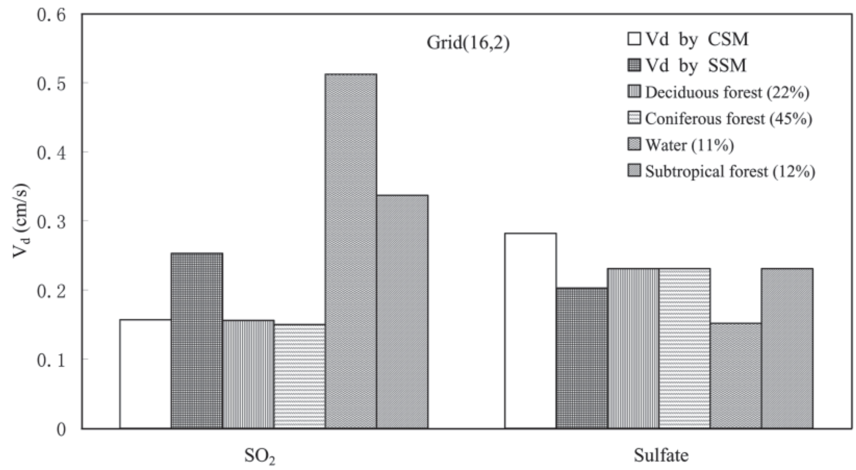

Fig. 2. Comparison of V_d for SO_2 and Sulfate by CSM and SSM in representative grid (16, 2).

by water the overall resistance in the deposition process decreases and V_d is 60% larger than one in coarse grid. Based on these findings, SSM can replace traditional CSM effectively; meanwhile, its precision is close to FSM, and can significantly save computing time compared with FSM.

3.3 Regional Deposition of Sulfur in Eastern China

The distribution of annual sulfur deposition in Eastern China in 2000 was simulated by RegADMS. The emission inventory used contains anthoropogenic emissions from area sources and point sources, which are based on the East Asia emission inventory and are partly improved (Wang et al. 2003a). RegADMS with CSM and SSM were applied to model sulfur deposition. Dry deposition over different land-use types by CSM and SSM is listed in Table 3. For sulfur dry deposition, more than 90% of sulfur deposits on soil-plant ecosystems and plays a key role in the sulfur cycle. Total sulfur deposition by SSM in Eastern China was 2.22×10^6 ton yr^{-1}, 55% of which was dry deposition (1.18 ton yr^{-1}). For most land-use types, except for water surfaces, sulfur dry depositions by SSM were larger than the ones by CSM, by about 1.5 times. Also the gap between dry sulfur deposition fluxes by CSM and SSM in Fig. 3 shows evident regional discrepancy distribution with -0.10 to1.09 g S m^{-2} yr^{-1} and relative change ratio of -9% to 57% statistically. It is noteworthy that to get highly distinguished regional distribution in the focus domain, sulfur depositions were presented in just six provinces including: Shandong, Anhui, Jiangsu, Zhejiang, Jiangxi, and Shanghai city as Eastern China in a narrow sense. It shows that SSM performed efficiently in estimating dry deposition with higher-resolution abilities in land-use types.

Table 3. Sulfur dry deposition to land-use types in Eastern China from CSM and SSM (10^6 ton yr^{-1}).

Surface	CSM	SSM	Ratio
Agriculture land	0.566	0.885	1.6
Coniferous forest	0.098	0.110	1.1
Water body	0.101	0.090	0.9
Tropical forest	0.016	0.021	1.3
Savannah	0.029	0.037	1.3
Total	0.810	1.176	1.5

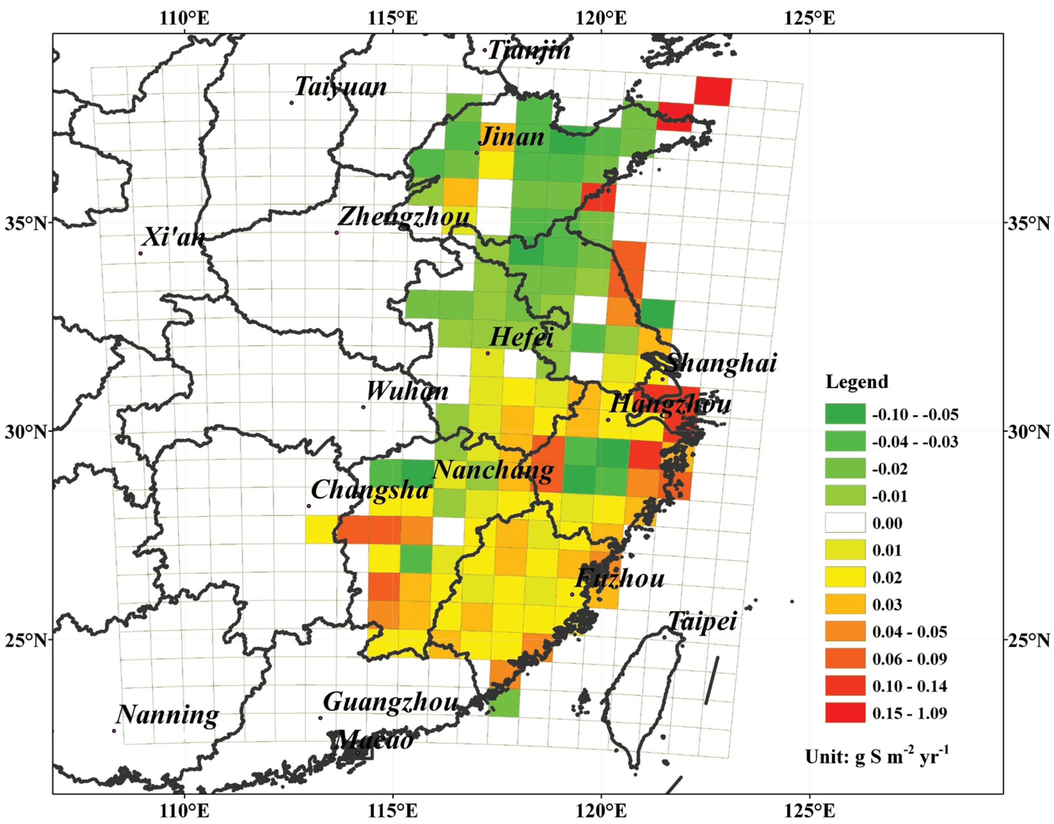

Fig. 3. Discrepancy in Sulfur deposition fluxes by CSM and SSM in Eastern China (g S m^{-2} yr^{-1}).

In addition, regional distribution of total sulfur deposition by SSM is shown in Fig. 4. The sulfur deposition flux is greatest in Shandong province, reaching 5.0 g m^{-2} yr^{-1} in the middle of the region. The modeled of sulfur deposition in Anhui and Jiangsu provinces exceeded 4.0 g m^{-2} yr^{-1} whilst in Fujian and Zhejiang province, they are smallest at less than 3.0 g m^{-2} yr^{-1}. On the whole, the estimated sulfur deposition fluxes in Eastern China are less than observations at Chongqing site in South-Western China, where sulfur deposition was recorded at about 10 g m^{-2} yr^{-1} (Jin et al. 2006; Wang et al. 2006). However, compared with the sulfur deposition in 1990, about 1 ~ 3 g m^{-2} yr^{-1} in Eastern China (Hao et al. 1996), a significant increase has occurred over the decade.

4. SUMMARY

The sub-grid scale method was applied to describe the dry deposition process in RegADMS, and the distribution of sulfur deposition in Eastern China was estimated. Our conclusions are presented below:

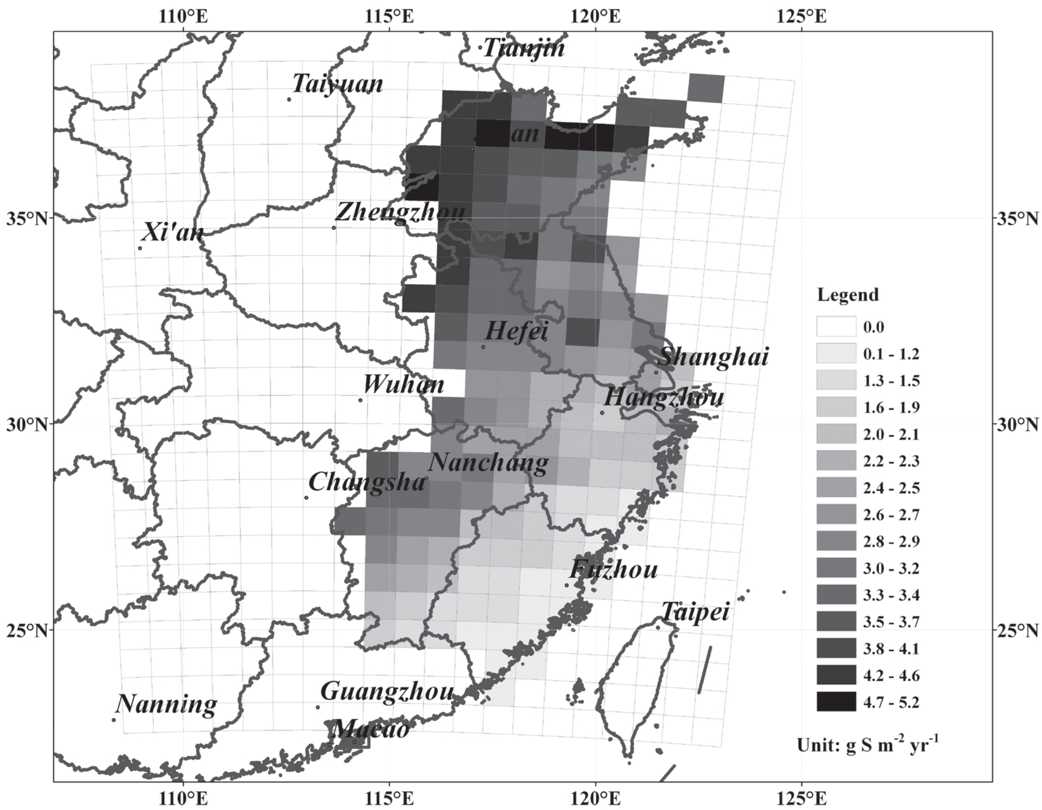

Fig. 4. Sulfur deposition flux distribution in Eastern China (g S m^{-2} yr^{-1}).

(1) Annual averaged V_d for SO_2 and SO_4^{2-} are 0.34 and 0.27 cm s^{-1} utilizing the fine grid scale method (FSM). For SO_2, V_d above water is larger compared to results for other land-use types, up to 0.66 cm s^{-1}. V_ds of SO_2 and SO_4^{2-} simulated by FSM agreed well with the semi-observation results at Yingtan Site and with the other research results.
(2) Comparison of root-mean-square of errors for the coarse grid scale method (CSM) and the sub-grid scale method (SSM) showed that the SSM was more precise and more reasonable than CSM, also involving much less computational time than for the FSM. SSM showed greater evident merit than CSM, especially for some specific grids.
(3) The total annual sulfur dry deposition for different land-use types by SSM was larger than that of CSM, except for over water and the evident regional discrepancy distribution showed SSM's efficiency in the regional deposition model. More than 90% of total sulfur is deposited on soil-plant ecosystems, and the atmosphere proves to be an important source of sulfur to these ecosystems. An evident regional gradient existed in total sulfur deposition, which was biggest for Shandong province and smallest for Fujian province. However, most sulfur deposition values in Eastern China surpassed 3.0 g m^{-2} yr^{-1}, which indicates an evident increase in recent years.

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