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Atmospheric sulfur deposition and the sulfur nutrition of crops at an agricultural site in Jiangxi province of China

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ABSTRACT

During the period From November 1998 to October 1999, the air sulfur dioxide (SO_2) and sulfate (SO_4^{2-}) concentrations were measured and rain water was collected on farmland at Yingtan, a typical red soil area in the Jiangxi province of China. Based on hourly meteorological data and surface resistance data from the literature, the dry deposition velocities of SO_2 and SO_4^{2-} were computed using a three-layer resistance analogy model, and sulfur dry deposition was calculated. The wet deposition was obtained from precipitation amount and sulfur concentrations in rainwater. The average dry deposition velocities of SO_2 and SO_4^{2-} on farmland were found to be 0.38 ± 0.16 cm s^{-1} (monthly average 0.16–0.55 cm s^{-1}) and 0.20 ± 0.12 cm s^{-1} (monthly average 0.15–0.27 cm s^{-1}), respectively. The annual total sulfur deposition for the study region is about 103 kg S ha^{-1}, of which 83% is dry deposition. The uncertainties due to measurement and the dry deposition model are less than 30%. It is also found that atmospheric deposition plays a key role in sulfur circulation within the agrecosystem, accounting for more than 90% of the total sulfur input to farmland

1. Introduction

Atmospheric sulfur deposition, including dry deposition and wet deposition, is one of the most important sulfur inputs to agrecosystems. The key sulfur compounds in the atmosphere are gaseous SO_2 (mostly emitted from anthropogenic sources) and aerosol SO_4^{2-} (transformed from SO_2 through gaseous and aqueous chemical reactions). Deposition of SO_2 and SO_4^{2-} can have significant influence on the circulation of sulfur in the soil–plant ecosystem, and many affect the fertilizer requirements of crops. There are therefore both academic and practical reasons to estimate the atmospheric deposition of sulfur to agrecosystems. There have been many studies on the measurement of wet and dry sulfur deposition in particular regions of China (Chen, 1993; Chen and Liu, 1997; Hong et al., 1987; Fan et al., 1993). For example, as reported by Chen (1993), the wet deposition ranges from 26 to 194 kg ha^{-1} a^{-1} and dry deposition from 17 to 397 kg ha^{-1} a^{-1} in Sichuan Province, where acid rain was found to be serious. The resistance analogy model performs well in calculating dry deposition velocities of gaseous species and particles (Voldner et al., 1986; Hicks et al., 1987; Walcek et al., 1986; Wang and Li, 1994; Smith et al., 2000). However, until now, few studies on atmospheric deposition in terms of its effect on sulfur circulation in farmland ecosystem have been reported (Fowler, 1989).

In this work we will focus on the estimation of sulfur deposition to a farmland ecosystem in a region of central China using combined field measurements and model calculations. Meteorological variables, including wind, temperature, pressure, radiation etc., were measured at an automatic weather station. These observations were applied to compute the turbulence parameters in the surface layer. The resistance analogy model was then used to calculate dry deposition velocities of atmospheric SO_2 and SO_4^{2-} particles.

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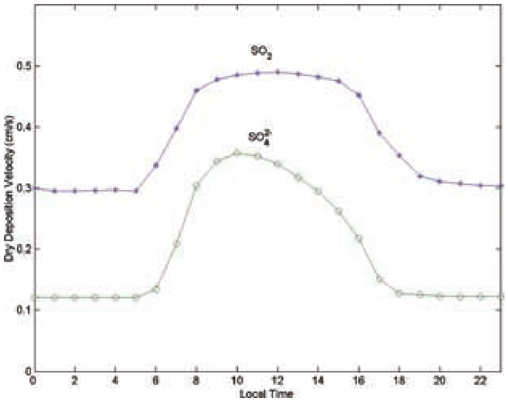

Fig. 1. The geographical location of the site.

The precipitation amount and sulfur concentrations in rainwater were combined to estimate the wet deposition. Total sulfur deposition is the sum of dry and wet deposition. Finally, the influence of atmospheric deposition on the sulfur balance in farmland ecosystem was investigated.

2. Methodology

2.1. *Site description*

From November 1998 to October 1999, field measurements were made over an ecology experimental station of the Chinese Academy of Science. The site is located far from the city of Yingtan in Jiangxi Province in central China (Fig. 1). The station is surrounded by farmland with red soil, which grows oilseed rape and paddy in different periods of the year. The pH of red soil is about 5.3–5.8, with a minimum of 4.5. The fraction of the time the land is bare soil is less than 10%. The growing seasons are June–August for paddy and other months except September for rape. Nothing grows during September. During 25–30% of the year the surface is wet due to precipitation, fog or dew. There are few obstructions and pollution sources within 10 km of the measurement site.

2.2. *Meteorological observations*

A 5.5-m high tower was set at the center of the farmland for microclimatic observations. The tower was equipped with automatic sensors at 5, 3.5, 2.0 and 1.0 m above the ground to measure wind speed/direction, temperature, pressure and humidity. Wind was observed by a magnetic suspended breeze sensor VF2 and temperature by an electrically heated thermometer HTF2. For pressure measurement, a vibrating drum barosensor ZGIV was used. In addition, a data logger DT500 was used to collect and analyze data every 60 min.

2.3. *Concentration measurements*

An air sampler TH110B made in TianHong Instrument Factory was applied to collect SO$_2$, using a bubbler method with H$_2$O$_2$ (pH 4.5, concentration 0.3%) as absorption liquid. The samples were collected at a frequency of 7 d per month and 8 times per day. At each survey, the sampling lasted 2 h with a flow rate of 0.8 L min^{-1}. Sulfur in the absorption and digestion liquid was analyzed by inductively coupled plasma spectroscopy (ICPAED). A TH150A sampler was applied to collect total suspended particles (TSP) with a flow rate of 100 L min^{-1}. The monthly sampling frequency for TSP was the same as that of SO$_2$, but only one sample was collected each day with a sampling time of 10 h. SO$_4^{2-}$ particulates were collected by a superthin fiberglass filter membrane (diameter <0.1 μm). After being digested in a 0.5 N HCl solution, SO$_4^{2-}$ was also analyzed by ICPAED. The container was closed to reduce evaporation. Rain water was collected continuously by a vessel within the sampling period of one month. The sulfur content was analyzed by ICPAED.

2.4. *Dry deposition model*

Since it is difficult to measure dry deposition directly, the observed concentrations and the resistance analogy model were combined to estimate dry deposition of sulfur.

Many factors can affect dry deposition, including atmospheric conditions, characteristics of the underlying surface and the deposited species itself. For particles, the dry deposition is influenced by Brown diffusion, inertia collision, gravitational setting etc. In order to take into account the main factors controlling the dry deposition process, a resistance model (Walcek et al., 1986) was applied to calculate dry deposition velocities of gaseous SO$_2$ and particle sulfate. Dry deposition velocities for gas and particles can be expressed as:

$$V_{\mathrm{d(g)}} = \frac{1}{R_\mathrm{a} + R_\mathrm{b} + R_\mathrm{c}}\ \text{(gas)} \tag{1a}$$

$$V_{\mathrm{d(a)}} = \frac{1}{R_\mathrm{a} + R_\mathrm{b}} + V_\mathrm{g}\ \text{(particles)}. \tag{1b}$$

In above equation, R_a, R_b and R_c are aerodynamic resistance, quasi-laminar resistance and underlying surface resistance, respectively. V_g is the gravitational setting velocity of particle, which can be estimated by the Stokes formula. Here, the gravitational setting velocity was taken into account for SO_4^{2-} particles with radii of 0.1, 2 and 10 μm. The final V_g for SO_4^{2-} is obtained from the simple arithmetic average over the three:

$$V_g = \frac{2r_p^2 g(\rho_p - \rho)}{9\rho\nu_a} \tag{2}$$

where r_p is the particle radius, ρ_p is the particle density. g is the acceleration due to gravity, ρ is the air density and γ_a is the air kinematical viscosity.

R_a is the resistance caused by turbulence when gas is transported from a reference height to the ground. Assuming that mass transfer is similar to heat transfer in the surface layer, R_a can be written as

$$R_a = \frac{\ln(Z_r/Z_0) - \psi_c}{\kappa u_*} \tag{3}$$

where Z_r is the reference height when calculating V_d; u_* is the friction velocity, k is the Von Karman constant (0.4), ψ_c is a function related to atmospheric stability and; Z_0 is the roughness length of the underlying surface. Since the dry deposition velocities for rape and paddy are expected to be different to a certain extent, in our calculations the roughness length was set to be seasonally in order to accommodate this varied effect.

R_b is the resistance when the deposited gas passes through the quasi-laminar layer. Based on a series of experiments, Wesely and Hicks (1977) recommended the following formula for estimating R_b, where Sc is the Schmidt number (the ratio of air kinematical viscosity γ_a to gaseous molecular diffusivity D_g):

$$R_b = \frac{2}{\kappa u_*} Sc^{2/3}. \tag{4}$$

For SO_4^{2-} particles, the formula given by Wesely et al. (1985) was adopted:

$$R_b = \begin{cases} \dfrac{1}{0.002u_*} & (L \geq 0) \\[2ex] \dfrac{1}{0.002u_*\left[1 + \left(-\dfrac{300}{L}\right)^{2/3}\right]} & (L < 0). \end{cases} \tag{5}$$

When the atmosphere is in a very unstable state ($Z_i/L < -70$), strong convection will increase the rates of vertical exchange and enhance the dry deposition velocity. Under this condition, the influence of mixing layer height Z_i is included in R_b (Wesely et al., 1985):

$$R_b = \frac{1}{0.002u_*\left[1 + \left(-0.3\dfrac{Z_i}{L}\right)^{2/3}\right]}. \tag{6}$$

R_c relates to interactions between the contaminant and the underlying surface. According to the studies of Wesely (1989) as well as Walmsley and Wesely (1996), surface resistance for SO_2 deposition varies under different conditions, such as land use, season, solar radiation and wetness of the surface. Here, surface resistance values for agriculture are selected and are given in Table 1.

The monthly dry deposition flux $F_d(Z_r)$ can be estimated from:

$$F_d(Z_r) = V_{d1}(Z_r) \times C_1(Z_r) + V_{d2}(Z_r) \times C_2(Z_r) \tag{7}$$

where V_{d1} and V_{d2} are the monthly average dry deposition velocities of SO_2 and SO_4^{2-}, respectively. C_1 and C_2 are the average concentration of SO_2 and SO_4^{2-} in the air measured over periods of a month. The annual deposition is obtained by summing the monthly depositions.

Table 1. *Surface resistance for SO_2 deposition to farmland at different conditions (s m^{-1}) (from Wesely, 1989)*

			Solar radiation (W m^{-2})				
Season	Roughness length (cm)	Albedo (%)	>400	200–400	0–200	Night	Wet surface
Spring	8	17	50	60	75	100	0
Summer	15	17	70	120	200	500	0
Early fall	10	23	500	500	500	500	100
Late fall	7	23	50	50	50	50	50
Winter	5	23	100	100	100	100	100

2.5. *Boundary layer parameterization*

For calculations of three components of resistance determining the dry deposition velocities, the turbulence parameters, such as u_*, θ_*, L and Z_i, are needed. Here, the boundary layer parameterization is on basis of the studies of Holtslag and Van Ulden (1983) as well as Van Ulden and Holtslag (1985), in which the energy budget method and routine meteorological data were adopted. Considering the surface energy balance, the net radiation (R) at the ground can be written as follows:

$$R_n = C + \lambda E + G \tag{8}$$

where C is the sensible heat flux, λE is the latent heat flux and G is the soil heat flux.

$$C = -\rho C_p u_* \theta_* \tag{9}$$

where $\rho = 1.293$ kg m^{-3} and $C_p = 1004$ J kg^{-1} K^{-1}.

The temperature scale θ_* for turbulent heat transfer is calculated from eqs. (8) and (9):

$$\theta_* = \frac{R_n - \lambda E - G}{-\rho C_p u_*}. \tag{10}$$

The parameterizations of R_n, λE and G are based on works by Holtslag and Van Ulden (1983), Van Ulden and Holtslag (1985) and Hanna and Chang (1992).

The Monin–Obukhov length L is calculated by eq. (11):

$$L = -\frac{u_*^3 T \rho C_p}{k g H_g}. \tag{11}$$

The height of mixing layer Z_i is parameterized using eq. (12) under stable, neutral and unstable conditions suggested by Zilitinkevich (1975):

$$Z_i = \begin{cases} \mathrm{Min}\left[ku_*/f,\ ku_*/f(u_*/fL)^{-0.5}\right] & \text{(stable)} \\ ku_*/f & \text{(neutral)} \\ \mathrm{Max}\left[ku_*/f, ku_*/f(-u_*/fL)^{0.5}\right] & \text{(unstable)} \end{cases} \tag{12}$$

where f denotes the Coriolis parameter.

3. Results and discussion

3.1. *Dry deposition velocity*

Based on the resistance model and meteorological data obtained from the automatic microclimatic station, hourly dry deposition velocities of SO_2 and SO_4^{2-} for the farmland were calculated. The statistical average dry deposition velocities during the study period are shown in Fig. 2.

The figure illustrates the distinct diurnal variations of the dry deposition velocities of SO_2 and SO_4^{2-} at the

Fig. 2. Hourly dry deposition velocities of SO_2 and SO_4^{2-} (cm s^{-1}).

Table 2. *The average dry deposition velocities of SO_2 and SO_4^{2-} for farmland*

	SO_2 (cm s^{-1})		SO_4^{2-} (cm s^{-1})	
Period	Average	Range	Average	Range
Nov 1998	0.55 ± 0.18	0.39–0.93	0.18 ± 0.10	0.12–0.21
Dec 1998	0.43 ± 0.13	0.35–0.70	0.20 ± 0.11	0.17–0.24
Jan 1999	0.37 ± 0.10	0.33–0.53	0.22 ± 0.15	0.17–0.25
Feb 1999	0.35 ± 0.06	0.34–0.36	0.24 ± 0.16	0.20–0.27
Mar 1999	0.44 ± 0.14	0.42–0.51	0.27 ± 0.19	0.19–0.31
Apr 1999	0.45 ± 0.11	0.43–0.48	0.24 ± 0.15	0.17–0.29
May 1999	0.46 ± 0.11	0.44–0.57	0.21 ± 0.11	0.16–0.25
Jun 1999	0.32 ± 0.16	0.30–0.34	0.17 ± 0.08	0.12–0.25
Jul 1999	0.31 ± 0.16	0.30–0.33	0.16 ± 0.07	0.12–0.23
Aug 1999	0.37 ± 0.16	0.37–0.38	0.15 ± 0.06	0.13–0.20
Sep 1999	0.16 ± 0.01	0.15–0.18	0.15 ± 0.05	0.13–0.20
Oct 1999	0.39 ± 0.26	0.16–0.88	0.17 ± 0.07	0.13–0.22
Annual	0.38 ± 0.16	0.15–0.93	0.20 ± 0.12	0.12–0.31

farmland, where V_d values are higher at during the day than at night. Usually, since there is no solar radiation and the atmosphere is in a more stable state at night, the turbulent resistance is relatively higher, leading to a lower dry deposition velocity. Additionally, for SO_2, the surface resistance always decreases during the day under clear sky conditions. However, it might be smaller at night if the surface is wet. For SO_4^{2-}, the quasi-laminar resistance is prevalent compared to the aerodynamic resistance; thus the former is the major factor controlling the diurnal variability of SO_4^{2-}. The dry deposition velocity of SO_2 is nearly double that of SO_4^{2-}, and both are stable at night.

The monthly and annual average dry deposition velocities are listed in Table 2. It shows that the seasonal variation of V_d for SO_2 is stronger than that for SO_4^{2-}. The dry deposition velocity of SO_2 exhibits a maximum (0.55 cm s^{-1}) in November and a minimum (0.16 cm s^{-1}) in September. For SO_4^{2-}, V_d changed little with the season, although a small peak at March can be found. The difference in seasonal variability for SO_2 and SO_4^{2-} is mainly due to the influence of surface resistance, which varies significantly with season for SO_2 and is zero for SO_4^{2-}. As stated in the section above, surface resistance plays an important role in determining V_d of SO_2, while seasonal variation of SO_4^{2-} is mainly controlled by the quasi-laminar resistance. The annual average dry deposition velocities of SO_2 and SO_4^{2-} at farmland are 0.38 ± 0.16 cm s^{-1} and 0.20 ± 0.12 cm s^{-1}, respectively. These results are in line with the estimates of Xu and Carmichael (1998), who have given 0.2–0.5 cm s^{-1} for SO_2 and 0.1–0.3 cm s^{-1} for SO_4^{2-} deposition over cultivation.

3.2. *SO_2 and SO_4^{2-} concentrations*

The monthly and annual average concentrations of SO_2 and SO_4^{2-} in air are given in Table 3. The annual average loadings for SO_2 and SO_4^{2-} are 64 μg S m^{-3} and 10.7 μg S m^{-3}, respectively. SO_2 and SO_4^{2-} exhibit distinct seasonal variations, which are affected by transformation, dry deposition and wet scavenging processes. The SO_4^{2-} concentration in the study region is much smaller than that of SO_2.

Table 3. *Observed SO_2 and SO_4^{2-} concentrations in air*

Period	SO_2(μg S m^{-3})	SO_4^{2-}(μg S m^{-3})
Nov 1998	60	11.8
Dec 1998	163	6.8
Jan 1999	155	30.4
Feb 1999	65	8.0
Mar 1999	53	7.4
Apr 1999	72	6.2
May 1999	9	5.9
Jun 1999	47	5.7
Jul 1999	53	14.7
Aug 1999	29	13.0
Sep 1999	21	11.1
Oct 1999	38	7.9
Annual	64	10.7

Table 4. *Precipitation amount and sulfur concentration in rainwater*

Period	Precipitation (mm)	S concentration (μg S L^{-1})
Nov 1998	9.4	7.766
Dec 1998	76.5	1.608
Jan 1999	67.2	2.887
Feb 1999	48.1	0.915
Mar 1999	79.6	4.648
Apr 1999	401.0	0.668
May 1999	488.8	0.241
Jun 1999	370.0	0.318
Jul 1999	209.4	0.589
Aug 1999	285.8	0.587
Sep 1999	106.1	0.868
Oct 1999	33.7	0.901
Annual	2175.6	0.791

Table 4 shows the monthly precipitation and sulfur concentration in rainwater. The annual precipitation during the study period is 2175.6 mm, with much raining falling during the spring and summer seasons. High sulfur concentrations are usually accompanied by small rainfall, indicating that precipitation has a significant influence on ion concentration in rainwater. The statistical analysis shows that the sulfur concentration is negatively correlated with precipitation, with a coefficient of −0.51.

3.3. *Sulfur deposition*

SO_2 and SO_4^{2-} dry deposition were calculated on the basis of formula (7). The results, together with the observed wet deposition, are listed in Table 5.

During the observational period, total sulfur deposition to farmland is 103 kg S ha^{-1}. Dry deposition is much greater than wet deposition, and accounts for 83% of the total deposition with a monthly range of 56–94%. The percentage of SO_2 dry deposition in total S dry deposition averages 92%, with a monthly range of 67–98%. Evidently, dry deposition, especially SO_2 deposition, plays the major role in total sulfur deposition due to heavy SO_2 pollution at the study region.

3.4. *Influence of atmospheric sulfur deposition on agrecosystem*

In order to understand the influence of atmospheric deposition on sulfur circulation in the agrecosystem, the input, output and balance of sulfur at two kinds of farmland were investigated. The preliminary results can be seen in Table 6. The estimates of sulfur in paddy and rape are based on the dry weight of root, stalk and grain and their sulfur content (Hu and Shen, 1998). In Jiangxi Province, the root and stalk are usually removed from the soil and are used as fuel when they have been dried. Runoff water was sampled by drain collectors; the sulfur content was then measured using the specific turbidity method (Liu et al., 1999).

Table 5. *SO_2 and SO_4^{2-} dry and wet deposition fluxes on farmland (kg S ha^{-1})*

Period	SO_2 dry deposition	SO_4^{2-} dry deposition	S dry deposition	S wet deposition	S total deposition
Nov 1998	8.5 (93.8%[a])	0.6	9.1 (92.5%[b])	0.7	9.8
Dec 1998	18.7 (98.1%)	0.4	19.1 (94.0%)	1.2	20.3
Jan 1999	15.2 (89.2%)	1.8	17.0 (89.8%)	1.9	18.9
Feb 1999	5.5 (92.2%)	0.5	6.0 (93.2%)	0.4	6.4
Mar 1999	6.2 (92.1%)	0.5	6.7 (64.5%)	3.7	10.4
Apr 1999	8.4 (95.6%)	0.4	8.8 (76.5%)	2.7	11.4
May 1999	1.1 (76.4%)	0.3	1.4 (55.0%)	1.2	2.6
Jun 1999	3.9 (94.0%)	0.3	4.1 (77.8%)	1.2	5.3
Jul 1999	4.4 (87.8%)	0.6	5.0 (80.4%)	1.2	6.2
Aug 1999	2.9 (84.6%)	0.5	3.4 (67.1%)	1.7	5.1
Sep 1999	0.9 (66.7%)	0.4	1.3 (58.8%)	0.9	2.2
Oct 1999	3.9 (91.4%)	0.4	4.3 (93.5%)	0.3	4.6
Annual	79.6 (92.3%)	6.7	86.3 (83.4%)	17.2	103.5

[a] Percentage of SO_2 dry deposition in total S dry deposition.
[b] Percentage of S dry deposition in total S deposition.

Table 6. *Sulfur balance at two kinds of farmland covered with rape and paddy in turn*

Items	Middle–low-production farmland (paddy 13.5 t ha^{-1} rape 3 t ha^{-1})	High-production farmland (paddy 17.4 t ha^{-1} rape 6 t ha^{-1})
Input (kg S ha^{-1})		
Dry deposition	86.3 (78.0%)	86.3 (78.0%)
Wet deposition	17.2 (15.3%)	17.2 (15.3%)
Irrigation	5.2 (4.7%)	5.2 (4.7%)
Organic fertilizer	3.4 (3.0%)	3.4 (3.0%)
Total	112.1	112.1
Output (kg S ha^{-1})		
Paddy	17.5	22.5
Rape	32.5	65.0
Leaching	22.1	22.1
Runoff	11.3	11.3
Volatilization	3.5	3.5
Total	86.8	124.3
Balance (kg S ha^{-1})		
Gap between input and output	25.3	−12.2

Loss of sulfur by leaching was determined by using lysimeter tubes. The water balance was estimated and the sulfur concentration determined. Loss of sulfur by volatilization was estimated by a closed or open chamber to determine the concentration of sulfur.

Table 6 illustrates that sulfur deposition has strong influence on the sulfur balance (input minus output) in the farmland ecosystem. Atmospheric deposition accounts for 93% (of which 78% is dry deposition and 15% wet deposition) of sulfur input needed by the farmland ecosystem. For farmland with middle to low production the sulfur balance is positive, indicating that sulfur is sufficient for plant nutrients. In contrast, for farmland with high production, the sulfur balance is negative, which suggests that supplemental sulfur in various forms, including fertilizer, is necessary in order to obtain persistent high productivity. Therefore, it can be concluded that much atmospheric sulfur-deposition, even at the quite high levels found at this site, may be beneficial to farmland with high productivity, although there may also be unwanted effects From these levels of pollution.

3.5. Uncertainty analysis

The estimates of sulfur deposition and the corresponding sulfur balance at the agrecosystem are highly dependent on the measurement technique and the dry deposition model used. Measurement of SO_2 concentration in the air by using a bubbler method will result in random errors due to artificial manipulation. In addition, it is appropriate that the dry deposition model has adopted the literature of surface resistance for calculating dry deposition velocity of SO_2. In order to estimate the error quantitatively, we double the surface resistance and calculate V_d again. The dry deposition velocity of SO_2 was found to decrease in the range -24 to -31% in different months. If the surface resistance were reduced by 50%, the monthly average dry deposition velocity of SO_2 would increase from 18 to 33%. Therefore, it can be concluded that the uncertainty in the estimate of sulfur dry deposition is less than 30%. In addition, it seems that the sulfur dry deposition has been overestimated due to the uncertainties in measurement of SO_2 concentration. In the future we will measure concentration gradients of SO_2 using an automatic analyzer and calculate surface resistance using a "big-leaf" model, so that the estimations of sulfur dry deposition will be improved. On the other hand, it should be noted that the measurements of sulfur due to runoff, leaching and volatilization are relatively rough, which will lead to an uncertainty of about 10–20%.

4. Summary and conclusions

In this paper, the atmospheric sulfur deposition on farmland was investigated. SO_2 and SO_4^{2-} concentrations in air and rainwater were measured for one year at the red soil ecology station in Yingtan, central China. By use of a resistance analogy model, dry deposition velocities of SO_2 and SO_4^{2-} were calculated, and then sulfur deposition to farmland was analyzed. Results show that dry deposition velocities have diurnal variations for SO_2 and SO_4^{2-} particles with higher values during the dayt than at night. The annual average dry deposition velocities on farmland are 0.38 ± 0.16 cm s^{-1} for SO_2 and 0.20 ± 0.12 cm s^{-1} for SO_4^{2-}, respectively. During the study period, total annual deposition to the farmland ecosystem is about 103 kg S ha^{-1}, of which 83.4% is dry deposition. Further investigations indicate that atmospheric deposition plays an important role in the sulfur balance of a farmland ecosystem. Thus high sulfur-deposition from the atmosphere seems to have advantages for crop yields from farmland with high production, although it may not generally be good for the environment.

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