

High solubility of SO₂

Q. Zhang and X. Tie

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High solubility of SO₂: evidence in an intensive fog event measured in the NCP region, China

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Abstract

A field experiment was conducted in a heavy SO₂ pollution area located in north China plain (NCP). During the experiment, SO₂ and other air pollutants, liquid water content (LWC) of fog droplets, and basic meteorological parameters were measured. During the experiment, an intensive fog event occurred between 5 and 8 November 2009. During the fog period, the concentrations of SO₂ showed a strong variability, and the variability was closely correlated to the appearances of fogs and LWC. For example, the averaged concentration of SO₂ during the non-fog period was about 25 ppbv. By contrast, during the fog period, the concentration of SO₂ reduced to about 4–7 ppbv. The large reduction of SO₂ suggests that a majority of SO₂ (about 70–80%) had been converted from gas-phase to aqueous-phase, showing a high solubility of SO₂. However, according to the value of Henry Law constant, the solubility of SO₂ is modest, which cannot explain the measured large reduction of SO₂. This study highlights that the aqueous reactions of SO₂ in the droplets of fogs play important roles to enhance the solubility of SO₂. To account for the effect of aqueous reactions on the solubility of SO₂, an “effective” Henry Law constant of SO₂ is proposed in this study. The study shows that without considering aqueous reactions of SO₂ in fog droplets, the estimate of the partitioning of SO₂ in droplets is significantly lower than the measured values. By contrast, when the “effective” Henry Law constant is applied in the calculation, the calculated SO₂ concentrations are significantly improved, showing that the aqueous reactions of SO₂ play important roles in controlling the solubility of SO₂, and should be considered in model calculations.

1 Introduction

Sulfur dioxide (SO₂) can be released to the atmosphere due to both natural and anthropogenic emissions. The natural emissions include volcanic eruptions and biomass burnings. The anthropogenic emissions are mainly due to coal and oil burning

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processes, accounting for more than 75% of global emissions (Chin et al., 2000). Coal accounts for about 70% of China's total energy consumption, and the consumption of coal increases rapidly in the recent years due to the aggressive economical development in the country, with the consumption of coal of 2.01 billion metric tons in 2009 (IEA, 2010). The large consumption of coal in China leads to high SO₂ emissions, resulting in the high concentrations of SO₂ (Kawamoto et al., 2004). One of the major fates of SO₂ gas is to form sulfate particles, and understanding the formation of sulfate particles in the troposphere is critical because of their effects on human health and climate (Zhao et al., 2006; Tie et al., 2003, 2009). Thus, studying the budget (such as emission, transformation, and deposition) of SO₂ becomes a crucial environmental issue in China.

Sulfur dioxide is a soluble gas in water, and can be oxidized within cloud and fog droplets, producing sulfuric acid. In this study, a field experiment was conducted in a heavy SO₂ pollution area which is located in north China plain (NCP). During the experiment, SO₂ and other air pollutants, liquid water content (LWC) of fog droplets, and basic meteorological parameters were measured. A large variability of SO₂ and liquid water content of fog droplets were observed, providing a good opportunity to study the solubility of SO₂ in the in-situ field measurement. The study of solubility of SO₂ has important implication for understanding the aqueous oxidation of SO₂ in fog droplets, which significantly influence the acidity in fogs. Because the acidity in clouds, precipitation, and fogs is a central environmental focus in China due to its rapidly economical development, the direct measurement of solubility of SO₂ can provide important information for the environmental issues mentioned above.

The paper is organized as the following way. In Sect. 2, we describe the field experiment, including the instruments and the data. In Sect. 3, we analyze of the result of the experiment to study solubility of SO₂ in fog droplets.

2 Description of the experiment

2.1 Information of the experiment

The field experiment took place in the north edge of Tianjin City (39.4° N in latitude and 117.05° E in longitude) at a surface site. The surface site is about 30 km from the center of the city. The city of Tianjin is a mega city with a population of 10 million. The city is located in the heart of North China Plain (NCP), and it is one of the populated and polluted regions in China (see Fig. 1). During the recent years, the rapid increase of economical development results in heavy atmospheric pollution, especially SO₂ pollution in this region (Tie and Cao, 2009; Zhang et al., 2010).

Figure 1 shows the horizontal distribution of SO₂ concentrations measured by the SCIAMACY (SCanning Imaging Absorption spectroMeter for Atmospheric Cartography), and the horizontal distribution of fogs measured by MODIS (Moderate Resolution Imaging Spectroradiometer). Figure 1 shows that the fog formation is closely correlated to the high concentrations of SO₂ in the NCP region. The coherence between the high SO₂ concentrations and the frequent fog formation provides a good opportunity to study SO₂ solubility by analyzing in-situ measurements of SO₂ concentrations and fog droplets in the NCP region. Thus, an intensive field measurement was conducted during a period of the fog season (from 5 to 8 November 2009) to study the variability of SO₂ and its correlation to LWC during the formation of fogs. Several instruments were deployed during the field experiment, including; (1) Fog Droplet Measurement Technology (FDMT) for fog droplet measurement, with diameters ranging from 2 to 32 μm (15 size bins), (2) Scan Mobility Particle Sizer (SMPS) for aerosol particle measurement, with particle diameters ranging from 10 to 662 nm (71 size bins), (3) the instruments of air pollutants (O₃, CO, NO_x, and SO₂), and (4) the instruments for basic meteorological fields (such as temperature, relative humidity, air pressure, winds, etc). These integrated instruments provide necessary information to study the solubility of SO₂ in fog droplets. In order to insure the quality of the instruments (QA/QC issue), all the instruments were calibrated before the measurement. The detailed information of calibration is described by Zhang et al. (2010).

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2.2 Background of meteorological conditions during experiment

During the experiment, an intensive fog event occurred between 5 and 8 November 2009. During the fog event, the NCP region was under the effect of a weak low pressure system, with calm winds. The averaged surface wind speed was only 1.1 m/s during the event. The averaged surface temperature was low (6.8 °C) and humidity was high with a large variation. These weather and meteorological conditions were very favorable for the formation of fogs. After the fog event (the late of 8 November), the low pressure system moved out of the NCP region. As a result, the averaged surface wind speed rapidly increased to about 4 m/s, and the averaged surface temperature increased to about 15 °C.

During the fog event, a periodic formation of fog occurred. Based on the duration of fog occurrences in the event, three fog periods are defined according to the combined characteristics of observed relative humidity (RH), ambient temperature (T), fog liquid water content (LWC), and the range of visibility (VIS). The first fog period (Fog-1) occurred from 03:45 to 10:42, 6 November. The second fog period (Fog-2) was appeared from 19:10, 6 November to 12:27, 7 November, and the third fog period (Fog-3) occurred from 16:29, 7 November to 05:40, 8 November (see Fig. 2). The detailed analysis of the effects of meteorological conditions on the formation of fogs is shown in Quan et al. (2011).

3 Result and discussion

3.1 Measurement of air pollutants

One of the important information for occurrence of fogs is the appearance of liquid water. The measurement shows that liquid water appeared during the 3 fog periods, with the averaged liquid water content (LWC) of 0.23, 0.35, and 0.35 g/m³, respectively (as indicated in Fig. 2). Figure 2 also shows the variability of visibility, CO, and SO₂

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before, during, and after the 3 fog appearances. During the three fog periods, there are several important aspects which are highlighted as follows. (1) The visibility range rapidly changed during the occurrence of fogs. For example, the mean visibility range was about 870 m during the non-fog periods, which was relatively low, indicating that the concentrations of aerosols were high during the entire measurement (a heavy haze event). During the 3 fog periods, the visibility ranges rapidly reduced to 20–50 m, suggesting that the fog occurrence resulted in an extremely low visibility event. Because the occurrence of extremely low visibility events was closely correlated to the formation of fogs, it can be used as an indirect evidence for the occurrence of fogs in the NCP region. (2) The measured SO₂ concentrations showed a strong variability, and the variability was closely correlated to the appearance of fogs and LWC, which shows the strong effect of LWC on the soluble gas of SO₂. For example, the averaged concentration of SO₂ during the non-fog period was about 25 ppbv. By contrast, the concentrations of SO₂ during the 3 fog periods were rapidly reduced to 6.8, 4.4, 7.2 ppbv, respectively. (3) In order to better understand the causes of the large reduction of SO₂ concentrations during the appearance of fogs and LWC, the measured variability of CO is analyzed (in the middle panel of Figure 2). The main difference between CO and SO₂ is that CO is not a soluble gas in water, while SO₂ is a soluble gas. Unlike the variability of SO₂, there was no clear evidence that CO variability was correlated to the occurrence of fogs and LWC. For example, the averaged concentration of CO during the non-fog period was 5.2 ppmv. Unlike the concentrations of SO₂, during the 3 fog periods, the changes in the concentrations of CO were not significant, with the mean concentrations of 4.8, 5.9, 7.6 ppmv, respectively. In addition, there was also an indication that the variability of CO concentrations was more sensitive to wind speeds rather than the formation of fogs. For example, after the fog period, the averaged wind speed increased from 1.1 to 4.1 m/s, leading to a sharp decrease of CO concentration (from 10 to 2 ppmv in a few hours). The complete different behaviors between the variability of CO and SO₂ during the fog periods suggest that SO₂ significantly reduced by dissolving gas-phase SO₂ in fog droplets during the formation of fogs.

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More quantitative analysis of the effect of fog water on SO₂ concentrations is shown in Fig. 3. It shows that during the 3 fog periods, the visibility ranges reduced by 94, 96, and 96%, respectively. There were no consistent changes in CO concentrations, with −8, +13, and +46% variations during the 3 fog periods, respectively. By contrast, the concentrations of SO₂ consistently reduced by −72, −82, and −71% during the 3 fog periods, respectively. The large reduction of SO₂ indicated that a majority of SO₂ (about 70–80%) had been converted from gas-phase to aqueous phase, showing a high solubility of SO₂. However, according to the value of Henry Law constant (the measure of solubility) of SO₂, the solubility of SO₂ is modest (with the Henry Law constant of 10²–10³ dependent on pH values), which is much less than the highly soluble species such as H₂O₂ (the Henry Law constant is 2.4 × 10⁵). As a result, the measured high solubility of SO₂ during the fog periods cannot be explained by the Henry Law constant. Other processes (such as aqueous reactions of SO₂ in water) could play important roles in controlling the solubility of SO₂, which need to be carefully studied.

3.2 SO₂ solubility

As suggested by Ravishankara (1997), the actual or effective solubility of chemical species in liquid water of clouds/fogs are determined by the following factors; (1) the diffusion rate (*k_a*) for transport gas-phase molecules into water, which is the first step to determine the solubility of chemical species; (2) the disassociation rate of a chemical species in droplets, which is a factor to enhance the solubility of chemical species; and (3) the rate of aqueous reaction of chemical species in droplets, which can further increase the solubility of chemical species.

Figure 4 shows the schematic description of the enhancement of solubility of SO₂ due to aqueous phase reactions. The Henry Law constant of chemical species is a combination result of the diffusion rate (*k_a*) and composition of a chemical species, which is modest for SO₂. The Henry Law constant combined with aqueous phase reactions can enhance the solubility of SO₂, which can be referred as an “Effective

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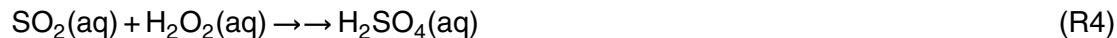


Henry Law constant” as indicated in Fig. 4. Figure 4 shows that when the molecules of SO₂ are transported (diffused) into a liquid droplet, aqueous reactions convert SO₂ into other chemical species. As a result, more molecules of SO₂ are transported into water droplets, and less molecules of SO₂ are returned back to the atmosphere, leading to the enhancement of solubility of SO₂. The quantitative calculation of the solubility of SO₂ with the consideration of aqueous phase reaction is described by the following method.

If no aqueous reaction is considered, the solubility of SO₂ is determined by the Henry Law constant, and the partitioning between gas and aqueous phase concentrations is expressed by the following equation;



Where SO₂(g) and SO₂(aq) represent SO₂ concentrations in gas and aqueous phases, respectively. However, if the aqueous phase reactions of SO₂ in droplets are considered, the calculation of solubility of SO₂ becomes to be complex. According to the study by Seinfeld and Pandis (1998), there are two important aqueous-phase reactions for SO₂. After SO₂ resolved in water, it can either react with H₂O₂ or O₃. Because O₃ is a very low soluble species (with the Henry Law constant of 0.02), the aqueous-phase reaction with O₃ is not significant compared to the reaction with H₂O₂. Thus, the calculation of solubility of SO₂ is mainly determined by the following expressions;



Where H₂O₂(g) and H₂O₂(aq) represent H₂O₂ concentrations for gas and aqueous phase, respectively. These expressions suggest that the solubility of SO₂ depends on three quantities; (1) the aqueous concentrations of SO₂(aq) in droplets, (2) the aqueous concentrations of H₂O₂(aq) in droplets, and (3) the rate of aqueous reaction

(*k*) between SO₂(aq) and H₂O₂(aq). Because the rate of the aqueous reaction between SO₂(aq) and H₂O₂(aq) is very fast (Seinfeld and Pandis, 1998), it requires more SO₂(g) to be converted to SO₂(aq), and the value of SO₂(aq) is strongly dependent upon the availability of H₂O₂(aq). As a result, the solubility of SO₂ is enhanced and the “effective”

Henry Law constant of SO₂ can be expressed;

$$H'_e(\text{SO}_2) = H_e(\text{H}_2\text{O}_2) \times \text{H}_2\text{O}_2(\text{g})/\text{SO}_2(\text{g}) \quad (\text{R5})$$

Where $H'_e(\text{SO}_2)$ represents the “effective” Henry Law constant of SO₂, and $H_e(\text{H}_2\text{O}_2)$ represents the Henry Law constant of H₂O₂.

The hypothesis of the “effective” Henry Law constant of SO₂ is tested by comparing the calculation with measurement of the aqueous phase partitioning of SO₂ in fog droplets. For the calculation, the measured values of LWC are used (see Fig. 2). The initial values of SO₂ are set to be the measured values before the occurrences of the fogs. During the fog periods, the concentration of SO₂(g) is set to be 5.0 ppbv which is similar to the measured SO₂ concentrations during the 3 fog periods, and the concentration of H₂O₂(g) is set to be 1.0 ppbv which is often measured in the atmosphere. With the above considerations, the partitioning between gas and aqueous phase concentrations of SO₂ are calculated. The calculation shows that without the consideration of aqueous phase reaction, the calculated concentrations of SO₂ are significantly over-estimated the measured values (as indicated in the red lines of Fig. 5). For example, the measured concentrations of SO₂ during the 3 fog periods are about 5 ppbv, while the calculated concentrations are about 20 ppbv that is about 4 times higher than measured values. By contrast, when the “effective” Henry Law constant is applied in the calculation, the calculated SO₂ concentrations are significantly improved. For example, the calculated minimum concentrations of SO₂ are close to the measured values (as indicated in the blue lines of Fig. 5), showing significant reduction compared to the case when the aqueous phase reaction is not considered. Although the calculation with the “effective” Henry Law constant considerably improves the calculated concentrations of SO₂ during fog periods, there are still some discrepancies between the calculation and measurement. For instant, there is a tendency that the calculated SO₂ concentrations

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have an earlier increase (with a time lag of 2 h) compared to the measured values. In the calculation, the reduction of SO₂ concentration is mainly due to the appearance of LWC. Thus, any uncertainty of measured LWC could cause the errors in the calculation of SO₂ concentration during fog periods.

4 Summary

Sulfur dioxide (SO₂) can be released to the atmosphere due to both natural and anthropogenic emissions. The anthropogenic emissions are mainly due to coal and oil burning processes, and coal accounts for about 70% of China's total energy consumption. The large consumption of coal in China results in high concentrations of SO₂ in the north China plain (NCP) region, leading to severely environmental problems in the region. Thus, studying the budget (such as emission, transformation, and deposition) of SO₂ becomes a crucial environmental issue in China. Sulfur dioxide is a soluble gas in water, and can be oxidized within cloud and fog water droplets, producing sulfuric acid. In this study, a field experiment was conducted in a heavy SO₂ pollution area in NCP. During the experiment, gas-phase SO₂ and other gas-phase air pollutants, water content of fog droplets, and basic meteorological parameters were measured. A large variability of SO₂ and liquid water content (LWC) of fog droplets were observed, providing a good opportunity to study the solubility of SO₂ in the field measurement.

The measurement shows that during 5 to 8 November 2009, there were 3 fog periods, with the averaged liquid water content (LWC) of 0.23, 0.35, and 0.35 g/m³, respectively. During the fog periods, the measured SO₂ concentrations showed a strong variability and the variability was closely correlated to the appearance of fogs and LWC. For example, the averaged concentration of SO₂ during the non-fog period was about 25 ppbv. By contrast, the concentrations of SO₂ during the 3 fog periods were rapidly reduced to 6.8, 4.4, 7.2 ppbv, respectively. The large reduction of SO₂ concentrations suggests that SO₂ was significantly reduced by dissolving gas-phase SO₂ into fog droplets during the formation of fogs. The large reduction of SO₂ (about 70–80%)

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during the fog periods also suggests that the solubility of SO₂ is considerably high. However, according to the value of Henry Law constant, the solubility of SO₂ is modest, which cannot explain the large reduction of SO₂. This measurement result suggests that the aqueous reactions of SO₂ play important roles to enhance the solubility of SO₂. Thus, an “effective” Henry Law constant of SO₂ is proposed in this study, which accounts for the aqueous reactions in the solubility of SO₂. The study shows that the calculated concentrations of SO₂ are significantly overestimated the measured values without considering the aqueous reactions. By contrast, when the “effective” Henry Law constant is applied in the calculation, the calculated SO₂ concentrations are significantly improved, showing that the aqueous reactions of SO₂ play important roles in controlling the solubility of SO₂, and should be considered in model calculations.

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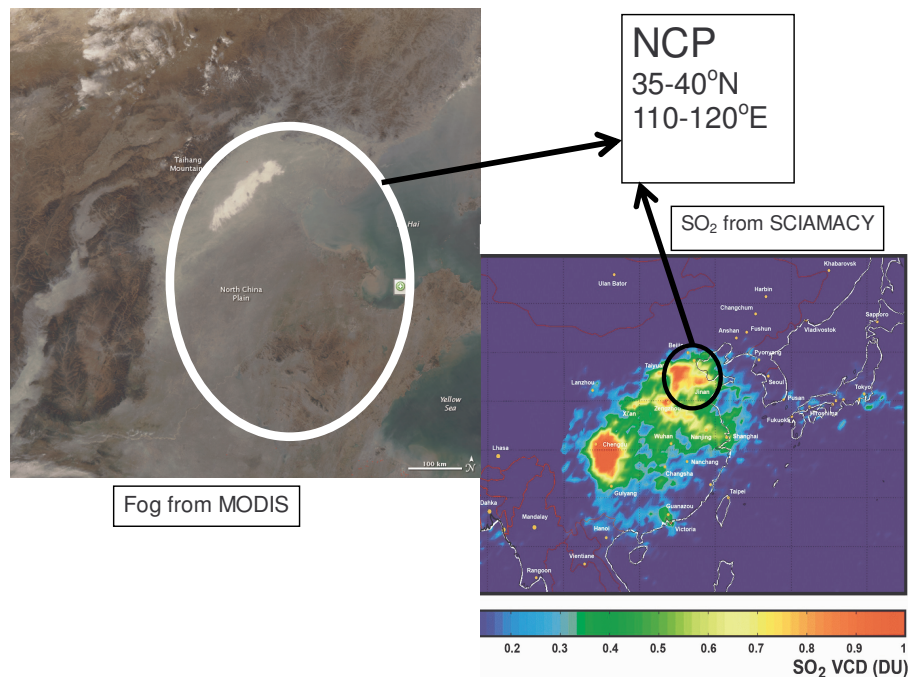


Fig. 1. The horizontal distribution of SO_2 concentrations (averaged in 2003) measured by the SCIAMACY satellite (right panel), and the horizontal distribution of fogs measured by the MODIS satellite on 6 November 2009 (left panel). The circles show the location of the NCP region.

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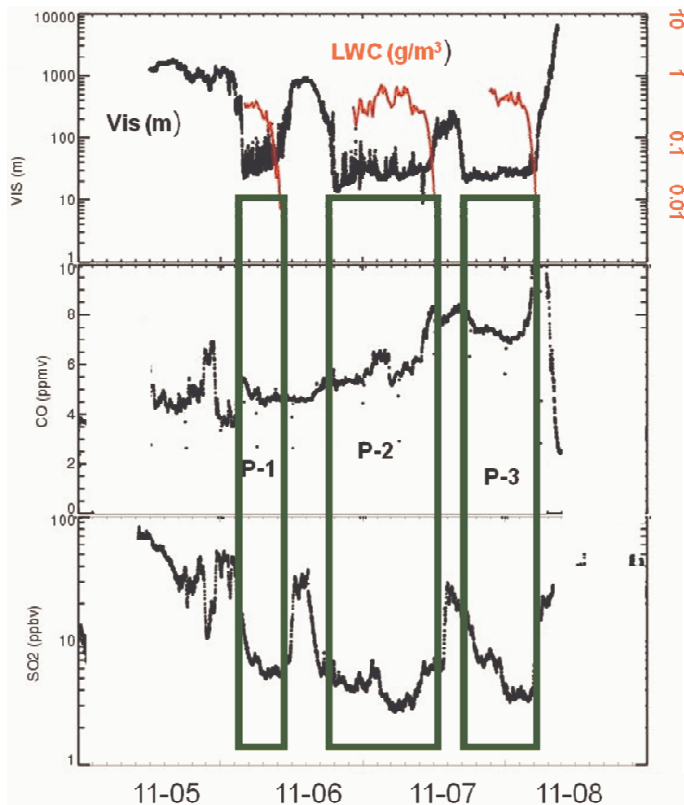


Fig. 2. The measured fog liquid water content (red lines in upper panel), the range of visibility (black lines in upper panel), the concentrations of CO (middle panel), and the concentrations of SO₂ (lower panel). The green boxes indicate the 3 fog periods.

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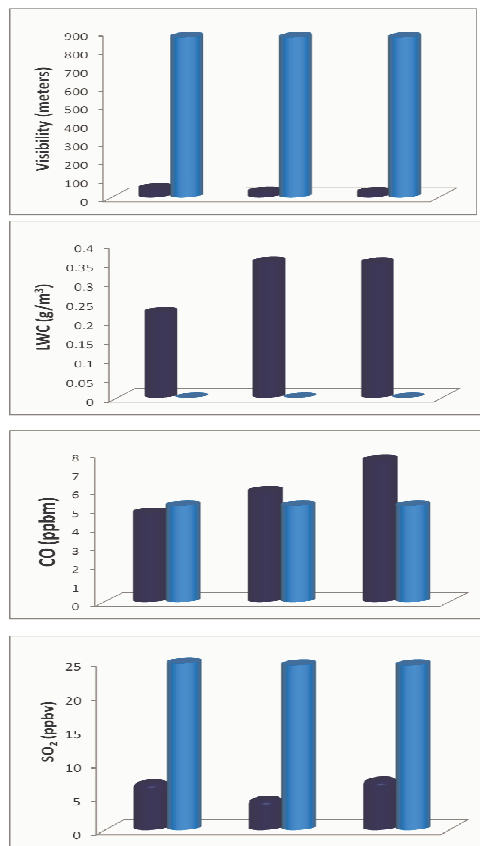


Fig. 3. The effects of fog formation on visibility (panel 1), LWC (panel 2), CO concentrations (panel 3), and SO₂ concentrations (panel 4). The dark blue columns show the mean values during the 3 fog periods, and the light blue columns show the mean values during non fog periods.

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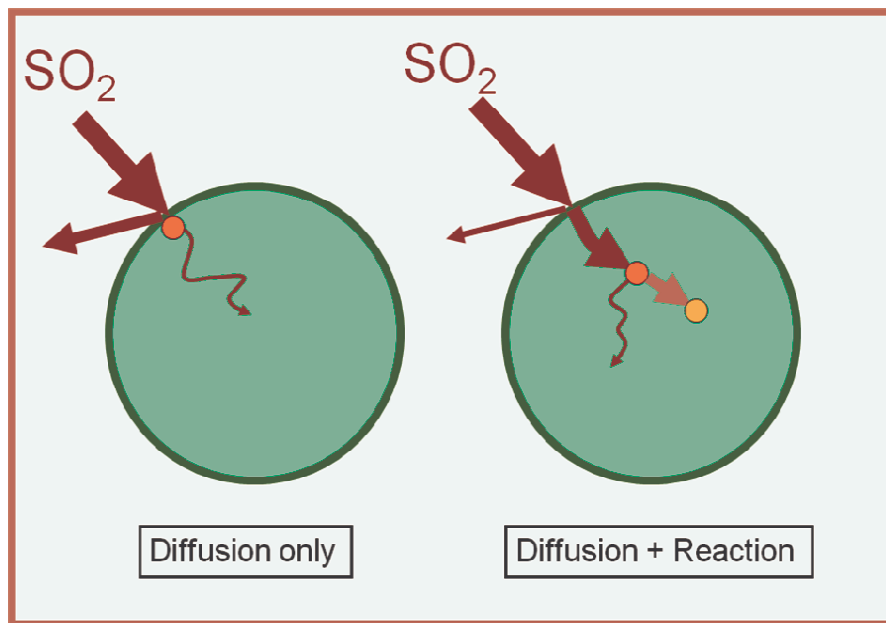


Fig. 4. The schematic description of the enhancement of solubility of SO₂ due to aqueous phase reactions. The left panel shows the processes which affect solubility of SO₂ without considering aqueous reactions. The right panel shows the processes which affect the solubility of SO₂ with considering aqueous reactions, which lead to the enhancement of the solubility of SO₂.

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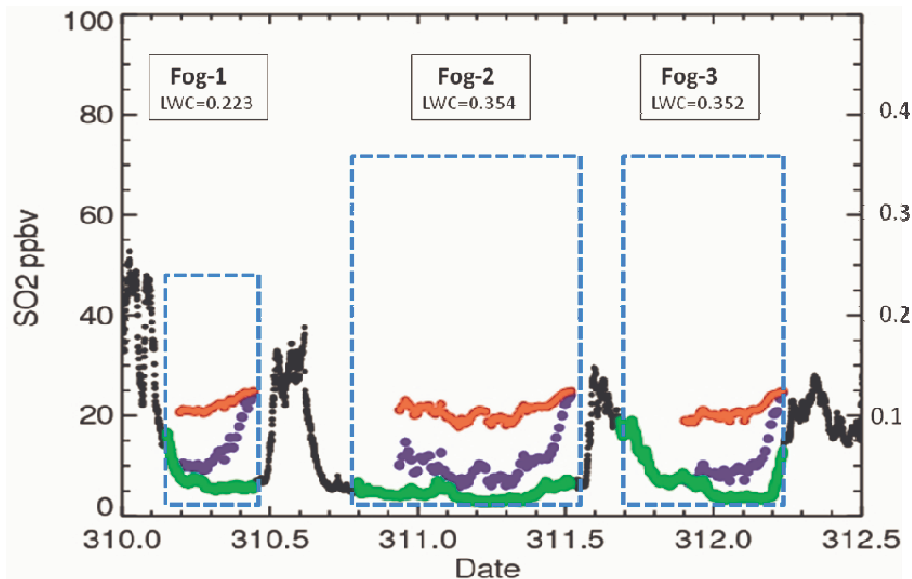


Fig. 5. The measured SO₂ concentrations (black lines during the non-fog periods, and green lines during the 3 fog periods), and the calculated SO₂ concentrations (red lines – without considering the aqueous phase reactions; blue lines – with considering the aqueous phase reactions).

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