In-situ Measurement and CMAQ Simulation of SO_2 over central China

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Background

As a booming economy relying on coal, China has emitted ~ 22 Tg SO_2 in 2009 compared to ~ 5.7 Tg in the U.S. 2009. With SO_2 emission reduction measures in the 11th Five-Year Plans of China (2006-2010) and the clean air action for Beijing Olympics, the total SO_2 emission has decreased from ~ 26 Tg (2006) to ~ 22 Tg (2009).

In the atmosphere, SO_2 is oxidized to sulfuric acid (H_2SO_4) and sulfate aerosols (SO_4^{2-}) mainly through the following reactions:

Gas phase: $OH + SO_2 + M \rightarrow HOSO_2 + M$

Aqueous: $SO_2 \cdot H_2O(aq) + H_2O_2(aq) \rightarrow H_2SO_4(aq) + H_2O(aq)$

SO_2 and its descendent sulfate aerosols not only have profound impacts on the local environment and human health, but also cause regional problems due to long range transports. It is essential to study the SO_2 distribution and evolution in China to improve our understanding of air quality issue in the East Asia.

SO_2 Measurement

In-situ measurement

UV fluorescence Technology: SO_2 molecules are first excited to a high energy level by photon ($h\nu_1$) and decay to a lower energy state with releasing the fluorescence ($h\nu_2$).

$$SO_2 + h\nu_1 \rightarrow SO_2^* \rightarrow SO_2 + h\nu_2$$

The intensity of fluorescence is proportional to the SO_2 concentration.

Instrument: modified TECO 42C trace level SO_2 analyzer, detection limit 0.3 ppbv for 10 seconds average.

Remote sensing

Aura OMI products: The Band Residual Difference (BRD) algorithm is developed at NASA-GSFC/UMBC SO_2 group. Daily SO_2 products are created and published (http://so2.umbc.edu/omi). These products have 4 estimates of SO_2 colum density: Planetary Boundary Layer (PBL), Lower Tropospheric (TRL), Middle Tropospheric (TRM), and Upper Tropospheric and Stratospheric (STL) SO_2 column. In this study, OMI SO_2 PBL products are used.

Spring 2008 Campaign in Central China

A month-long aircraft campaign was conducted in spring 2008 (March 25 to April 22, 2008), Henan province of central China (Figure 1 right). Henan is the most populous region in China with more than 100 million residents, and has heavy industries including coal mines and power plants. Coal is also widely used for domestic cooking and heating.

A Y-7 turboprop transport aircraft (Figure1 left) was used as the airborne measurement platform, and located at Zhengzhou Xinzheng International Airport (IATA code: CGO 34.52N, 113.84E). The ambient air inlet was installed on the rack to the left of fuselage. The cruise speed was ~ 400km/h and spirals from 900 to the 4000 m were conducted during research flights. SO_2 information within the PBL (lower than 1000 m) was retrieved from the measurements during the gradual descending to land at CGO.

Figure 1 Y-7 aircraft at CGO and the location of central China

Summary of Research Flights

Figure 2 demonstrates an example of SO_2 measurements during the flight in 04/05/2008. One spiral over Changyuan (114.68E, 32.20N) is conducted from 1000 to 4000 m showing up to 5 ppbv SO_2, and one descending over Zhengzhou (CGO) observes up to 18 ppbv SO_2 within the PBL (lower than 1000 m).

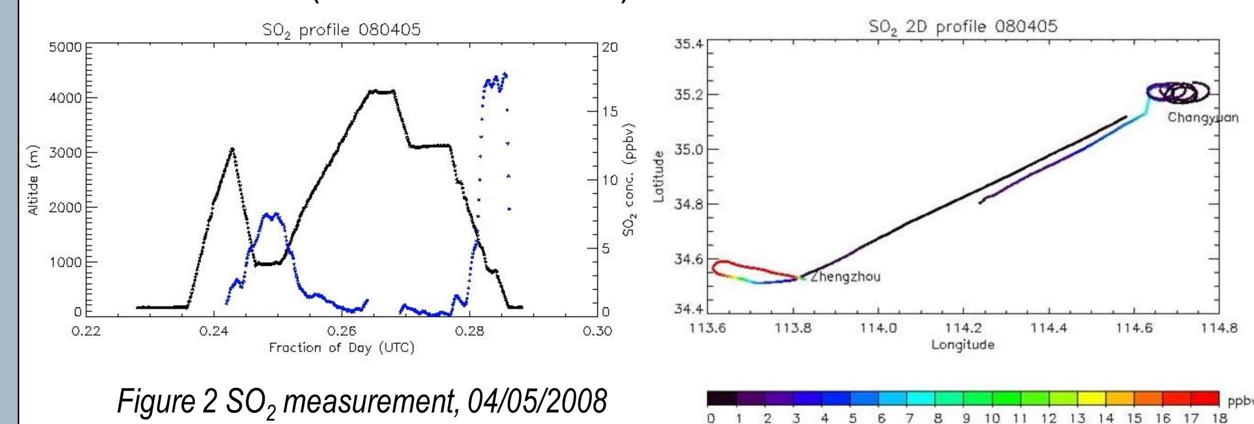

Figure 2 SO_2 measurement, 04/05/2008

During the campaign, total 8 flights have been carried out over different locations such as Changyuan (strong OMI SO_2 signal region) and Yexian (weak OMI SO_2 signal region). Here is the summary of major features of each flight:

Date	Location	Spiral	Descend (CGO)
04/04	Changyuan	2 ppbv SO_2 (1500 m)	15 ppbv SO_2 (1000 m)
04/05	Changyuan	7 ppbv SO_2 (1000~2000m)	15 ppbv SO_2 (<1000 m)
04/09	Huojia	1 ppbv SO_2 (>3000 m)	N/A
04/15	Shangcai	1 ppbv SO_2 (1000~4000 m)	10 ppbv SO_2 (<1000 m)
04/16	Suiping	0.5 ppbv SO_2 (1000~4000 m)	5 ppbv SO_2 (<1000 m)
04/18	Yexian	~0 ppbv SO_2 (1500~4000 m)	3 ppbv SO_2 (<1000 m)
04/20	Weishi	3 ppbv SO_2 (4000 m)	7 ppbv SO_2 (<1000 m)
04/22	Yexian	1 ppbv SO_2 (2500~3500 m)	4 ppbv SO_2 (<1000 m)

* Coordinates: Changyuan (114.68E, 35.20N), Huojia (113.63E, 35.27N), Shangcai (114.26E, 33.25N), Suiping (113.95E, 33.15N), Yexian (113.35E, 33.62N) and Weishi (114.17E, 34.41N)

The SO_2 distribution has highly temporal and spatial variability. Figure 3 demonstrates the average SO_2 profile with high error bar near the surface. The average SO_2 column content is 0.78 Dobson Unit (DU).

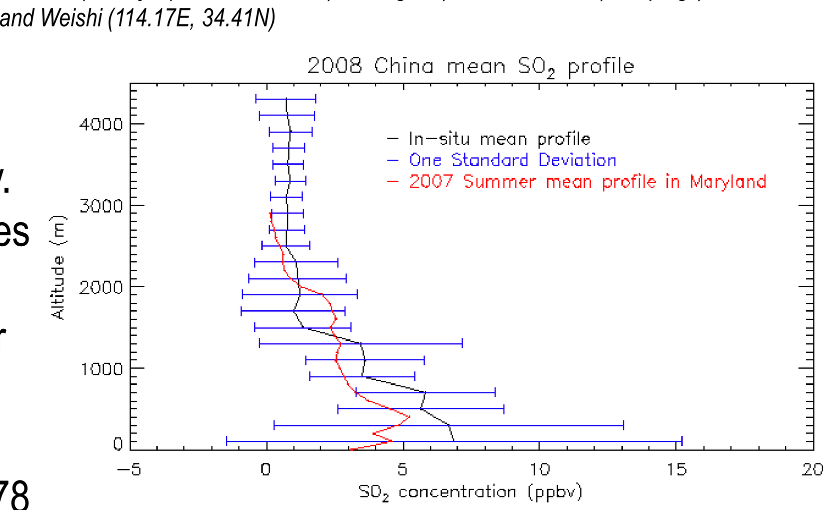

Figure 3 Average SO_2 profile of 2008 China campaign

CMAQ Simulation and INTEX-B Emission Inventory

Numerical simulation of this campaign is conducted using the CMAQ V4.6 model, driven by meteorological fields generated by WRF V3.1. Two nested WRF domains are created with 30 km and 10km resolution respectively (Figure 1 right).

NASA INTEX-B emission inventory is applied to provide emission input data for the CMAQ model. INTEX-B emission data have the resolution of 0.5° x 0.5°, and are interpolated to 30/10 km grids. The SO_2 emissions are allocated into two category, power sector and others (including industrial, residential and transportation emission).

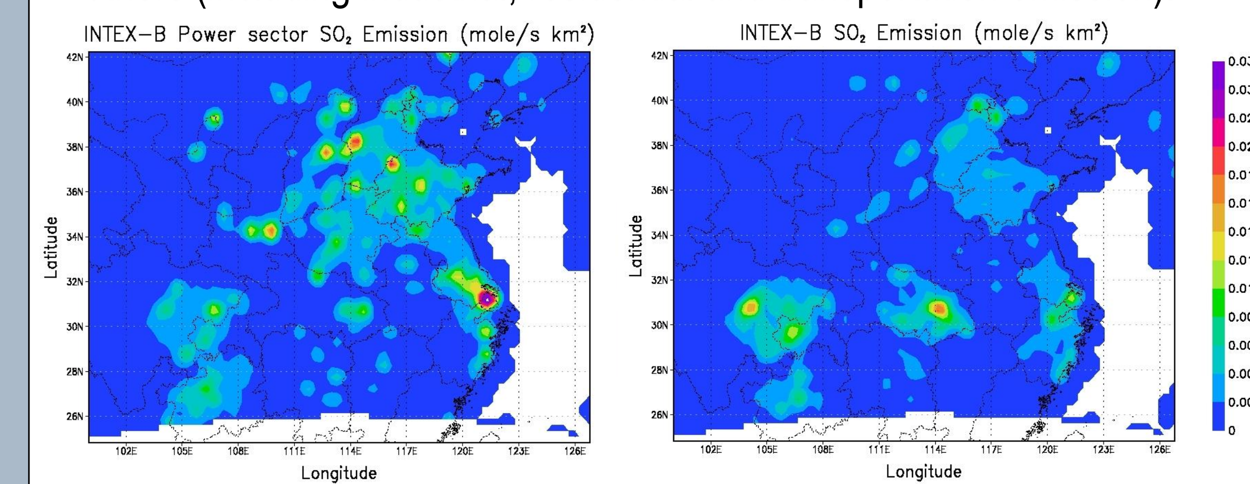

Figure 4 INTEX-B SO_2 emission map (left: power plants, right: others)

The time period of CMAQ simulation is 45 days (03/13 to 04/26) with the first 15 days as spin –up. Three sensitivity runs have been done with different mesophyll resistance (NoMR to MR) value to optimize the SO_2 dry deposition velocity, and different advection (HYAMO to HPPM) of CMAQ model for the calculation of horizontal advections.

List of runs: CMAQ_NoMR_HYAMO, CMAQ_NoMR_HPPM, and CMAQ_MR_HPPM

** NoMR = no MR (MR = 0), MR = MR is set to 8000 s/cm*

CMAQ Results

Figure 5 shows an case of the *in-situ* measurement and CMAQ simulation comparison. CMAQ tends to capture the profile well but fails to reproduce the high SO_2 concentration with the PBL.

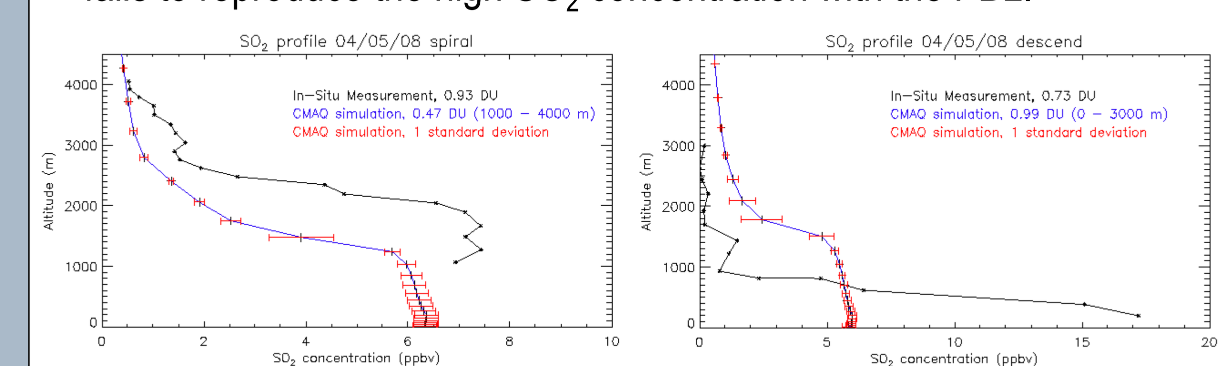

Figure 5 Comparison of in-situ measurement and CMAQ simulation

The modification of SO_2 mesophyll resistance (MR) decreases the SO_2 dry deposition velocity leading to the reduction of dry deposition rate from 600 $g/km^2\ h^{-1}$ to 430 $g/km^2\ h^{-1}$ on 04/18/2008, with decrease in the plains of central domain and increases in the west mountainous area.

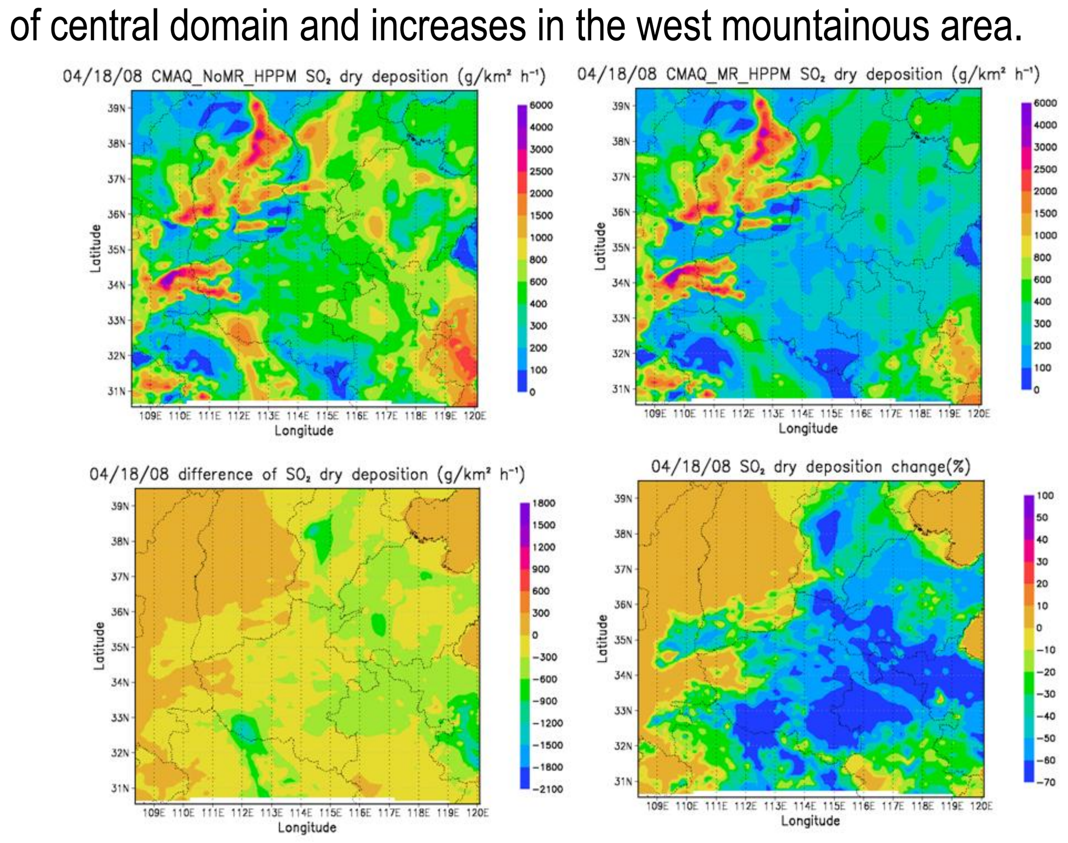

Figure 6 SO_2 dry deposition rate 04/18/2008

The monthly average CMAQ SO_2 profile is calculated based on 3 sensitivity runs (Figure 7). All the runs underestimate the SO_2 concentration within the PBL and overestimate the SO_2 in the free troposphere (FT). And the run with MR correction has better evaluation of surface SO_2 concentration.

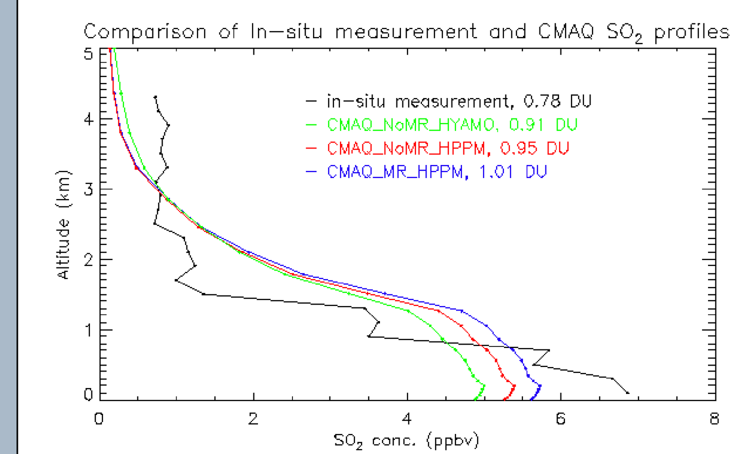

Figure 7 Average in-situ and CMAQ SO_2 profiles

The budget of sulfur compounds from CMAQ silumation is calculated (Figure 8), assuming all SO_2 left in atmosphere is 100% converted to sulfate aerosols.

It reveals that ~11.6 kt of SO_2 (5.8 kt S) and ~4.5 kt of sulfate aerosols (1.5 kt s) are transported daily out of the domain to the East China Sea. The budget is balanced with error less than 3%.

The lifetime of SO_2 is computed as 36.5 hours

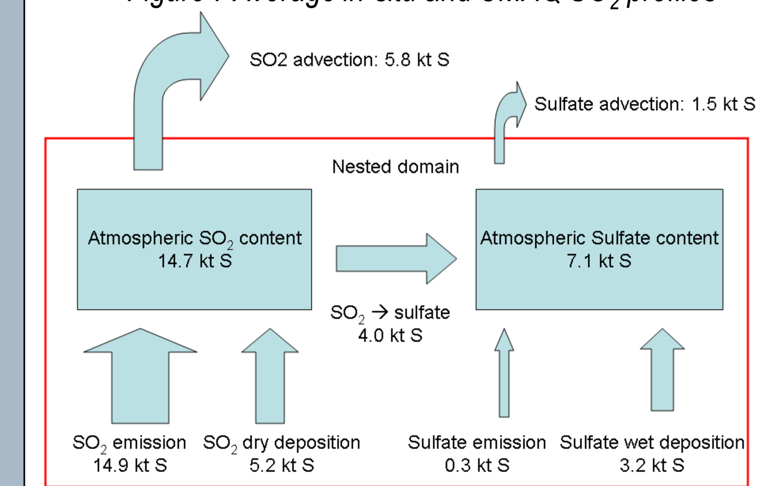

Figure 8 Budget of Sulfur Compounds (CMAQ_MR_HPPM)

The comparison of OMI (SO_2 burden 86.9 kt in 4.60 mkm^2) and CMAQ (SO_2 burden 70.1 kt in 4.48 mkm^2) column map demonstrates CMAQ captures the location of SO_2 plumes well but underestimates the burden by ~20%. ** kt = 10^3 tons, $mkm^2 = 10^6\ km^2$*

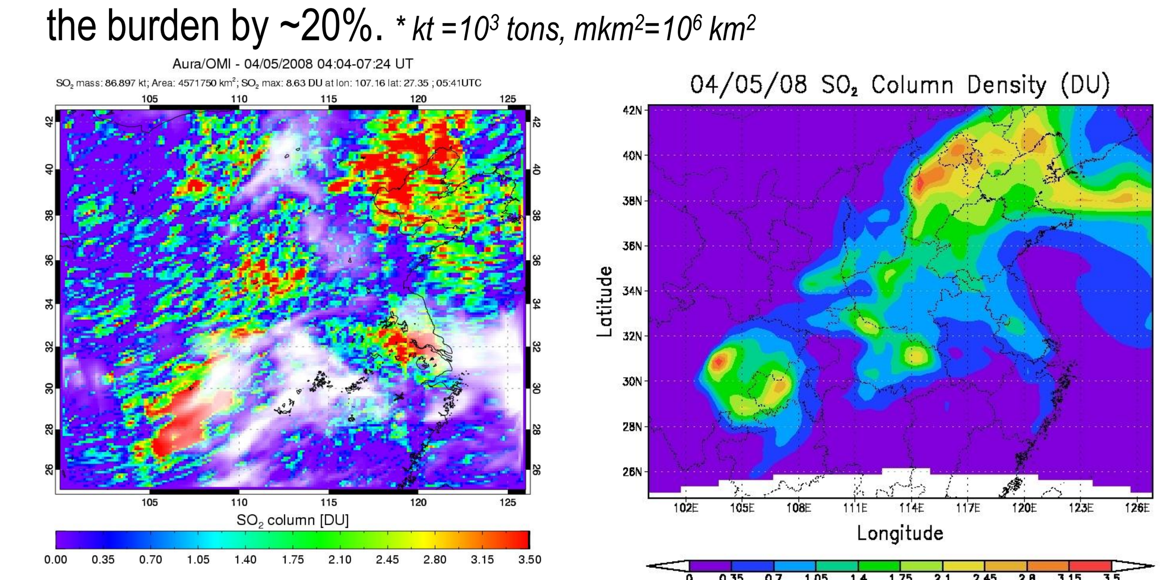

Figure 9 Comparison of OMI and CMAQ SO_2 burden

Conclusion

We simulated SO_2 over central China using CMAQ v4.6 with improvements of the SO_2 dry deposition rate and advection scheme. The comparison of *in-situ* and remotely-sensed measurements with CMAQ simulation shows a reasonably consistent picture of the tropospheric SO_2. WRF-CMAQ has trouble with mixing leading to overestimate of SO_2 in the PBL and underestimate in the FT. On average, *in-situ* observations show more SO_2 aloft (above 3000 m altitude) than CMAQ simulation, indicating substantial export of S from central China. The model also indicates that ~ 50% of the S is exported and the SO_2 lifetime is ~ 1.5 days. In-situ measurements of 2008 demonstrate relatively low SO_2, probably due to emission controls instituted for the 2008 Beijing Olympic Games.

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