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Effects of meteorological conditions on sulfur dioxide air pollution in the North China plain during winters of 2006–2015

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HIGHLIGHTS

- Meteorological conditions with OMI-based high SO_2 days in China are studied.
- Climatological anomaly of meteorological variables in high SO_2 days are quantified.
- Year-to-year winter change of columnar SO_2 distribution is mostly due to SO_2 emission.
- Surface SO_2 distribution has stronger dependence on meteorology than columnar SO2.
- Columnar SO_2 climatology is not representative to surface SO_2 climatology.

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ABSTRACT

The last decade has seen frequent occurrences of severe air pollution episodes of high loading in SO_2 during winters in the North China Plain (NCP). Using satellite data from the Ozone Monitoring Instrument (OMI), chemistry transport model (GEOS-Chem) simulations, and National Center for Environmental Predication (NCEP) meteorological reanalysis, this study examines meteorological and synoptic conditions associated with air pollution episodes during 2006–2015 winters. OMI-based SO_2 data suggest a large decrease (~30% in area average) of SO_2 emissions since 2010. Statistical analysis shows that meteorological conditions associated with the top 10% of OMI-based high SO_2 days are found on average to be controlled by high pressure systems with 2 m s^{-1} lower wind speeds, slightly warmer, 1–2 °C, temperatures and 10–20% higher relative humidities from the surface to 850 hPa. Numerical experiments with GOES-Chem nested grid simulations at 0.5° × 0.667° resolution are conducted for winters of 2009 as a control year, and 2012 and 2013 as years for sensitivity analysis. The experiments reveal that year-to-year change of winter columnar SO_2 amounts and distributions in first order are linearly proportional to the change in SO_2 emissions, regardless of the differences in meteorological conditions. In contrast, the surface SO_2 amounts and distributions exhibit highly non-linear relationships with respect to the emissions and stronger dependence on the meteorological conditions. Longer data records of atmospheric SO_2 from space combined with meteorological reanalysis are needed to further study the meteorological variations in air pollution events and the air pollution climatology in the context of climate change.

1. Introduction

Sulfur dioxide (SO_2) gas is emitted both naturally and anthropogenically through volcanic eruptions and fossil fuel combustion. Estimates by the World Health Organization (WHO, 2001) show that economic health impact (excess mortality and morbidity) due to air pollution of SO_2 is ~43.8 billion RMB Yuan (or ~6.5 billion $) in China. Smith et al. (2011) found that annual emissions of SO_2

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topped ~35 Teragrams (Tg) in the US and Canada, and ~41 Tg SO_2 in Western and Central Europe during the 1970s. However, in the last two decades, North America (United States and Canada) and Europe have been steadily reducing their emissions from 24 Tg to 31 Tg, respectively in 1990 to 17 Tg and 14 Tg, respectively in 2000, and to 15 Tg and 11 Tg, respectively in 2005. These decreasing trends contrast with the increasing trend of SO_2 in many developing countries; annual emissions by sector and fuel types calculated from satellite data show an increasing trend of SO_2 during 1996–2008 and decreasing thereafter in China, with a range of 30–40 Tg per year (Lu et al., 2010).

The distribution of atmospheric SO_2 not only depends on the emission of SO_2, but also is affected by meteorological conditions. Xue and Yin, 2013 found that at Shanghai, SO_2 amounts were negatively correlated with temperature, dew point, relative humidity, wind speed and positively correlated with pressure from October 2004 to September 2012. Bridgman et al. (2002) found that SO_2 surface concentrations in the Czech Republic can be influenced by strong variations of wind direction, wind speed and temperature within the seasons. In Trabzon City, Turkey, Cuhadaroglu and Demirci (1997) found that SO_2 surface concentrations when compared with humidity, wind and temperature have moderate relations in November and December while having weaker relations during January–April. However in Balikesir, Turkey, SO_2 was highly correlated with lower temperatures and lower wind speeds, and less correlated with relative humidity (Ilten and Selici, 2008).

This study investigates how meteorological factors favor high episodes of SO_2 pollution events in China through a combined use of a chemistry transport model (CTM), satellite products of SO_2, and meteorological reanalysis from the National Center for Environmental Predication (NCEP). Many satellite sensors have the capability to monitor atmospheric SO_2 from space, including Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Afe et al., 2004; Richter et al., 2006; Lee et al., 2008, 2009; Zhang et al., 2012), Ozone Monitoring Instrument (OMI) (Krotkov et al., 2008; Carn et al., 2015; Yang et al., 2007; He et al., 2012), and most recently Ozone Mapping and Profiler Suite (OMPS) (Yang et al., 2013). However, these satellite-based SO_2 data in the past have been primarily used for estimating SO_2 emissions and to some extent to evaluate and improve CTM simulations of atmospheric SO_2 (Lee et al., 2009, 2011; Wang et al., 2013). Yang et al. (2013) is among the few studies that have combined meteorological data and satellite SO_2 data from OMPS, to study the role of the atmosphere in an air pollution event for the 2013 winter in China. Numerous studies have also conducted ground-based observations and modeling analyses of air pollution events in China (Chan and Yao, 2008; Lu et al., 2010, 2011; Zhang et al., 2015), just to name a few.

Our study area focuses on the North China Plain (NCP) where the SO_2 emissions have changed rapidly due to the combination of fast economic growth and implementation of air pollution control policies in the last decade for this region (Li et al., 2010). However, these rapid changes of SO_2 emission, together with frequent SO_2 pollution episodes also make the NCP a unique place to combine both satellite data and CTM results to study air pollution meteorology (Yang et al., 2013). Past studies of air pollution meteorology have primarily relied on ground-based observations and numerical models (Fiore et al., 2012). Hence, our joint and new analysis of satellite and model data can reveal (to some extent) how the changing climate (including meteorological conditions) may affect SO_2 air quality, and thus have important implications for predicting future air quality as the climate continues to change (Fiore et al., 2012). The study period of focus is the meteorological winter (December, January and February) during 2006–2015. We describe the data and model in Section 2, model experiment design and approaches in Section 3, results in Section 4 and conclude the paper in Section 5.

2. Datasets and study area

Data used in this study over the NCP (110°E–125°E, 30°N-42°N, Fig. 1) include: (1) Level 3 OMI-best pixel scans, (2) hourly data from a CTM driven by the meteorology from the Goddard Earth Observing System (GEOS); and (3) reanalysis meteorological data from NCEP.

2.1. OMI SO_2

The Ozone Monitoring Instrument (OMI) is a sun-synchronous polar orbiting Dutch/Finnish sensor on the AURA satellite launched on 15 July 2004. OMI is a nadir viewing imaging spectrograph that measures backscattered solar radiation over the 264–504 nm wavelength. The first UV band is from 263 to 311 nm while the second band is from 307 to 383 nm. The absorption spectrum of SO_2 is typically between 305 and 330 nm. OMI uses the 310.8–314.4 nm wavelength to capture the SO_2 in the atmosphere. OMI's pixel size is 13 km (along the orbit) and 24 km (across the orbit) at nadir. (Levelt et al., 2006). OMI's field view of 114° corresponds to a 2600 km wide swath on the surface, which enables daily global coverage. It uses a 2-D Charge Coupled Device (CCD) that can obtain spatial and spectral data simultaneously. Beginning on 25 June 2007, the OMI sensor has been flagged for row anomalies, of which, changes over time. Row anomalies are an anomaly that affects the radiance data in all wavelengths in a particular view direction. Through 28 February 2015, rows 21–54 have been affected by this anomaly which accounts for 39% of the data.

For this study, we used the planetary boundary layer (PBL) SO_2_PBL data from the level-3 OMI/AURA SO_2 data product, OMSO2e (Version 003) (Krotkov et al., 2015) retrieved 4 May 2015, for the meteorological winter time range of 1 December 2005 to 28 February 2015. The current version of the data in the SO_2_PBL is based on a principal component analysis (PCA) of the OMI radiance data (Li et al., 2013). This differs from the band residual difference (BRD) retrieval (Krotkov et al., 2005), as data in high latitudes tend to have larger noise and biases. The OMI SO2 product has been validated over China (Krotkov et al., 2015). The current version of OMI SO2e contains the best pixel of the data. These data have been screened for OMI row anomaly and other data quality flags. During the available 900 days of data, only 870 days contained data. SO_2 accuracy depends on two components: the uncertainty in slant column density (SCD) and the average photon path, characterized by the error in assumed air mass factor (AMF). Also, depending on vertical distribution, aerosols and sub pixel clouds affect AMF.

2.2. GEOS–Chem model

The global chemical transport model GEOS-Chem (v9-01-03) is driven by the Goddard Earth Observing System (GEOS) (Bey et al., 2001) of the NASA Global Modeling and Assimilation Office (GMAO). We use GEOS-Chem for a three-month simulation during the winter months of sulfate aerosols with a spin up of one month with full chemistry. Assimilated meteorological fields from GEOS of NASA Global Modeling Assimilation Office are used to drive GEOS-Chem (Park et al., 2004). The model uses GEOS-5 meteorological fields with 47 vertical levels initially ran at 4° latitude by 5° longitude and then in a nested run at 0.5° latitude by 0.667° longitude. The bottom model layer is ~100 m thick above the surface. The temporal resolution is a six hour average for 3-D meteorological variables and a three hour average for 2-D variables.

The global anthropogenic emissions for NO_x, SO_x, and CO are