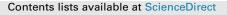
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Anthropogenic sulphur dioxide load over China as observed from different satellite sensors



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^a Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki, Thessaloniki, Greece

^b Royal Netherlands Meteorological Institute (KNMI), De Bilt, The Netherlands

^c Belgian Institute for Space Aeronomy, Brussels, Belgium

^d German Aerospace Center, Remote Sensing Technology Institute, Germany

^e Institute of Environmental Physics, University of Bremen, Bremen, Germany

f NASA Goddard Space Flight Center, Greenbelt, MD, USA

^g Earth System Science Interdisciplinary Center, College Park, MD, USA

HIGHLIGHTS

• SCIAMACHY/Envisat, OMI/Aura and GOME2/MetopA SO₂ columns over China are presented.

• Monthly mean time series of megacities and known power plant locations are examined.

• 90% of the locations studied show a sharp decline in SO₂ emissions this past decade.

• 70% of the locations have a statistically significant annual cycle with highs in winter.

• The implementation of government desulphurisation legislation over China is effective.

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ABSTRACT

China, with its rapid economic growth and immense exporting power, has been the focus of many studies during this previous decade quantifying its increasing emissions contribution to the Earth's atmosphere. With a population slowly shifting towards enlarged power and purchasing needs, the ceaseless inauguration of new power plants, smelters, refineries and industrial parks leads infallibly to increases in sulphur dioxide, SO₂, emissions. The recent capability of next generation algorithms as well as new space-borne instruments to detect anthropogenic SO₂ loads has enabled a fast advancement in this field. In the following work, algorithms providing total SO₂ columns over China based on SCIA-MACHY/Envisat, OMI/Aura and GOME2/MetopA observations are presented. The need for postprocessing and gridding of the SO₂ fields is further revealed in this work, following the path of previous publications. Further, it is demonstrated that the usage of appropriate statistical tools permits studying parts of the datasets typically excluded, such as the winter months loads. Focusing on actual point sources, such as megacities and known power plant locations, instead of entire provinces, monthly mean time series have been examined in detail. The sharp decline in SO₂ emissions in more than 90% -95% of the locations studied confirms the recent implementation of government desulphurisation legislation; however, locations with increases, even for the previous five years, are also identified. These belong to provinces with emerging economies which are in haste to install power plants and are possibly viewed leniently by the authorities, in favour of growth. The SO₂ load seasonality has also been examined in detail with a novel mathematical tool, with 70% of the point sources having a statistically significant annual cycle with highs in winter and lows in summer, following the heating requirements of the Chinese population.

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* Corresponding author. *E-mail address:* mariliza@auth.gr (M.E. Koukouli).

1. Introduction

Sulphur dioxide emissions in China have long interested the scientific community, both from the modelling aspects as well as the study of the remote sensing observations of SO₂. In the work of Lu et al., 2010, it was shown that from 2000 to 2006, the total SO₂ emissions in China increased by 53%, with power plants contributing around half of that increase. Furthermore, a clear North-South gradient was revealed, with Northern provinces increasing their emissions by 85%, whereas the Southern values increased by only 28%. In the same work, it was shown that the growth rate in SO₂ emissions slowed around year 2005 and exhibited a decrease after 2006, mainly due to the widespread application of flue-gas desulfurization devices (FGD) in power plants in response to a new policy of the Chinese government (Lu et al., 2010; Klimont et al., 2013), also reported that, while China is the largest single contributor of SO₂ emissions (supplying around 30% of total global emissions), Chinese SO₂ emissions peaked around 2006 followed by a decline towards 2011. Satellite observations have since verified these findings, as discussed in the work of Li et al., 2010, showing substantial increases in SO₂ and tropospheric column NO₂ OMI/ Aura observations from 2005 to 2007 over several areas in northern China where large coal-fired power plants were built during this period (Zhang et al., 2009), as well as dramatic reductions in SO₂ for 2008. These reductions further confirm the effectiveness of FGD devices, as also extensively discussed by Lu et al., 2010. The recent work of Krotkov et al., 2016, has extended the time period examined to 2014, confirming the continued decrease in SO₂ emissions from 2010 onwards in the context of a further reduction associated with the ongoing global economic crisis.

The inherent potential of different satellite instruments to sense strong SO₂ sources has been long demonstrated in literature; from the era of Nimbus 7/TOMS and GOME/ERS-2 where volcanic eruptions and outgassing (Krueger, 1983; Eisinger and Burrows, 1998) as well as lignite burning from Eastern European power plants (Zerefos et al., 2000) were identified, all the way to OMI/ Aura monitoring of the air quality over Canadian oil sands (McLinden et al., 2012, 2014). Anthropogenic sources such as copper smelters, fires in sulphur plants, coal-fired power plants, heavy metal smelting and so on, have been identified in numerous works and a variety of satellite sensors, see for e.g. the works of Carn et al., 2004, 2007; De Foy et al., 2009; Fioletov et al., 2013; Bauduin et al., 2014, among others. Similarly, volcanic outgassing and eruptive events have been revealed for e.g. by Khokhar et al., 2005, Lu et al., 2013, Carn et al., 2015, among others. In this work, the SO₂ atmospheric load over China as reported by SCIA-MACHY/Envisat, OMI/Aura and GOME2/MetopA is presented. The funding behind this line of investigation originates in the FP7 European Union Marco Polo/Panda project which aims to study air quality over China using a variety of space-born observational tools and modelling techniques [http://www.marcopolo-panda.eu/]. Scientists, local authorities, municipal and provincial level decision-makers are to benefit from the latest that air quality monitoring technology has to offer. The only disadvantage of current satellite instrumentation in air quality science is the fact that the aforementioned sensors all fly on polar orbiting platforms, hence providing, at best, two measurements per location per day. This fact makes it impossible to study the daily variability of the tropospheric pollutants of interest to air quality, an important piece of information for the short lived species, such as the NOx family of gases. However, a number of geostationary orbiting satellites carrying air quality morning instruments are due to become operational during the next decade; Korea with GEMS (Geostationary Environment Monitoring Spectrometer; Lasnik et al., 2014) to be flown on GeoKOMPSAT-2B, Europe (ESA and EUMETSAT) with the UVNS (UV NIR Spectrometer; http://esamultimedia.esa.int/docs/ EarthObservation/Sentinel4_facts_2015.pdf) on Sentinel-4 and NASA with TEMPO (Tropospheric Emissions: Monitoring of Pollution; Zoogman et al., 2016) will cover Asia, Europe and America respectively with hourly measurements on a fine spatial resolution. Those instruments will bring inumerous new possibilities in the field of emission monitoring from space. The work reported here does not aim to inter-compare, evaluate or perform intersatellite validation of the SO₂ estimates obtained by different sensors and algorithms. The paper wishes to present the vertical column density (VCD) data reported by each algorithm and assess their individual strengths and shortcomings in particular for studying the SO₂ atmospheric load over China. The main premise is that almost all these are official datasets that an independent user should be able to simply download and post-process for her/his scientific needs, and this paper will proceed with this line of enquiry in mind. To be more specific, the main goals of this paper are the following:

- i. Firstly to demonstrate the potential of the SCIAMACHY/ Envisat, OMI/Aura and GOME2/MetopA satellite instruments and algorithms to observe the anthropogenic SO₂ load over China. The focus is on presenting the optimum spatial and temporal scales needed in order to achieve an adequate signal-to-noise ratio for each sensor.
- ii. Secondly, to identify SO₂ emitting point sources separately for each instrument and algorithm. Here, attention is given to pixel size and the effect of data gridding onto regular maps. The effect of local point sources on surrounding regions is also discussed.
- iii. Thirdly, to demonstrate that significant trends per point source can be observed when the appropriate postprocessing has been performed. Furthermore, special cases of locations with positive trends, possible dust incursion effects and/or strong seasonal signals, are identified and discussed.

The datasets are presented in Section 2 with the GOME2/ MetopA GDP4.7 SO₂ product, the SCIAMACHY/Envisat SGP5.02 product, the OMI/Aura NASA PCA product and the OMI/Aura BIRA algorithm product discussed in Sections 2.1, 2.2, 2.3 and 2.4 respectively. In the various sub-sections of Section 3, we describe the analysis and associated findings. Finally, the main findings and conclusions are summarized in Section 5.

2. The datasets

In this work, sulphur dioxide columns reported as Vertical Column Densities, *VCDs*, by the SCIAMACHY/Envisat, OMI/Aura and GOME2/MetopA instruments are studied. Traditionally, in comparison and evaluation works such as the one by Fioletov et al. (2013), the Slant Column Density [SCD] is examined so that the application of a common air mass factor [AMF] will lead to comparable results. However, as stated in the introduction, in this work we aim to demonstrate the benefits that each reported VCD provides without delving into the intricacies of each algorithm.

The domain considered extends from 60° to 135°E and from 20° to 55°N and covers all of China as well as parts of the Far East. The data were filtered for high Solar Zenith Angle, *SZA*, of >70°, cloud fraction, of >0.2 and also SO₂ algorithm flagging. The filtered data were then averaged onto a 0.25° × 0.25° monthly grid using a 0.75° smoothing average box.

The provenance of each dataset is discussed in the following four sub-sections.

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