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Natural and anthropogenic contributions to long-term variations of SO_2, NO_2, CO, and AOD over East China

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ABSTRACT

Concentrations of atmospheric pollutants over East China have varied considerably during the past decades. These variations are partly due to variations of human activities, e.g., increasing energy consumption and implementation of government emission control policies, and partly to natural fluctuations. This study aims to separate the effects of natural processes and anthropogenic activities on the increase/decrease of the concentrations of some of the most important pollutants (SO_2, NO_2, CO and aerosols) over East China in the last decade. This was achieved by the comparison of the temporal variations in long-term time series of satellite-retrieved aerosol optical depth (AOD) and vertical column densities (VCDs) of SO_2, NO_2, and CO, with those in model-simulated time series of the natural variations only. The latter were created by the use of the same anthropogenic emissions throughout the whole simulation, while using re-analysis data (MERRA) to describe meteorological processes and natural emissions. Thus, the comparison between observed and simulated temporal variations reveals the effects due to anthropogenic emissions only, assuming that the atmospheric processes affect natural and anthropogenic species in the same way. In the analysis, a Kolmogorov–Zurbenko (KZ) filter is used to extract long-term components from both the observed and simulated data and normalization to the situation at a certain reference point is used to eliminate bias between observations and simulations. By this new method, natural and anthropogenic contributions to long-term variations of trace gases and AOD are quantitatively estimated. The results show that NO_2 VCDs increased from 2004 to 2011 by 76% and of the overall increase in this period only 1% ± 1% was attributed to natural factors, 99% ± 1% attributed to anthropogenic factors. AOD increased by 24% between 2001 and 2011 and of the overall increase 24% ± 32% was due to natural factors and 76% ± 32% was due to anthropogenic factors. SO_2 VCDs decreased by 15% from 2007 to 2013, natural and anthropogenic factors contributed respectively 16% ± 14% and 84% ± 14% to the overall decrease in this period. CO decreased since 2003 with 13% and of the overall decrease 6% ± 6% was due to natural factors and 94% ± 6% was due to anthropogenic factors.

1. Introduction

China has been experiencing increasing air pollution during several decades, especially in densely-populated areas such as the Beijing–Tianjing–Hebei (BTH) region and the Yangtze River Delta (YRD) region. Most of the heavily polluted regions are located in East China, which is experiencing rapid industrialization and urbanization (Huang et al., 2013; Lin et al., 2010). The Chinese government has implemented legislation to reduce the emission of air pollutants by application of improved combustion technologies, promoting

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renewable energy technologies, and tightening emission control policies (Xu, 2011; ChinaFAQs project, 2012). However, increasing energy demands together with adverse effects of meteorological conditions and synoptic situations on air quality (Miao et al., 2017) render it difficult to assess the effects of government regulations to improve air quality in China. van der A et al. (2017) suggested that without air quality regulations, sulphur dioxide (SO_2) concentrations would be almost three times higher and nitrogen dioxide (NO_2) concentrations would be at least 30% higher than they are today. Although much progress has been made, improvement of China's air quality still has a long way to go.

The most common pollutants relevant to air quality and human health include SO_2, NO_2, carbon monoxide (CO), ozone (O_3), and aerosols. These pollutants are mainly emitted by anthropogenic activities, except for O_3, whose variation depends on multiple factors such as emission of precursors from anthropogenic sources, atmospheric oxidation and stratospheric intrusion (Lelieveld and Dentener, 2000; Monks et al., 2015; Sillman, 1995; Sun et al., 2016; Sun et al., 2017). Satellite instruments can monitor atmospheric species from space and provide spatial and temporal information about their concentrations and, for short-lived species, their source regions. SO_2 and NO_x data derived from the Ozone Monitoring Instrument (OMI) are used to investigate their long-term variations in China and the effectiveness of emission control policies on the concentrations of these species (Fioletov et al., 2016; Krotkov et al., 2016; van der A et al., 2017). They found that the increase of SO_2 and NO_2 concentrations in China has been significantly constrained by air quality regulations. Gu et al. (2013) found a significant increase in tropospheric NO_2 from 2005 to 2010 over East China using satellite observations. Liu et al. (2011) used CO column data from the Scanning Imaging Absorption SpectroMeter for Atmospheric ChartographY (SCIAMACHY) and Measurements of Pollution in the Troposphere (MOPITT) to evaluate model results for eastern China, and found that model simulations systematically underestimate the true atmospheric CO concentrations. In addition, OMI, MODerate Resolution Imaging Spectroradiometer (MODIS), multi-angle imaging spectroradiometer (MISR), and along-track scanning radiometer (ATSR-2/AATSR) aerosol optical depth (AOD) data were used to investigate long-term variations of aerosols over China and found an increasing trend of AOD before 2011 then followed by a decreasing trend (de Leeuw et al., 2018; Qu et al., 2016; Xin et al., 2011; Zhang et al., 2017; Zhao et al., 2017).

Previous studies have revealed that air pollutant concentrations in China vary considerably over time. Factors that cause these variations include short term, seasonal and long term meteorological influences, and variations in both natural and anthropogenic emissions (Pearce et al., 2011; van der A et al., 2008; Zhang et al., 2016b). Natural factors that influence long-term variations of air pollutants mainly include long-term variations in meteorological conditions (such as decadal variations in precipitation and monsoon strength, causing wet removal, and wind speed facilitating dispersion as well as generation and transport) (Zhao et al., 2010), climate change (global warming) (Cai et al., 2017; Zhang et al., 2018; Zhao et al., 2018), and long-term natural emission variations (Li and Xie, 2014), while anthropogenic factors include changes in the amount of emissions induced by human activities (such as economic development, increasing demand of energy, urbanization processes and government emission control measures) (Wang et al., 2017). However, few studies have quantitatively separated natural and anthropogenic contributions to the temporal variations of trace gases and aerosols which affect air quality.

Government emission control policies should be established based on a quantitative understanding of the contributions of natural processes and anthropogenic activities to air quality. Quantifying the impacts of natural and anthropogenic factors on long-term variations of air pollutants is critical for guiding emission control measures. Fu et al. (2016) reported that from 1980 to 2010, climate change alone could lead to a decrease in wintertime $PM_{2.5}$ concentrations by 4.0–12.0 $\mu g\,m^{-3}$ in northern China, and an increase in summertime $PM_{2.5}$ concentration by 6.0–8.0 $\mu g\,m^{-3}$ in the same region. Mao et al. (2016) indicated that the decadal trend of black carbon concentrations from 1980 to 2010 was mainly driven by changes in emissions, while interannual variations were dependent on variations of both meteorological parameters and emissions.

Modelling is a useful tool to investigate factors controlling the variations in air pollutants in China, but it is difficult to get accurate frequently updated emission data. Models depend on emissions, but these emissions vary in time, and emission inventories are updated at intervals of years during which emissions may change substantially with changing economic conditions, meanwhile emissions are also affected by meteorological factors. Emissions of pollutants over China vary in terms of amount and trends among various emission inventories. For example, following the EDGAR emission inventory, the SO_2 emission increased between 2001 and 2008, while, in contrast, the REAS inventory shows a decrease since 2006 (Wang et al., 2017). Top-down emissions derived from satellite observations provide information reflecting the actual state on a time scale of about 1 month (Ding et al., 2017; Koukouli et al., 2018; Mijling and van der A, 2012). However, this information does not discriminate between natural and anthropogenic sources, which is the focus of the current study where these contributions are investigated by comparing satellite-observed column amount of trace gases and AOD with model simulations in which emission rates are fixed.

In this study, we use time series of SO_2, NO_2, and CO VCDs and AOD over East China, retrieved from several satellite sensors for a period of 10–15 years. The same quantities were simulated with the Model for Ozone and Related Chemical Tracers version 4 (MOZART-4) (Emmons et al., 2010). However, to simulate effects of natural variations on these concentrations, the anthropogenic emissions used in the simulations were the same for the whole period, i.e. fixed at their values in 2006, but actual meteorological parameters were used. Hence, any variations in the simulated species are due to variations of natural factors influencing emissions and subsequent processes. Natural factors are assumed to influence model simulations and satellite observations in the same way. In that case the influence of natural factors can be derived from the model simulations as described above and used to separate effects of natural and anthropogenic factors on long-term variations in satellite observations of trace gases and aerosols by comparison of the observed and modeled variations. Long-term variations in the observed and simulated data were then extracted using the low-pass Kolmogorov–Zurbenko (KZ) filter (Rao and Zurbenko, 1994). In the analysis, only data were used after normalization to the values in a certain reference situation, i.e. anomalies were used to determine the anthropogenic and natural contributions from the combined satellite observations and model-simulated natural variations.

2. Data and methods

2.1. Satellite SO_2 data

For this study, we use the monthly OMI level-3 aggregated tropospheric column SO_2 retrieved by the BIRA-IASB (Belgian Institute for Space Astronomy) which are calculated using an advanced Differential Optical Absorption Spectroscopy (DOAS) scheme combined with radiative transfer algorithm (Theys et al., 2015). It should be noted that since June 2007, the radiance data of OMI for some particular viewing directions have been corrupt, a feature known as the OMI row anomaly. Hence, the suggested row anomaly filtering (http://projects.knmi.nl/omi/research/product/rowanomaly-background.php, last access: 26 June 2018) has been used in SO_2 retrieval. The horizontal grid size of the OMI level-3 SO_2 VCDs is 0.125° × 0.125° and the precision of the data is within 12% over East China (Theys et al., 2015). The SO_2 VCDs data were acquired by personal communication. Data for the period of 2005–2014 were analyzed.