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 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–Tianjin–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$_2$ 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 model satellite 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; Xiong 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 μg m$^{-3}$ in northern China, and an increase in summertime PM$_{2.5}$ concentration by 6.0–8.0 μg 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; Koukoulis 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.
2.2. Satellite NO2 data

In this study, satellite-based monthly tropospheric NO2 columns for the period 2002–2014 are used. Data are derived from the level-2 product of the SCIAMACHY (2002–2012) and the Global Ozone Monitoring Experiment 2 (GOME-2) (2007–now), which are retrieved by KNMI and BIRA-IASB with the DOAS technique (Boersma et al., 2004). The data can be downloaded from the website of the Tropospheric Emission Monitoring Internet Service (TEMIS, http://www.temis.nl). For GOME-2 data only pixels measured in the forward mode are used. Retrieval algorithms for NO2 and its expected uncertainty for SCIAMACHY are similar to those for GOME-2 (Irie et al., 2012; Richter et al., 2011). The biases in SCIAMACHY and GOME-2 NO2 data over China are −5 ± 14% and +1 ± 14%, respectively, which are small and insignificant (Irie et al., 2012). The overpass times of SCIAMACHY (1000 LT) and GOME-2A (0930 LT) are close enough to avoid potential bias between these data (Irie et al., 2012). In Section 3.4, we also confirmed that bias between the two instruments can be efficiently eliminated by normalization. We averaged the data from the two instruments for the overlapping period 2007–2012. The grid size for these data is 0.25° × 0.25°.

2.3. Satellite CO data

Monthly CO columns are obtained from the version 3, level-3 product of the MOPITT instrument. MOPITT is a multi-channel thermal-infrared (TIR) and near-infrared (NIR) instrument on board the EOS-Terra satellite with horizontal spatial resolution of 22 km × 22 km and swath width around 640 km. In this study we use CO total columns derived from MOPITT TIR observations using CO absorption lines at 4.6 μm. The retrievals are performed using an optimal estimation technique, the maximum a posteriori solution (MAP) (Deeter, 2003; Emmons et al., 2004). MOPITT-retrieved CO data have been extensively validated by Emmons et al. (2004, 2007) and are within the target accuracy and precision (10%) for much of the planet. The grid resolution of the MOPITT level-3 CO product is 1° × 1°. In this study, monthly CO data for the period of 2000–2014 are used (https://eosweb.larc.nasa.gov/project/mopitt/mopitt_table).

2.4. Satellite AOD data

Monthly AOD data were obtained from the MODIS/Terra level-3 dark target (DT) product of Collection 6 (C6) (Levy et al., 2013) with a spatial resolution of 1° × 1°. The C6 DT expected error is ±(0.05 + 0.15%AERONET) over land and + (0.04 + 0.15%AERONET), −(0.02 + 0.15%AERONET) over sea relative to the AERONET optical thickness (%AERONET) (Levy et al., 2013). Validation over China, using AERONET AOD as reference, shows that the good performance of the MODIS DT and DB (deep blue) merged data set for AOD up to 1.8 with a light overestimation (de Leeuw et al., 2018; Zhang et al., 2016a). The DB and merged (DT + DB) AOD products have compared with DT AOD data and the results show that differences between natural/anthropogenic contributions derived from the three datasets (DT, DB, and DT + DB merged) are negligible. The MODIS AOD data were acquired from NASA Goddard Earth Sciences Distributed Active Archive Center through the website of NASA’s Goddard Space Flight Center (http://ladsweb.nascom.nasa.gov/). In this study, we used AOD data for the period of 2000–2014.

To improve the quality of SO2, NO2, CO, and AOD data, we excluded data with a solar zenith angle larger than 75° or with a relatively high cloud radiance fraction (>50%).

2.5. MOZART-4 model

We performed a model experiment to simulate SO2, NO2, CO, and AOD using the MOZART-4 global chemical transport model with assimilated meteorology. To be consistent with satellite observational data, tropospheric aggregated (from surface to ~13 km altitude) concentrations of SO2 and NO2 were used, while total column aggregated concentrations of CO and AOD were used. A detailed description of the standard version of MOZART-4 is given by Emmons et al. (2010). Model simulated concentrations have been widely evaluated through the comparison with satellite observations (Cao et al., 2013; Emmons et al., 2010; Su et al., 2012). These evaluations show that MOZART-4 can reproduce well the spatial and temporal variability of tropospheric chemical composition. It is suitable for many tropospheric investigations on the regional to global scale. MOZART-4 is an offline model, in this study, it is driven by the assimilated meteorological data from NASA’s Modern-Era Retrospective Analysis for Research and Applications (MERRA) with a resolution of 1.9° × 2.5° (latitude × longitude), and 56 vertical levels. The top of the model level is located at about 2 hPa. We conducted a 17-year simulation (1998–2014) with the first two years (1998–1999) as spin-up time.

In the experiment, anthropogenic emissions over Asia are from the Regional Emission inventory for Asia (REAS) (Kurokawa et al., 2013) and fixed to the 2006 level, while the inputs of meteorological parameters and natural emissions are changing over time. Biogenic emissions are calculated online based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). Biomass burning emissions from the Global Fire Emissions Database, version 2 (GFEDv2) (van der Werf et al., 2006) are used. Emissions of NOx from soil (Yienger and Levy, 1995) and lightning (Emmons et al., 2010; Price et al., 1997), as well as emissions of sea salt (Mahowald et al., 2006a) and dust (Mahowald et al., 2006b) are calculated online. Thus, the model results as they are used here show the natural effects of changing meteorological conditions and larger scale synoptic conditions on the VCDs of trace gases and AOD for a situation in which the anthropogenic contributions to the emissions are fixed to the level in 2006.

2.6. Kolmogorov–Zurbenko filter

A KZ(13,3) filter is used to extract long-term variations of air pollutant concentrations over East China. The KZ filter is based on the assumption of Rao and Zurbenko (1994) that a time-series of atmospheric concentration data (in their case O3) can be represented by:

\[ X(t) = \epsilon(t) + S(t) + W(t) \]

where \( X(t) \) is the original time series, \( \epsilon(t) \) is the long-term trend component, \( S(t) \) is the seasonal change, \( W(t) \) is the short-term variation, and \( t \) is time. The long-term trend component is attributable to long-term changes in meteorological conditions as well as changes in emissions in response to policy measures and economic conditions. The seasonal component is a result of changes in the solar angle, and the short-term component results from meteorological and short-term emission fluctuations (Rao and Zurbenko, 1994; Rao et al., 1997; Wise and Comrie, 2012).

The KZ(m,p) filter is defined as \( p \) applications of a simple moving average of \( m \) points. The moving average (each iteration) can be expressed as:

\[ Y_i = \frac{1}{m} \sum_{j=-k}^{k} X_{i+j} \]

where \( X \) is the original time series, \( k \) is the number of values included on each side of the targeted value \( i \), the window length \( m = 2k + 1 \) (Milanclus et al., 1998). The output of the first pass \( Y_1 \) then becomes the input for the second pass, and so on. Adjusting the window length \( m \) and the number of iterations \( p \) makes it possible to extract different scales of temporal variations (Eskridge et al., 1997). To filter periods of less than \( N \) months, the following criterion is used:

\[ m \times p^{1/2} \leq N \]
Fig. 1. Multi-year averaged tropospheric SO$_2$ (2005–2014), tropospheric NO$_2$ (2002–2014), total column CO (2000–2014), and AOD (2000–2014) in China derived from satellite observations (left panel) and MOZART-4 simulations (right panel). The elevated VCDs over the region in East China marked by the red box are further investigated as regards temporal variations and controlling factors. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
In this study, a $KZ_{(13,3)}$ filter (13-month length $[m]$ with three iterations $[p]$), which can remove cycles of $<23$ months ($13 \times 3^1 < 23$ months, i.e. $\sim 1.9$ years), is used to extract the long-term trend from both satellite-observed and model-simulated $SO_2$, $NO_2$, CO, and AOD data. The result is the long-term component of the original data:

$$s(t) = KZ_{(13,3)}$$  

(4)

3. Results and discussion

3.1. Spatial distribution of air pollutants over East China

The satellite-derived and model-simulated multi-year averaged columns of $SO_2$ (2005–2014), $NO_2$ (2002–2014), CO (2000–2014), and AOD (2000–2014) are shown in Fig. 1. High pollutant VCDs are visible mainly over East China (denoted by the red box, latitude: 27.5°–42°N, longitude: 110°–125°E), especially for short-lived species such as $SO_2$ and $NO_2$. In addition, the Sichuan Basin and Pearl River Delta are also heavily polluted areas, albeit that the VCDs are substantially lower than over East China. In this study, we will focus on East China.

In East China, $SO_2$ and $NO_2$ are mainly concentrated over the Beijing–Tianjin–Hebei region, northwest Shandong, northeast Henan, and the YRD region, but with substantial differences between the spatial variations of the $SO_2$ and $NO_2$ VCDs. Because $SO_2$ is mainly discharged by coal combustion, $SO_2$ VCDs are highest over the North China Plain, where numerous power plants and coal-consuming industries exist (Krotkov et al., 2016; van der A A et al., 2017). $NO_2$ VCDs over the YRD are comparable to those over the North China Plain, mainly because there is a large number of vehicles in the YRD region. Besides, the lifetimes of $SO_2$ and $NO_2$ are relatively short due to their high chemical reactivities or fast dry deposition velocities (Seinfeld and Pandis, 2006). Due to these short lifetimes, $SO_2$ and $NO_2$ are not transported over long distances, and the highest concentrations are observed. The spatial distribution of CO is more uniform because of its relatively longer lifetime. High AOD values (> 0.8) occur over roughly the same regions where $NO_2$ columns are high, but due to the much longer atmospheric residence time of aerosol particles, several days to a week, as opposed to a few hours for $NO_2$ and $SO_2$, they are transported over larger distances. High AOD values are also observed over the eastern parts of Hubei, Hunan and Sichuan provinces and Chongqing.

The spatial distributions of the simulated $SO_2$, $NO_2$, and CO VCDs are similar to those retrieved from the satellite observations. However, the VCD values are quite different, likely because the simulation does not use actual emissions. Meanwhile, the lower spatial resolution of the model simulations as compared to that of the satellite data also contributes to the bias of model simulations. It is noted that the modeled spatial distribution of AOD, with high values in the central part of the study area, deviates from that observed by MODIS. However, these biases will not affect our discussion about the controlling factors on the long-term variation of pollutants in Sections 3.2 and 3.3, since the model simulation aims to derive the influence of natural factors on long-term variation of species concentrations from the temporal variations of the differences with respect to a reference situation which are independent of this bias.

Since we fixed the anthropogenic emissions to 2006, model simulations were compared with satellite observations for this year. Correlation coefficients ($R$) between model simulations and satellite observation for $SO_2$, $NO_2$, CO, and AOD in 2006 are 0.96, 0.94, 0.9, and 0.62, respectively. Normalized mean biases (NMB) for these species are 276%, −25%, 12%, and −44%. Results show that the model is able to reproduce the variations of trace gases and AOD, but significant biases exist between model and observation. The large NMB for $SO_2$ is probably because the REAS $SO_2$ emission was overestimated in 2006, it is the highest emission among several emission inventories (Wang et al., 2017). However, these biases will be excluded by normalization. In Section 3.4, we will use an example to demonstrate how to eliminate bias by normalization. The NMB was calculated by:

$$NMB = \frac{\sum_{i=1}^{N} (M_i - O_i)}{\sum_{i=1}^{N} O_i} \times 100\%$$  

(5)
where $M_i$ represents the simulated value, $O_i$ represents the observational data, and $N$ denotes the number of data pairs.

3.2. Temporal variations of pollutants over East China

The time series of the satellite retrieved and model-simulated VCDs of SO$_2$, NO$_2$, CO, and AOD over East China were obtained by spatially averaging for the predefined East China region. Long-term variations of these trace gases and AOD were extracted by $KZ_{13,3}$ filter (described in Section 2.6) and they were normalized with respect to the values at the start of each dataset (Fig. 2), which is taken as a reference situation. Variations in the normalized model results can be attributed to long-term changes in natural factors as compared to the reference situation, since anthropogenic emissions were fixed at the 2006 level and the $KZ_{13,3}$ filter removes short-term and seasonal variations. Therefore, the larger the deviations from 1, the more the modeled concentrations are affected by changes in natural conditions as compared to the reference situation. On the other hand, normalized satellite retrievals reveal the actual deviation of the long-term concentration with respect to the reference condition, which can be attributed to variations in both natural and anthropogenic factors. Hence, the difference between the variations in the satellite-observed and the modeled normalized time series provides information on the contributions of anthropogenic factors.

Normalized SO$_2$ simulations show a small increase since 2009 (Fig. 2a, red line). On the other hand, the normalized SO$_2$ observations (dark blue line in Fig. 2a) show an overall decrease between 2007 and 2014, with an increase between early 2009 and the end of 2011, followed by a further decrease, indicating a decreasing trend of anthropogenic SO$_2$ emissions since 2007 (van der A et al., 2017). The satellite observed SO$_2$ reduction rate from 2007 to 2013 is 15%, which is comparable to the reduction rate of 12.1% and 17.5% from 2010 to 2014 found by Wang et al. (2014) and Xia et al. (2016). This decrease is likely due to the installation of desulphurization devices in many power plants to reduce SO$_2$ emissions since the start of the desulphurization program of China’s 11th Five-Year Plan in 2005/2006 (Lu et al., 2010).

The normalized tropospheric NO$_2$ data merged from two satellite instruments (SCIAMACHY and GOME2) show an obvious increase between 2004 and 2011 (Fig. 2b, dark blue line), which agrees fairly well with other studies (Irie et al., 2016; Krotkov et al., 2016; Wang et al., 2017; Xia et al., 2016). The NO$_2$ VCDs increased 76% from 2004 to 2011, close to the increase rate of 71% from 2005 to 2011 by Xia et al. (2016). However, the simulated NO$_2$ VCDs (red line) show very little variation during the whole simulation period. This indicates that natural factors have little effect on long-term variations of NO$_2$, as opposed to the stronger influence of anthropogenic activities on NO$_2$ columns. The increase in NO$_2$ VCDs is attributed to fast urbanization in East China (Huang et al., 2013), which leads to rapid increases in the number of vehicles, power plants and industrial facilities, which are important sources of NO$_2$. Following the Multi-resolution Emission Inventory for China (MEIC) data, 25% of NO$_2$ was released by vehicles, 32% by power plants and 39% by industry in 2010 (Zhao et al., 2015). Following the regulation of the 12th Five-Year Plan and the “Action Plan”, NO$_2$ filtering systems have been installed at power plants and heavy industrial installations. Such policies contribute largely to the observed NO$_2$ decrease since 2012 (Liu et al., 2016; van der A et al., 2017; Wang et al., 2017). The decreasing rate of NO$_2$ VCDs from 2011 to 2013 was 24% in this study, comparable to 18% from 2011 to 2014 by Xia et al. (2016).

The normalized CO satellite observations (Fig. 2c, dark blue line) show a decreasing trend from 2003 to 2013 with a reduction rate of 13%, whereas those from the MOZART-4 simulation (red line) are fairly constant (except from minor year-to-year fluctuations), indicating a decrease of anthropogenic CO during this period. The downward trend of CO emissions over China in recent years has been confirmed by both in situ and satellite observations (Li et al., 2017; Worden et al., 2013; Yin et al., 2015; Yumimoto et al., 2014). This is mostly due to improvements in combustion technologies, recycling of industrial coal gases, strengthened vehicle emission standards, and energy structure adjustments (Li et al., 2017; Yin et al., 2015; Yumimoto et al., 2014). Besides, clean energies such as hydroelectric, wind and nuclear energy are increasingly used in China. In contrast, CO emissions in India, South, Southeast, Central, and Russian Asia increased significantly (Li et al., 2017) which indicates that the decrease in CO VCDs over East China was mostly influenced by the decrease of local emissions. The simulated CO VCDs show relatively small variation, which indicates that natural factors cause short-term fluctuations of CO VCDs but do not increase or decrease CO VCDs on longer timescale.

The normalized satellite-derived AOD data show an overall increase between 2001 and 2011, with an initial increase followed by periods with decreasing and increasing AOD (Fig. 2d, dark blue line), consistent with the conclusion by de Leeuw et al. (2018). The increase rate of AOD from 2001 to 2011 is 24%. Since simulated results (Fig. 2d, red line) show little variation during this period, the varying observations probably indicate an increase of anthropogenic aerosols. It is worth noting that variations of AOD may not be consistent with aerosol mass concentrations. AOD is the column-integrated aerosol extinction, which is affected by many factors such as boundary layer height, relative humidity, temperature, wind speed and direction and long-range transport (Crumeyrolle et al., 2014; Guo et al., 2017; Huang et al., 2014; Miao et al., 2017), whereas aerosol is characterized by PM$_{2.5}$, i.e. the integrated dry mass of aerosol particles with an ambient aerodynamic diameter of 2.5 μm, measured near the surface. Hence AOD and PM$_{2.5}$ are very different parameters which may be related through multi-parameter models (Ma et al., 2016) but have different temporal behaviour. Satellite-observed AOD decreased from 2012 to 2013, with a decrease rate of 11%, which is probably related to government emission control policies. A drastic decrease of observed AOD as well as gas species (SO$_2$, NO$_2$, and CO) appeared around 2008, whereas model simulations did not deviate largely during this period. This implies that the decrease of AOD and trace gas columns was caused by the decrease of anthropogenic emissions rather than natural effects. This decrease was attributed to the economic recession (Lin et al., 2010; Schneider and van der A, 2012).

3.3. Contributions of natural and anthropogenic factors to pollutant variations

In the previous section, we have qualitatively discussed the impact of both natural factors and human activities on VCDs of SO$_2$, NO$_2$, CO, and AOD. In this section, we quantify these contributions to the long-term variations of those species. From the time series of the normalized long-term components of satellite observations (Fig. 2), we can divide these long-term variations into two periods. One is the increasing period, when VCDs and AOD increase due to the increase of anthropogenic emissions; the other is the decreasing period, when VCDs and AOD decrease because government emission control policies take effect. Natural and anthropogenic contributions are calculated for each of these two periods. For each period, species concentrations are normalized with respect to the start of that period (Fig. 2), which is defined as the reference situation. The method we use is based on the hypothesis that natural factors have the same influence on both model simulations and satellite observations. For example, if natural factors increase model-simulated concentrations by 10% because of less precipitation, natural factors will also increase satellite-observed concentrations by 10%. However, satellite observations are also affected by anthropogenic influences. Assuming that natural and anthropogenic effects are additive, the latter can be extracted from the anomaly of the normalized satellite observations by subtraction of the natural relative impact. As a result, the average contributions of natural factors and human activities in a certain time period can be derived:
where $C_{\text{nature}}$ is the contribution of natural factors, $C_{\text{anthro}}$ is the contribution of anthropogenic activities, $M$ is the normalized long-term component of the simulations, $O$ is the normalized long-term component of the observations and $n$ is the number of months (the satellite observations and model simulations are monthly averages; note that the reference month is not used in eqs. 6 and 7 and thus the time step observations and model simulations are monthly averages). Note that the reference month is not used in eqs. 6 and 7 and thus the time step of the contributions from each step is the number of months (the satellite observations and model simulations are monthly averages; note that the reference month is not used in eqs. 6 and 7 and thus the time step observations and model simulations are monthly averages).

According to this method, the accuracy of the long-term variations of meteorological conditions are the most important in separating the contributions of natural and anthropogenic factors. Short-term signals of meteorological conditions are the most important in separating the contributions of natural and anthropogenic factors.

$$C_{\text{anthro}} = \frac{1}{n-1} \sum_{i=2}^{n} \frac{O_i - M_i}{M_i(O_i - 1)} \times 100\%$$

$$C_{\text{nature}} = \frac{1}{n-1} \sum_{i=2}^{n} \frac{O_i(M_i - 1)}{M_i(O_i - 1)} \times 100\%$$

Fig. 3. Contributions of natural and anthropogenic factors to long-term variations of SO$_2$, NO$_2$, CO, and AOD in increasing (a) and decreasing (b) periods as defined in Fig. 2.
filters are able to extract the long-term component from monthly time-series data.

In the previous section, we presumed that the KZ(13,3) filter can extract the long-term component from both satellite observations and model simulations and that normalization can efficiently eliminate the bias between model simulations and satellite observations. In order to evaluate the efficiencies of the KZ(13,3) filter and normalization, a series of sensitivity tests was conducted.

In this study, the satellite NO$_2$ data are derived from the products from SCIAMACHY (2002–2012) and GOME-2 (2007–2014) by averaging the data for the overlapping period 2007–2012. The spatially averaged monthly NO$_2$ VCDs from SCIAMACHY, GOME-2, and the average of the two instruments (AVERAGE) from 2007 to 2012 are shown in Fig. 4. The correlation coefficient (R) and NMB between SCIAMACHY NO$_2$ and GOME-2 NO$_2$ are 0.96 and $-13\%$, respectively. Since we only care about the long-term variation, the KZ(13,3) filter was used to extract the long-term components from the three datasets. After filtering, the R between SCIAMACHY NO$_2$ and GOME-2 NO$_2$ increased to 0.99 (Fig. 5), indicating a high agreement between long-term variations of NO$_2$ from the two instruments. However, the obvious NMB between the two instruments even increased from $-13\%$ to $-18\%$.

Then we normalized the filtered NO$_2$ with respect to the start time of each dataset (Fig. 6). The three lines overlap and appear as one. The R between SCIAMACHY and GOME-2 $\approx 1$ and the NMB decreased from $-18\%$ to $-85\%$, $0.8\%$. The NMB between AVERAGE and SCIAMACHY/GOME-2 NO$_2$ is $\approx 0.5\%$. As a result, the bias between the two instruments is efficiently removed by normalization. In the same way, the biases between satellite observations and model simulations can also be eliminated.

4. Conclusions

In this study, the temporal patterns of SO$_2$, NO$_2$, and CO columns and AOD in East China were investigated using satellite data and MOZART-4 simulations for $>1$ decade. We quantified the contributions from natural variations and anthropogenic factors on the increase/decrease of these species. The KZ(13,3) filter is used to extract long-term trends from both satellite-observed and model-simulated concentrations in East China. Normalized satellite retrieval data indicate that atmospheric SO$_2$ columns decreased by 15% since 2007, which can be mostly attributed to the government emission control policies. NO$_2$ columns have increased significantly (76%) from 2004 to 2011 and this increase is attributed to the rapid increase in the number of vehicles, power plants and industrial installations. The NO$_2$ columns started to decrease since 2012 due to national regulations. CO levels decreased by 13% since 2003, which may be due to improvements in combustion technology and energy structure adjustments. AOD increased by 24% before 2012 over the study area in East China, which is probably due to the increase of anthropogenic aerosol emissions combined with secondary aerosol formation from precursor gases such as SO$_2$ and NO$_2$, although emission abatement policy has a complicated effect on AOD (Lin et al., 2010). The observed decrease of gas species and AOD around 2008 is attributed to economic recession. Contributions of natural factors to the increase of SO$_2$ (2006–2007), NO$_2$ (2004–2011), CO (2001–2003) VCDs, and AOD (2001–2011) are 61% ± 2%, 1% ± 1%, 11% ± 7%, and 24% ± 32%, respectively. For anthropogenic factors, these contributions are 39% ± 2%, 99% ± 1%, 89% ± 7%, and 76% ± 32%, respectively. Natural contributions to the decrease of SO$_2$ (2007–2013), NO$_2$ (2012–2013), CO (2003–2013) VCDs, and AOD (2012–2013) are 16% ± 14%, 17% ± 9%, 6% ± 6%, and 0, respectively. For anthropogenic factors, these contributions are 84% ± 14%, 83% ± 9%, 94% ± 6%, and 100%. Our results provide valuable information for the quantification of natural and anthropogenic effects on air pollution in East China. For East China, the strongest effects are from anthropogenic origin, but natural effects cannot be ignored in a careful analysis of air quality regulations.

Fig. 4. Time series of spatially averaged monthly NO$_2$ VCDs from SCIAMACHY, GOME-2, and the average of the two instruments (AVERAGE) during 2007–2012.

Fig. 5. Time series of KZ(13,3) filtered monthly NO$_2$ VCDs from SCIAMACHY, GOME-2, and AVERAGE during 2007–2012.
Fig. 6. Normalization of K_{2,113} filtered NO\textsubscript{2} with respect to the start time of SCIAMACHY, GOME-2, and AVERAGE, respectively, during 2007–2012.

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