On the next pages follow the:

- Response to referee report #1

- Response to referee report #1

- Revised manuscript with track-changes

Response to referee report #1

We thank Referee #1 for the valuable comments. Below we give a point-to-point response to comments by referee. The referee's comments are given in italic font, while our response is given in bold font.

While I find the topic of the manuscript timely and intriguing, I think that the paper is too thin on content and there is little independent information presented or literature citations given to support their weak conclusions.

In our paper we conclude that NOx emissions would have been 30 % higher and SO2 concentrations would be 3 times higher as they are today without the strong air quality regulations taken in the last 10 years. This is not a weak conclusion for a country with such serious air pollution problems. The conclusions come from careful analysis of satellite data as shown in our paper. All independent sources are properly referenced.

For instance, the authors do not really discuss or quantify the "effectiveness of air quality policy", as given in the title, beyond saying that the trends in space-based air quality data appear somewhat consistent.

The effectiveness is literally quantified as 30 % and factor 3 for respectively NOx and SO2. As shown in the paper this reduction is only an effect of air quality regulations and excluding all economic effects. It is not clear to us what text the referee refers to with "*trends in space-based air quality data appear somewhat consistent.***".**

Further analysis is needed before this manuscript should be published or before it can live up to what is promised by the title.

Please, specify what analysis you feel is missing.

Comments

Abstract. Please clearly say in the abstract what is new and interesting about your work.

New in this paper is that the effect of air quality measures is quantified by dividing the air pollutant emissions by the fossil fuel use, which leads to a trend in emission factor measured from space. We will emphasize this in the update of the paper. This can be done successfully because satellite products and algorithms have improved greatly. Satellite measurement indicate independently that NOx emissions in China peaked in 2012 and are greatly reduced since.

"unprecedented accuracy" – you didn't show this or even really discuss this.

The term "unprecented" indeed sounds somewhat dramatic, but it only states that our new data set improves over old ones. We will replace this with the more neutral "improved". The improved accuracy is mentioned and further discussed in Theys et al. (2015) and Ding et al. (2015, 2016).

The abstract does not have any interesting or strong conclusions.

See comments above.

Introduction. 1st paragraph. 1st sentence. Why are satellite instruments "especially effective"?

We agree the wording is not entirely logic and we will change the sentence into:

" Satellite instruments can monitor air quality from space by mapping e.g. aerosols and tropospheric ozone, but are especially useful for emission estimates in observing the relatively short-living gases nitrogen dioxide (NO2) and sulphur dioxide (SO2)"

2nd sentence. What improved datasets? 4th sentence. How is this data set "improved"? Accuracy?

The Improved datasets are directly discussed after this sentence in the same paragraph. SO2 from OMI described in Theys et al. (2015), and NOx emissions from OMI with DECSO v4, described in Ding et al. (2015, 2016). The accuracy is described in these papers. For SO2 the column concentrations are on average within 12 % in agreement with ground observations. NOx emissions have an accuracy of about 20% in each grid cell. We will add this information to the text.

Please put your work into the context of other studies of emissions and trends over China. There are quite a few recent ones to discuss that use satellite data. Please include the new paper by McLinden et al. (Nature). Many of these discuss individual emission sources, such as power plants. However, you do not, which would be necessary to estimate the effectiveness of environmental regulations.

McLinden et al. (Nature) was published 1 week after we submitted this paper, but we will of course refer and discuss this work in the update of our paper. Other studies about emissions and trends over China that are referred or discussed in our paper are Richter et al. (2005), van der A et al. (2006), Stavrakou et al. (2008), Kurokawa et al. (2009), Lee et al. (2010), Li et al (2011), He H. et al. (2012), Zhang et al. (2012), Yang et al. (2013), Mijling et al. (2013), Fioletov et al. (2015), Krotkov et al. (2015), and Liu et al. (2016).

Indeed we discuss the emissions on a provincial level to give an overview for a country which harbours at least 1000 power plants. A single power plant is therefore not representative for the air quality regulations. That is why we use the total fossil fuel use in combination with satellite observations. We present a new method to estimate the total effectiveness of environmental regulations, by directly relating the emissions to the fossil fuel consumption in the country. This top-down approach saves us the effort of evaluating all individual contributions.

Section 3.2. There are many speculations here. Back up them up with provincial data. Please overplot fuel consumption data in Figure 3.

Section 3.2 Unfortunately the fuel consumption data for Chinese provinces is not publicly available, which limits our possibilities. However, many regulations are nation-wide as are our main analysis and conclusions. The fuel consumption data is shown in Figure 5.

Section 3.3. Are the trends in OMI NO2 consistent with the provincial emissions data? Please plot.

Section 3.3 We have done our analysis on the NOx emission data from inversions and not on NO2 concentrations, since emissions are clearly localised while concentrations are strongly affected by transport and meteorology. The quality of the inversion has been assessed in Mijling et al. [2013]. Also emissions are directly affected by air quality regulations.

Section 3.4. Again, I'm interested in the provincial data.

Unfortunately the fuel consumption data for provinces is not publicly available. Perhaps the reviewer is aware of sources we overlooked?

Figures 1 & 2. Need to have a map of provinces for the reader to refer to with major cities. Most readers will not know provincial names. It may also help to plot the locations of major power plants.

We will add a map with the provinces of China (and power plant density) for convenience of the reader.

Response to referee report #2

We thank Referee #2 for the valuable comments. Below we give a point-to-point response to comments by referee. The referee's comments are given in italic font, while our response is given in bold font.

The authors present a study of temporal trends of NO2 and SO2 derived from satellite observations over China, and relate them directly to changes in fossil fuel consumption in order to investigate the effectiveness of environmental regulations. While the former has been done in several studies before, the latter is to my knowledge new and provides an interesting approach. The study thus matches the scope of ACP and should generally be published.

However, the provided material is rather sparse (in particular as the introduction let the reader expect to see an analysis on provincal level, which is not given), and the results for NOx are not convincing; the authors try to interprete some local maxima by some reason, but I see no consistent explanation for the whole, rather complex, temporal pattern.

Thus, the study needs major revisions, in particular for NOx, providing additional information which either substantiates the discussion of trends or let the authors be more cautious with their statement about NOx concentrations being 30% higher without regulations.

The temporal patterns in NOx emissions can be explained by the (lack of) regulations on NOx emissions, a shift in fuel consumption patterns and other economic factors. We hope to have explained the trends and our conclusions better in the revised version of our paper by adding more discussion on the results. To support our conclusions and analysis we have added in the revised version the provincial data for SO2 concentrations and NOx emissions per year and province. In order to clarify our findings we have revised the Figures of the time series by adding the provincial time series of the 10 most emitting provinces. More details about our revisions follow below.

Major concerns:

- Provincial levels The authors point out that it is great to have improved NO2 and SO2 datasets on high spatial resolution, which allow the analysis of time series on provincial level. Thus, the authors should indeed investigate the trends of SO2, NOx, fossil fuel, and ratios on provincial levels, which probably will provide valuable further information and help to understand/assess the NOx-per-fuel trend (see below).

The NOx and SO2 datasets are indeed analysed on a provincial level. To summarize our findings we decided not to show the individual time series for 30 provinces, but instead show an average and range of the timeline for the 10 most polluted provinces. Individual time series have now been added in tables for SO2 and NOx with the concentration and emissions per year and per provinces. In the text we elaborated more on the provincial time series.

The fuel consumption per province was not publicly available at the time of submitting our paper. Only recently the provincial data became available for most provinces, although we have no indication of the uncertainty of these numbers, especially for the smallest provinces. The ratio per province will combine the uncertainties on these coal/oil consumption numbers with the errors in the satellite data. This makes it difficult to draw any conclusions out of these ratios and therefore we have decided to ignore the provincial fuel numbers in this study.

On the other hand, we found evidence in the papers of Guan et al. (2012) and Hong et al. (2016) that the sum of the <u>coal</u> consumption of all provinces is more accurate than the number provided for the whole of China, that is why we have updated Figure 5 and 6 with the total coal consumption of the provinces, which only slightly changes the results. For oil we have not enough provincial data to do the same.

Guan, D., Z. Liu, Y. Geng, S. Lindner and K. Hubacek, The gigatonne gap in China's carbon dioxide inventories, Nature Climate Change, 2, 672–675, (2012), doi:10.1038/nclimate1560

Hong, C., Zhang, Q., He, K., Guan, D., Li, M., Liu, F., and Zheng, B.: Variations of China's emission estimates response to uncertainties in energy statistics, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-459, in review, 2016.

- Annual vs. semi-annual means: For SO2, only April-September is considered "due to a lower accuracy at higher latitudes" (3.2) For NO2, information on the kind of averaging is missing in 3.3, but later it is stated that total annual emissions are used (3.4). Why? The lower accuracy and snow/ice argument holds as well for NO2. As the trends for both SO2 and NO2 are compared to fossil fuel and to each other, the period for calculating means has to be consistent. In any case, the authors should also provide the "winter"-trend for SO2, despite the lower accuracy. Is it similar (with higher noise) or significantly different from Fig. 3?

It is true that both NO2 and SO2 are irregularly and sparsely sampled by the satellite at high latitudes in wintertime. This lowers the accuracy and potentially introduces biases when averages are calculated for these periods based on the available satellite measurements. The SO2 analysis in this paper depends on such averages. NOx emissions, however, are derived using an inversion algorithm insensitive to data gaps.

. The SO2 data from OMI is worse in winter than in summer, but the annual data is good enough and not significantly different from Figure 3. The reason we switched to summer means is that the SCIAMACHY SO2 observations are very scarce in winter, since in the most optimal situation SCIAMACHY has global coverage only once every 6 days. On top of that the SCIAMACHY data is more noisy due to the lower spatial resolution. To keep consistency for all SO2 data we had done all analysis for summer means. However, triggered by and based on the remarks of the reviewer about consistency, the authors came to the conclusion that it is better to use annual averages for both OMI products, since this is the main focus of our study and these data are compared to annual coal/oil consumptions.

For SCIAMACHY and GOME-2 we still use summer values. If we switch to annual means the GOME-2 line does not change a lot, but for SCIAMACHY the changes are much larger, although the decreasing trend clearly remains. For SCIAMACHY there is practically no data above 45 degree North and in other regions the number of data point is still scarce due to the global coverage once every 6 days. This makes the SCIAMACHY data very sensitive to weather effects.

We have adapted the text accordingly in section 3.2 and 3.3 and made new Figures 3 and 6.

- NOx regulations Almost no information is provided about the concrete NOx regulations. Please discuss the different possibilities in general, and the taken measures in detail, for reducing NOx over China, and provide a table similar as for SO2. From what I learned from the media, there were different measures taken during the Olympic games, like shutting off power plants nearby and building new ones more remotely (which would change the local, but not the total trends, underlining the need for investigations on provincal levels), or reduction of traffic (which would affect the NOx, but not the NOx per fuel). These (and other) different measures and their effect on NOx vs. NOx per fuel trends have to be discussed.

The measures mentioned by the reviewer for the Olympic Games and other events like World Expo, Youth Olympic Games, EPAC meeting, etc. are mostly of a temporary nature as shown by Mijling et al. (2009). They showed that since most measures were cancelled after the Olympic Games the NO2 levels were back to normal in a couple months. There have been less permanent regulations for NOx emissions (besides traffic emission regulations) than for SO2, which had for some time a higher priority in China. That is why we have mentioned the NOx regulations in the text, but not made a table for these regulations. However, in the revised version we added a table for the NOx regulations and added some discussion on the traffic regulations and events.

Please note that that we relate NOx emissions to Standard Coal Equivalents (SCE). The reduction of traffic will reduce the NOx emissions but also the NOx per fuel (i.e. NOx per SCE), since NOx is emitted by two different source sectors: (1) traffic and (2) industry including energy production. Of those two source sectors traffic has a higher emission factor (i.e. NO2 per fuel) (see Zhao et al., 2013), thus a shift in source sectors result in different NOx per fuel values. Note that for SO2 this is no issue. To explain this better we have adapted the text (see the next review item).

Zhao, B., Wang, S. X., Liu, H., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NOx emissions in China: historical trends and future perspectives, Atmos. Chem. Phys., 13, 9869-9897, doi:10.5194/acp-13-9869-2013, 2013.

- NOx per fossil fuel While the SO2 per fuel significantly decreases over the years, in accordance to regulations, the situation is less clear for NOx. The authors state that already 2008 the regulations worked out (explaining the 2009 minimum). But why is NOx per fuel increasing again (by 20%!) in the following years? Have the measures been cancelled? The attempt to explain Fig. 6 by shipping is pure speculation, as it is not supported by any data. The conclusion that NO2 would be 30% higher today without the measures taken is not convincing unless the decline in 2015 compared to the high plateau 2011-2014 is explained; or were all measures concerning NOx just taken in 2015? In any case, the 30% is overestimated as it compares the minimum to the maximum of the timeseries, completely ignoring statistical fluctuations.

Important for NOx emissions is the growth of the transport sector in China. In the last 10 years the amount of freight transport, expressed in tonnes*km, is approximately doubling every 6 years according to the official statistical information from NBSC (2016). This means that the transport sector is faster growing than the industrial/energy sector, resulting in a gradual shift in the relative shares of the source sectors, which results in gradually higher NOx per fuel in time because of the higher emission factor of traffic (see Zhao et al., 2013). Exception of this gradual growth is the year 2009 because of the following reasons:

- The global economic crisis affected especially the transport sector, which led to a shift in source sector and a reduction of NOx per fossil fuel. See De Ruyter de Wildt et al. (2012)

- The global economic crisis also led to the practice of slow steaming for international ship transport. See Faber et al. (2012) and Boersma et al. (2015).

This explanation has been added to the text. The text has been changed to:

"The year 2009 coincides with the global economic crisis when there was less export of goods from China. This affected especially the transport sector, mostly transport over water, as shown in De Ruyter de Wildt et al. (2012). Faber et al. (2012) and Boersma et al. (2015) showed that the economic crisis also resulted in a significant reduction of the average vessel speed to save fuel used by ship transport. This caused not only a shift in source sectors but in general led to lower NOx per fuel values. This explains the dip in pollution per fuel unit in 2009. After 2012 the gradual increase of NOx per fuel (as a result of the strongly growing transport sector) slowly stops, and the year 2015 shows a sharp decline in NO_x per fossil fuels unit. This can be directly related to the rapidly growing installation of Selective Catalytic Reduction (SCR) equipment at power plants since 2012 and new emission standards for cars as shown by Liu et al. (2016). "

Boersma, K.F., G.C.M. Vinken, J. Tournadre, Ships going slow in reducing their NOx emissions: changes in 2005–2012 ship exhaust inferred from satellite measurements over Europe, Environ. Res. Letters, 10, 7, doi.10.1088/1748-9326/10/7/074007, 2015

De Ruyter de Wildt, M., H. Eskes, K. F. Boersma, The global economic cycle and satellite-derived NO2 trends over shipping lanes, Geophys. Res. Letters, doi:10.1029/2011GL049541, 2012

Faber J, Nelissen D, Hon G, Wang H and Tsimplis M, Regulated slow steaming in maritime transport—an assessment of options, costs and benefits CE Delft (The Netherlands: Delft) (www.cedelft.eu/publicatie/regulated_slow_steaming_in_maritime_transport/1224) p 117 , 2012

The conclusion of a reduction of NOx of 30% as a result of air quality regulations is based on the following:

- Without any air quality regulations we expect a gradual growth in NOx per fossil fuel as a result of the relatively faster growing transport sector that has a higher emission factor. The growth stopped in the year 2012 as a result of the mandatory installation of Selective Catalytic Reduction (SCR) equipment at power plants since 2012 and new emission standards for cars (see Table 2). When comparing the year 2015 with the 2012 level we calculate that NOx would be at least (assuming no growth after 2012) of 25% higher without air quality measures. This is different from the earlier 30% because of the new coal numbers we are using in the revised version of the paper. We claim at least 25 % because in this calculation we don't account for the further growth of the transport sector after 2012, while in fact the fast growth of transport still continued.

Minor comments:

Page 1 Line 13: What does "spatially consistent" mean?

We mean that they have about the same quality around the world, but since that is not entirely true, we changed this to "global".

Page 1 Line 25: The factor of 3 for SO2 is different from the statement in 3.4, and not supported by the presented data.

We changed the text in both cases to "about 2.5 times higher". In 2015 the normalized emission factor is 0.4 compared to around 1 in the period 2003-2007.

Page 1 Line 31: "concentrations" should be "column densities"

We have made this correction.

Page 2 Line 11: Please provide the full name of "He K."

We included the full name Kebin He.

Page 3 Line 2: Add a reference to Liu et al., 2016: http://www.atmos-chem-phys.net/16/5283/2016/

We agree and added the reference.

Page 3 Line 12: Please provide some information how and how far the new retrieval improves the quality of SO2.

We have added that the new retrieval algorithm "improves the accuracy of the SO_2 data for OMI with a factor 2"

Page 5 Line 15: NO2 changes during the Olympic games (or during the Shanghai Expo) have been discussed before; please add references.

We have added a reference to Mijling et al. (2009), who discuss the effect of the Olympic Games on NOx.

Page 6 Line 25: "by definition not sensitive": This is a too strong statement which only holds under the assumption that CHIMERE is doing everything right within the DESCO algorithm.

We changed "by definition" into "in general".

Page 7 Line 6: There is no "Fig. 5b”.

We removed the reference.

Cleaning up the air: Effectiveness of air quality policy for SO_2 and NO_x emissions in China

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Abstract. Air quality observations by satellite instruments are global~~spatially consistent,~~ and have a regular temporal resolution, which make them very useful in studying long-term trends in atmospheric species. To monitor air quality trends in China for the period 2005-2015 we derive SO_2 columns and NO_x emissions on a provincial level with ~~an unprecedented~~ improved accuracy. To put these trends into perspective they are compared with public data on energy consumption and the environmental policies of China. We distinguish the effect of air quality regulations from economic growth by comparing them relatively to fossil fuel consumption. Pollutant levels, per unit of fossil fuel, are used to assess the effectiveness of air quality regulations. We note that the desulphurisation regulations enforced in 2005-2006 only had a significant effect in the years 2008-2009 when a much stricter control of the actual use of the installations began. For national NO_x emissions a distinct decreasing trend is only visible since 2012, but the emission peak year differs from province to province. Unlike SO_2, emissions of NO_x are highly related to traffic. Furthermore, regulations for NO_x emissions are partly decided on a provincial level. The last three years show both a reduction in SO_2 and NO_x emissions per fossil fuel unit, since the authorities have implemented several new environmental regulations. Despite an increasing fossil fuel consumption and a growing transport sector, the effects of air quality policy in China are clearly visible. Without the air quality regulations the concentration of SO_2 would be about 2.5~~almost 3~~ times higher and the NO_2 concentrations would be at least 25~~30~~% higher than they are today in China.

1. Introduction

Satellite instruments can monitor air quality from space by mapping e.g. aerosols and tropospheric ozone, but are especially useful for emission estimates~~effective~~ in observing the relatively short-living gases nitrogen dioxide (NO_2) and sulphur dioxide (SO_2). For these ~~-~~two trace gases improved data sets recently became available, enabling analysis of air quality time series on a national or provincial level with ~~an unprecedented~~improved accuracy. Theys et al. (2015) presented a new data set of SO_2 ~~concentrations~~column densities derived from the Ozone Monitoring Instrument (OMI) satellite instrument (Levelt et al., 2006). They conclude that the SO_2 concentrations derived from OMI agree on average within 12% with ground observations. This data set strongly improves on earlier SO_2 data sets from satellites, which motivated this study. For NO_2, instead of using concentration data, we assess directly the emission data of nitrogen oxides ($NO_x = NO_2 + NO$) that was derived from satellite observations by Mijling and Van der A (2012) and removes the meteorological influences. The precision of the derived NO_x emissions per grid cell of 0.25 x 0.25 degree is estimated as 20% (Ding et al., 2016).

China is one of the biggest emitters of SO_2 and NO_2 into the atmosphere because its large economy depends heavily on fossil fuels as an energy source. China alone is responsible for about 30 % of the global total emissions of SO_2 into the atmosphere (Klimont et al., 2013), while over 90% of the SO_2 emissions are caused by coal consumption in China (Chen and Xu, 2010). Coal is mainly used by thermal power plants and energy-intensive industry (e.g. steel, cement and glass), and to a lesser extent by residential use. SO_2 is also released by the use of oil and natural gas, but the sulphur content in these fuel types is much lower. Of these sources power plants are responsible for about 30-40 % of all emissions and industry for another 50-60 % (He K. et al., 2012, ChinaFAQs project, 2012). According to the Multi-resolution Emission Inventory for China (MEIC) (http://www.meicmodel.org/) the source of SO_2 emissions in 2010 was 29.4 % from power plants, 57.7 % from industry and 11.7 % residential and 1.2 % from transport. Figure 1 shows the location of the 600 biggest thermal power plants on the map of China including a list of provinces mentioned in this study. At a global scale, volcanic activity is another important source of atmospheric SO_2. However, plumes of active volcanoes are seldom observed over China.

NO_x is released by more or less the same anthropogenic sources, i.e. the burning of coal or oil. The main difference with SO_2 is that traffic is a much more important source for NO_x. NO_x emission factors (i.e. emissions per fossil fuel unit) in the transport sector are generally much higher than emission factors in energy and industry, which makes traffic one of the major sources of NO_x in China. According the MEIC inventory, 25% of NO_2 in 2010 was released by traffic, 32% by power plants, 4% by residential sources and 39% by industry, with the cement industry being the biggest emitter in this sector.

To reduce SO_2 in China, the authorities have implemented several environmental regulations. The most important regulation was the desulphurization of coal-fired power plants in 2005/2006 (Xu, 2011). This was later followed in the 12^{th} five-year plan (2011-2015) by stricter control on the implementation of the regulations, additional filtering efforts, switching to low-sulphur coal and gasoline, phasing out obsolete capacity in coal-using industry, phasing out of small-scale coal mining, and gradually using more oil, gas and renewable energies instead of coal since 2011. An overview of all regulations related to SO_2 is shown in Table 1, which includes the year of the beginning of the implementation.

The regulation of NO_x was started much later than for SO_2. Although the 12^{th} five-year plan already mentioned the intention to reduce NO_2 by 10 % (target) (ChinaFAQs project, 2012), only from 2012 onward NO_x filtering systems were installed, mainly at power plants but also for heavy industry. These regulations for NO_x were announced in 2013 in the Air Pollution Prevention and Control Action Plan (CAAC, 2013) for the coming 5 years. Gradually China has implemented several new national emission standards for cars during the time period of our study (see Table 2). Some Rregulations for road vehicles (e.g. ban on older polluting cars) have been introduced in China on a provincial or even city level, rather than nationwide. Strong regulations have also been enforced during specific events like the Olympic Games in 2008, Shanghai World Expo in 2010, Nanjing Youth Olympic Games in 2014, APEC meeting in 2014, etc. but those regulations are mostly of a temporary nature as shown by e.g. Mijling et al. (2012) for the Olympic Games in 2008.

To study the efficiency of the environmental policies, we analysed satellite observations of SO_2 and tropospheric NO_2 of the last 11 years. SO_2 satellite observations over China have been studied earlier by Lee et al. (2010), Li et al (2011), He H. et al. (2012), Yang et al. (2013), Fioletov et al. (2015), and Krotkov et al. (2015). Satellite observations are very useful for SO_2 trend studies, as recently McLinden et al. (2016) showed that bottom-up inventories are underestimating SO_2 emissions worldwide with about 0-10%. NO_2 satellite observations over China have been evaluated by e.g. Richter et al. (2005), van der A et al. (2006), Zhang et al. (2012), and Krotkov et al. (2015). Emission estimates of NO_x over China have been analysed by Stavrakou et al. (2008), Kurokawa et al. (2009), and more recently by Mijling et al. (2013) and by Liu et al. (2016a). In these studies linear trends of the concentration of air pollutants are often used. Here, however, we will relate changes derived on a provincial level for China with the energy consumption and the environmental policies of the country. This gives insight in the efficiency of the applied air quality policies and regulations. The comparison of SO_2 trends with those of NO_x emissions enables us to distinguish environmental policies specifically applied on coal-based industry and power plants with general environmental measures and trends in traffic.

2 Observational data

2.1 Satellite observations of SO_2

SO_2 is observed in the UV spectral range of satellite observations of SCIAMACHY (on Envisat), GOME-2 (on METOP-A) and OMI (on EOS-AURA). SO_2 retrieval algorithms have been earlier developed for GOME-1 by Eisinger and Burrows (1998), for SCIAMACHY (Lee et al., 2008), GOME-2 and for OMI by Krotkov et al (2006). Recently a new retrieval algorithm has been developed (Theys et al., 2015) that ~~clearly~~ improves the precision~~quality~~ of the SO_2 data for OMI with a factor 2, allowing us to derive more accurate trends based on OMI. The retrieval method is based on a Differential Optical Absorption Spectroscopy (DOAS) scheme to determine the slant columns from measured spectra in the 312-326 nm spectral range, which are then background corrected and converted to vertical columns using an Air Mass Factor (AMF). The AMF is calculated with the radiative transfer model LIDORT (LInearized~~-~~ Discrete Ordinate Radiative Transfer model). More details about the retrieval procedure are described in Theys et al. (2015). Also the operational algorithm of NASA for SO_2 from OMI has recently been improved. This algorithm and the algorithm of Theys et al. (2015) have a very comparable performance as shown by Fioletov et al. (2016).For this study, the algorithm of Theys et al. (2015) has been applied on the observations of the OMI instrument (Levelt et al., 2006) for its whole mission from 2004 onwards~~.~~

To improve the quality of the OMI SO_2 data we exclude observations with a cloud fraction of more than 50 percent or with a fitting chi-square higher than 1. The solar zenith angle is limited to 75° and the viewing angle to 50°. Since the OMI instrument is suffering from the so-called row anomaly since 2007 (KNMI, 2012), we filter the affected rows (24-49, 54-55) in the same way for all years in the time series.

As we focus on anthropogenic SO_2, the SO_2 data for 15 June - 9 July 2011 have been removed because of its contamination with volcanic SO_2 from the eruption of the Nabro volcano in Africa and the transport of its plume to China (Brenot et al., 2014).

As a first step in our study we have made monthly means for the whole data set by averaging and gridding the data to a resolution of 1/8° by 1/8°. The gridding algorithm takes into account the area of each satellite footprint overlapping the grid cell. The resulting data set is a time series of monthly means for the time period October 2004 to December 2015.

For comparison we also use the official ESA SCIAMACHY/Envisat SO_2 product version SGP 5.02 and the standard data product from the GOME-2/METOP-A version GDP 4.7, as developed within the EUMETSAT Satellite Application Facility for Atmospheric Composition and UV radiation, O3MSAF, project and distributed by http://atmos.caf.dlr.de/gome2/. The data of these instruments are noisier than the OMI datasets because of the lower spatial coverage, different fit window and the lower signal-to-noise ratio of the SCIAMACHY and GOME-2 instruments. Therefore, their quality-controlled monthly mean SO_2 data have been recalculated by spatially averaging for each grid cell the data from the eight surrounding neighbouring cells, hence creating a smoothed SO_2 field. For details on the methodology and findings refer to Koukouli et al. (2016).

2.2 NO_x emission estimates from satellite observations

For NO_x emission data we use the results of an update (version 4) of the DECSO (Daily Emission estimates Constrained by Satellite Observation) algorithm developed by Mijling and van der A (2012). DECSO calculates emissions by applying a Kalman filter for the inversion of satellite data and a regional Chemical Transport Model (CTM) for the forward model calculation. It takes transport from the source into account with a semi-Lagrangian approach. The CTM we use is CHIMERE v2013 (Menut et al., 2013) with meteorological information from the European Centre for Medium-range Weather Forecasts (ECMWF) with a horizontal resolution of approximately 25x25 km^2. The DECSO algorithm is applied to OMI NO_2 observations derived by the DOMINO v.2 algorithm (Boersma et al., 2011). The latest improvements of the DECSO algorithm resulting in version 4 are described by Ding et al. (2015, 2016). The monthly average emission data over China we use is available on 0.25 degree resolution for the period 2007-2015 on the web-portal www.globemission.eu.

3 Temporal analysis over China

3.1 Sources of SO_2 and NO_x in China

The multi-annual mean of SO_2 for 2005-2015 is shown in Figure ~~1~~2. As the lifetime of SO_2 is relatively short (typically 4-48 hours) (Lee et al., 2011, Fioletov et al., 2015), the observed SO_2 concentrations are a good proxy for the location of SO_2 emissions. Regions with large SO_2 concentrations are South Hebei, the province Shandong (around the city Zibo) and the region around Chongqing. South Hebei is a region with many power plants just east of the mountainous coal-mining area in Shanxi. The hot spot in the province Shandong is related to a strongly industrialized area with lots of coal-using industry. In the Chongqing region both coal mines and heavy industry are located.

Rather than located at hot spots, high NO_2 concentrations are more distributed over the East of China, mainly because traffic is an important source of NO_x emissions (see Figure ~~2~~3a). The underlying NO_x emissions are shown in Figure ~~2~~3b. Like for the SO_2 concentrations, NO_x emission spots can be found at the location of big power plants. Also clearly visible are the megacities of China, and ship tracks along the coast and sources along the big rivers.

3.2 SO_2 trends over China

To construct time series of SO_2 we have averaged the data to ~~months April-September to semi-~~annual means of the vertical columns derived from OMI. ~~The remaining monthly means are excluded from the analysis due to a lower accuracy at higher latitudes and part of the higher latitudes is missing due to snow cover.~~ From these ~~semi-~~annual mean SO_2 data we constructed time series for each province (see Table A.1). Figure 4~~3~~ shows the mean normalized time series for the 10 provinces with the highest total SO_2 column densities (i.e. Tianjin, Shandong, Hebei, Shanxi, Henan, Beijing, Jiangsu, Shanghai, Anhui and Liaoning), together responsible for 60% of all ambient SO_2 in China. The individual time series are drawn as thin black lines. The minimum and maximum of these time series for each year are shown in the grey shaded area to indicate the variability. The time series of Shanghai is the lowest black line of the 10 series, thus the reductions have been strongest in this province since 2005. Apart from Ningxia province, all provincial time series show very similar patterns ~~with a clear exception for the province Ningxia, whose time series is added to Figure 3 for comparison~~. In general, the SO_2 concentrations were at a maximum in the year 2007, when the start of decreasing trend is visible in China. Despite some fluctuations the SO_2 concentrations remain relatively constant from 2010 till 2013, where after they are decreasing again.

A different trend is observed for Ningxia, a province in the mid-north of the country with a relative low population density and large coal resources. ~~For Ningxia~~Here an increasing trend emerges for the years starting from 2010 when several new coal power plants were put into operation. A list of largest power plants (with a capacity of more than 600 MW) and the start year of their operation is shown in Table ~~2~~3. From 2012 onward, the more stringent SO_2 emission regulations also started to have effect in Ningxia.

3.3 NO_x emission trends over China

National NO_x emission trends show a different pattern than those of SO_2. We observe an increasing trend till about 2012, with an exception of the year 2009 which is related to regulations started at the Olympic Games in 2008 (Mijling et al., 2008) and the global economic crisis which shortly slowed down the Chinese economic growth. Total NO_x emissions in East China reached their peak levels in 2012, and have been decreasing since. While the economy kept growing after 2012, the emission of NO_x slowly decreases again as a result of the air quality regulations described in Section 1. According to the DECSO emission inversion, in 2015 the NO_x emissions were 4.9 Tg N/yr, which is 22.8% lower than in the peak year 2012.

However, the 2015 emissions were still 14.1% higher than in the reference year 2007. The trends per province (see Table A.2) show very similar patterns with only the starting year (the year with maximum NO_x emissions) of the decrease in emissions varying over the provinces. In Figure 54a, the normalized (to the year 2007) time series of annual NO_x emissions for East China (102-132°E, 18-50°N) is shown in similar way as for SO_2 in Figure 34. The mean, minimum and maximum of the 10 provinces with highest NO_x emission are shown (Shandong, Hebei, Henan, Jiangsu, Guangdong, Shanxi, Zhejiang, Anhui, Sichuan and Hubei), together responsible for 65% of all Chinese NO_x emissions. The thin black lines show the times series for the individual 10 provinces, where the lower line represents Guangdong. Figure 54b shows this the peak year for each province. Provinces where air pollution regulations, for e.g. traffic, got a lot of attention at an early stage, like Beijing and Shanghai, have reached their maximum before 2011. Most industrialised regions show their peak in the years 2011-2013. Some of the less developed and populated provinces show a maximum in 2014, which means that their decrease in NO_x emissions is very recent. Regional variations are mainly due to the fact that regulations for the NO_x emission reductions, for instance in traffic or power plants, are determined and implemented on a provincial level (Liu et al., 2016b). For the province Ningxia we see a very similar pattern occurring as for SO_2, which shows that for this low-densely populated province traffic plays a small role and the trend is determined by the operation of newly-built power plants.

3.4 Air pollution in relation to fossil fuel consumption

To relate the observed SO_2 and NO_x reduction to environmental regulations we have to take into account the coal and oil consumption in the same time period. The total coal consumption in Standard Coal Equivalent (SCE) units per year for China and the total oil consumption (also in SCE units) are shown in Figure 56, based on data of NBSC (2015). According to Guan et al. (2012) and Hong et al. (2016) the sum of the coal consumption of all provinces is more accurate than the number provided for the whole of China, thus we use the provincial totals for coal consumption. For NO_x emissions the transport sector plays an important role, especially ships are one of the largest NO_x emitters per fuel unit in the transport sector. The total freight transport almost doubles every 6 years in China.

Since the burning of coal and oil are the dominant sources of SO_2 and NO_x emissions, we can consider the total emissions of these air pollutants as the product of the national use of coal and oil (activity) and the average emission factor of one unit coal/oil. The effectiveness of environmental regulation will be reflected in a decrease of this emission factor. Therefore, we divide the annual SO_2 column measured from satellites and the annual NO_x emissions by the annual coal and oil consumption in China. In this way we get a measure of the emitted SO_2 or NO_x per unit (SCE) of fossil fuel consumption reflecting the Chinese environmental policy. The results are shown in Figure 67. One might argue that SO_2 is more related to coal than oil, but division by only coal yields the same results. In our analysis we omit gas consumption since this is very limited in China and hence does not affect the results significantly.

We focus here mainly on the results for OMI, because of the instrument's high spatial resolution and lack of instrumental degradation. However, SO_2 data of the SCIAMACHY and GOME-2 instrument are also added in Figure 67 to be able to further look into the past (starting in 2003) and to verify the results of OMI. The SO_2 data of SCIAMACHY and GOME-2 are averaged over the summer months (April-September). The remaining monthly means are excluded from the analysis due to a lower accuracy at higher latitudes and a large part of the higher latitudes is missing due to snow cover. For OMI Eeach data point is averaged over 6 (for SO_2) or 12 (for NO_x) months and the total area of China, which reduces the root-mean-square error to a negligible level. Biases among all instruments are removed by normalizing the values to those in reference year 2007. Up to 2009, the results agree fairly well. After 2009, we see the results of GOME-2 and OMI for SO_2 slowly diverge in time, which might be result of the instrument degradation of the UV spectra of GOME-2 after 2009 (Munro et al., 2016).

Changing weather conditions from year-to-year can affect the results for SO_2 concentrations and when these weather conditions are different ~~in the morning~~during the ~~(~~overpass of SCIAMACHY and GOME-2 (around 9:30 local time)~~)~~ and ~~afternoon (~~overpass of OMI (around 13:30 local time)~~)~~, this can lead to differences between the instruments. The global coverage of SCIAMACHY is once every 6 days and for GOME-2 and OMI almost daily. The limited number of samples for SCIAMACHY makes this data more sensitive to weather conditions. Note that due to the nature of the inversion algorithm the NO_x emission data is ~~by definition~~in general not sensitive to meteorological variability.

For SO_2 we see a big decrease in the years 2008 and 2009, while the desulphurization program of the 11th five-year plan started already in 2005/2006, when the authorities begin to reduce SO_2 emissions by installing desulphurization devices in many power plants (Lu et al., 2010). In 2006 SO_2 monitoring devices were also installed in the chimneys of the power plants. This resulted in a decrease in SO_2 emissions from 2006, while the much bigger decrease of SO_2 in 2008-2009 reflects the stronger government control at that time on the actual use of the equipment (Xu et al., 2011). After 2009, the SO_2 content per consumed coal unit only slowly decrease until 2011. From 2012 onwards we see a stronger annual decrease in SO_2. This coincides with the 12th five-year program when new measures were taken to upgrade the coal quality, to modernize the industry and to put more effort on law enforcement. Especially the law enforcement in the last years concerning the prohibition of flue gas bypass and the use of desulphurization devices in the steel industry played an important role.

For the NO_x emissions the total annual emissions ~~(summer and winter)~~ are used and divided in the same way as for SO_2 by the total coal and oil consumption. Here, however we should keep in mind that the transport sector (especially by shipping) emits much more NO_x per fuel unit than the power and industrial sectors (see e.g. Zhao et al., 2013). Thus the percentage of the total fuel used by transport is relevant for the graph of NO_x ~~(see Figure 5b)~~. In the early years we see in general a small increase in NO_x emissions per fuel unit due to the increasing fraction of the transport sector in the fuel use. Exceptions are the year~~s~~ 2009 and the recent year~~s~~ ~~2013 and~~ 2015. The year 2009 coincides with the global economic crisis when there was less export of goods from China. This affected especially the transport sector, mostly transport over water, as shown in De Ruyter de Wildt et al. (2012). ~~-~~ Faber et al. (2012) and Boersma et al. (2015) showed that the economic crisis also resulted in a significant reduction of the average vessel speed to save fuel used by ship transport. This caused not only a shift in source sectors but in general led to lower NO_x per fuel values. This~~which~~ explains the dip in pollution per fuel unit in 2009. After 2012 the gradual increase of NO_x per fuel (as a result of the strongly growing transport sector) slowly stops, and ~~The reduction in the year 2013 can still be seen as it was caused by a smaller fraction of shipping transport in that year, but~~ the year 2015 shows a sharp decline in NO_x per fossil fuels unit. This can be directly related to the rapidly growing installation of Selective Catalytic Reduction (SCR) equipment at power plants since 2012 and the introduction of new emission standards for cars, as shown by Liu et al. (2016). This strong reduction in NO_x for 2015 and the equally strong reduction for SO_2 in 2014 and 2015 are a result of very effective recent environmental regulations in the last years in China. By comparing the efficiency level in 2015 with earlier levels we can conclude from Figure ~~6~~7 that without these air quality regulations SO_2 concentrations would nowadays be about ~~more than~~ 2.5 times higher and the NO_2 concentrations would be at least ~~30~~25% higher in China today.

4 Discussion

The current developments in data products derived from satellite observations provide high quality time series of the air pollutants NO_x and SO_2. Although the mean of observed SO_2 columns are not linearly related to the SO_2 emissions because of the influence of the weather, it can still be argued that these satellite data products, whether concentrations or emissions, provide a fair comparison over the various regions from year to year. By comparing these time series with fossil fuel energy consumption the economic growth is removed from the equation and we can monitor the effectiveness of air quality policies. We foresee that this method will become a valuable tool for policy makers concerning air quality regulations.

For China we see similar patterns in the trends of SO_2 per province. In 2006 a nation-wide implementation of desulphurisation installations started. However, the effects are only visible in 2008 and 2009 when a strict control by the Chinese authorities on the actual use of the desulphurisation installations started. In 2009, we see the effect of the air quality regulations for SO_2 and NO_x resulting from the ~~Olympic Games~~global economic recession at the end of 2008. The increasing relative contribution of the transport sector to the NO_x emission slowly increases the amount of NO_x per fossil fuel unit after 2009. After 2011 we see a steadily decreasing SO_2 pollution per fossil fuel unit caused by various Chinese environmental regulations. In the last year of our time series, 2015, a clear effect becomes visible of very recent regulations for NO_x emissions from power plants and heavy industry. The fit of linear trends often used in earlier studies is therefore no longer applicable to the Chinese situation.

The availability of high quality satellite data for the last ten years is especially interesting for China where the situation is rapidly changing. For instance in Europe and Japan desulphurisation started much earlier when these satellite data were not yet available. On the other hand, in India SO_2 and NO_x emissions are still growing and possible new regulations can be monitored in the years to come with an even better quality using forthcoming sensors as e.g. TROPOMI ~~onboard~~on-board Sentinel-5 Precursor.

Despite the growing use of coal and oil in the last ten years in China we see reduced emissions per fuel unit in the past few years. This decreasing trend in both SO_2 and NO_x for China is likely to continue in the coming years for which the Chinese national government has announced less use of coal, more environmental regulations for SO_2 and NO_x and stricter reinforcement of control of environmental policies.

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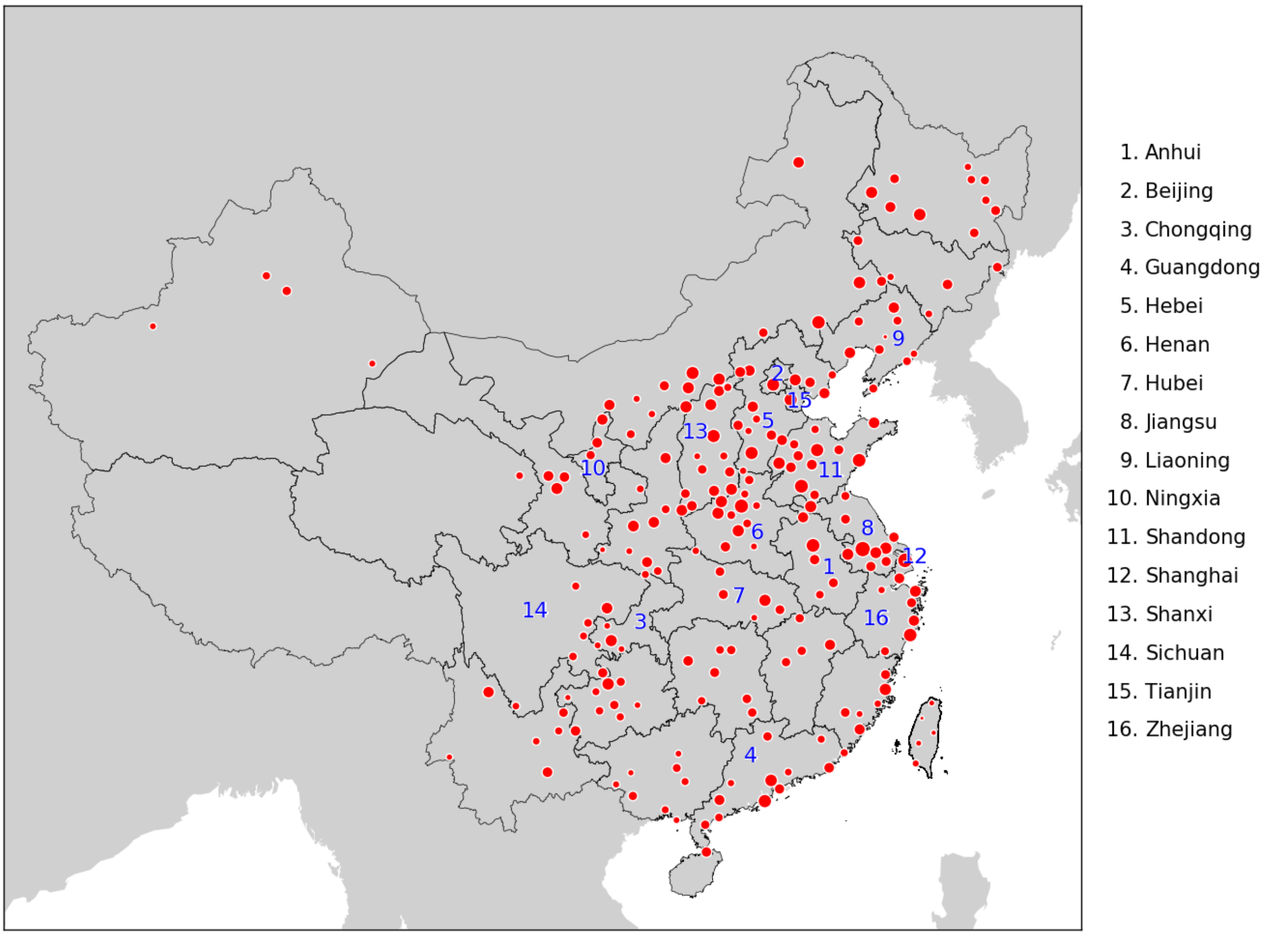


Figure 1: Location of power plants in China according to REAS v.2 (Kurokawa et al., 2013). The size of each dot indicates the emission of the power plants (Power plants in close proximity are combined in a single dot). In addition, a list is given of the provinces mentioned in this study.

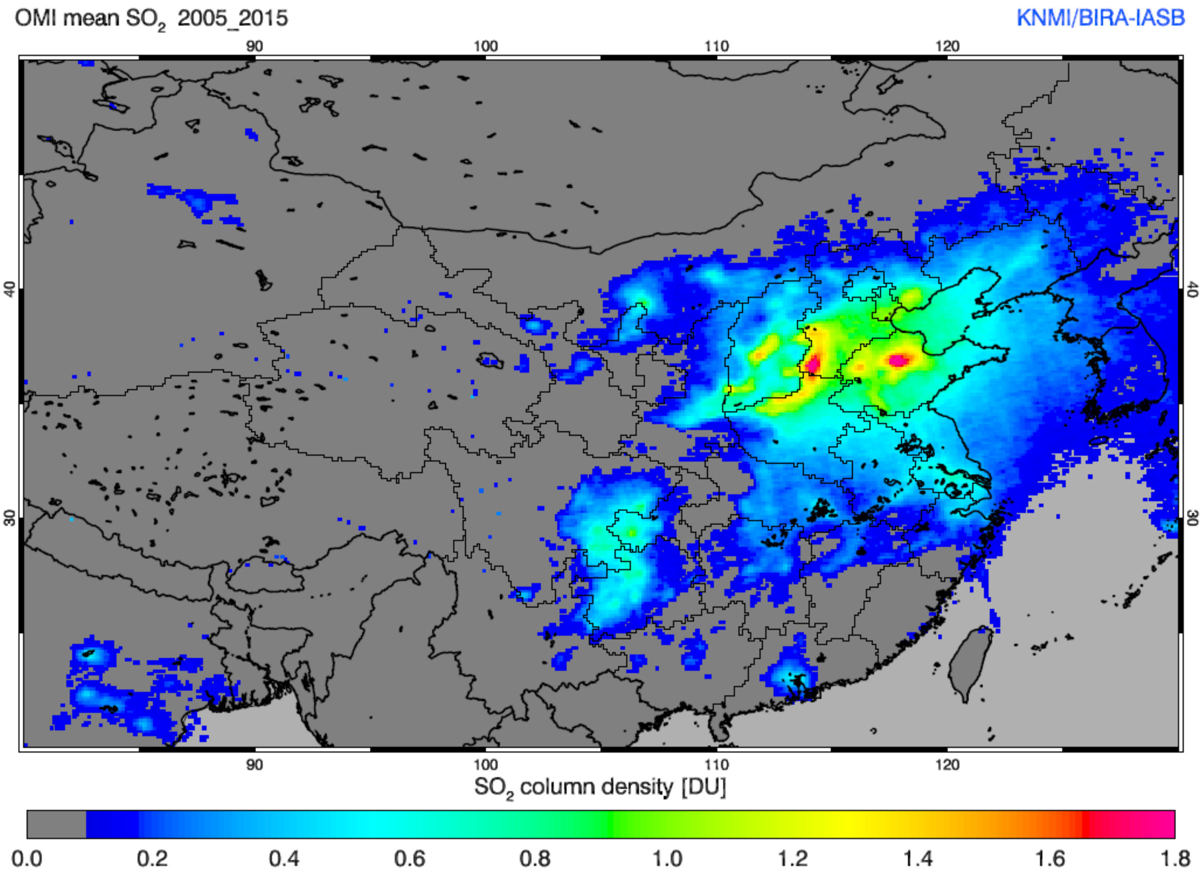


Figure ~~1~~2: Average SO_2 concentrations for the period 2005 to 2015 as observed by the OMI satellite instrument. Data below 0.1 DU is masked (grey colour).

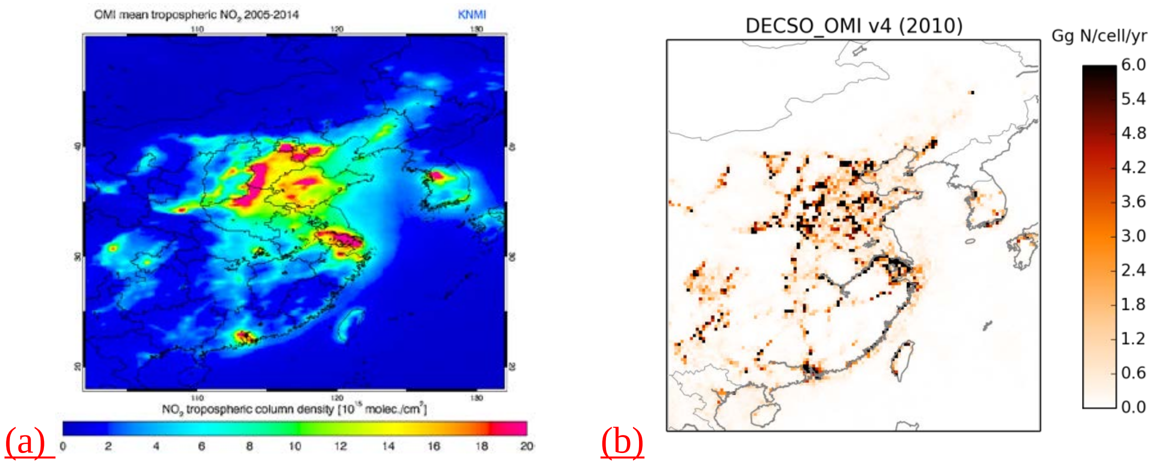


Figure ~~3~~2: (a) The averaged tropospheric NO_2 concentrations over China measured by OMI in the period 2005-2014. (b) The NO_x emissions in the year 2010 derived from the OMI satellite observations.

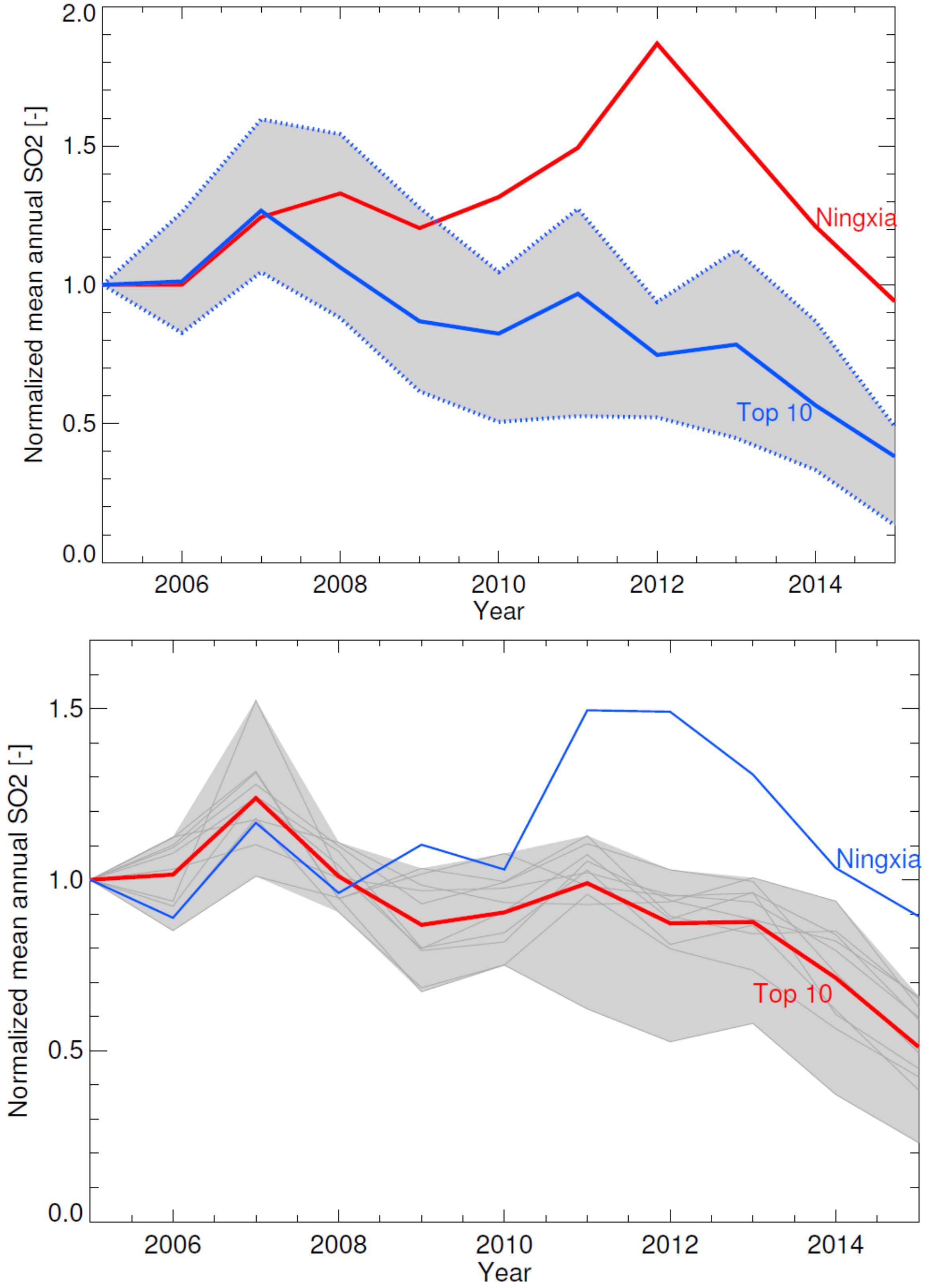
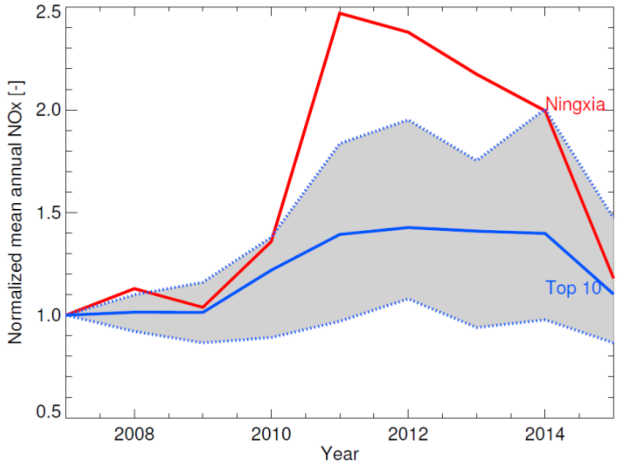
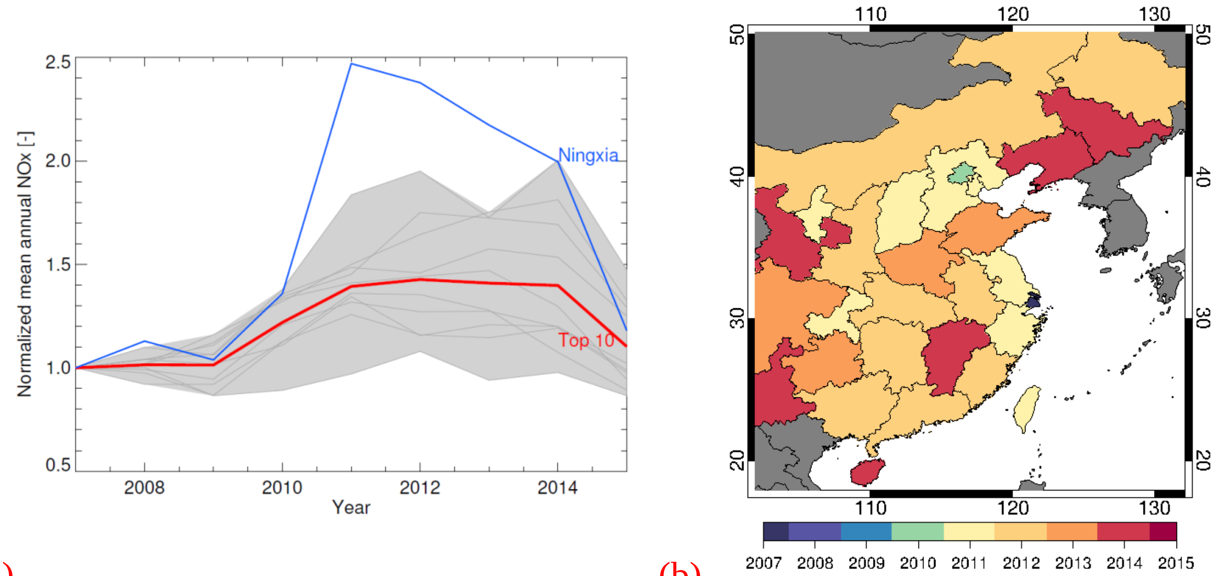


Figure ~~3~~4: Time series (red~~blue~~ line) of the ~~semi-~~annual mean ~~(April-September)~~ of the 10 provinces with the highest SO_2 concentrations derived from the OMI satellite observations. The time series are normalized to their value in 2005. The grey area indicates the maximum range of the individual values of the times series of each of the 10 provinces. The thin black lines show the individual time series of those provinces.~~.~~ The province of Ningxia has a distinct deviating trend, here shown in blue~~red~~.





(a) (b)

Figure 45: (a) Shown are the annual total NO_x emission estimates for the last 9 years for ~~East China~~the top 10 of highest NO_x emitting provinces in East China. Emissions are derived with DECSO V4 using OMI observations. The thin black lines show the individual time series of those provinces. (b) Peak year of the NO_x emissions per province.

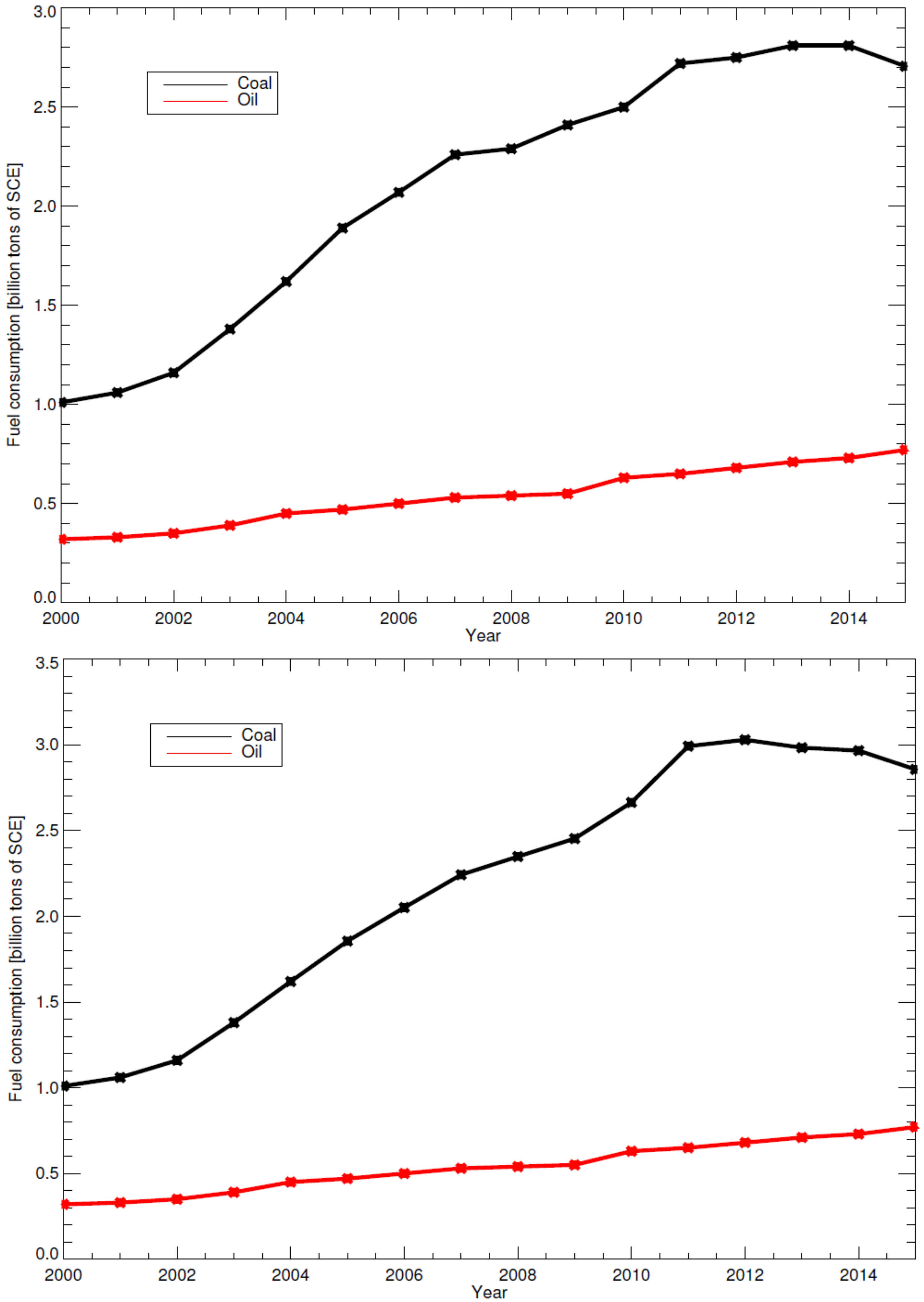


Figure ~~5~~6: In black the annual ~~oil~~ coal consumption and in red the annual oil~~coal~~ consumption for China is shown.

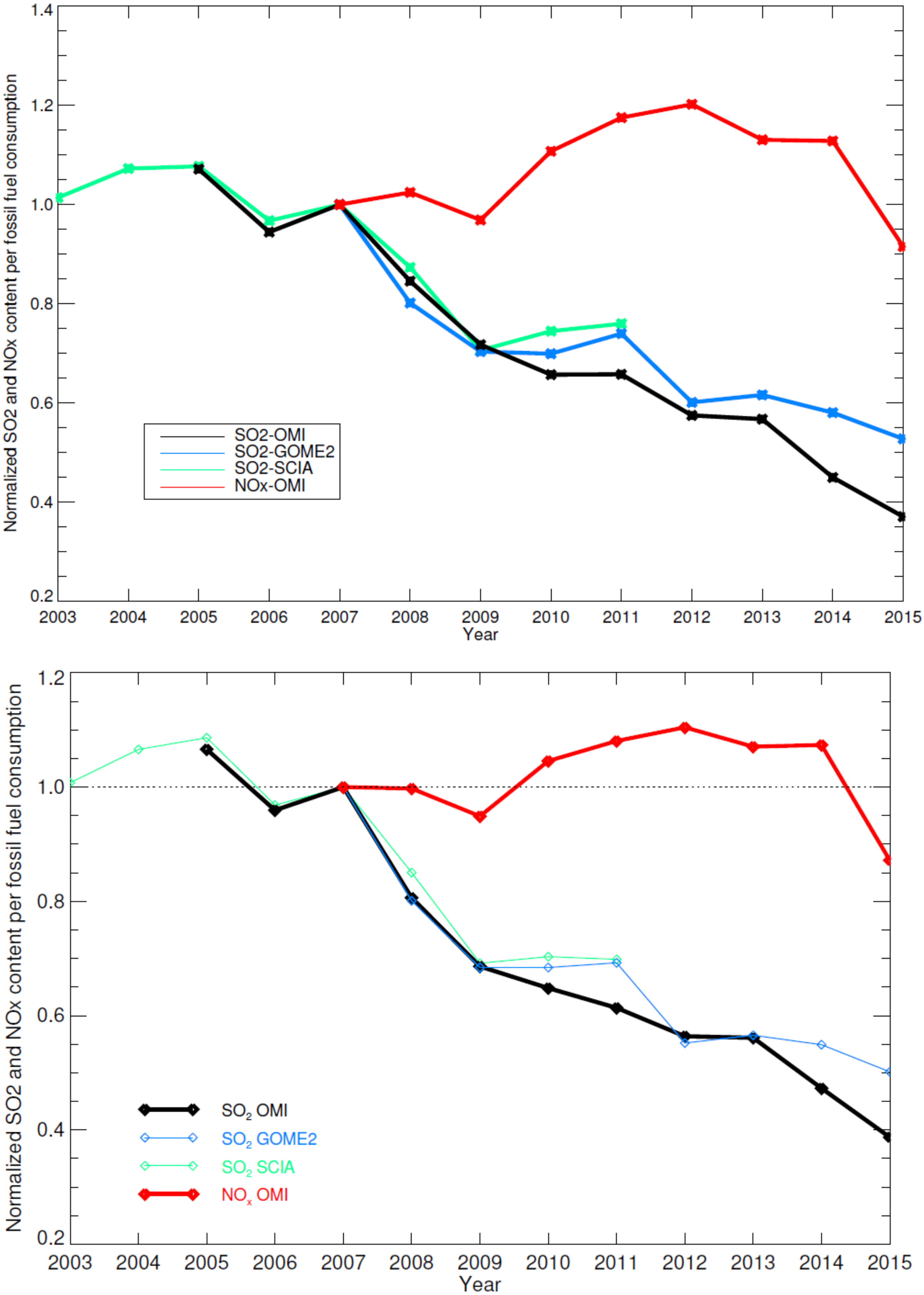


Figure 67: Time series of the ratio of the mean SO_2 columns and the fossil fuel consumption in China based on observations of OMI (black), SCIAMACHY (green), and GOME-2 (blue). The ratios of the annual NO_x emissions and the fossil fuel consumption is based on observations of OMI (red). All time series are normalized to the year 2007.

Table 1: Environmental regulations of the Chinese national government to reduce SO_2 in the air.

Start year of implementation	**Regulation**	**Reference**
2005-2006	Desulphurization techniques in power plants.	Li et al., 2010
2005-2012	Closure of several of the most polluting power plants	Liu et al., 2015
2008	Stricter control of implementation of desulphurization in power plants	Xu et al., 2011 Liu et al., 2015
2011	Use of more gas and renewable energies instead of coal	NBSC, 2015
January 2012	New emission standard of air pollutants for thermal power plants	MEP, 2015
2013	Mandatory SO_2 filtering of small-scale coal-fired industry	Zhang, 2013, NDRC, 2013
End of 2013	Stricter control of environmental policy	CAAC, 2013, State Council, 2014
End of 2013	Further desulphurization in industry	CAAC, 2013, NDRC, 2013
2014	Phasing out small-scale coal-fires boilers	CAAC, 2013, State Council, 2014
2014	Closure of 2000 small-scale coal mines	Zhu, 2013
End of 2014	Use of low-sulphur coal	State Council, 2014
End of 2014	Cap on coal consumption	State Council, 2014

Table 2: Environmental regulations of the Chinese national government to reduce NOx emissions.

Year of implementation	**Regulation**	**Reference**
2011-2015	Installation of Selective Catalytic Reduction (SCR) equipment at power plants	Liu et al., 2016, CAAC, 2013
2007	China 3 (Euro 3) emissions standards for cars, nationwide	www.dieselnet.com
2011	China 4 (Euro 4) emissions standards for gasoline cars, nationwide.	www.dieselnet.com
2015	China 4 (Euro 4) emissions standards for diesel cars, nationwide.	www.dieselnet.com

Table ~~2~~3: Main power plants in Ningxia province (> 600 MW). Data collected from www.sourcewatch.org.

Power plant	Capacity (MW)	In operation since	Remark
CPI Linhezhen	700	unknown	
Daba-1	1200	< 2000	
Daba-2	1100	unknown	An extension of Daba-1
Ningxia Zhongning-2	660	2005-2006	
Guodian Shizuishan-2	1980	2006	
Ningdong Maliantai	660	2006	
Huadian Ningxia Lingwu units 1&2	1200	2007	
Guodian-Dawukou	1100	2010	Extension of the original 440 MW plant
Guohua Ningdong	660	2010	
Ningxia Liupanshan	660	2010	
Huadian Ningxia Lingwu units 3&4	2120	2010-2011	
Shenhua Yuanyang Lake	1320	2010-2011	
Shuidonggou	1200	2011	
Ningdong Younglight	660	2013	

Appendix A

Table A1 Annual SO_2 column densities (DU/grid cell) per province observed by OMI

Province	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015
Anhui	0.625	0.586	0.953	0.637	0.499	0.567	0.671	0.553	0.602	0.379	0.280
Beijing	0.753	0.829	0.989	0.711	0.778	0.749	0.850	0.673	0.634	0.640	0.491
Chongqing	0.514	0.509	0.530	0.567	0.580	0.580	0.492	0.370	0.469	0.269	0.136
Fujian	0.099	0.123	0.196	0.135	0.104	0.113	0.112	0.080	0.107	0.076	0.064
Gansu	0.144	0.136	0.150	0.130	0.135	0.123	0.134	0.127	0.131	0.105	0.103
Guangdong	0.251	0.257	0.280	0.239	0.171	0.177	0.138	0.095	0.118	0.086	0.080
Guangxi	0.199	0.203	0.270	0.236	0.127	0.190	0.179	0.092	0.134	0.091	0.072
Guizhou	0.424	0.478	0.532	0.516	0.418	0.424	0.357	0.261	0.345	0.167	0.100
Hainan	0.098	0.086	0.091	0.092	0.060	0.090	0.106	0.027	0.087	0.055	0.000
Hebei	0.903	0.931	0.996	0.908	0.874	0.881	0.922	0.863	0.844	0.716	0.540
Heilongjiang	0.134	0.141	0.144	0.142	0.124	0.138	0.154	0.162	0.125	0.134	0.135
Henan	1.036	0.920	1.222	0.938	0.709	0.778	0.992	0.827	0.762	0.585	0.439
Hubei	0.487	0.477	0.603	0.490	0.342	0.386	0.479	0.365	0.378	0.288	0.176
Hunan	0.364	0.330	0.448	0.371	0.270	0.281	0.320	0.240	0.259	0.180	0.112
Jiangsu	0.847	0.782	1.054	0.917	0.678	0.716	0.871	0.687	0.735	0.524	0.326
Jiangxi	0.272	0.278	0.373	0.267	0.202	0.222	0.244	0.197	0.230	0.184	0.136
Jilin	0.205	0.233	0.260	0.259	0.191	0.207	0.201	0.187	0.203	0.192	0.156
Liaoning	0.512	0.576	0.602	0.568	0.504	0.478	0.475	0.479	0.515	0.480	0.321
NeiMongol	0.154	0.170	0.200	0.174	0.175	0.180	0.191	0.178	0.177	0.186	0.152
Ningxia	0.234	0.208	0.273	0.225	0.258	0.241	0.350	0.349	0.306	0.242	0.209
Qinghai	0.079	0.085	0.077	0.080	0.079	0.088	0.083	0.091	0.096	0.082	0.086
Shaanxi	0.357	0.301	0.401	0.324	0.261	0.269	0.338	0.304	0.315	0.246	0.224
Shandong	1.197	1.309	1.531	1.315	1.113	1.188	1.323	1.232	1.191	0.870	0.592
Shanghai	0.874	0.744	0.883	0.828	0.588	0.656	0.544	0.460	0.507	0.325	0.202
Shanxi	0.748	0.806	0.928	0.779	0.593	0.612	0.789	0.703	0.661	0.614	0.493
Sichuan	0.429	0.429	0.513	0.376	0.415	0.427	0.394	0.293	0.350	0.198	0.123
Taiwan	0.089	0.071	0.081	0.085	0.074	0.090	0.074	0.055	0.086	0.052	0.051
Tianjin	1.197	1.344	1.577	1.132	1.217	1.289	1.176	1.140	1.150	1.005	0.708
XinjiangU.	0.073	0.074	0.087	0.093	0.088	0.094	0.090	0.090	0.111	0.101	0.084
Xizang/Tibet	0.080	0.097	0.086	0.091	0.094	0.111	0.096	0.097	0.083	0.087	0.087
Yunnan	0.140	0.159	0.182	0.147	0.144	0.153	0.149	0.131	0.127	0.100	0.085
Zhejiang	0.383	0.337	0.452	0.395	0.297	0.316	0.403	0.258	0.321	0.212	0.158
P.R.China	0.397	0.392	0.444	0.373	0.330	0.342	0.358	0.335	0.332	0.280	0.225

Table A2 Annual NO_x emissions (Gg N/year) per province derived from OMI observations

Province	2007	2008	2009	2010	2011	2012	2013	2014	2015
Anhui	167	169	187	224	242	292	288	282	215
Beijing	91	62	90	107	88	80	89	74	64
Chongqing	54	57	70	75	87	95	96	100	70
Fujian	96	114	100	114	161	162	153	167	137
Gansu	31	38	37	42	61	73	60	78	52
Guangdong	383	383	331	341	371	413	360	374	331
Guangxi	118	148	118	145	152	224	224	200	157
Guizhou	107	130	142	154	131	194	191	180	122
Hainan	8	13	11	17	23	22	25	37	30
Hebei	427	423	403	515	563	543	544	511	436
Heilongjiang	33	36	30	25	43	54	40	49	31
Henan	334	347	370	445	470	481	491	433	315
Hubei	135	140	144	186	248	263	233	270	199
Hunan	109	112	124	162	163	216	184	218	187
Jiangsu	374	344	344	421	470	433	428	445	365
Jiangxi	51	58	65	73	85	105	111	150	112
Jilin	30	22	18	20	45	50	43	48	43
Liaoning	122	128	124	169	205	225	178	199	173
NeiMongol	98	116	117	156	215	215	169	142	111
Ningxia	30	34	32	41	75	72	66	61	36
Qinghai	6	6	6	10	14	13	15	13	13
Shaanxi	118	118	113	181	216	222	196	208	158
Shandong	464	510	493	629	689	677	731	712	580
Shanghai	96	103	101	95	109	75	83	93	84
Shanxi	292	284	253	328	397	395	373	313	260
Sichuan	155	158	179	204	232	254	271	280	205
Taiwan	100	106	98	106	113	118	106	114	111
Tianjin	77	86	99	136	152	114	102	97	88
Yunnan	83	109	105	97	118	159	144	176	126
Zhejiang	243	253	247	270	327	281	294	292	240
EastChina	4332	4502	4454	5382	6150	6402	6179	6201	4941