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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 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 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 almost 3 times higher and the NO_2 concentrations would be at least 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 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 accuracy. Theys et al. (2015) presented a new data set of SO_2 concentrations derived from the

Ozone Monitoring Instrument (OMI) satellite instrument (Levelt et al., 2006). 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.

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. 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 12th 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 12th 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. Regulations 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.

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). 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

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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). 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 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 quality of the SO_2 data for OMI, 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). For this study, the algorithm 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

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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 NOx 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. 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 2a). The underlying NO_x emissions are shown in Figure 2b. 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 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. Figure 3 shows the mean normalized time series for the 10 provinces with the highest total SO_2 columns (i.e. Tianjin, Shandong, Hebei, Shanxi, Henan, Beijing, Jiangsu, Shanghai, Anhui and Liaoning). The minimum and maximum of the time series for each year are shown in the grey shaded area to indicate the variability. 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 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. 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 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 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 4a, 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 3. 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). Figure 4b shows this 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., 2016). For the province Ningxia we see a very similar pattern occuring 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.

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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 5, based on data of NBSC (2015). 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 6. 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 6 to be able to further look into the past (starting in 2003) and to verify the results of OMI. Each 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 (overpass of SCIAMACHY and GOME-2) and afternoon (overpass of OMI), this can lead to differences between the instruments. Note that the NO_x emission data is by definition 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

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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. 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 years 2009 and the recent years 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, which explains the dip in pollution per fuel unit in 2009. 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 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 level we can conclude from Figure 6 that without these air quality regulations SO_2 concentrations would nowadays be more than 2 times higher and the NO_2 concentrations would be at least 30% 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 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 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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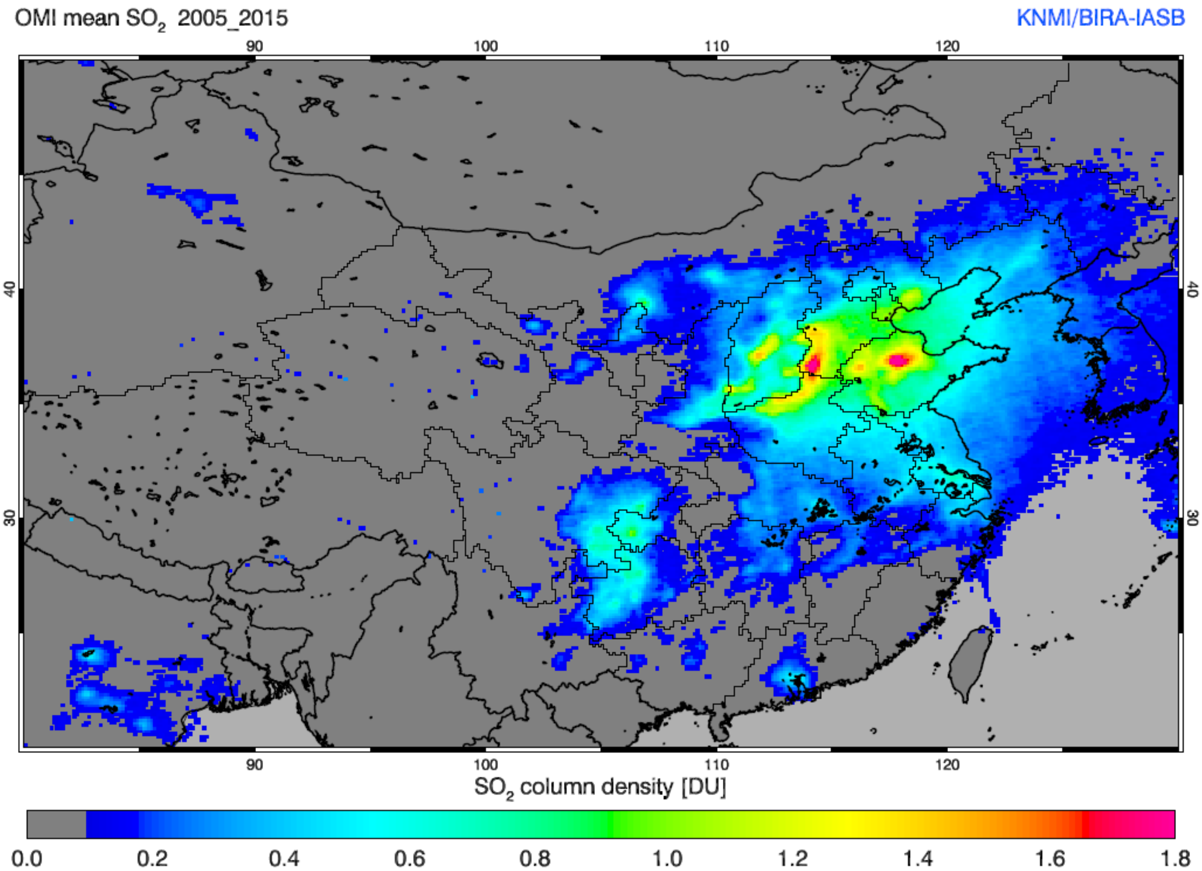

Figure 1: Average SO_2 concentrations for the period 2005 to 2015 as observed by the OMI satellite instrument. Data below 0.1 DU is masked (grey colour).

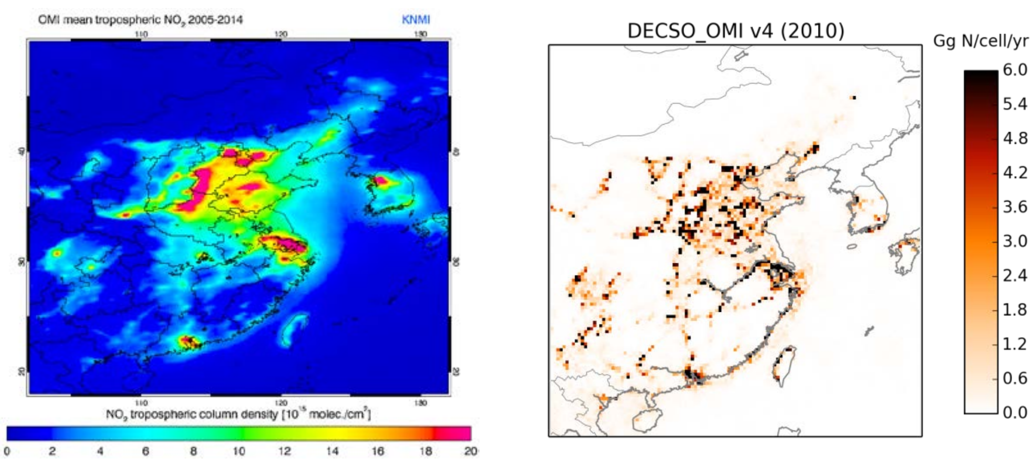

Figure 2: (a) The averaged tropospheric NO_2 concentrations over China measured by OMI in the period 2005-2014. (b) The NO_x emissions in the year 2010 derived from the OMI satellite observations.

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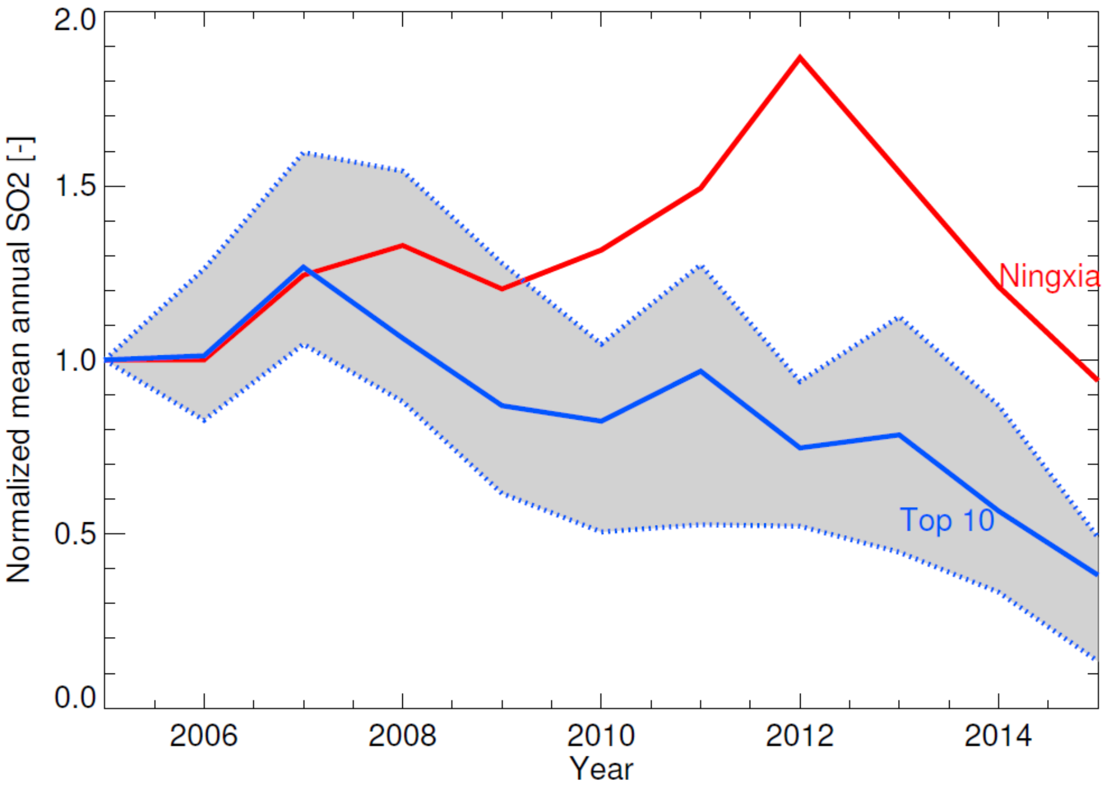

Figure 3: Time series (blue line) of the semi-annual mean (April-September) of the 10 provinces with the highest SO_2 concentrations derived from the OMI satellite observations. The time series are normalized to the value in 2005. The grey area indicates the maximum range of the individual values of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

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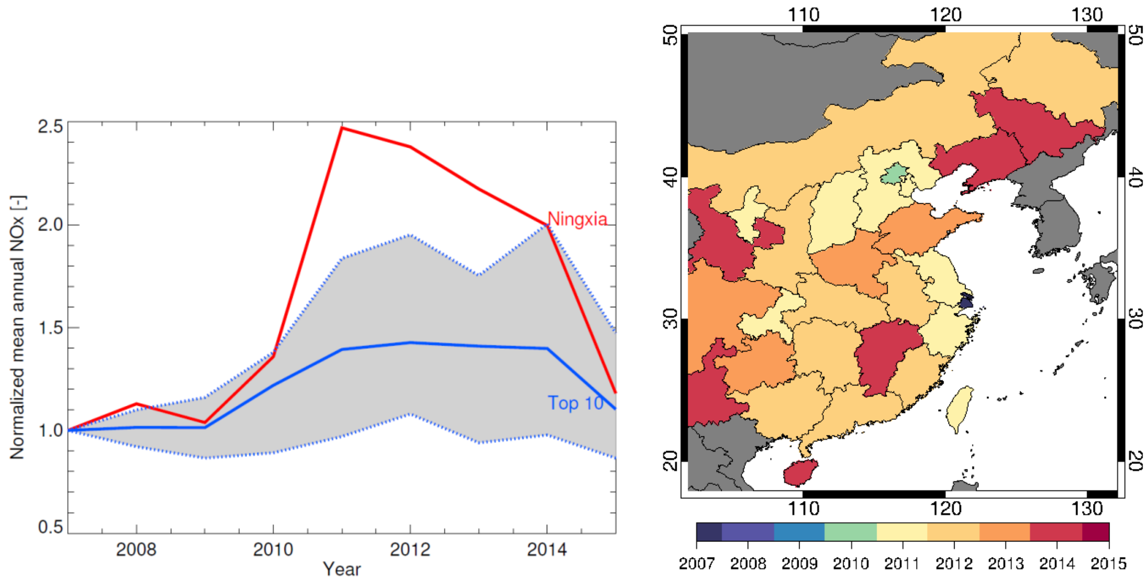

Figure 4: (a) Shown are the annual total NO_x emission estimates for the last 9 years for East China. Emissions are derived with DECSO V4 using OMI observations. (b) Peak year of the NO_x emissions per province.

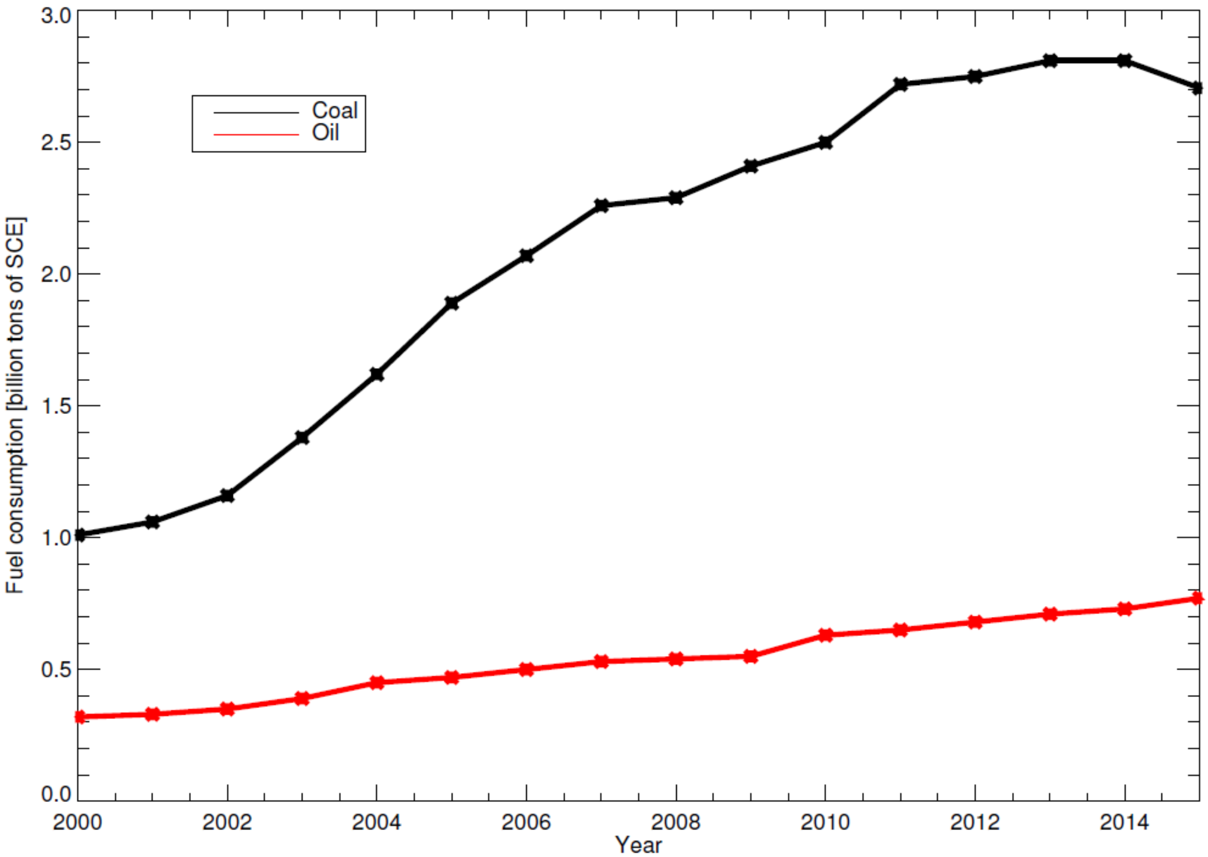

Figure 5: In black the annual oil consumption and in red the annual coal consumption for China is shown.

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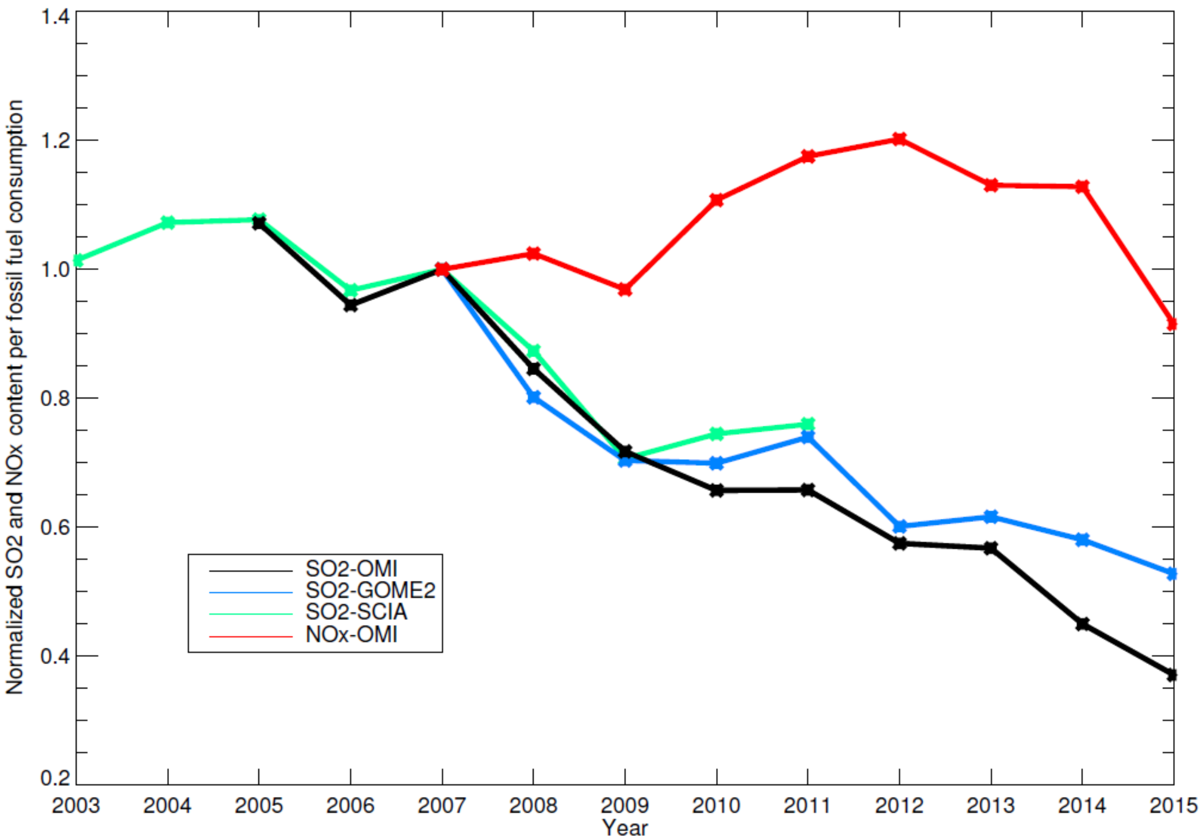

Figure 6: Time series of the ratio of the mean SO_2 columns and the fossil fuel consumption in China based on observations of OMI (black), SCIAMACHY (green), and GOME-2 (blue). The ratios of the annual NO_x emissions and the fossil fuel consumption is based on observations of OMI (red). All time series 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

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Table 2: 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	