#### 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.

- My main concern is that the authors fail to put their work into context with the recent publications on the decrease in 5 Chinese emissions. Several of these publications actually present city level trends in pollution and discuss the environmental policy at the city/province level. And several discuss changes in pollutant levels over time. There are also several papers on bottom-up emission inventories that should be reviewed. I recommend that the authors do a thorough search of the literature and discuss the major conclusions of these papers, so as to give credit to the work that has come before. In the process, the importance of your work will be put into perspective.
- 10 We are aware of the extensive literature on the topic of trends of emissions in China and indeed our discussion on these publications has been deliberately brief. On the other hand the aim of this paper is not to give an review of the existing literature on this topic, but more to focus on literature discussing similar approaches: papers related to satellite-derived trends of concentrations and papers discussing effects of policy making. Note that four of the recent papers on this topic were only published after our original submission of the paper.
- 15 However, we have expanded the paragraph discussing earlier work in the paper to include more studies on regional or city-scale trends and more recent papers on the decrease in NOx in the last years : "To study the efficiency of the environmental policies, we analysed satellite observations of SO<sub>2</sub> and tropospheric NO<sub>2</sub> of the last 11 years. SO<sub>2</sub> satellite observations over China have been studied earlier by Lee et al. (2010), Li et al (2011), He (2012), Yang et al. (2013), Fioletov et al. (2015), and Krotkov et al. (2015). Satellite observations are very
- 20 useful for SO<sub>2</sub> trend studies, as recently McLinden et al. (2016) showed that bottom-up inventories are underestimating SO<sub>2</sub> emissions worldwide with about 0-10%. NO<sub>2</sub> 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). These studies all showed a strong increase in NO2 over East China. On a city-scale or regional level in China trends are analysed and reported by Gu et al.( 2013), Schneider et al. (2015), and Duncan et al. (2016). Although some cities
- 25 showed already a decreasing trend, notably in the Pearl river delta. An overall decrease in NO<sub>2</sub> concentrations in China is only recently observed by Irie et al. (2016) Liu et al., (2016) and de Foy et al. (2016). To avoid the effects of transport and lifetime of NO2 several authors evaluated NOx emissions instead. Emission estimates of NO<sub>x</sub> 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)."
- 30 Additional references that are used are:

Duncan B N, Lamsal L N, Thompson A M, Yoshida Y, Lu Z, Streets D G, Hurwitz M, M and Pickering K E, 2016, A space-based, high-resolution view of notable changes in urban NOx pollution around the world (2005–2014),J. Geophys. Res. 121, 976–96

Gu D, Wang Y, Smeltzer C and Liu Z, 2013, Reduction in NOx emission trends over China: regional and seasonal variations, Environ. Sci. Technol., 47, 12912–9

Schneider, P., W.A. Lahoz and R.J. van der A, Recent Satellite-based Trends of Tropospheric Nitrogen Dioxide over Large Urban Agglomerations Worldwide, Atm. Chem. Phys., 2015, 15, 1205-1220, doi:10.5194/acp-15-1205-2015.
B. de Foy, Z. Lu and D. G. Streets, Satellite NO2 retrievals suggest China has exceeded its NOx reduction goals from

the twelfth Five-Year Plan, Scientific Reports 6, Article number: 35912 (2016) doi:10.1038/srep35912

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#### 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 regular font.

5 The authors have modified the original manuscript, but several of the requested "major revisions" have not been resolved. I cannot recommend publication before the following issues have been adressed adequately:

1. My main concern about the study is that the authors are quite bold with there statement that Chinese NO2 levels would be higher by 30% (now 25%) without regulations.

This is quite speculative and not supported by the shown data: on the one hand, the authors claim that regulations were increasingly taken over the last years, while on the other hand the 25% reduction is just estimated based on the value in 2015. So if the authors would have had written their study one year earlier, based on data for 2005-2014, they would have had to conclude that the many regulations taken so far are useless!

So unless the authors provide a good (and quantitative) explanation on why the NOx per fossil fuel ratio kept almost constant in 2010-2014, despite regulations, they cannot claim that the observed reduction in 2015 allows for a direct interpretation as "this is the accurately quantified benefit of regulation efforts".

- We claim that the reduction is at least 25 % (actually 27%) but we are also considering errors in the fossil fuel consumption numbers. This is a cautious estimate based on the years 2011 (the last year that is still not much affected by SCR installations) and the last year 2015. Without regulations we expect a continuous steady growth after 2012 in the NOx per fossil fuel consumption, because the percentage of the transport sector is increasing. (The transport
- 20 sector is emitting much more NOx per fossil fuel unit than the energy or industrial sectors.) This conclusion is based on the statistics of the NBSC (2015) and Wu et al. (2017), which shows that the transport is growing with a factor two every 5 years. This is much faster than any other sector is growing.

We have adapted the text to clarify this point at several places in the text at the end of section 3.

Important point in our conclusion is that regulations for NOx, unlike for SO2, were effective not earlier than 2012 and especially a big effort is made in the last two years as shown in Liu et al., (2016). We see the growth in NOx per fossil fuel stop and slowly decrease after 2012.

Interesting to see is that the decrease in NOx emission seems to be behind the actual installation of SCR technology. We saw something similar for SO2 where not the actual implementation of filtering was important but the control of the use of these filters a few years later made the change (see Xu, 2011). But if this is also the case for NOx filters is

30 pure speculation at this point.

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I already made this point in my first review. The reply given by the authors and their modifications in the text expand the discussion of (a) the changing role of transport and shipping, and (b) the effectiveness of SCR equipment.

35 However, (a) is provided qualitatively (which is fine for undestanding the general pattern, but far too vague to assess the robustness of the 25% reduction estimate),

Please note that it is very hard to get reliable statistics (or numbers at all) related to emissions in China. The exact numbers for transport and shipping are not available to us, however information on the relative growth of traffic is given in NBSC and Wu et al. (2017) and amount to a doubling every 5-6 years. However, this is not enough for

40 quantitatively analysis, which is the reason that we say the reduction is "at least 25 %", while due to higher contribution of traffic this should even be higher. We have given some clarification on this point in the text at the end of section 3.

while (b) refers to a submitted paper, not accessible yet, and now suddenly claims SCR equipment playing a key role (something not mentioned at all in the first version nor in the introduction of the second version).

Meanwhile, the paper has been published on 24 October in Environmental Research Letters. We hope that this take away the objection of the referee on this point. We have added in the introduction:

- 5 "According to Liu et al. (2016) Selective Catalytic Reduction (SCR) equipment was installed in this period and growing from a penetration of about 18% in 2011 until 86% in 2015. SCR equipment in power plants are expected to reduce the emissions of the power plant with at least 70% (ICAC, 2009). The SCR installation is the most significant measure taken to reduce the NO<sub>x</sub> emissions and it largely coincides with the peak year of observed NO<sub>2</sub> concentrations (Liu et al., 2016)."
- 10 "To our knowledge no regulations for ship emissions have been announced."

In addition I would like to point out that if you have to consider changes of the relative role of traffic (without having a specific number for this), the concept of the simple NOx/fuel ratio is meaningless.

15 Originally this concept was designed for SO2, while for NOx it is somewhat more difficult to interpret. However, we certainly do not see it as meaningless since based on the limited information we have we can still conclude that the environmental regulations, especially regarding SCR, are very effective recently.

Note that we really see a strong decrease in observed NO2 concentrations over China. By dividing it by fossil fuel we put it in perspective: in this way we see a trend in emission factor instead of emissions themselves. Whether this is

20 caused by shifts in the importance of certain sectors or by air quality regulations has been verified by additional information.

So the authors have to actually explain the years 2010-2014 as well as the year 2015 with data for (ship) traffic, SCR, etc. (including some error estimate of the NOx/fuel ratio, which is affected by uncertainties in both NOx emissions and fossil fuel inventory), or have to weaken their statements on NOx reduction.

We have included explanations for the growth and reduction of NOx per fossil fuel in the entire period of our research. The growth can be explained by the growing transport sector, the reduction in 2009 is a result of less transport due to the global economic recession and the last period of strong decrease of NOx emission is due to

- regulations for power plants (installing SCR) and on-road vehicle emission regulations. This has been explained in the text with related references. In addition we added the following remarks:
  "After 2009 the NO<sub>x</sub> per fossil fuel is slowly increasing because the transport sector is growing faster than the energy sector and has a higher emission factor. Statistics of the NBSC (2015) show that the transport is growing with a factor two every 5-6 years (Wu et al., 2017)."
- 35 "For NO<sub>x</sub> per fossil fuel we were expecting a gradual growth after 2012 because of the continuing relative growth of the transport sector. Keeping this in mind we compare the years 2015 with 2012 and conclude that without air quality regulations the NO<sub>2</sub> concentrations would be at least 25% higher in China today"

The uncertainty in NOx emissions is reduced to a negligible level as explained in the text due to the averaging over the province area and averaging over a time period of 12 months. The uncertainty of the fossil fuel numbers is much

40 higher but difficult to quantify. Papers on this topic are analysing earlier versions of the fossil fuel inventory. They show that the uncertainty varies over time and per province. Since the uncertainty of our fossil fuel numbers is unknown, we decided not to discuss this in the paper.

Wu, Y., S. Zhang, J. Hao, H. Liu, X. Wu, J. Hu, M. P. Walsh, T. J. Wallington, K. M. Zhang, S. Stevanovic, On-road vehicle emissions and their control in China: A review and outlook, Science of The Total Environment, 574, 332-349, doi:10.1016/j.scitotenv.2016.09.040, 2017

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#### 2. The topic of the paper is regulation, but the reader learns almost nothing about it.

In my first review, I have asked to "discuss the different possibilities in general, and the taken measures in detail". While the authors have addressed the second point in a new table, the general possibilities/techniques for reducing NOx emissions for cars, PPs etc. are not discussed in the introduction. This has to be added (including SCR!) with appropriate references.

10 We have added in the introduction of the revised version more details on the air quality regulations concerning the SCR equipment installations and the new car standards. Also some discussion on the expected NOx reductions is included. This resulted in the following new text:

" (.....) According to Liu et al. (2016) Selective Catalytic Reduction (SCR) equipment was installed in this period and growing from a penetration of about 18% in 2011 until 86% in 2015. SCR equipment in power plants are expected to

- 15 reduce the emissions of the power plant with at least 70% (ICAC, 2009). The SCR installation is the most significant measure taken to reduce the  $NO_x$  emissions and it largely coincides with the peak year of observed  $NO_2$ concentrations (Liu et al., 2016). At the same time China has implemented several new national emission standards for cars during the time period of our study (see Table 2). The change from China 3 to China 4 standard for cars in the period 2011-2015 reduces the maximum allowed amount of  $NO_x$  emissions for on-road vehicles with 50% (Wu et
- 20 al., 2017). More strict regulations for on-road vehicles (e.g. ban on older polluting cars) have been introduced in China on a provincial or even city level, e.g. in Beijing, rather than nationwide. To our knowledge no regulations for ship emissions have been announced.(....)"

3. Finally, I have asked for analysis on province level, as well as the other reviewer. While the authors now provide provincial data, they do not actually provide provincial analysis. This, however, would provide a qood opportunity to shed light on the discussion of the effectiveness of regulations. So, how does the NOx per fuel ratio look like for different provinces? Is it different for coastal provinces (with significant ship emissions)? Can you relate the temporal patterns per province to SCR equipment? Are the temporal patterns for Shanghai Expo/Olympic games/APEC matching our expectations?

30 Such an in-detail analysis on provincal level would reveal how robust the presented ratio actually is.

Unfortunately the provincial data on coal and oil consumption are not reliable enough to do these analyses for all provinces. Most uncertainties occur in the numbers related to oil, although also coal consumption numbers are not always correct. However, for the coal-consumption dominated provinces an analysis is possible, and that is what we added to the paper with the following discussion:

- 35 "On a provincial scale we can in principle do similar analyses, but unfortunately the provincial energy consumption related to coal and oil have a very high uncertainty due to inconsistencies in interprovincial imports and exports (Hong et al., 2016). We see this reflected in a high variability in the annual provincial data and sometimes missing data. Especially for the oil consumption the data have high uncertainties (Guan et al., 2011, Hong et al., 2016). Therefore, we have only analysed the 5 provinces with dominating coal consumption as shown in Figure 8. In this
- 40 graph we excluded Guizhou province because of its difficult to interpret coal consumption in 2011 as a result of large power shortages (NBSC, 2015, Xinhua, 2011). For SO2 per fossil fuel unit we see that all provinces follow the national trend. For NOx per fossil fuel we see more variation per province depending on the role of transport. Most of these by coal consumption dominated provinces start their decreasing trend from 2011 reflecting the national program on

SCR installations starting that same year. It is interesting to see that the commissioning of new power plants in 2011 causes a strong increase in both SO2 and NOx in the province Ningxia (see Figure 4 and 5a). However, when compensating for fossil fuel usage, one can see that the same national air quality regulations are applied here, as the trend in Figure 8 shows the same pattern as for other provinces. This show the strength of the presented method to

5 assess the efficiency of air pollution regulations."

The SCR and other air quality measures are clearly visible in the added Figure 8.

We are not focusing too much on a discussion on the temporary air quality regulations for certain cities since these measures are lasting for a few days or a couple of months at most and it is not the purpose of this paper to analyse these events. We have (co-)authored several other papers on this topic. Only for small provinces and big events like

10 the Olympic Games in Beijing and World Expo in Shanghai it enters our present results. Therefore we have only added the following sentence:

"Events like the Olympic Games in Beijing in 2008 and the World Expo in Shanghai in 2010, when temporary strict air quality regulations have been enforced, can be recognised in this Table as years with significant lower emissions for these provinces."

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#### Additional comments:

- Section 3.3: "Total NOx emissions in East China reached their peak levels in 2012, and have been decreasing since". From Figure 5, I would rather claim that overall emissions have stayed constant for 2012-2014, and decreased in 2015.

20 We have changed this sentence to "Total NO<sub>x</sub> emissions in East China reached their peak levels in 2012, and have stopped increasing since this year"

- Table 2: Please include a column on the expected NOx reduction.

We have added in the text in the introduction some discussion on the expected NOx reductions (see answer to point 2)

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## The next pages contain the marked-up manuscript

# Cleaning up the air: Effectiveness of air quality policy for SO<sub>2</sub> and NO<sub>x</sub> emissions in China

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Abstract. Air quality observations by satellite instruments are global 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 improved accuracy. To put these trends 15

- 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<sub>x</sub> emissions a distinct decreasing trend is 20 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<sub>x</sub> emissions are partly decided on a provincial level. The last three years show both a reduction in SO<sub>2</sub> and NO<sub>x</sub> 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<sub>2</sub> would be about 2.5 25 times higher and the NO<sub>2</sub> concentrations would be at least 25% 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 in observing the relatively short-living gases nitrogen dioxide (NO<sub>2</sub>) and sulphur dioxide (SO<sub>2</sub>).

- For these two trace gases improved data sets recently became available, enabling analysis of air quality time series on a 30 national or provincial level with improved accuracy. Theys et al. (2015) presented a new data set of SO<sub>2</sub> column densities derived from the Ozone Monitoring Instrument (OMI) satellite instrument (Levelt et al., 2006). They conclude that the SO<sub>2</sub> 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 35 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<sub>2</sub> into the atmosphere

40 (Klimont et al., 2013), while over 90% of the SO<sub>2</sub> 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

- 5 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<sub>2</sub>. However, plumes of active volcanoes are seldom observed over China.
- NO<sub>x</sub> is released by more or less the same anthropogenic sources, i.e. the burning of coal or oil. The main difference with SO<sub>2</sub> is that traffic is a much more important source for NO<sub>x</sub>. NO<sub>x</sub> 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<sub>x</sub> in China. According the MEIC inventory, 25% of NO<sub>2</sub> 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<sub>2</sub> 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<sup>th</sup> 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<sub>2</sub> is shown in Table 1, which includes the year of the beginning of the implementation.
- The regulation of NO<sub>x</sub> was started much later than for SO<sub>2</sub>. Although <u>T</u>the 12<sup>th</sup> five-year plan already-mentioned the intention to reduce NO<sub>2</sub> by 10 % (target) (ChinaFAQs project, 2012), only from 201<u>1</u>2 onward NO<sub>x</sub> filtering systems were installed, mainly at power plants but also for heavy industry. These regulations for NO<sub>x</sub> were announced in 2013 in the Air Pollution Prevention and Control Action Plan (CAAC, 2013) for the coming 5 years. According to Liu et al. (2016) Selective Catalytic Reduction (SCR) equipment was installed in this period and growing from a penetration of about 18% in 2011 until 86% in 2015. SCR equipment in power plants are expected to reduce the emissions of the power plant with at least 70%
- 25 (ICAC, 2009). The SCR installation is the most significant measure taken to reduce the  $NO_x$  emissions and it largely coincides with the peak year of observed  $NO_2$  concentrations (Liu et al., 2016). Gradually–At the same time China has implemented several new national emission standards for cars during the time period of our study (see Table 2). The change from China 3 to China 4 standard for cars in the period 2011-2015 reduces the maximum allowed amount of  $NO_x$  emissions for on-road vehicles with 50% (Wu et al., 2017). SomeMore strict regulations for <u>on-road</u> vehicles (e.g. ban on older
- 30 polluting cars) have been introduced in <u>China</u> on a provincial or even city level, <u>e.g. in Beijing</u>, rather than nationwide. <u>To</u> our knowledge no regulations for ship emissions have been announced. 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, <u>and the</u> APEC meeting in 2014, <u>etc.</u> but those regulations <u>wea</u>re 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 (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). <u>All these studies showed a strong increase in NO<sub>2</sub> over East China.</u>
   <u>On a city-scale or regional level trends are analysed and reported by Gu et al.(2013)</u>, <u>Schneider et al. (2015)</u>, and <u>Duncan et al. (2016)</u>. <u>Although some cities showed already a decreasing trend, notably in the Pearl river delta</u>, an overall decrease in <u>NO<sub>2</sub> concentrations in China is only recently observed by Irie et al. (2016) Liu et al., (2016) and De Foy et al. (2016).</u>

<u>exclude meteorology as a factor for variability in NO<sub>2</sub> several authors evaluated NO<sub>x</sub> emissions instead.</u> Emission estimates of NO<sub>x</sub> 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, whether of concentrations or emissions, linear trends of the concentration of air pollutants are often used.

5 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.
 We apply this to NO<sub>x</sub> emissions instead of concentrations for the period 2007 until 2015. The comparison of SO<sub>2</sub> trends with those of NO<sub>x</sub> emissions enables us to distinguish environmental policies specifically applied on coal-based industry and power plants with general environmental measures and trends in traffic.

#### 10 2 Observational data

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#### 2.1 Satellite observations of SO<sub>2</sub>

SO<sub>2</sub> 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<sub>2</sub> 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

- 15 algorithm has been developed (Theys et al., 2015) that improves the precision of the SO<sub>2</sub> 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
- 20 about the retrieval procedure are described in Theys et al. (2015). Also the operational algorithm of NASA for SO<sub>2</sub> 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<sub>2</sub> 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  $1/8^{\circ}$  by  $1/8^{\circ}$ . 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<sub>2</sub> 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<sub>2</sub> data have been recalculated by spatially averaging for each grid cell the data from the eight surrounding
- 40 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<sub>x</sub> 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

- 5 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<sup>2</sup>. The DECSO algorithm is applied to OMI NO<sub>2</sub> 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
- 10 use is available on 0.25 degree resolution for the period 2007-2015 on the web-portal <u>www.globemission.eu</u>.

#### 3 Temporal analysis over China

#### 3.1 Sources of SO<sub>2</sub> and NO<sub>x</sub> in China

The multi-annual mean of  $SO_2$  for 2005-2015 is shown in Figure 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  $SO_2$ 

15 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<sub>2</sub> concentrations are more distributed over the East of China, mainly because traffic 20 is an important source of NO<sub>x</sub> emissions (see Figure 3a). The underlying NO<sub>x</sub> emissions are shown in Figure 3b. Like for the SO<sub>2</sub> concentrations, NO<sub>x</sub> 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<sub>2</sub> trends over China

To construct time series of SO<sub>2</sub> we have averaged the data to annual means of the vertical columns derived from OMI. From

- 25 these annual mean  $SO_2$  data we constructed time series for each province (see Table A.1). Figure 4 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,
- 30 thus the reductions have been strongest in this province since 2005. Apart from Ningxia province, all provincial time series show very similar patterns. In general, the SO<sub>2</sub> concentrations were at a maximum in the year 2007, when the start of decreasing trend is visible in China. Despite some fluctuations the SO<sub>2</sub> 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. 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 3. From 2012 onward, the more stringent  $SO_2$  emission regulations also started to have effect in Ningxia.

#### 3.3 NO<sub>x</sub> emission trends over China

National NO<sub>x</sub> emission trends show a different pattern than those of SO<sub>2</sub>. 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<sub>x</sub> emissions in East

- 5 China reached their peak levels in 2012, and have <u>stopped increasingbeen decreasing</u> since <u>this year</u>. 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)
- 10 of the decrease in emissions varying over the provinces. Events like the Olympic Games in Beijing in 2008 and the World Expo in Shanghai in 2010, when temporary strict air quality regulations have been enforced, can be recognised in this Table as years with significant lower emissions for these provinces. In Figure 5a, the normalized (to the year 2007) time series of annual NO<sub>x</sub> emissions for East China (102-132°E, 18-50°N) is shown in similar way as for SO<sub>2</sub> in Figure 4. The mean, minimum and maximum of the 10 provinces with highest NO<sub>x</sub> emission are shown (Shandong, Hebei, Henan, Jiangsu,
- 15 Guangdong, Shanxi, Zhejiang, Anhui, Sichuan and Hubei), together responsible for 65% of all Chinese NO<sub>x</sub> emissions. The thin black lines show the times series for the individual 10 provinces, where the lower line represents Guangdong. Figure 5b shows 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
- 20 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.

#### 25 **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 6, 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

30 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

- 35 coal/oil. The effectiveness of environmental regulation will be reflected in a decrease of this emission factor. Therefore, we divide the annual SO<sub>2</sub> column measured from satellites and the annual NO<sub>x</sub> emissions by the annual coal and oil consumption in China. In this way we get a measure of the emitted SO<sub>2</sub> or NO<sub>x</sub> per unit (SCE) of fossil fuel consumption reflecting the Chinese environmental policy. The results are shown in Figure 7. One might argue that SO<sub>2</sub> 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 7 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 each data point is averaged over -12 months and the total area of China, which reduces the root-mean-square error to a negligible

- 5 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<sub>2</sub> 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<sub>2</sub> concentrations and when these weather conditions are different during the overpass of SCIAMACHY and GOME-2 (around 9:30 local time) and 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<sub>x</sub> emission data is in general not sensitive to meteorological variability.
- For SO<sub>2</sub> we see a big decrease in the years 2008 and 2009, while the desulphurization program of the 11<sup>th</sup> five-year plan
  started already in 2005/2006, when the authorities begin to reduce SO<sub>2</sub> emissions by installing desulphurization devices in many power plants (Lu et al., 2010). In 2006 SO<sub>2</sub> monitoring devices were also installed in the chimneys of the power plants. This resulted in a decrease in SO<sub>2</sub> emissions from 2006, while the much bigger decrease of SO<sub>2</sub> 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<sub>2</sub> content per consumed coal unit only slowly decrease until 2011. From 2012 onwards we see a stronger annual decrease in SO<sub>2</sub>. This coincides with the 12<sup>th</sup> 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<sub>x</sub> emissions the total annual emissions are used and divided in the same way as for SO<sub>2</sub> 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<sub>x</sub>
- 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<sub>x</sub>. In the early years we see in general a small increase in NO<sub>x</sub> emissions per fuel unit due to the increasing fraction of the transport sector in the fuel use. Exceptions are the year 2009 and the recent year 2015. The year 2009 coincides with the global economic crisis (Lin and McElroy, 2011) 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.
- 30 (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<sub>x</sub> per fuel values. This explains the dip in pollution per fuel unit in 2009. <u>After 2009 the NO<sub>x</sub> per fossil fuel is</u> <u>slowly increasing because the transport sector is growing faster than the energy sector and has a higher emission factor</u>. <u>Statistics of the NBSC (2015) show that the transport is growing with a factor two every 5-6 years (Wu et al., 2017)</u>. After
- 35 2012 the gradual increase of  $NO_x$  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 to a lesser account to 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.
- 40 By comparing the efficiency level in 2015 with earlier levels we can conclude from Figure 7 that without these air quality regulations SO<sub>2</sub> concentrations would nowadays be about 2.5 times higher. For NO<sub>x</sub> per fossil fuel we were expecting a gradual growth after 2012 because of the continuing relative growth of the transport sector. Keeping this in mind<del>and</del> we

compare the years 2015 with 2012 and conclude that without air quality regulations the  $NO_2$  concentrations would be at least 25% higher in China today.

On a provincial scale we can in principle do similar analyses, but unfortunately the provincial energy consumption related to coal and oil have a very high uncertainty due to inconsistencies in interprovincial imports and exports (Hong et al., 2016).

- 5 We see this reflected in a high variability in the annual provincial data and sometimes missing data. Especially for the oil consumption the data have high uncertainties (Guan et al., 2011, Hong et al., 2016). Therefore, we have only analysed the 5 provinces with dominating coal consumption as shown in Figure 8. In this graph we excluded Guizhou province because of its difficult to interpret coal consumption in 2011 as a result of large power shortages (NBSC, 2015, Xinhua, 2011). For SO<sub>2</sub> per fossil fuel unit we see that all provinces follow the national trend. For NO<sub>x</sub> per fossil fuel we see more variation per
- province depending on the role of transport. Most of these by coal consumption dominated provinces start their decreasing trend from 2011 reflecting the national program on SCR installations starting that same year. It is interesting to see that the commissioning of new power plants in 2011 causes a strong increase in both SO<sub>2</sub> and NO<sub>x</sub> in the province Ningxia (see Figure 4 and 5a). However, when compensating for fossil fuel usage, one can see that the same national air quality regulations are applied here, as the trend in Figure 8 shows the same pattern as for other provinces. This show the strength of the presented method to assess the efficiency of air pollution regulations.

#### **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

- 20 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 25 desulphurisation installations started. However, the effects are only visible in 2008 and 2009 when a strict control by the 26 Chinese authorities on the actual use of the desulphurisation installations started. In 2009, we see the effect of the air quality 27 regulations for  $SO_2$  and  $NO_x$  resulting from the global economic recession at the end of 2008. The increasing relative 28 contribution of the transport sector to the  $NO_x$  emission slowly increases the amount of  $NO_x$  per fossil fuel unit after 2009. 2009 After 2011 we see a steadily decreasing  $SO_2$  pollution per fossil fuel unit caused by various Chinese environmental
- 30 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

35 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 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

40 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 2: Average SO<sub>2</sub> 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:(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 4: Time series (red line) of the annual mean 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.



Figure 5: (a) Shown are the annual total  $NO_x$  emission estimates for the last 9 years for 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 6: In black the annual coal consumption and in red the annual oil consumption for China is shown.



Figure 7: 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.



Figure 8: Same graph as Figure7 but with time series for the provinces Hebei, Henan, Nei Mongol, Ningxia and Shanxi included. Time series per province of the ratio of the mean SO<sub>2</sub> columns and the fossil fuel consumption are drawn in blue. The ratios of the annual NO<sub>x</sub> emissions and the fossil fuel consumption per province are shown in red. All time series are normalized to the year 2007 and based on OMI observations.

Start year of	Regulation	Reference		
implementation				
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	Xu et al., 2011		
	power plants	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	MEP, 2015		
	plants			
2013	Mandatory SO <sub>2</sub> 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 1: Environmenta	l regulations of the	Chinese national	government to re	educe SO <sub>2</sub> in the air.
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Table 2: Environmental regulations of the Chinese national government to reduce NOx emissions.

Year of	Regulation	Reference
implementation		
2011-2015	Installation of Selective Catalytic Reduction (SCR) equipment	Liu et al., 2016,
	at power plants. In 2013 the SCR equipment was installed in	CAAC, 2013
	about 50% of all power plants.	
2007	China 3 (Euro 3) emissions standards for cars, nationwide	<u>Wu et al.,</u>
		2017www.dieselnet.com
2011	China 4 (Euro 4) emissions standards for gasoline cars,	<u>Wu et al.,</u>
	nationwide.	2017www.dieselnet.com
2015	China 4 (Euro 4) emissions standards for diesel cars,	Wu et al.,
	nationwide.	2017www.dieselnet.com

Table 3: Main newer plants in Ningvia province (> 600 MW). Data collected from www.sourcewatch.org
Table 5. Main power plants in Angxia province (> 000 MW). Data conected from www.sourcewatch.org.

Power plant	Capacity	In operation	Remark
	(MW)	since	
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

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

# Table A2 Annual NO<sub>x</sub> emissions (Gg N/year) per province in the domain of DECSO (in parenthesis the fraction of provincial area <u>considered</u>) 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 <u>(61%)</u>	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(74%)	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 (83%)	98	116	117	156	215	215	169	142	111
Ningxia	30	34	32	41	75	72	66	61	36
Qinghai	6	6	6	<del>10</del>	-14	13	45	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 (51%)	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 <u>(36%)</u>	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