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Detection from space of a reduction in anthropogenic emissions of nitrogen oxides during the Chinese economic downturn

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Abstract

Rapid economic and industrial development in China and relatively weak emission controls have resulted in significant increases in emissions of nitrogen oxides (NO_x) in recent years, with the exception of late 2008 to mid 2009 when the economic downturn led to emission reductions detectable from space. Here vertical column densities (VCDs) of tropospheric NO_2 retrieved from satellite observations by SCIAMACHY, GOME-2 and OMI (both by KNMI and by NASA) are used to evaluate changes in emissions of NO_x from October 2004 to February 2010 identifying impacts of the economic downturn. Data over polluted regions of Northern East China suggest an increase of 27–33% in annual mean VCD of NO_2 prior to the downturn, consistent with an increase of 49% in thermal power generation (TPG) reflecting the economic growth. More detailed analysis is used to quantify changes in emissions of NO_x in January over the period 2005–2010 when the effect of the downturn was most evident. The GEOS-Chem model is employed to evaluate the effect of changes in chemistry and meteorology on VCD of NO_2. This analysis indicates that emissions decreased by 20% from January 2008 to January 2009, close to the reduction of 18% in TPG that occurred over the same interval. A combination of three relatively independent approaches indicates that the economic downturn was responsible for a reduction in emissions by 9–11% in January 2009 with an additional decrease of 10% attributed to the slow-down in industrial activity associated with the coincident celebration of the Chinese New Year.

1 Introduction

The Chinese economy grew rapidly over the past decades, resulting in significant increases in energy demand met primarily by increases in the use of coal and other fossil fuels (Chinese Statistical Yearbook, 2009). This, together with relatively weak emission controls, has resulted in significant growth in emissions of nitrogen oxides ($NO_x \equiv NO + NO_2$) (Zhang et al., 2007, 2009a,b; Zhao et al., 2009; Lei et al., 2010;

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Lin et al., 2010a). Understanding the magnitude of emissions of NO_x in China and its recent trend has been a challenge, however. The bottom-up approach to estimating emissions of NO_x relies on information on emission activities and emission factors (Streets et al., 2003; Zhang et al., 2007, 2009a; Zhao et al., 2009; Lei et al., 2010). Inadequacies and inaccuracies both in the statistics of emission activities and in measurements of emission factors have resulted in large uncertainties in bottom-up estimates (Streets et al., 2003; Zhang et al., 2007, 2009a; Zhao et al., 2010).

Vertical column densities (VCDs) of tropospheric nitrogen dioxide (NO_2) are retrieved from multiple satellite instruments, including GOME, SCIAMACHY, GOME-2, and OMI (Boersma et al., 2004, 2007; Richter et al., 2005; Bucsela et al., 2008; Mijling et al., 2009). This information has been exploited to evaluate emissions of NO_x in China from the top-down perspective as an alternative to the bottom-up approach (Martin et al., 2003, 2006; Jaegle et al., 2005; Wang et al., 2007; Stavrakou et al., 2008; van der A et al., 2008; Lin and McElroy, 2010; Lin et al., 2010b). There are large uncertainties, however, associated with VCDs inferred for NO_2 as a consequence of the variety of assumptions and errors introduced at different stages of the retrieval process (Boersma et al., 2004, 2007, 2009; Bucsela et al., 2008; Zhou et al., 2009; Hains et al., 2010; Lamsal et al., 2010; Lin et al., 2010b) (see Sect. 3). Lin et al. (2010b) proposed a methodology to reduce the effect of retrieval errors on the derivation of emissions by combining data from multiple satellite instruments. This procedure results in a more realistic estimate of emissions of NO_x for different seasons (Lin and McElroy, 2010; Lin et al., 2010b). Questions remain however concerning the true magnitude of emissions and the effect of retrieval errors on top-down estimates.

Despite large uncertainties, retrieved VCDs of NO_2 have been used extensively to evaluate variations in emissions of NO_x in China that have occurred over the past 15 or so years. Combining retrievals from GOME and SCIAMACHY, Richter et al. (2005) showed that VCDs of NO_2 increased by about 50% over industrial regions of China between 1996 and 2004. Their findings were supported by subsequent studies (He et al., 2007; Zhang et al., 2007; Stavrakou et al., 2008; van der A et al., 2008; Yue

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et al., 2009). Zhang et al. (2007) compared changes in emissions of NO_x from 1995 to 2004 inferred from space with changes derived using the bottom-up methodology. Stavrakou et al. (2008) applied an assimilation approach to calculate changes in emissions of NO_x in China corresponding to changes in VCDs of NO_2 inferred from GOME and SCIAMACHY. They found an increase by about 66% from 1997 to 2006. Zhang et al. (2009b) employed changes in VCDs of NO_2 retrieved from OMI to pinpoint the significant expansion of the thermal power sector in Inner Mongolia between 2005 and 2007. Mijling et al. (2009) found a notable decrease in VCDs of NO_2 over North China based on retrievals from GOME-2 and OMI during the 2008 Beijing Olympics in response to restrictions imposed on traffic and mandated reductions in industrial activities.

The latest economic downturn of China that set in around late 2008 and persisted until late 2009 provides a good opportunity to evaluate the sensitivity of satellite retrievals to emissions of NO_x associated with variations in economic activities (Lin et al., 2010a). The downturn resulted in a significant slowdown in energy demand that is clearly reflected in power generation statistics (available at http://www.stats.gov.cn/tjsj/). In this study, reductions in emissions of NO_x as a result of the economic downturn are estimated based on changes in tropospheric VCDs of NO_2 retrieved from SCIAMACHY, GOME-2 and OMI. The inferred emission reductions are evaluated using data for thermal power generation (TPG) in China, and are contrasted to the general increase from October 2004 to February 2010. Particular efforts are made to analyze emissions of NO_x in January from 2005 to 2010 when the effect of economic activities on VCDs of NO_2 is most significant. Simulations using the global chemical transport model (CTM) GEOS-Chem are applied to identify the effect of changes in chemistry and meteorology as compared with changes in emissions on observed values for the VCDs of NO_2.

The present analysis is focused in Northern East China (108.75° E–123.25° E, 29° N–41° N; see Fig. 1). Covering approximately 17% of the land area in China, this region is characterized by intensive industrial activity and dense population. It is estimated to have contributed to as much as 55% of anthropogenic emissions of NO_x in China in

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2006 (Zhang et al., 2009a), resulting in large-scale high values for the VCDs of NO_2 (Richter et al., 2005; Stavrakou et al., 2008; van der A et al., 2008; Lin et al., 2010b). In addition, data coverage is relatively good for satellite retrievals in this region (Fig. 1), an important factor contributing to a reduction in the uncertainties associated with the representativeness of the data applicable to the analysis of changes in emissions of NO_x.

Sections 2 and 3 present Chinese TPG data and four retrieval products for tropospheric VCDs of NO_2 employed in this study. Section 4 analyzes seasonal and interannual variations in TPG and VCDs of NO_2 from October 2004 to February 2010 in response to the changing pace of economic development. Section 5 describes a detailed calculation of the changes in emissions of NO_x in January from 2005 to 2010 based on the TPG data and retrieved values for the VCDs of NO_2, further quantifying the effect of the economic downturn on emissions in January 2009. In this section, simulations of GEOS-Chem are used to evaluate the effect of variations in chemistry and meteorology on VCDs of NO_2. Section 6 concludes the present study.

2 Thermal power generation

Growth in TPG is a relatively good proxy for growth in anthropogenic emissions of NO_x in China. TPG is estimated to have accounted for approximately 44% of anthropogenic emissions of NO_x in 2006 (Zhang et al., 2009a). It is highly correlated with industrial activity which was responsible for ~26% of NO_x emissions in 2006 (Zhang et al., 2009a): close to 70% of total electricity generation was consumed by industry in recent years. According to Zhang et al. (2007), TPG in China grew by a factor of 1.64 from 2000 to 2004, as compared to an increase of 1.48 in anthropogenic emissions of NO_x. The difference reflects effects of changes in emission factors and changes in emissions from the transportation and residential sectors, responsible for 24% and 6% of NO_x emissions in 2006, respectively (Zhang et al., 2009a).

The unpublished dataset of the National Bureau of Statistics of China for monthly TPG in China from October 2004 to February 2010 were employed in this study as a guide to our estimate of changes in emissions of NO_x. Also included are monthly datasets for total power generation in China from all sources and total power generation for the three regional electricity grids serving the domain of this study (East China Grid, Central China Grid and North China Grid; see McElroy et al., 2009 for geographical specifications of the grids). As shown in Fig. 2, changes in annual power generation for the three regional grids are in excellent agreement with changes in total power generation in China. Thus relative changes in total TPG in the three grids were assumed to be the same as relative changes in total TPG in China. This information will be used to evaluate changes in emissions of NO_x estimated from interannual variations of VCDs of NO_2 retrieved from satellite instruments.

3 Retrievals of VCDs of tropospheric NO_2

VCDs of tropospheric NO_2 retrieved from SCIAMACHY, GOME-2 and OMI were used in this study to evaluate changes in emissions of NO_x from October 2004 to February 2010. Four retrieval products were employed (Table 1), including one for each of SCIAMACHY and GOME-2 derived by the Royal Netherlands Meteorological Institute (KNMI) (Boersma et al., 2004; Mijling et al., 2009) and two retrievals for OMI implemented independently by KNMI (Boersma et al., 2007) and by the National Aeronautics and Space Administration (NASA) Goddard Space Flight Center (GSFC) (Bucsela et al., 2008). (The two OMI products are identified as OMI_KNMI and OMI_NASA hereafter.) Data for GOME-2 are available since March 2007. SCIAMACHY and GOME-2 pass over China in the morning; OMI in the afternoon. OMI has the largest viewing swath (2600 km) and the highest horizontal resolution at nadir view (13 km × 24 km), followed by GOME-2 and SCIAMACHY. OMI_NASA uses less strict criteria for cloud screening than OMI_KNMI, resulting in greater data coverage (Fig. 1). Detailed descriptions of key parameters of the retrievals are presented in Table 1. The level-2

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data of the three KNMI retrievals were gridded to daily level-3 datasets at a horizontal resolution of 0.25° longitude × 0.25° latitude; for OMI_NASA, the conversion was implemented by NASA.

Tropospheric VCDs of NO_2 are retrieved in three steps. Satellite instruments observe the UV/VIS radiance reflected from the atmosphere. The reflectance is reduced due to absorption by NO_2 in the slant column along the pathway of light. This information is used to derive the slant column density (SCD) of NO_2. The SCD is partitioned then into separate contributions from the stratosphere and troposphere. Finally, the tropospheric SCD is divided by an air mass factor (AMF) to obtain the tropospheric VCD of NO_2. The SCD is derived by both KNMI and NASA using a Differential Optical Absorption Spectroscopy (DOAS) technique. In deriving tropospheric SCDs, an assimilation approach utilizing the global CTM TM4 is employed by KNMI to account for the dynamic variability of NO_2 in the stratosphere (Boersma et al., 2004, 2007). With respect to OMI_NASA, the contribution from stratospheric NO_2 is derived by selecting SCDs at locations where the influence of tropospheric NO_2 is not expected to be significant, smoothing meridionally with a boxcar function, followed by a zonal planetary wave analysis up to wave-2 (Bucsela et al., 2008). The calculation of AMF also differs significantly between the approaches by KNMI and by NASA, differences which contribute significantly to resulting uncertainties in the ultimately retrieved values for VCDs of NO_2. Errors in AMF result from inaccuracies in assumptions concerning surface albedo, cloud fraction, cloud pressure, and the a priori vertical profile adopted for tropospheric NO_2. The three KNMI retrievals employ information on surface albedo derived from TOMS (Herman and Celarier, 1997) and GOME (Koelemeijer et al., 2003), while OMI_NASA utilizes information based on GOME alone. For cloud parameters, the methods of FRESCO (Koelemeijer et al., 2001), FRESCO+ (Wang et al., 2008) and O_2–O_2 (Acarreta et al., 2004) are used for SCIAMACHY, GOME-2 and OMI (both by KNMI and by NASA), respectively. In calculating the AMF, the vertical profile of NO_2 has to be predetermined. The KNMI retrievals make use of real-time profiles simulated by TM4 (Boersma et al., 2004, 2007). In OMI_NASA, a constant annually-averaged

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vertical profile simulated by GEOS-Chem is used for each location (Bucsela et al., 2008). More analyses and validations of the retrievals are discussed in previous studies (Boersma et al., 2004, 2007, 2009; Bucsela et al., 2008; Zhou et al., 2009; Hains et al., 2010; Lamsal et al., 2010; Lin et al., 2010b).

Systematic biases have been identified in the retrieval products (Boersma et al., 2008, 2009; Bucsela et al., 2008; Zhou et al., 2009; Hains et al., 2010; Lamsal et al., 2010; Lin et al., 2010b). It is estimated that VCDs retrieved by KNMI may be overestimated by up to 40% based on in situ measurements in Europe and the US (Boersma et al., 2009; Zhou et al., 2009; Hains et al., 2010; Lin et al., 2010b). Lamsal et al. (2010) found significant errors over the US in the seasonality of VCDs in OMI_NASA attributed partly to the use of time independent a priori vertical profiles for NO_2. Retrieval errors are expected also for China, though they cannot be quantified currently due to lack of accurate in situ measurements.

4 Changes in TPG and VCDs of NO_2 over October 2004–February 2010

4.1 Changes in TPG

As shown in Fig. 2, Chinese TPG grew significantly and relatively linearly until the economic downturn that set in around late 2008. The annual average TPG increased by 49% from 5.23 terawatt hours per day (TWh day^{-1}) between October 2004 and September 2005 to 7.79 TWh day^{-1} between October 2007 and September 2008. Under the influence of the downturn, it decreased by as much as 6% from the prior maximum. Correspondingly, the annual average total power generation decreased by about 3%. The decrease in TPG was more significant than the decrease in total power generation reflecting the preference to reduce thermal power rather than cleaner sources of energy (hydro power, nuclear power, etc.). Starting from mid 2009, growth of power generation resumed in pace with the recovery of the economy.

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In addition to the general interannual trend, the TPG varies month to month in response to seasonal variation in energy demand (Fig. 2a). It increases in summer and winter to meet the demand for air conditioning and heating, respectively. It minimizes normally in February in conjunction with the Chinese New Year (CNY) which results in large-scale reductions in industrial activities and energy demand. The effect of the CNY is compared to the effect of the economic downturn in Sect. 5.3.

4.2 Changes in retrieved VCDs of NO_2

4.2.1 Seasonal variability

The seasonal variation of VCDs of NO_2 differs significantly from that of power generation (Fig. 2a). VCDs of NO_2 maximize in winter due to increases in anthropogenic emissions associated with domestic heating and, more importantly, increases in the atmospheric lifetime of NO_x as a result of reduced photochemical activity. In particular, the lifetime of NO_x in winter is 3–5 times longer than in summer (Martin et al., 2003; Lin and McElroy, 2010). VCDs of NO_2 minimize in summer when NO_x is removed rapidly through reaction with the hydroxyl radical. Emissions of NO_x from lightning and soil are greatest in this season and mixing in the PBL is most intense. NO_x emitted from surface is lofted to higher altitudes where satellite instruments are more sensitive to the presence of NO_2. These factors tend to increase values for retrieved VCDs of NO_2, while their effects are more than offset by the negative impact of the shorter lifetime. In general, VCDs of NO_2 decrease by a factor of 3–7 from winter to summer (Fig. 2a).

The seasonality of VCDs of NO_2 differs between the four retrieval products (Fig. 2a). OMI_KNMI suggests the strongest seasonal variation of NO_2. Values retrieved by OMI_NASA exceed OMI_KNMI in summer, while they are much smaller than OMI_KNMI in winter. The differences are attributed in part to the fact that OMI_NASA uses an annually-averaged a priori vertical profile for NO_2 for all seasons and does not account for increases in mixing in the PBL from winter to summer (Lamsal et al., 2010). As the sensitivity of satellite instruments increases with the height of NO_2 in the troposphere,

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a given amount of radiance received by the instruments can be interpreted by the retrieval algorithm as either a lower concentration of NO_2 at higher altitudes or a higher concentration at lower altitudes. Use of an annual average a priori vertical profile for NO_2 is expected to result in an overestimate of tropospheric VCDs retrieved in summer with an underestimate in winter. Another difference between the two retrievals is associated with the allocation of NO_2 to the stratosphere and troposphere. OMI_KNMI attributes more NO_2 to the stratosphere than OMI_NASA, and the difference is greatest in winter (Lamsal et al., 2010). This factor, however, cannot account for the larger seasonality of VCDs of NO_2 in OMI_KNMI as compared to OMI_NASA.

For the three KNMI products, the seasonality of VCDs of NO_2 retrieved from SCIAMACHY and GOME-2 is smaller than that from OMI (Fig. 2a). This may be due in part to the fact that SCIAMACHY and GOME-2 pass over China in the morning when the variation of the lifetime of NO_x with season is less than the change in the afternoon during the overpass of OMI, as indicated by our model simulations.

4.2.2 Interannual trend

Despite differences in seasonality between retrieved VCDs of NO_2 and TPG, interannual trends of VCDs were generally consistent with trends in TPG (Fig. 2b). From October 2004 to September 2008, annual mean VCDs of NO_2 increased by 33%, 30% and 27% for SCIAMACHY, OMI_KNMI and OMI_NASA, respectively, as compared to the increase of 49% in TPG. The smaller increases in VCDs may be attributed in part to the overall reduction in emission factors during the time period, as expected from the reduction inferred for the prior decade (Zhang et al., 2007). Other factors may include changes in chemical and meteorological conditions affecting the lifetime of NO_x, and/or natural sources of NO_x that did not change significantly over the years.

Both OMI retrievals indicate that, prior to the downturn, the magnitude of VCDs of NO_2 increased most significantly in winter due to the longer lifetime of NO_x (Fig. 2b). Changes in VCDs of NO_2 retrieved from SCIAMACHY were more variable than those inferred from OMI due likely to much lower data coverage (Fig. 1).

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The decline of industrial activities due to the economic downturn resulted in a reduction in VCDs of NO_2 during the interval from late 2008 to mid 2009 (Fig. 2). The four retrieval products suggest that the temporary minimum in annual mean VCDs of NO_2 associated with the downturn was 12–14% lower than the peak prior to the onset of the downturn. The reductions were about twice as large as the reduction in annual TPG, as a result of the timing of the economic downturn. The downturn was most intense during the 2008–2009 winter, the season in which the lifetime of NO_x is longest and the magnitude of VCD of NO_2 is most sensitive to changes in emissions. Thus the impact is exaggerated for a given amount of emission reduction on the annual average VCD of NO_2.

5 Quantifying the effect of the economic downturn on emissions of NO_x in winter

As discussed in Sect. 4, the extent of the economic downturn was greatest during the winter of 2008–2009 (Fig. 2). In addition, emissions of NO_x from other sources (lightning, soil and biomass burning) are negligible in winter, allowing for a more direct analysis of the relation between anthropogenic emissions of NO_x and VCDs of NO_2 retrieved from space. Here the six January months over the period 2005–2010 were selected for purposes of quantifying the effect of the downturn on emissions of NO_x based on retrieved values of VCDs for NO_2. Results are compared with independent analysis exploiting the TPG data.

5.1 TPG-inferred changes in emissions of NO_x

The TPG in January increased between 2005 and 2010 except for the reduction from January 2008 to January 2009 (Fig. 3). It increased by as much as 51% from 2005 to 2008, decreased by 18% from 2008 to 2009, and increased by 43% from 2009 to 2010.

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The relatively low TPG in January 2009 was a result of the economic downturn compounded by the impact of the CNY. The CNY is associated with an officially recommended 7-day holiday. Industrial activities are reduced significantly nation-wide during the holiday, and certain industries may be affected for a longer period (i.e., from several days before to several days after the holiday). The timing of the CNY is based on the Chinese calendar. Thus its exact date on the Gregorian calendar varies from one year to another. The holiday is held normally in February. In 2009, however, it ocurred during the period 25–31 January (Table 2).

Emissions of NO_x from TPG are estimated to have decreased from January 2008 to January 2009 by the same amount as the reduction in TPG. This is a reasonable approximation as emission factors should not change significantly over the 1-year period. The low TPG in January 2009 was driven mainly by reduced demand from industry for electricity associated with the economic downturn and the CNY, while demand for electricity for residential may have remained relatively constant. Thus reductions of emissions from industry (residences) may be larger (smaller) than 18%, provided that emission factors for these two sectors remained relatively constant from January 2008 to January 2009. Meanwhile, emissions from transportation may be affected to some extent both by the economic downturn and by the CNY. Considering the major contribution of TPG and industry to emissions of NO_x, it is estimated that total anthropogenic emissions of NO_x decreased by 18% from January 2008 to January 2009. The effect of the economic downturn is distinguished from the effect of CNY in Sect. 5.3.

5.2 Changes in emissions of NO_x in January derived from satellite retrievals

Changes in VCDs of NO_2 are determined by changes in emissions of NO_x as well as by changes in meteorology and chemistry affecting the atmospheric lifetime of NO_x and the partitioning between NO_2 and nitric oxide (NO). The impact of changes in emissions is identified in this study by separating the effect of changes in chemical and meteorological conditions as simulated by the global CTM GEOS-Chem.

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5.2.1 GEOS-Chem simulations

GEOS-Chem (http://wiki.seas.harvard.edu/geos-chem/index.php/Main_Page) is a global CTM used worldwide to study tropospheric chemistry. Version 8.1.1 of the model was used in this study. The model was run with the full O_x-CO-NO_x-VOC-HO_x chemistry at a resolution of 2.5° longitude × 2° latitude with 47 vertical layers. It was driven by the assimilated meteorological fields of GEOS-5 produced by the NASA Global Modeling and Assimilation Office (GMAO). The non-local scheme formulated by Holtslag and Boville (1993) was used to simulate mixing in the PBL. Anthropogenic emissions in Asia representative of 2006 taken from the INTEX-B mission were used for NO_x, carbon monoxide (CO) and non-methane volatile organic compounds (NMVOC) (Zhang et al., 2009). They were held constant in simulations for all years. Detailed model setup was described by Lin and McElroy (2010) and Lin et al. (2010b).

In calculating modeled VCDs of NO_2 corresponding to the three KNMI retrievals, the averaging kernel (AK) was applied to simulated vertical distributions of NO_2 to eliminate the effect of the a priori vertical profiles of NO_2 assumed in the retrieval process (Fig. 3; red solid lines); for OMI_NASA, the AK is not available for this calculation. For comparison purposes, modeled VCDs of NO_2 without applying the AK are shown also in Fig. 3 (green solid lines). Unless stated otherwise, AK's were accounted for in all of the modeled VCDs employed in the subsequent analysis.

Model values for the VCDs of NO_2 are much smaller than retrieved values, as shown in Fig. 3a. For January 2006, model results are about 50%, 49% and 76% of the retrievals from SCIAMACHY, OMI_KNMI and OMI_NASA, respectively. By January 2010, model VCDs amount to only about 31%, 24%, 33% and 49% of values derived from SCIAMACHY, GOME-2, OMI_KNMI and OMI_NASA, respectively. The large and increasing differences are attributed to several factors. Aside from the short-term reduction due to the economic downturn, emissions of NO_x are expected to have increased in recent years as a consequence of rapid industrial development and inadequate emission controls (Zhao et al., 2009; Lei et al., 2010; Lin et al., 2010a). Another factor

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relates to the systematic biases in the retrievals discussed in Sect. 3. Errors in GEOS-Chem may also be an important source of the model-retrieval differences (Lin and McElroy, 2010). In particular, the heterogeneous reaction of N_2O_5 with liquid water on the surface of aerosols is a major loss pathway of NO_x in winter. The current model uses the rate of uptake of N_2O_5 by aerosols compiled by Evans and Jacob (2005). More recent measurements (Bertram et al., 2009; Brown et al., 2009) suggested that the actual rate of uptake may be an order of magnitude lower than assumed by Evans and Jacob (2005). Our preliminary analysis for January 2009 using the nested version of the model (0.667° longitude × 0.5° latitude) showed that reducing the rate of uptake by a factor of 10 resulted in an increase by 13–18% in modeled regional mean VCDs of NO_2 across Northern East China. To derive changes in emissions of NO_x using satellite retrievals and model simulations, we assumed that the overall systematic bias from the two sources is constant throughout the years, as discussed below.

5.2.2 Interannual trend of emissions of NO_x

Interannual changes in retrieved VCDs of NO_2 relative to a reference year, R, can be decomposed approximately in terms of a factor representing the effect of changes in emissions, R_e, a factor reflecting the effect of changes in chemistry and meteorology (that is simulated by GEOS-Chem), R_m, and a factor representing the effect of changes in systematic errors in retrievals and CTM simulations, R_b. In other words, $R=R_e \cdot R_m \cdot R_b$, where $R_e \cdot R_m$ represents changes in the actual VCDs of NO_2. Assuming errors in retrievals and simulations relative to their true values are constant over time, i.e., $R_b=1$, then $R=R_e \cdot R_m$. Thus changes in emissions can be calculated as $R_e=R/R_m$. Here the reference year was chosen to be 2009 in order to better define the impact of the economic downturn.

Changes from January 2005 to January 2010 in monthly mean VCDs of NO_2 derived from OMI_KNMI are generally consistent across Northern East China (Fig. 4). Averaged over the region, the VCD decreased by 9% from January 2005 to January 2006, increased by 51% from January 2006 to January 2008, and then decreased by

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25% from January 2008 to January 2009 (Fig. 3). Changes in simulated VCDs (with the AK applied) from 2005 to 2009, while of a smaller magnitude, indicate a pattern similar to results from the retrieval (Fig. 3; red solid lines). The changes reflect the effect of variations in chemistry and meteorology that cannot be neglected. The impact of the choice of a priori vertical profiles for NO_2 is evident from the differences between simulated VCDs with and without applying the AK (Fig. 3; red solid lines versus green solid lines). Due to the existence of clouds, ice, snow and instrument limitations (e.g., row anomalies in OMI measurements), retrievals are not available for all locations in all days. The effect of data availability is apparent in representing changes in monthly mean VCD of NO_2 by using data in days with valid retrievals versus data in all days (Fig. 3, green solid lines versus blue solid lines).

Changes in monthly emissions of NO_x in January derived from OMI_KNMI are shown in Fig. 3b (orange dotted line) and Fig. 5. As compared to changes in TPG, increases in emissions are more moderate from 2005 to 2008 attributed likely to decreases in emission factors (Zhang et al., 2007; Zhao et al., 2009). From 2008 to 2009 and then from 2009 to 2010, emissions of NO_x first decreased by 20% and then re-grew by 43%, close to changes observed for TPG (Fig. 3b; pink solid line).

Under specified meteorological conditions, changes in emissions may affect the lifetime of NO_x and its partitioning into NO_2 and NO through the nonlinear photochemistry, which may lead to relative changes in VCDs of NO_2 that are not simply proportional to relative changes in emissions. This effect was evaluated by feeding into the model simulation for January 2010 the 47% increase in anthropogenic emissions of NO_x from January 2006 to January 2010 derived based on OMI_KNMI. A sensitivity simulation for January 2010 driven by the enhanced emissions resulted in an increase of about 47% in monthly mean VCDs of NO_2. This suggests that changes in emissions of NO_x derived in this study were affected insignificantly by the nonlinear photochemistry.

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Changes in emissions of NO_x relative to OMI_NASA can be derived using the same approach. This retrieval product assumes time-invariant a priori vertical profiles for NO_2 and does not provide the AK. Thus, in deriving the emissions, simulated VCDs of NO_2 both with and without applying the AK taken from OMI_KNMI were used to approximate the effect of chemistry and meteorology. As shown in Figs. 3a and 4, VCDs for NO_2 in January derived from OMI_NASA varied differently from OMI_KNMI throughout the years. VCDs decreased more significantly from 2005 to 2006, decreased slightly from 2007 to 2008, and decreased by 12% only from 2008 to 2009. As a result, changes in emissions estimated from OMI_NASA differ significantly from those derived from OMI_KNMI as well as from the changes indicated by the TPG data (Figs. 3b and 5). In particular, there is a slight reduction from 2007 to 2009 (Fig. 3b; purple dotted and purple dashed lines).

Emissions of NO_x in January derived from GOME-2 decreased by 10% from 2008 to 2009 and increased by 35% from 2009 to 2010 (Figs. 3b and 5). Emissions of NO_x relative to SCIAMACHY were nearly constant between 2007 and 2009 (Figs. 3b and 5). The lack of variation is inconsistent with changes in TPG and is attributed likely to errors associated with the low data coverage of SCIMACHY.

Overall, changes in emissions of NO_x derived from OMI_KNMI seem to best capture changes in emissions inferred from changes in TPG. Therefore OMI_KNMI is used in the following section to quantify the individual effects of the economic downturn and CNY on emissions of NO_x in January 2009.

5.3 Effect of the economic downturn versus CNY

Three approaches were proposed to separate the impact of the downturn from that of CNY in January 2009. The latter can be estimated from the TPG data. CNY occurred in February in the four years of 2005, 2007, 2008, and 2010 (Table 1). In these years, CNY was assumed to be the sole cause of differences in daily TPG between January and February. Its effect was calculated then as the reduction of daily TPG from January to February. The reduction is about 8% for 2005, 15% for 2007, and 13% for both 2008

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and 2010. The effect of CNY is estimated accordingly as the average of these values, 12%.

The CNY holiday in 2009 took place between 25–31 January with large impacts on economic activities. Industrial production may not have fully resumed by early February. For this year, the effect of CNY may be allocated mainly to January and to a lesser extent to February. Assuming CNY affected January 2009 alone and that its impact on industrial activities was independent of the economic downturn, it is estimated that the downturn led to a reduction of 7% in daily TPG. Assuming a smaller impact of CNY in January 2009, at 10%, the effect of the downturn is estimated at 9%.

A similar analysis was applied to emissions of NO_x derived from OMI_KNMI, assuming that CNY affected daily emissions in the same way as it impacted daily TPG. Based on the retrieval, the effect of the downturn on emissions of NO_x in January 2009 was calculated as 9% (11%) corresponding to the effect of the CNY assumed as 12% (10%).

A third approach was implemented by calculating the emissions corresponding to OMI_KNMI using retrieval data for days of January that were not influenced significantly by CNY. In January 2009, retrieved VCDs declined rapidly after 20 January (Fig. 6a). A minimum value was observed on 23 January, coincident with the passage of a cold front (with sharp increases in surface air pressure and decreases in surface air temperature) bringing cleaner air from the north. This cold event was captured by the model, including the timing of the local minimum in VCDs of NO_2 (Fig. 6). However, modeled VCDs experienced much weaker day-to-day variation after 20 January as compared to retrieved data. Therefore it is concluded that emissions of NO_x were affected significantly by CNY after 20 January 2009.

In the third approach, the "monthly mean" VCD in January 2009 that would be independent of CNY was approximated by the mean of VCDs during 1–20 January. This approximation was made both for OMI_KNMI and for model results in deriving the emissions. The calculation was unchanged for monthly mean VCDs in other months. Under this assumption, it is found that emissions of NO_x decreased by about 11% from

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January 2008 to January 2009. This reflects the effect of the economic downturn alone, and is consistent with results from previous approaches assuming an impact from the CNY of 10%.

Overall, our best estimate suggests that the CNY and the economic downturn were responsible for reductions in emissions of 10% and between 9 and 11%, respectively.

6 Conclusions and discussion

Tropospheric VCDs of NO_2 over the period October 2004 to February 2010 retrieved from SCIAMACHY, GOME-2 and OMI (both by KNMI and by NASA) were used to evaluate recent changes in emissions of NO_x in industrialized and densely populated Northern East China and to identify the effect of the economic downturn during late 2008–late 2009. On an annual basis, retrieved VCDs of NO_2 indicate a general increase from late 2004 to late 2008, a subsequent significant reduction during the downturn, and a resumption of growth since mid 2009. The interannual variations are generally consistent with changes in thermal power generation used as a proxy for anthropogenic emissions of NO_x. Prior to the downturn, annual mean VCDs of NO_2 increased by 27–33% for retrievals from SCIAMACHY and OMI (both by KNMI and by NASA), compared to the increase by 49% in thermal power generation.

More detailed analysis was conducted to quantify changes in emissions of NO_x in January over 2005–2010 when the effect of the downturn was most evident. The global CTM GEOS-Chem was used to identify the effect of chemistry and meteorology on interannual variations of VCDs of NO_2. A sensitivity analysis suggests a relatively linear relationship between emissions of NO_x and VCDs of NO_2 under given meteorological conditions with an insignificant effect of changes in emissions on the lifetime of NO_x and on partitioning between NO_2 and NO.

Interannual changes in emissions of NO_x in January derived based on the OMI product retrieved by KNMI best capture changes in emissions inferred from the thermal power generation data. Emissions estimated from the particular retrieval decreased

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by 20% from January 2008 to January 2009, consistent with the decrease of 18% in thermal power generation.

The decline in emissions of NO_x from January 2008 to January 2009 is found to be a consequence both of the economic downturn and of decrease in industrial activity during the Chinese New Year. The two factors were separated by three approaches based on data for thermal power generation and VCDs of NO_2 retrieved from OMI by KNMI. It is concluded that the economic downturn led to a reduction of between 9 and 11% in emissions of NO_x, as compared to a reduction of 10% associated with celebration of the Chinese New Year.

The present study suggests that satellite retrievals of tropospheric VCDs of NO_2, while subject to significant uncertainties, provide meaningful information for evaluating relative changes in emissions of NO_x. This is consistent with previous studies (Richter et al., 2005; He et al., 2007; Stavrakou et al., 2008; van der A et al., 2008; Mijling et al., 2009; Yue et al., 2009; Zhang et al., 2009b). Furthermore, since emissions of NO_x are tied closely to economic and industrial activities, retrievals of VCD of NO_2 may be used to provide an indirect proxy for changes in the economy – a valuable contribution particularly when official economic data are not readily available.

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Table 1. Properties of NO_2 retrievals.

Instrument	SCIAMACHY	GOME-2	OMI	OMI
Institute	KNMI	KNMI	KNMI	NASA
Retrieval version	TM4NO2A v1.10 (level-2)	TM4NO2A v1.10 (level-2)	DOMINO v1.0.2 (level-2)	OMNO2e (daily level-3)
Onboard satellite	Envisat	MetOp-A	EOS-Aura	EOS-Aura
Overpass time over China	~10:30 a.m.	~10:00 a.m.	~02:00 p.m.	~02:00 p.m.
Nadir view resolution	$30\times60\,km^2$	$40\times80\,km^2$	$13\times24\,km^2$	$13\times24\,km^2$
Viewing swath	960 km	1920 km	2600 km	2600 km
Global coverage	6 days	~1 day	1 day	1 day
Spectral window	220–2380 nm	425–450 nm	405–465 nm	405–465 nm
Spectral resolution	0.44 nm	0.50 nm	0.63 nm	0.63 nm
Stratospheric contribution	TM4 assimilation	TM4 assimilation	TM4 assimilation	Simplified (see text)
Cloud scheme	FRESCO	FRESCO+	O_2–O_2	O_2–O_2
Cloud screening	Cloud radiance<50% (Cloud fraction<15%)	Cloud radiance<50% (Cloud fraction<15%)	Cloud radiance<50% (Cloud fraction<15%)	Cloud fraction<30%
Surface reflectivity	TOMS+GOME	TOMS+GOME	TOMS+GOME	GOME
A priori vertical profile of NO_2	TM4 simulations	TM4 simulations	TM4 simulations	Time independent
Data source	www.temis.nl	www.temis.nl	www.temis.nl	mirador.gsfc.nasa.gov
Reference	Boersma et al. (2004)	Boersma et al. (2004); Mijling et al. (2009)	Boersma et al. (2007)	Bucsela et al. (2008)

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Table 2. Officially recommended holidays associated with Chinese New Year.

Year	2005	2006	2007	2008	2009	2010
Dates of holiday	9 Feb–15 Feb	29 Jan–4 Feb	18 Feb–24 Feb	6 Feb–12 Feb	25 Jan–31 Jan	13 Feb–19 Feb

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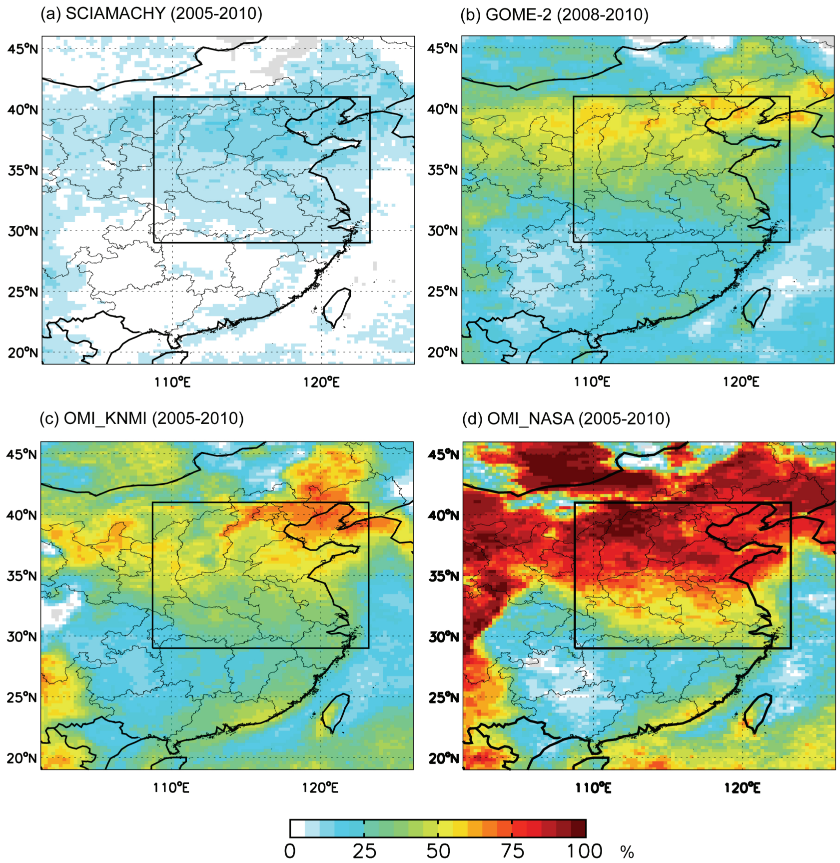

Fig. 1. The percentage of days in January with valid data averaged over 2005–2010 in the four retrieval products. OMI_NASA has more coverage than OMI_KNMI because of weaker criteria for cloud screening. Also shown is boundary specifications (black lines) of Northern East China analyzed in this study.

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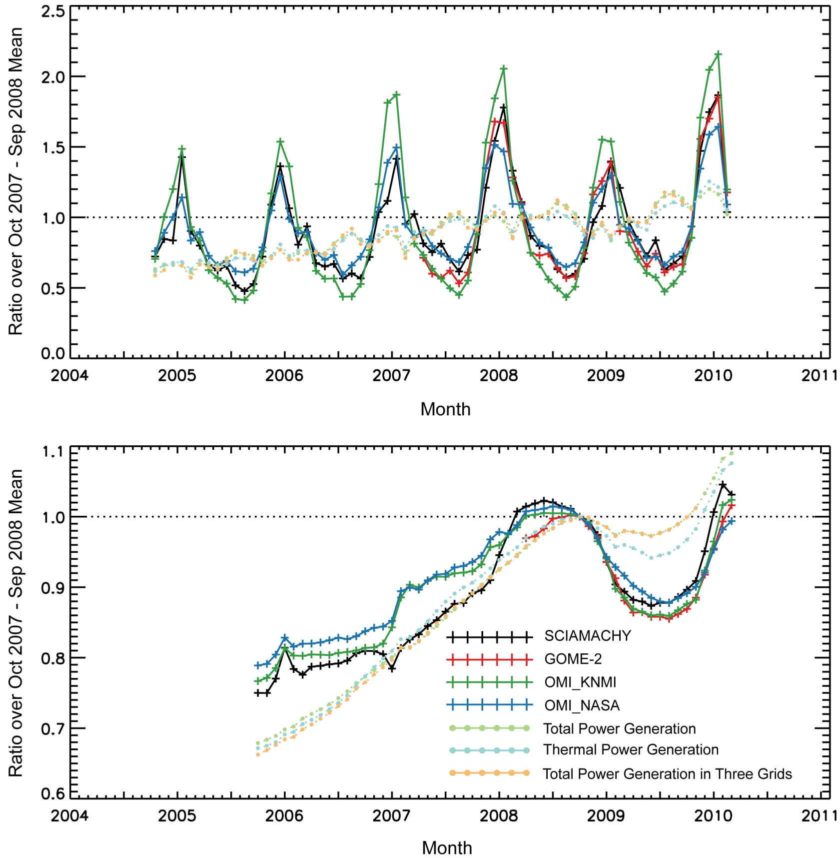

Fig. 2. Monthly variations in regional mean VCDs of NO_2 and power generation in China relative to values averaged over October 2007–September 2008 (top panel), and corresponding variations after a 12-month moving average (bottom panel; each point represents the mean over prior 12 months).

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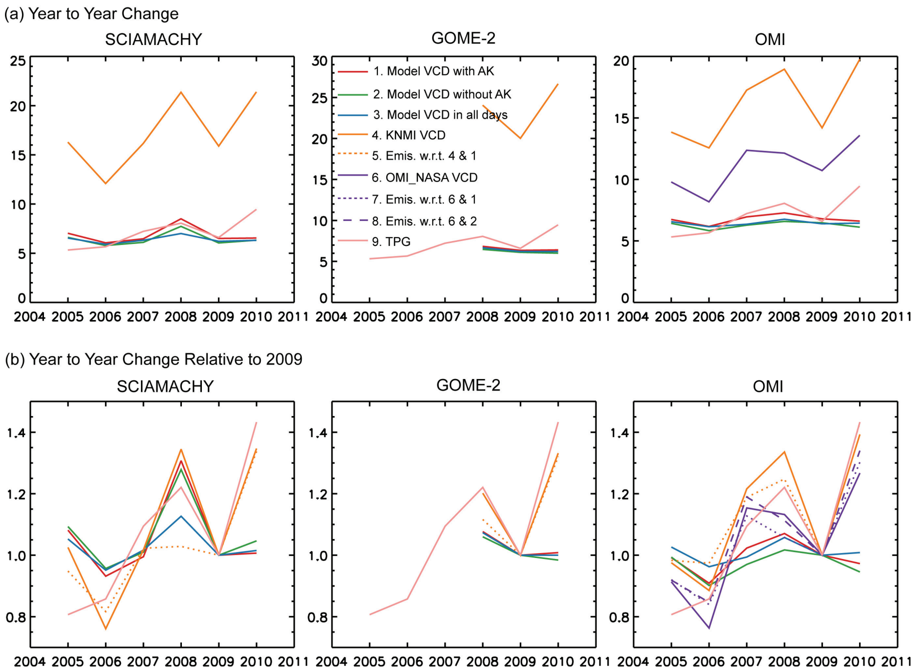

Fig. 3. Changes in TPG, VCDs of NO_2 and derived emissions of NO_x in January from 2005 to 2010. **(a)** Changes in the magnitude of TPG ($TWh\,day^{-1}$) and VCDs of NO_2 (10^{15} $molec\,cm^{-2}$). **(b)** Changes in TPG, VCDs of NO_2 and emissions of NO_x relative to values in January 2009. See text for the derivation of emissions of NO_x.

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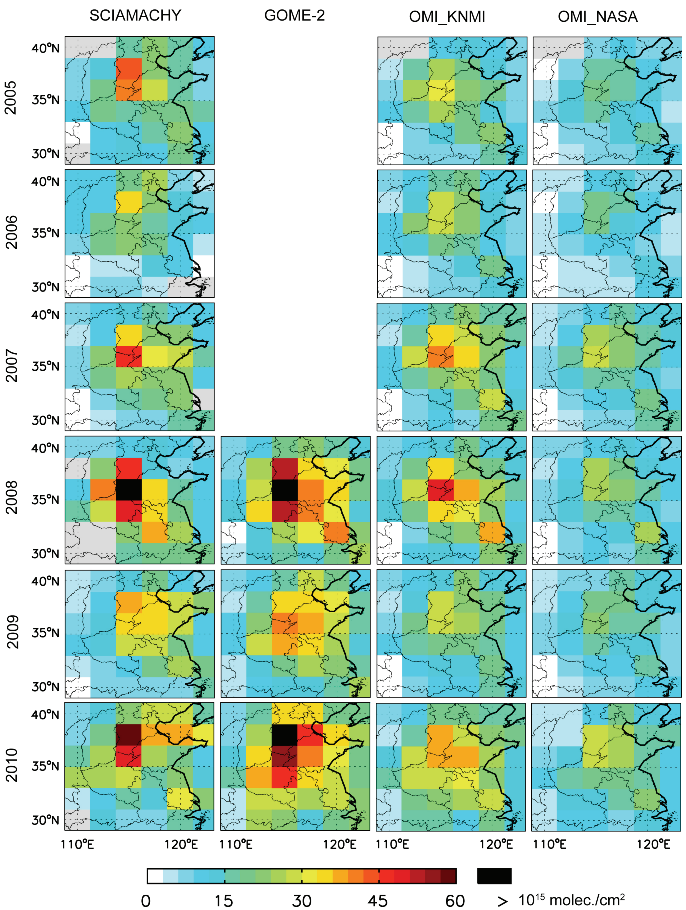

Fig. 4. Spatial distributions of retrieved VCDs of NO_2 in January over Northern East China. Data are presented at the model resolution of 2.5° longitude × 2° latitude. Areas in grey color do not have valid retrievals.

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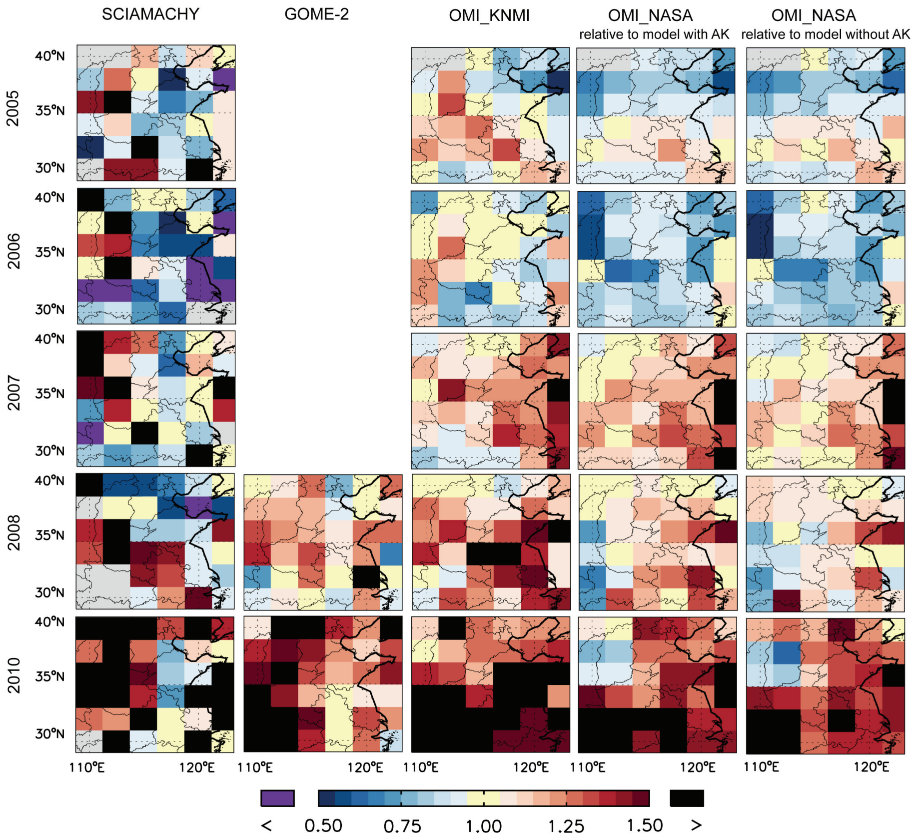

Fig. 5. Derived emissions of NO_x in January of 2005–2010 relative to emissions in January 2009 over Northern East China. Data are presented at the model resolution of 2.5° longitude × 2° latitude. Areas in grey color do not have valid retrievals. See text for the derivation of emissions of NO_x.

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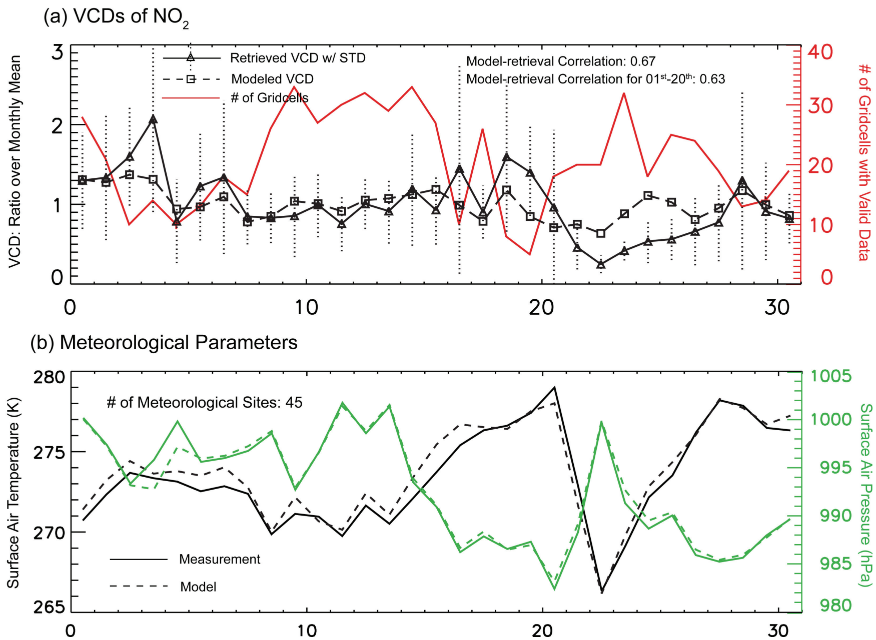

Fig. 6. Daily variations of regional mean VCDs of NO_2 and meteorological parameters in January 2009. **(a)** Variations of retrieved and modeled VCDs of NO_2 relative to regional means. Data in a given day are derived as the mean of the ratios of VCDs in gridcells with valid retrievals in that day over monthly mean VCDs in corresponding gridcells. Also shown is the number of gridcells out of the total of 36 over the region in each day that contain valid retrieval data. **(b)** Variations of measured and modeled daily average surface air temperature and surface air pressure. Meteorological measurements are taken from the global hourly dataset (DS3505) archived in the National Oceanic and Atmospheric Administration (NOAA) National Climatic Data Center (NCDC) (http://www7.ncdc.noaa.gov/CDO/cdo; see Lin et al., 2010a). Daily average values for air temperature (air pressure) are calculated to be means over 3-hourly (6-hourly) data in correspondence to the temporal resolutions of individual model parameters.

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