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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 12-month 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 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; errors in the estimate are most likely less than 3.4 %.

1 Introduction

Nitrogen oxides ($NO_x \equiv NO + NO_2$) are an important constituent in the troposphere, affecting the formation of ozone and aerosols and acid deposition. They are emitted from anthropogenic combustion sources, biomass burning, soil and lightning. In the air, they are destroyed mainly through the formation of nitric acid under two major pathways, including reaction of NO_2 with the hydroxyl radical (OH) and heterogeneous reaction of nitrogen pentoxide (N_2O_5) with liquid water on aerosol surfaces (Seinfeld and Pandis, 2006; Martin et al., 2003; Lamsal et al., 2010). They may also be converted to reservoirs like peroxyacetyl nitrate (PAN) and other organic nitrates (Seinfeld and Pandis, 2006). The partitioning of NO_x into NO and NO_2 is controlled by photolysis of NO_2 and reactions of NO with ozone and the hydroperoxyl radical (HO_2) (Seinfeld and Pandis, 2006).

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, 2009). This, together with relatively weak emission controls, has resulted in significant growth in emissions of NO_x (Zhang et al., 2007, 2009a, b; Zhao et al., 2009; Lei et al., 2010; 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; Richter et al., 2005; Boersma et al., 2007; 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; Jaeglé et al., 2005; Martin et al., 2006; 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 a 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 (van der A et al., 2006; He et al., 2007; Zhang et al., 2007; Stavrakou et al., 2008; van der A et al., 2008; Yue 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 anthropogenic 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 year-on-year (i.e., value compared to the
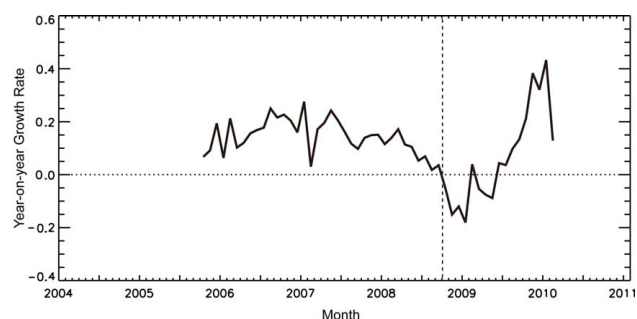

Fig. 1. Year-on-year growth rate in monthly thermal power generation in China. The value for a particular month is calculated as its percentage difference from the same month of the previous year. The vertical dashed line denotes the start of negative growth in thermal power generation.

same period in the previous year) changes of power generation (Fig. 1). Figure 1 shows that the rate of year-on-year growth in monthly thermal power generation (TPG), an indication of China's booming economy, started to decline in 2008 summer and became negative by October 2008. The growth rate stayed negative, with the lowest values over the 2008–2009 winter, until mid 2009 when TPG started to resume its growth. (The exception in February 2009 was a result of different timing of the Chinese New Year between 2008 and 2009; see below.) In this study, changes in tropospheric VCDs of NO_2 retrieved from SCIAMACHY, GOME-2 and OMI are used to evaluate the impact of the economic downturn on emissions of NO_x. The inferred emission reductions are evaluated using the TPG data, and are contrasted to the general increase from October 2004 to February 2010.

Particular efforts are made to quantify changes in emissions of NO_x in January from 2005 to 2010 based on retrieved VCDs of NO_2, by identifying the effect of changes in chemistry and meteorology on VCDs of NO_2 using simulations of the global chemical transport model (CTM) GEOS-Chem. Retrievals of NO_2 in wintertime may be affected by larger solar zenith angle and snow/ice on the ground. Choosing such months, however, is based on considerations for a larger signal of the economic downturn, a better relation between anthropogenic sources of NO_x and VCDs of NO_2, as well as an interesting timing of the Chinese New Year affecting also economic activities. In the 2008–2009 winter, the economic downturn was at its most intense phase with largest reductions in TPG (Fig. 1), therefore the signal of changing economy can be better captured from space with smaller interference from retrieval errors and other contaminations. In winter, emissions of NO_x from biomass burning, soil and lightning are very small in China (Jaeglé et al., 2005; Lin and McElroy, 2010), therefore a direct relation is easier to obtain between VCDs of NO_2 and anthropogenic sources of NO_x. For other seasons, soil and lightning sources contribute to a
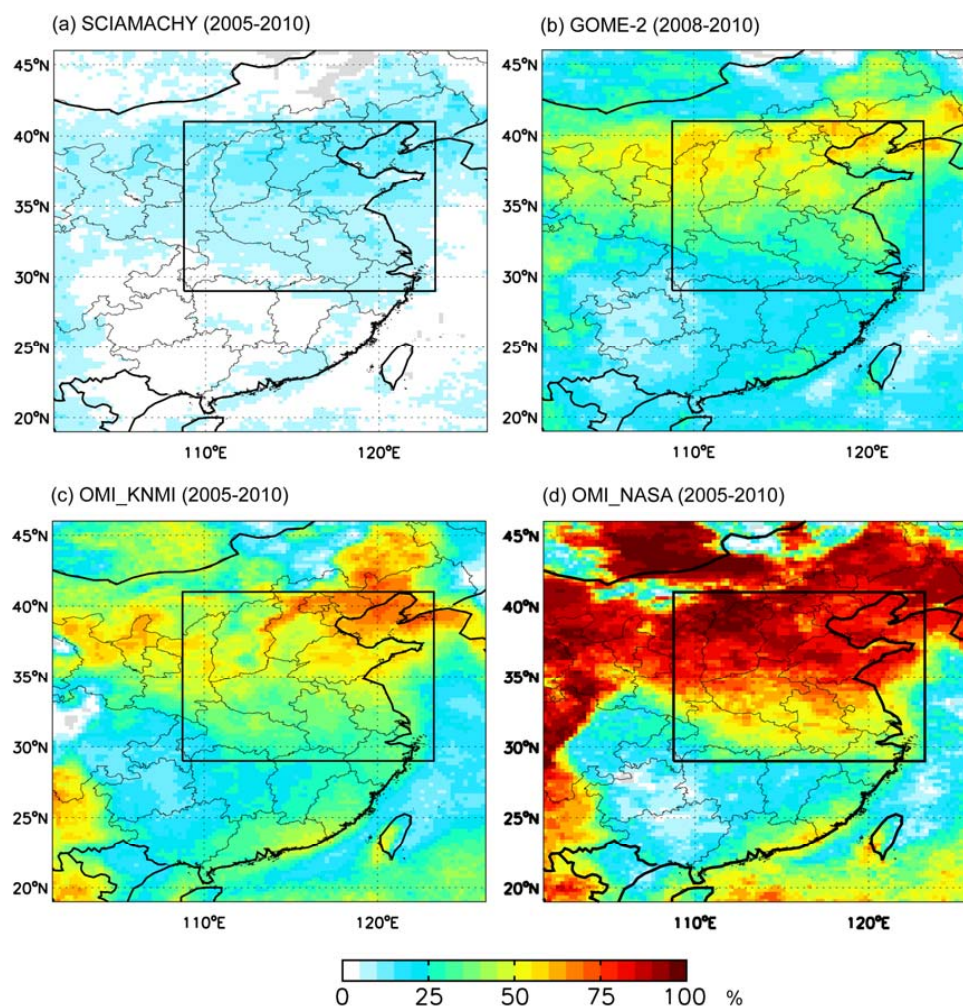
Fig. 2. The percentage of days in January with valid data averaged over 2005–2010 in the four retrieval products. OMI_NASA has more coverage than OMI_KNMI because of weaker criteria for cloud screening. Also shown is boundary specifications (black lines) of Northern East China analyzed in this study.

large portion of total emissions (summer in particular; see Jaeglé et al., 2005; Wang et al., 2007; Lin et al., 2010b), introducing interference in relating VCDs of NO_2 and anthropogenic sources of NO_x.

Furthermore, emissions of NO_x in January 2009, differing from the same period in other years, are of particular interest also because of the coincident occurrence of the Chinese New Year (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); providing another opportunity to evaluate satellite retrievals in detecting short-term emission reductions. Following the CNY, the dates of the holiday on the Gregorian calendar vary from one year to another: the holiday is held in February in most years while it occurred during 25–31 January for 2009 (Table 1).

The holiday, together with the economic downturn, resulted in the largest year-on-year decline in monthly TPG in January 2009 (Fig. 1). In the present study, the signal in satellite retrievals for holiday-associated reductions in emissions of NO_x in January 2009 is identified, and is compared with the signal for emission reductions due to the economic downturn.

The present analysis is focused in Northern East China (108.75° E–123.25° E, 29° N–41° N; see Fig. 2). 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 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

Table 1. Officially recommended holidays associated with Chinese New Year.

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

in this region (Fig. 2), 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 introduce Chinese TPG data and four satellite products for VCDs of tropospheric NO_2 employed in this study. Section 4 briefly describes the GEOS-Chem model. Section 5 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 6 presents a detailed calculation of the changes in emissions of NO_x in January from 2005 to 2010 based on the TPG data and satellite products, further quantifying the effects both of the economic downturn and of the CNY 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 7 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., 2007, 2009a).

The unpublished dataset of the National Bureau of Statistics of China for monthly TPG in China from October 2004 to February 2010 are 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. 3b, changes in 12-month 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 are 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 are used in this study to evaluate changes in emissions of NO_x from October 2004 to February 2010. Four products are employed (see Table 2 for detailed descriptions), including the SCIAMACHY and GOME-2 products retrieved by the Royal Netherlands Meteorological Institute (KNMI) (Boersma et al., 2004; Mijling et al., 2009) and two OMI products retrieved 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 x 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. 2). Detailed descriptions of key parameters of the retrievals are presented in Table 2. The level-2 data of the three KNMI retrievals are gridded to daily level-3 datasets at a horizontal resolution of 0.25° long×0.25° lat; for OMI_NASA, the conversion is 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. For each product, the SCD is derived with the 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; Boersma et al., 2007; Dirksen et al., 2011). With

Table 2. Properties of NO_2 retrievals.

Instrument	SCIAMACHY	GOME-2	OMI	OMI
Institute	KNMI	KNMI	KNMI	NASA
Retrieval version	TM4NO$_2$A v1.10 (level-2)	TM4NO$_2$A v1.10 (level-2)	DOMINO v1.0.2 (level-2)	OMNO$_2$e (daily level-3), a.k.a. "Standard Product"
Onboard satellite	Envisat	MetOp-A	EOS-Aura	EOS-Aura
Overpass time over China	~10:30 LT	~10:00 LT	~14:00 LT	~14:00 LT
Nadir view resolution	30×60 km^2	40×80 km^2	13×24 km^2	13×24 km^2
Viewing swath	960 km	1920 km	2600 km	2600 km
Global coverage	6 days	~1 day	1 day	1 day
Spectral window	426.3–451.3 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 %[a]	Cloud radiance <50 %[a]	Cloud radiance <50 %[a]	Cloud fraction <30 %
Surface reflectivity	TOMS + GOME	TOMS + GOME	TOMS + GOME[b]	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	http://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)

[a] (Over Northern East China in January months) For SCIAMACHY, the cloud fraction is always less than 25 %, and is less than 15 % (20 %) for 88 % (99.6 %) of the cases. For GOME-2, the cloud fraction is always less than 31 %, and is less than 15 % (20 %) for 74 % (94 %) of the cases. For OMI_KNMI, the cloud fraction is always less than 32 %, and is less than 15 % (20 %) for 76 % (93 %) of the cases.

[b] Since 17 Feburary 2009, the surface reflectance dataset based on OMI measurements was used.

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 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. SCIAMACHY and GOME-2 employ information on surface reflectance derived from TOMS (Herman and Celarier, 1997) and GOME (Koelemeijer et al., 2003). OMI_KNMI also employs the TOMS/GOME dataset prior to 17 Feburary 2009, since when a new reflectance dataset based on OMI measurements is used. OMI_NASA utilizes information based on GOME alone for surface reflectance. 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; Boersma et al., 2007). In OMI_NASA, a constant annually-averaged vertical profile simulated by GEOS-Chem is used for each location (Bucsela et al., 2008). More analyses and validations of the re-

trievals are discussed in previous studies (Boersma et al., 2004; Boersma et al., 2007; Bucsela et al., 2008; Boersma et al., 2009; 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; Bucsela et al., 2008; Boersma et al., 2009; 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), with a bias in winter higher than in summer (Boersma et al., 2011). 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.

Recently, several improvements have been made to reduce retrieval errors in OMI_KNMI (Boersma et al., 2011); to date most improvements have been applied only to data over 2004–June 2008 (see www.temis.nl). In particular, a new look-up table (LUT) for radiative transfer calculation is implemented, resulting in a 17 % reduction in retrieved VCDs of NO_2 over East Asia (Boersma et al., 2011). Inaccuracies in the old LUT (for retrievals analyzed here) are consistent in time and thus should not introduce significant errors in our trend analysis. Since 17 Feburary 2009, OMI_KNMI employs the surface reflectance dataset based
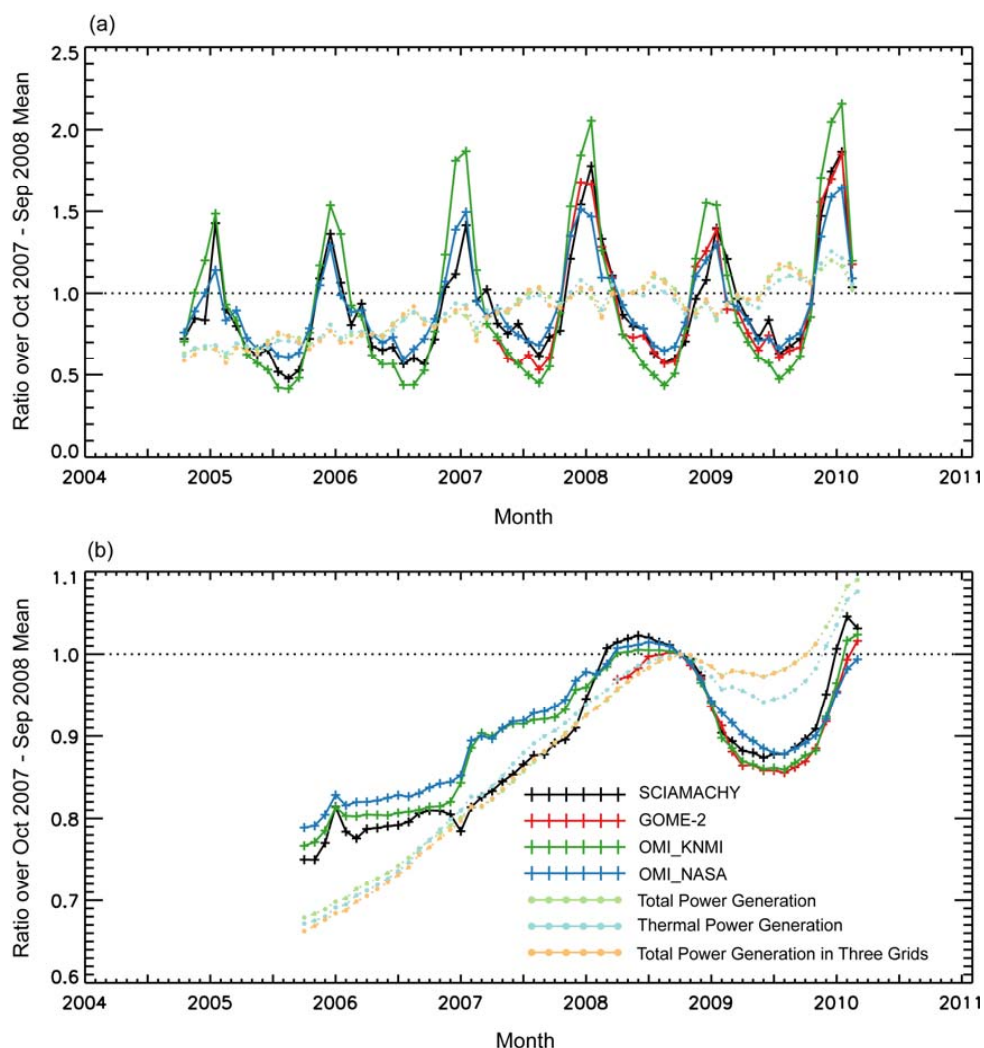
Fig. 3. (a) Monthly variations in regional mean VCDs of NO$_2$ and power generation in China relative to values averaged over the 12-month period of October 2007–September 2008. **(b)** Corresponding variations after a 12-month moving average: each point represents the mean over prior 12 months; and an increase (decrease) of value denotes a year-on-year increase (decrease) in VCDs of NO$_2$ or power generation.

on OMI measurements in place of the previous dataset from TOMS/GOME. The change results in a reduction of retrieved VCDs by about 5 % over East Asia (Boersma et al., 2011). It does not affect our analysis of the NO$_2$ trend prior to February 2009 and of the impacts of economic downturn and CNY for January 2009. The updated retrieval also reduces errors in the a priori vertical profile of NO$_2$ (Boersma et al., 2011). Errors in the a priori profile, however, are not important for quantifying emissions of NO$_x$ in the present analysis, since the (day-to-day varying) averaging kernel is employed in calculating modeled VCDs of NO$_2$ (see Sect. 4) to compare with retrieved values. Other improvements are minor and not important for the present analysis, including a cross-track stripe correction and a high-resolution surface height dataset (Boersma et al., 2011).

4 GEOS-Chem

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 is used in this study, with a major update by employing the non-local scheme (Holtslag and Boville, 1993) to simulate mixing in the PBL in place of the assumption of a fully-mixed PBL (This update has been standardized since Version 8.2.2). The model was run with the full Ox-CO-NO$_x$-VOC-HOx chemistry at a resolution of 2.5° long×2° lat 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). Anthropogenic emissions in Asia representative of 2006 taken from the INTEX-B mission are 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).

Model errors in simulating VCDs of NO_2 result from inaccuracies in the calculation of the lifetime of NO_x and the partitioning between NO_2 and NO (Martin et al., 2003; Martin et al., 2006). 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 of Bertram et al. (2009) and Brown et al. (2009) suggested that the actual rate of uptake may be lower than assumed by Evans and Jacob (2005), by up to an order of magnitude in some cases. The two studies differ, however, in the importance for the uptake of N_2O_5 of environmental parameters such as relative humidity and aerosol compositions (Bertram et al., 2009; Brown et al., 2009). Our preliminary sensitivity analysis for January 2009 using the nested version of the model (0.667° long×0.5° lat) 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 for Northern East China. Other sources of model errors relate to the concentration of OH affecting the $NO_2 + OH$ reaction, the amount of aerosols affecting the uptake of N_2O_5, the formation of organic nitrates, the concentration of ozone affecting the partitioning between NO_2 and NO, as well as likely errors in meteorological parameters. Overall, model errors are estimated to be about 30–40 %, based on previous studies (Martin et al., 2003; Wang et al., 2007; Lin et al., 2010b).

In calculating modeled VCDs of NO_2 for comparisons with the three KNMI retrievals, the averaging kernel (AK) was applied to simulated vertical distributions of NO_2 to eliminate the effect of errors in the a priori vertical profiles of NO_2 assumed in the retrieval process. For OMI_NASA, the associated AK is not available, thus retrieved VCDs are compared with modeled VCDs both with and without applying the AK taken from OMI_KNMI. Unless stated otherwise, AK's are accounted for in all of the modeled VCDs analyzed in the subsequent sections.

5 General variations in TPG and retrieved VCDs of NO_2 over October 2004–February 2010

This section analyzes general changes in thermal power generation (TPG) and retrieved VCDs of NO_2 over October 2004–February 2010, providing an overall picture of the seasonality and interannual variability of both data to facilitate the emission analysis in Sect. 6. Discussions for interannual variation are focused on the 12-month mean values, after implementing a 12-month moving average to both data series. A more detailed analysis is presented in Sect. 6 for January

months to quantify emissions of NO_x and effects of the economic downturn and the CNY.

5.1 Variations in TPG over October 2004–February 2010

As shown in Fig. 3b, Chinese TPG grew significantly and relatively linearly until the economic downturn that set in around late 2008. The 12-month 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. It then decreased by as much as 6 % under the influence of the downturn. Simultaneously, there was a reduction by about 3 % in the 12-month average total power generation. 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.

The TPG also varies month to month in response to seasonal variation in energy demand (Fig. 3a). It increases in summer and winter to meet the demand for air conditioning and heating, respectively (Zhang et al., 2007). The TPG minimum normally occurs in February in conjunction with the CNY which results in large-scale reductions in industrial activities and energy demand.

5.2 Variations in retrieved VCDs of NO_2 over October 2004–February 2010

5.2.1 Seasonal variability

The seasonal variation of VCDs of NO_2 differs significantly from that of power generation (Fig. 3a). VCDs of NO_2 reach a maximum 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). The minimum VCDs of NO_2 occur 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. The enhanced natural sources tend to increase VCDs of NO_2, while their effects are more than offset by the negative effect of the shorter lifetime. In general, VCDs of NO_2 decrease by a factor of 3–7 from winter to summer (Fig. 3a).

The seasonality of VCDs of NO_2 differs between the four retrieval products (Fig. 3a). OMI_KNMI suggests the strongest seasonality in NO_2. Values of OMI_NASA exceed OMI_KNMI in summer, while they are much smaller than OMI_KNMI in winter. The differences are in part because the overestimate of OMI_KNMI is larger in winter than in summer (Boersma et al., 2011). Furthermore, OMI_NASA

uses an annually-averaged a priori vertical profile for NO_2 for all seasons and does not account for increases in the PBL mixing from winter to summer (Lamsal et al., 2010). As the sensitivity of satellite instruments increases with the height of NO_2 in the troposphere, a given amount of radiance received by the instruments can be interpreted by the retrieval algorithm as either more NO_2 at a higher altitude or less NO_2 at a lower altitude. An annual average a priori vertical profile for NO_2 tends to result in an overestimate of tropospheric VCDs retrieved for summer with an underestimate for 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; Dirksen et al., 2011). This factor, however, cannot account for the larger seasonality of VCDs in OMI_KNMI as compared to OMI_NASA.

For the three KNMI products, the seasonality of NO_2 in SCIAMACHY and GOME-2 is smaller than OMI_KNMI (Fig. 3a). This may be partially because SCIAMACHY and GOME-2 pass over China in the mid morning while OMI passes in the early afternoon when the mechanisms for NO_x sink differ to some extent. For the two loss pathways of NO_x, the $NO_2 + OH$ reaction depends on solar radiation; thus it is more important in the afternoon than in the morning, and is subject to larger seasonal variation than the $N_2O_5 + H_2O$ pathway. Therefore the seasonality of the lifetime of NO_x in the afternoon is greater than that in the morning, as indicated by our model simulations.

5.2.2 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. 3b). From October 2004 to September 2008, 12-month 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 inferred from the findings of Zhang et al. (2007) for previous years. 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.

The effect of slowing industrial activities due to the economic downturn is evident in retrieved VCDs of NO_2 (Fig. 3b). The four retrieval products suggest that the 12-month mean VCD of NO_2 from mid 2008 to mid 2009 was 12–14 % lower than the mean value over mid 2007–mid 2008. The reductions were about twice as large as the reduction in TPG because of the timing of the economic downturn together with the differences in seasonality between TPG and VCDs. The downturn was most intense during the 2008-2009 winter (Fig. 1). In addition, the VCD of NO_2 reaches

a maximum in winter (Fig. 3a), thus its winter value contributes to the 12-month average more than values in other seasons. By comparison, the seasonality of TPG is much smaller (Fig. 3a), thus values in individual seasons contribute more evenly to the 12-month average. Therefore, a given relative increase of values in winter has a larger impact on the 12-month mean for VCDs of NO_2 and a smaller impact for TPG.

6 Quantifying year-on-year emission changes for January and effect of the economic downturn

In this section, a detailed derivation is provided to quantify year-on-year changes in emissions of NO_x for January over 2005–2010, identifying the causes of the emission anomaly in January 2009. In particular, the interannual variation of emissions for January is quantified in Sects. 6.1 and 6.2 by employing data for TPG, retrievals for VCDs of NO_2, and simulations of GEOS-Chem. In Sect. 6.3, the analysis is focused on emission reductions from January 2008 to January 2009, distinguishing the impact of the economic downturn for January 2009 from the effect of the CNY.

6.1 TPG-inferred changes in emissions of NO_x

The TPG in January increased between 2005 and 2010 except for the reduction from 2008 to 2009 (Fig. 4c). It increased by 51 % from 2005 to 2008, decreased by 18 % from 2008 to 2009, and increased again by 43 % from 2009 to 2010. The relatively low TPG in January 2009 was a result of the economic downturn compounded by the impact of the CNY.

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-yr period. The low TPG in January 2009 was driven mainly by reduced electricity demand from industry because of the economic downturn and the CNY, while electricity demand for residential use 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. This independent information is used to evaluate changes in emissions derived from satellite retrievals.
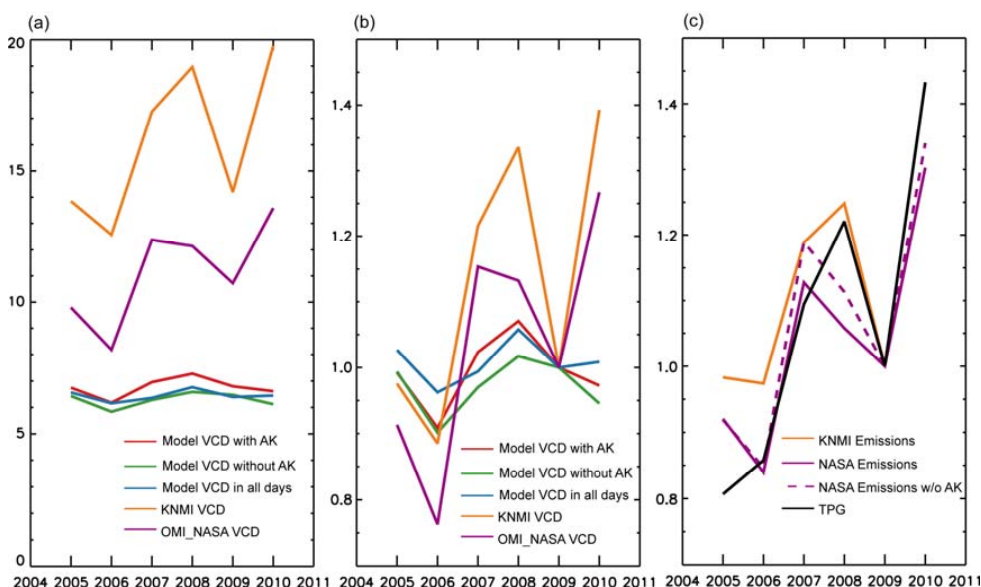
Fig. 4. **(a)** Changes in January mean VCDs of NO_2 (10^{15} molec cm^{-2}) from 2005 to 2010 with respect to OMI retrievals. **(b)** Corresponding changes in VCDs of NO_2 relative to values in January 2009. **(c)** Changes in emissions of NO_x derived from OMI relative to values in January 2009; also shown is changes in TPG (TWh day^{-1}) relative to January 2009. See Sect. 6.2 for the derivation of emissions of NO_x.

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

Emissions of NO_x can be derived from retrievals of NO_2 based on the mass balance approximation (Martin et al., 2003). Neglecting the effect of horizontal transport, the VCD of NO_2 is determined by emissions of NO_x, the atmospheric lifetime of NO_x, and the partitioning between NO_2 and nitric oxide (NO), i.e., $\Omega = E \cdot \alpha$, where Ω denotes the VCD of NO_2, E denotes emissions of NO_x, and α denotes the combined effect of the lifetime of NO_x and the partitioning between NO_2 and NO. Consequently, $E = \Omega/\alpha$. The reader is referred to Martin et al. (2003) and Lin et al. (2010b) for a detailed derivation.

The magnitude of α is determined by chemical and meteorological conditions. In this study, it is simulated by GEOS-Chem for each January: $\alpha = \Omega_m/E_m$. Here Ω_m indicates modeled VCD of NO_2 and E_m denotes emission inputs for the CTM. Consequently, emissions of NO_x can be derived from retrieved VCDs of NO_2: $E_r = \Omega_r/\alpha = \Omega_r/\Omega_m \cdot E_m$, where the subscript "r" denotes retrievals.

Of particular interest are variations in emissions of NO_x for January across the years. The ratio of emissions over a reference month is $R_e = E_r/E_{r0} = (\Omega_r/\Omega_{r0})/(\Omega_m/\Omega_{m0}) \cdot (E_m/E_{m0})$, where the subscript "0" denotes the reference month. In this study, the reference month is chosen to be January 2009 in order to better define the impact of the economic downturn. Furthermore, $E_m \approx E_{m0}$ since model inputs for anthropogenic emissions are the same for all months and other sources

of NO_x are negligible in January (Jaeglé et al., 2005; Lin and McElroy, 2010). Therefore one gets $R_e = R_r/R_m$, where $R_r = \Omega_r/\Omega_{r0}$ and $R_m = \Omega_m/\Omega_{m0}$ representing retrieved and modeled changes in VCDs of NO_2 relative to January 2009, respectively. To ensure consistency with R_r, R_m is calculated based on model results sampled from days with valid satellite data and applied with the averaging kernel. Analysis is then conducted for R_r, R_m and R_e in Sects. 6.2.1 and 6.2.2 for individual retrieval products.

As discussed in Sects. 3 and 4, errors exist in retrieval products and model simulations. These errors, together with inaccuracies in emission inputs for the CTM, result in large differences between retrieved and modeled VCDs of NO_2 for individual months (see Appendix A). Their impact on derived changes in emissions of NO_x is evaluated in Sects. 6.2.1 and 6.2.2 using the independent estimate based on data for TPG. In addition, the emission derivation here does not take into account the effects of changing emissions of NO_x, CO or NMVOC on the nitrogen chemistry under specified meteorological conditions. The effects, however, are not expected to be important in this study (see Appendix B). Furthermore, horizontal transport may affect the emission derivation for individual locations due to a relatively long lifetime of NO_x in January. However, the present study focuses mainly in Northern East China as a whole (with large area), and transport within the region does not significantly affect the total abundance of NO_x in the region. In this case, the effect of horizontal transport is expected to be small and can be neglected appropriately (Lin and McElroy, 2010).
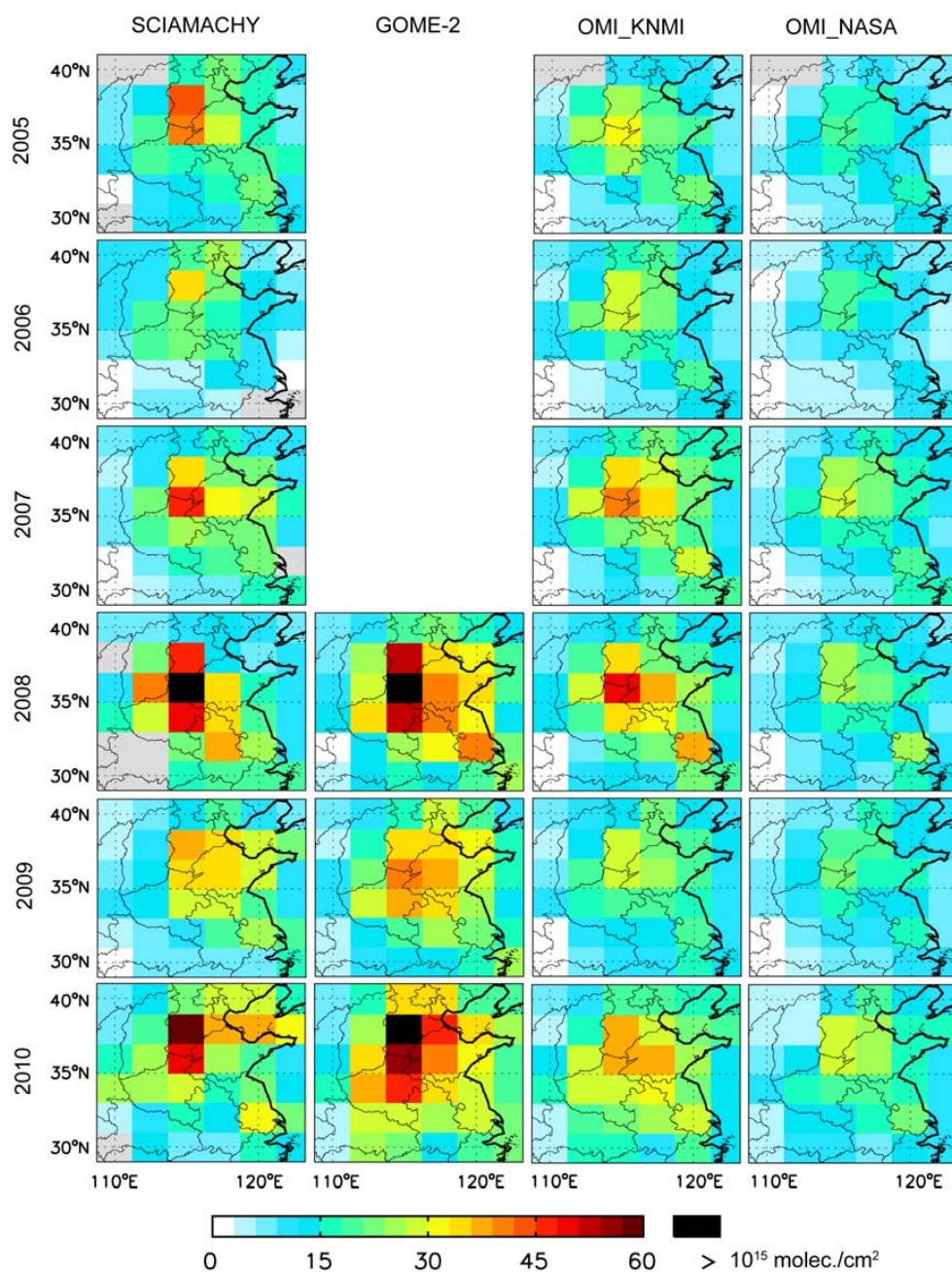
Fig. 5. Spatial distributions of retrieved VCDs of NO_2 in January over Northern East China. Data are presented at the model resolution of $2.5°$ long $\times 2°$ lat. Areas in grey color do not have valid retrievals.

6.2.1 Changes in emissions of NO_x derived from OMI_KNMI

Changes from 2005 to 2010 in January mean VCD of NO_2 for OMI_KNMI are generally consistent across Northern East China (Fig. 5). Averaged over the region, the VCD decreased by 9 % from 2005 to 2006, increased by 51 % from 2006 to 2008, and then decreased by 25 % from 2008 to 2009 (Fig. 4a, b). Values specified here are determined

by changes in emissions and many other factors (chemical/meteorological conditions, data availability, averaging kernel, etc.). Factors other than emissions are analyzed in detail in Appendix C.

Changes in January mean emissions of NO_x derived from OMI_KNMI are shown in Fig. 4c and Fig. 7. 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
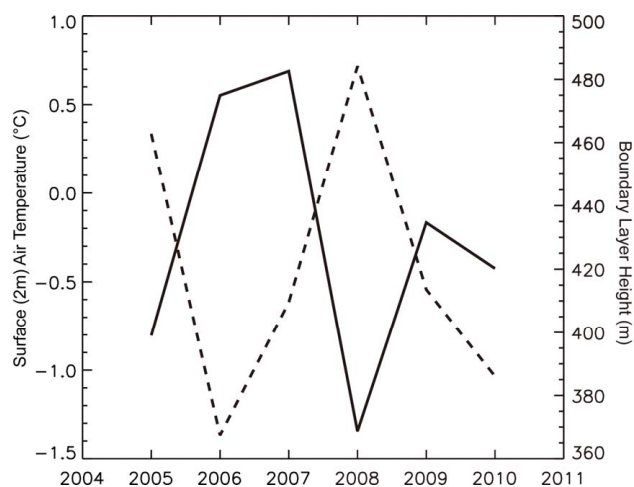
Fig. 6. Variations from 2005 to 2010 of January mean surface (2m) air temperature (solid line) and planetary boundary layer height (dashed line) over Northern East China.

to 2009 and then from 2009 to 2010, emissions of NO_x first decreased by 20 % and then rebounded by 43 %, close to the changes in TPG (Fig. 4c).

6.2.2 Changes in emissions of NO_x derived from other retrieval products

Changes in emissions of NO_x for January from 2005 to 2010 can be derived from the other three retrievals using the same approach as that for OMI_KNMI.

OMI_NASA 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 effects of chemistry and meteorology. As shown in Figs. 4a, b and 5, VCDs in OMI_NASA differ from OMI_KNMI throughout the years, as a result of differences both in retrieval methods and in criteria for valid data (see details in Sect. 3 and Table 2). Values of OMI_NASA 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 (Figs. 4c and 7). More importantly, they are not consistent with the changes indicated by the TPG data. In particular, there is a slight reduction from 2007 to 2009 (Fig. 4c; both purple lines) that is not seen in the TPG data.

Changes in GOME-2 and associated model VCDs are consistent with changes in OMI_KNMI and associated model values, although with different magnitudes (Fig. 8b). Derived based on GOME-2, emissions of NO_x in January decreased by 10 % from 2008 to 2009 and increased by 35 % from 2009 to 2010 (Figs. 7 and 8c). The sign of emission

changes is consistent with that for TPG while the magnitude is smaller, particularly for the reduction from 2008 to 2009.

VCDs retrieved from SCIAMACHY are influenced by variations in chemical and meteorological conditions across the years, as suggested by model results (sampled at the overpass time of the instrument from all days) showing that the minimum value in January 2006 differs from the maximum in January 2008 by about 18 % (Fig. 9b; blue line). This is consistent with the finding for OMI_KNMI in Appendix C. The SCIAMACHY product suffers greatly from the lack of valid data (Fig. 2), and the effect of incomplete sampling is much greater than the effects for OMI and GOME-2 that have better data coverage. This is evident by the large differences between model VCDs sampled from all days and those sampled from days with valid satellite data, particularly for January 2008 (Fig. 9a, b; blue lines versus green lines). As a result, emissions of NO_x derived from SCIAMACHY are also affected significantly by inadequate data coverage: they were nearly constant between 2007 and 2009 (Fig. 9c), inconsistent with the changes in TPG.

Overall, changes in emissions of NO_x derived from OMI_KNMI seem to best capture the changes inferred from 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.

6.3 Effect of the economic downturn versus CNY

Three approaches are proposed in the study to separate the impact of the downturn from that of CNY for January 2009. The impact of CNY can be estimated from the TPG data. CNY occurred in February in 2005, 2007, 2008, and 2010 (Table 1). For these years, CNY is assumed to be the sole cause of differences in daily TPG between January and February. Its effect is calculated then as the reduction of daily TPG from January to February, which amounts to about 8 % for 2005, 15 % for 2007, and 13 % for both 2008 and 2010. The effect of CNY is estimated accordingly as the average of these values, i.e., a reduction of 12 % with a standard deviation of 3 %.

The CNY holiday in 2009 took place over 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 the CNY affected January 2009 alone and that its impact on industrial activities was independent of the economic downturn, it is estimated that, of the total of 18 % reduction in daily TPG from January 2008 to January 2009, the CNY resulted in a reduction of 12 % while the downturn led to a reduction of 7 %. Assuming a smaller impact of CNY in January 2009, at 10 %, the effect of the downturn is estimated to be 9 %.

A similar analysis is applied to emissions of NO_x derived from OMI_KNMI, assuming that CNY affected daily emissions in the same way as it influenced daily TPG. For a
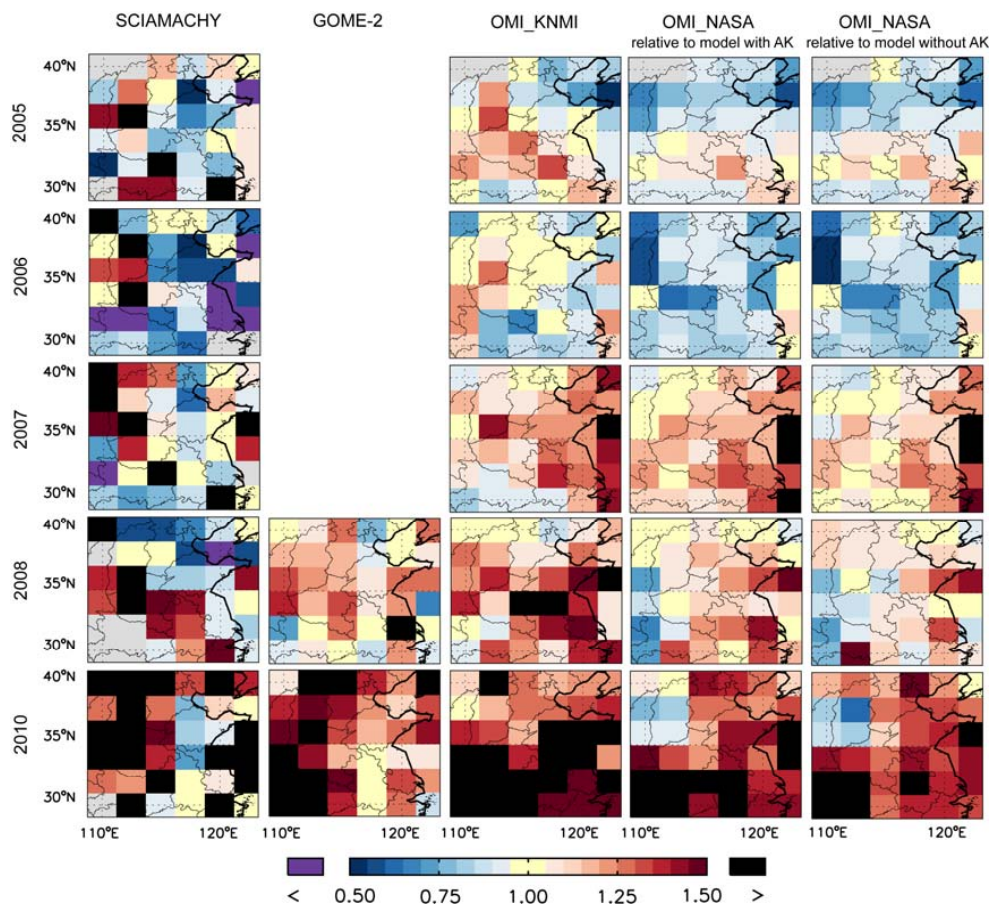
Fig. 7. Ratios of derived emissions of NO$_x$ in January of 2005–2010 over emissions in January 2009 in Northern East China. Plots for 2009 are skipped as the values are uniformly unity. Data are presented at the model resolution of 2.5° long×2° lat. Areas in grey color do not have valid retrievals. See Sect. 6.2 for the derivation of emissions of NO$_x$.
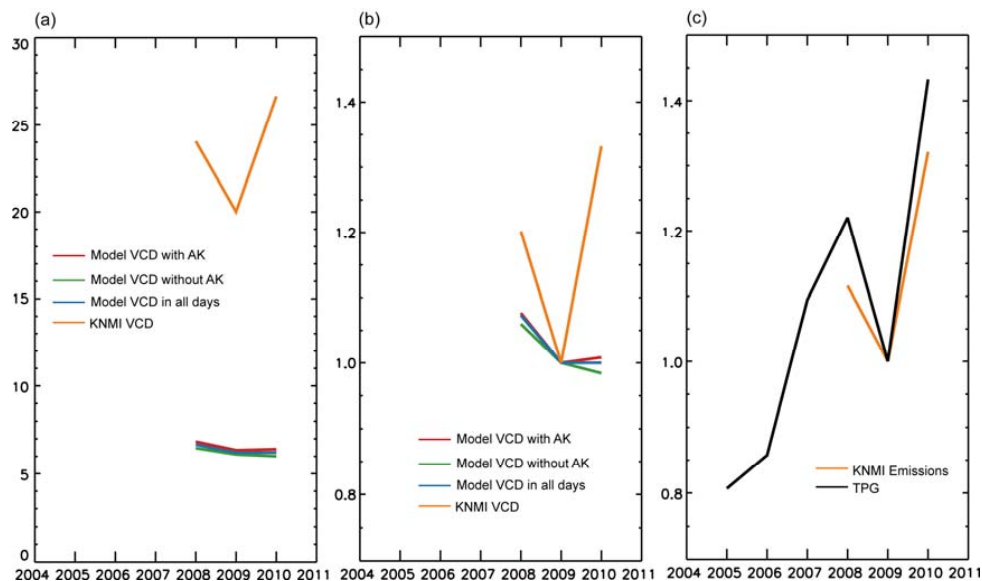

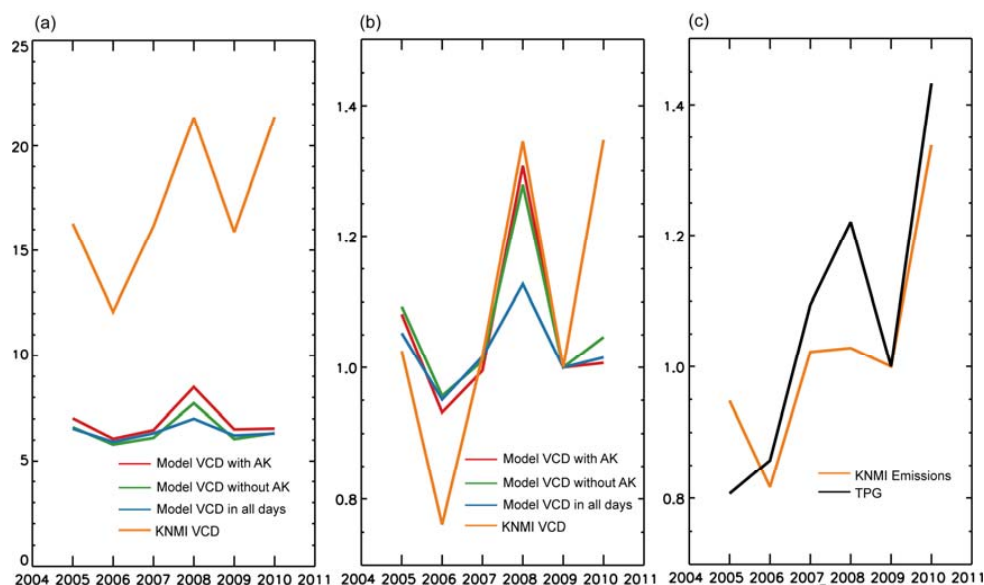**Fig. 8.** Similar to Fig. 4 but for GOME-2.

Fig. 9. Similar to Fig. 4 but for SCIAMACHY.

total emission reduction of 20 % from January 2008 to January 2009 derived from OMI_KNMI, the negative effect of the downturn was calculated as 9 % (11 %) corresponding to an assumption of 12 % (10 %) for the negative effect of the CNY.

A third approach is taken by calculating the emissions using data of OMI_KNMI for days of January that were not influenced significantly by CNY. In January 2009, retrieved VCDs declined rapidly after 20 January (Fig. 10a). A minimum value is 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 is captured by the model, including the timing of the local minimum in VCDs of NO_2 (Fig. 10). However, modeled VCDs experience 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 the CNY after 20 January 2009. The decline started earlier than the holiday partly because certain industries reduced production prior to the holiday (see the introduction).

In the third approach, the "monthly mean" VCD for January 2009 that would be independent of the CNY is approximated by the mean of VCDs during 1–20 January. This approximation is made both for OMI_KNMI and for model results in deriving the emissions. The calculation is unchanged for monthly mean VCDs in other months. Under this assumption, it is found that emissions of NO_x decreased by about 11 % from 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 of 10 % from the CNY.

Overall, our best estimate suggests that the CNY and the economic downturn were responsible for reductions in emissions of NO_x by 10 % and 9–11 %, respectively. The effects of CNY and downturn derived here are subject to errors in the year-on-year change of retrieved VCDs (R_r), errors in the change of modeled VCDs (R_m), errors associated with the data coverage, and errors in the TPG-based estimate of the effect of CNY for January 2009. At the moment, it is difficult to estimate these four errors accurately due to the lack of sufficient information. However, their individual impacts are not expected to exceed 1.5 %, 1.5 %, 1 %, and 2.5 %, respectively, for each of the estimated effects of the downturn and CNY for January 2009. Assuming these errors are independent, their overall impact (calculated by summation in quadrature) is not expected to exceed 3.4 %. Furthermore, since the three approaches adopted in this study lead to similar results, it seems that these individual errors are compensated significantly by each other. Thus a value of 3.4 % is most likely an upper limit for errors in the estimated effects of the downturn and CNY for January 2009.

7 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) are 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 a 12-month mean 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
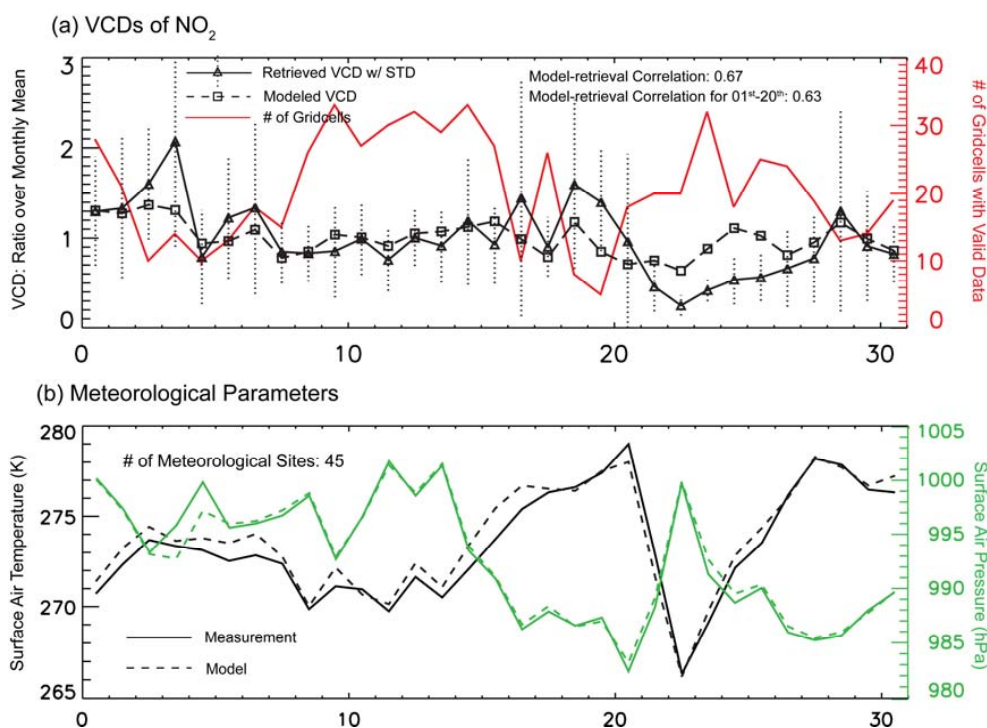
(a) VCDs of NO₂

(b) Meteorological Parameters

Fig. 10. Daily variations of regional mean VCDs of NO$_2$ and meteorological parameters in January 2009. **(a)** Variations of retrieved (OMI_KNMI) 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.

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, 12-month mean VCDs of NO$_2$ increased by 27–33 % (from October 2004–September 2005 to October 2007–September 2008) for SCIAMACHY and OMI (both by KNMI and by NASA), compared to the increase by 49 % in thermal power generation.

More detailed analysis is conducted to quantify changes in emissions of NO$_x$ for January over 2005–2010, in which season the effect of the downturn is most evident. The global CTM GEOS-Chem is used to identify the effect of chemistry and meteorology on interannual variations of VCDs of NO$_2$. Year-on-year changes in emissions of NO$_x$ for January derived from the OMI product produced by KNMI best capture changes in emissions inferred from the thermal power generation data. Estimated from the particular product, emissions decreased 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 eco-

nomic downturn and of decrease in industrial activity during the Chinese New Year. The two factors are separated by three approaches based on data for thermal power generation and the OMI data from 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 the celebration of Chinese New Year. Errors in the estimate are most likely less than 3.4 %.

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 VCDs 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.

Appendix A

Comparisons of retrieved and modeled VCDs of NO₂

Errors exist in both retrieved and modeled VCDs of NO_2, as discussed separately in Sects. 3 and 4. The overall effect of these errors can be estimated by comparing retrievals and model results for representative months.

For January 2006, model results are about 50 %, 49 % and 76 % of the retrievals from SCIAMACHY, OMI_KNMI and OMI_NASA, respectively (Figs. 4a and 9a). Reducing KNMI retrievals by a factor of 1.4 (corresponding to an assumption that the retrievals are overestimated by 40 %; see Sect. 3 for a detailed analysis of retrieval errors) results in model-retrieval ratios of about 70 % for both SCIAMACHY and OMO_KNMI. The remaining differences result from errors in retrievals and model simulations that have not been accounted for and inaccuracies in emission inputs of GEOS-Chem.

By January 2010, emissions of NO_x have grown significantly from January 2006 as a consequence of rapid industrial development and inadequate emission controls (Zhao et al., 2009; Lei et al., 2010; Lin et al., 2010a), despite the temporary reductions associated with the economic downturn. Meanwhile, inputs of emissions were held unchanged in the CTM simulation. As a result, model VCDs depart further from satellite retrievals: they amount to only about 31 %, 24 %, 33 % and 49 % of values derived from SCIA-MACHY, GOME-2, OMI_KNMI and OMI_NASA, respectively (Figs. 4a, 8a, and 9a). The model-retrieval ratios would be 43 %, 34 %, 46 % relative to the three KNMI products, respectively, if a factor of 1.4 is applied to scale down the retrieval values. The model-retrieval difference is much larger for GOME-2 than for the other two KNMI products. This appears to suggest that errors in the GOME-2 product may be greater than 40 %, but the exact cause has to be revealed by future research.

Appendix B

Effect of nonlinearity in nitrogen chemistry on derived emissions

The abundance of NO_2 in the troposphere is controlled by nonlinear photochemical processes affecting the lifetime of NO_x and the partitioning between NO_2 and NO. Here the sensitivity is evaluated for these two factors to changes in emissions of NO_x, CO and NMVOC under specified meteorological conditions.

Under given meteorological conditions, the lifetime of NO_x and the NO_2–NO partitioning may be affected by changes in emissions of NO_x through the nonlinear photochemistry. Stavrakou et al. (2008) found that an increase of

70–100 % in emissions of NO_x (from 1997 to 2006) results in an increase of 10 % in the lifetime of NO_x in winter over Northern East China due to reduced concentration of OH. This effect, however, is partly offset by a reduced NO_2:NO ratio (Martin et al., 2006). As a result, Martin et al. (2006) found that VCDs of NO_2 are highly proportional to emissions of NO_x for a wide range of emissions both in summer and in winter.

In this study, the effect of increasing emissions of NO_x on VCDs of NO_2 is evaluated further 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 from OMI_KNMI (see Sect. 6.2.1). 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 are affected insignificantly by the nonlinear photochemistry, consistent with the findings of Martin et al. (2006).

Changes in emissions of CO and NMVOC since 2005 may affect VCDs of NO_2 by affecting the abundances of ozone, OH, and other radicals. This effect was found by Lin et al. (2010b) to be insignificant for July 2008 employing a sensitivity test increasing emissions of CO and NMVOC by 50 %. In winter, the photochemical activity in the troposphere is much lower than that in summer. Therefore the effect of changing CO and NMVOC since 2005 is not expected to be important in deriving emissions of NO_x in winter.

Appendix C

Effects of non-emission factors on (OMI_KNMI associated) modeled year-on-year changes in VCDs of NO₂

A variety of factors other than emissions may affect the abundance of NO_2 in the troposphere observed by OMI. They are discussed here by employing results of GEOS-Chem simulations sampled at the overpass time of OMI.

Year-on-year changes in chemical and meteorological conditions have an influence on VCDs of NO_2, as simulated by the CTM with constant emissions of NO_x (and sampled from all days) (Fig. 4a, b; blue lines). The model VCD averaged over Northern East China varied by about 7 % year to year, with a minimum in January 2006 and a maximum in January 2008. The changes were contributed partly by variations in the planetary boundary layer height (PBLH). As shown in Fig. 6, the PBLH reached a minimum in January 2006 and a maximum in January 2008, with a difference of ∼30 %, affecting the vertical distributions of a variety of species important for the nitrogen chemistry. A sensitivity simulation increasing the PBLH by 30 % for January 2006 resulted in an increase of about 4 % in modeled VCDs of NO_2. Meanwhile, surface air temperature in January 2008 was about 2 °C lower

than January 2006, increasing the reaction rate of OH + NO$_2$ (a termolecular reaction) and decreasing the thermal decomposition of N$_2$O$_5$. The temperature difference thus tends to result in a lifetime of NO$_x$ in January 2008 shorter than that in January 2006, compensating for the effect of varying PBLH. Wind speed and precipitation are not found to be significant factors for the differences in modeled VCD of NO$_2$ between January 2006 and January 2008 (not shown). Other non-emission factors affecting the abundance of NO$_2$ include cyclone passages, clouds, aerosols, and stratospheric ozone (affecting the radiation into the troposphere). Further research is required for a more systematic analysis specifying the impacts of individual factors on VCDs of NO$_2$.

Satellite retrievals are not available for all locations for all days due to the existence of clouds, ice, snow and instrument limitations (e.g., row anomalies in OMI measurements) (Fig. 2). The effect of data availability can be evaluated by comparing simulated VCDs based on model results for all days with those sampled from days with valid satellite data (Fig. 4a, b; green lines versus blue lines). The reduced number of days for sampling results in a larger (smaller) difference in January mean VCD between 2006 (2008) and 2009.

Furthermore, satellite retrievals are subject to errors in the a priori vertical profiles of NO$_2$. The effect on retrieval-model comparisons for VCDs of NO$_2$ can be eliminated by applying the AK to model results. Year-on-year changes in January mean VCD (as simulated by the CTM) differ significantly between model results with and without applying the AK, particularly for 2007 and 2008 (Figs. 4a,b; green lines versus red lines). This indicates that the a priori vertical profile of NO$_2$ differs from the profile simulated by GEOS-Chem, and that the AK should be used (as in this study) for a proper model-retrieval comparison.

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