

Effect of emission changes in Southeast Asia on global hydroxyl and methane lifetime

By S. B. DALSRØREN^{1,4*}, I. S. A. ISAKSEN^{1,4}, L. LI^{1,2} and A. RICHTER³, ¹Department of Geosciences, University of Oslo, Norway; ²NILU, Norwegian Institute for Air Research, Kjeller, Norway; ³Institute of Environmental Physics, University of Bremen, Bremen, Germany; ⁴CICERO, Centre for International Climate and Environmental Research, Oslo, Norway

(Manuscript received 20 July 2008; in final form 18 May 2009)

ABSTRACT

We performed model studies on how anthropogenic emission changes in Southeast Asia (region between 60–150°E and 10°S–50°N) in the period 1980–2020 could contribute to changes in hydroxyl and methane lifetime on a global scale. From 1980 to 2000, we calculate small global OH and methane lifetime changes due to compensating effects by emission changes in Southeast Asia and emission changes in the rest of the world. There is no guarantee that this offset will persist in the future. Southeast Asia is going through rapid economic development and emission increases there may be a major driver for changes. The development of Asian emissions after year 2000 is under much debate and for this period we apply several emission scenarios. For most emission scenarios the simulated Southeast Asian induced changes in global hydroxyl and methane lifetime after year 2000 are moderate. However, an upper estimate assuming very high increases in NO_x emissions results in substantial increases of hydroxyl and corresponding reductions in global methane lifetime. Interestingly, for the high NO_x emission case our results fit very well with recent satellite observations on trends of NO₂ over central eastern China.

1. Introduction

Hydroxyl (OH) is the main oxidant in the troposphere (Levy, 1971; Thompson, 1992). This radical reacts with and removes several pollutants and greenhouse gases, one of them being methane (CH₄). The OH abundance itself is in turn highly dependent on some of these pollutants, in particular CH₄, NO_x and CO (Wang and Jacob, 1998; Lelieveld et al., 2002; Dalsøren and Isaksen, 2006). CO and CH₄ emission increase lead to an overall reduction in current global averaged OH levels. An increase in NO_x emissions increases global OH as long as it takes place outside highly polluted regions. Southeast Asia is a region going through rapid changes in emissions of the mentioned components. (Dalsøren and Isaksen, 2006) showed significant OH increases within this region from 1990 to 2001. The change in methane loss was also large compared to other regions. OH changes in low latitude regions could have significant effect on global methane levels since its loss with OH is favoured at high temperatures (Atkinson, 2007).

There is much debate how large the emission increase in southeast Asia and in particular China have been after 2000 [Akimoto et al., 2006; Streets et al., 2006; Ohara et al., 2007; Zhang et al., 2007; Lin et al., 2008; Streets et al., 2008; Zhang et al., 2008¹ (http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html)]. Until quite recently an accelerating increase in energy use is reported (Ohara et al., 2007; Raupach et al., 2007; Lin et al., 2008), but improved technology and regulations might have resulted in less steep emission increases (Ohara et al., 2007; Raupach et al., 2007; Lin et al., 2008). Observations of some atmospheric components show increases (Ding et al., 2008) and trends (Richter et al., 2005; Akimoto et al., 2006; Uno et al., 2007; Zhang et al., 2007; Van Der A et al., 2008; Zhang et al., 2008) that are larger than the trends in emission inventories. Model simulations using these inventories are for instance not able to reproduce the high increase in NO₂ observed from satellite (Akimoto et al., 2006; Uno et al., 2007; Zhang et al., 2007). For CO another key component for the OH budget the estimated increase in emissions (Ohara et al., 2007; Streets and Zhang et al., 2008;¹ Tanimoto et al., 2008) and observed concentrations (Tsutsumi et al., 2006; Tanimoto et al.,

*Corresponding author.

Forskningsparken, Gaustadalleen 21, 0349 Oslo, Norway.

e-mail: stigbd@geo.uio.no

DOI: 10.1111/j.1600-0889.2009.00429.x

¹Zhang, Q., Streets, D. G., He, K., et al., A new inventory of anthropogenic emissions in Asia for the year 2005/2006, Atmos. Chem. Phys. Discuss., manuscript in preparation, 2008.

2008) are smaller than for NO_2 and around 15%. Taken into account uncertainties and a possible positive bias in MOPITT satellite (Emmons et al., 2008) over time a positive trend is less clear. For NMVOCs the estimated size of the emission trend is quite similar to CO (Klimont et al., 2002; Ohara et al. 2007).

The further developments in the coming decades are also uncertain (Sheehan and Sun, 2006; OECD/IEA, 2007; Ohara et al., 2007). Whether the emission increase will continue with the same strength or not is highly dependent on large emitters like China and India (OECD/IEA, 2007).

2. Model and experiment

The calculations were performed with the OsloCTM2 model (Berglen et al., 2004; Isaksen et al., 2005; Dalsøren and Isaksen, 2006), run in T42 resolution with 40 vertical layers up to 10 hPa. A tropospheric version of the model was used with meteorology and stratospheric boundary conditions for the year 2000. The meteorological data is taken from the IFS forecast model (Gregory et al., 2000) at ECMWF. The stratospheric boundary conditions come from previous simulations with stratospheric versions of the model. The simulations in this study calculate the evolution of 59 hydrogen, oxygen, nitrogen, carbon and sulphur containing gaseous components. The calculations of changes in OH and methane lifetime are based on hourly output from the model. The model has been evaluated and shown to reasonably reproduce outflow and formation of secondary components in the vicinity of Asia in previous studies (Wild et al., 2003; Hsu et al., 2004). We have also in this study included a comparison of year 2000 measurements of CO, O_3 and HNO_3 at surface stations in or near Southeast Asia. However, we focus our comparison on the short-lived component NO_2 over the continent since reproducing this distribution is essential for the OH budget. Much of the validation of the model results is done by comparison with satellite retrieved NO_2 columns. Retrieval of tropospheric NO_2 columns from GOME measurements have been reported in a number of publications including (Leue et al., 2001; Martin et al., 2002; Richter and Burrows, 2002; Boersma et al., 2004). While the details of the different retrieval schemes vary in particular with respect to the assumptions made for the vertical distribution of NO_2 in the troposphere, the overall approach is similar in all studies. Data from the GOME and SCIAMACHY instruments have been applied in a large number of studies including several focusing on Asia (Richter et al., 2005; Van Der A et al. 2006, 2008; Ghude et al., 2008). The satellite retrieval used in this study has some inherent uncertainties important to be aware of. These may also in some cases explain parts of the discrepancy with the model results. Satellite remote sensing measurements of tropospheric NO_2 use absorption spectroscopy in the visible part of the spectrum. The stratospheric contribution is removed from the measurement by subtracting a measurement taken on the same day at the same latitude over the clean Pacific region.

The spectral retrieval and stratospheric removal have low uncertainties of a few percent over regions with a large signal as is found in China. In the last step of the retrieval, the vertical sensitivity of the measurements is accounted for using an a priori profile for NO_2 and aerosols as well as climatological values for surface albedo. As neither the aerosol distribution nor the NO_2 vertical profile is well known over China, this conversion introduces relatively large uncertainties of about 30%. A more detailed discussion of uncertainties in satellite measurements of tropospheric NO_2 can be found in (Boersma et al., 2004) and (Richter et al., 2005).

This study focused solely on anthropogenic emission changes. The natural (vegetation, soil and ocean) and biomass burning were taken from the RETRO database (Schultz et al., 2007, 2008). These emissions were kept fixed at their 2000 levels and seasonal cycles in all simulations. This was also the case for methane for which the surface model level was forced to zonal and monthly averaged observed concentrations for year 2000 available from the RETRO database (Schultz et al., 2007).

An overview of the applied anthropogenic emissions and model simulations can be found in Table 1. The anthropogenic emissions of CO and NO_x in Southeast Asia in the different simulations are shown in Fig. 1. For NMVOCs, NO_x and CO the RETRO emission inventory (Schultz et al., 2007) was applied in the time slice runs for 1980 and 2000. The sulphur emissions used are described in (Berglen et al., 2004) and based on GEIA for 1980 and AEROCOM for 2000. A sensitivity study was also performed for 2000 using the gridded REAS emission inventory (Ohara et al., 2007) for Southeast Asia (region between 60–150E and 10°S–50°N) for all components. After 2000 no global emission inventories exists nor a gridded dataset for the whole of Southeast Asia. For the 2006_REF study we scaled on a per country basis the year 2000 RETRO emissions in Southeast Asia with the reported 2000–2003 trends in the REAS study (Ohara et al., 2007) assuming that this trend continued until 2006. The Southeast Asian aircraft and ship emissions were also increased by 4%/year and 2%/year, which are conservative measures based on global average reported (ATA, 2007) or projected (Dalsøren et al., 2007) development. For the rest of the world we used the 2000 emissions levels for all emission sectors and all components. As discussed in the introduction the emission changes from 2000 to 2006 in Southeast Asia and in particular China are uncertain. The assumptions for the 2006_high_ NO_x simulation were made to obtain an upper estimate of OH changes. We assume a high increase in NO_x which for most conditions increase OH whereas we assume no increase (except for shipping and aircraft) in CO and NMVOCs that tend to reduce OH. In the 2006_high_ NO_x we assumed NO_x emissions changes in China equal to the increase in energy consumption for different emission sectors (China Statistic Administration, <http://www.stats.gov.cn/tjsj>). For the rest of Southeast Asia the 2006 NO_x emissions were

Table 1. Overview of model simulations and applied emission inventories

Simulation name	Basis year Southeast Asia	Emission inventory Southeast Asia	Emission China	Basis year rest of world	Emission inventory rest of world
1980	1980	RETRO 1980 (CO, NO _x , NMVOCs), GEIA (SO ₂)	RETRO 1980, GEIA	1980	RETRO 1980, GEIA
AS00W80	2000	RETRO 2000(CO, NO _x , NMVOCs), AEROCOM (SO ₂)	RETRO 2000, AEROCOM	1980	RETRO 1980, GEIA
AS80W00	1980	RETRO 1980, GEIA	RETRO 1980, GEIA	2000	RETRO 2000, AEROCOM
2000_RETRO	2000	RETRO 2000, AEROCOM	RETRO 2000, AEROCOM	2000	RETRO 2000, AEROCOM
2000_REAS	2000	REAS	REAS	2000	RETRO 2000, AEROCOM
2006_REF	2006	RETRO 2000 and AEROCOM scaled to REAS 2006	RETRO 2000, and AEROCOM scaled to REAS 2006	2000	RETRO 2000, AEROCOM
2006_High_NO _x	2006	RETRO 2000 and AEROCOM Scaled to REAS 2006 for NO _x and SO ₂ , RETRO 2000 for CO, NMVOCs (no change)	RETRO 2000 scaled increase energy consumption for NO _x , AEROCOM scaled to REAS 2006 for SO ₂ . RETRO 2000 for CO, NMVOCs (no change)	2000	RETRO 2000, AEROCOM
2020_REF	2020	RETRO 2000 and AEROCOM scaled to REAS reference scenario for 2020	RETRO 2000 and AEROCOM scaled to REAS reference scenario	2000	RETRO 2000, AEROCOM
2020_PSC	2020	RETRO 2000 and AEROCOM scaled to REAS reference scenario for 2020	RETRO 2000 and AEROCOM scaled to REAS PSC scenario for 2020	2000	RETRO 2000, AEROCOM
2020_PFC	2020	RETRO 2000 and AEROCOM scaled to REAS reference scenario for 2020	RETRO 2000 and AEROCOM scaled to REAS PFC scenario for 2020	2000	RETRO 2000, AEROCOM
2020_High_NO _x	2020	Same yearly rate of increase as in 2006_High_NO _x	Same yearly rate of increase as in 2006_High_NO _x	2000	RETRO 2000, AEROCOM

similar to those in the 2006_REF study. For sulphur emissions we used the REAS trend (2006_REF_study) for all countries. For CO and NMVOCs we assumed no changes. This is a low estimate based on the small emissions and concentrations changes of CO discussed in the introduction. Assuming that CO and NMVOCs have the same development of emission signature is also a simplification. Further there might be different changes for different sectors and geographical regions that we do not account for. However the OH response to CO concentration changes is quite linear and the effect of NMVOC emission changes moderate (Lelieveld et al., 2004). Recent regional changes in ship and aircraft emissions would be of importance

since these sources have high emission factors for NO_x compared to other chemical constituents. In the 2006_high_NO_x case we assumed a 10%/yr increase in emissions from ships for all components in accordance with Asian harbour statistics (<http://www.szport.net:8080/eng/Statistics/index.htm>) and statistics on seaborne trade (UNCTAD, 2006). The aircraft emissions in the region were augmented by 7%/yr (IATA, 2005).

We performed several simulations for 2020. Since we wanted to focus on the contribution from Southeast Asia the CO, NO_x and NMVOC emissions for the rest of the world were kept at 2000 level. The methane concentrations were also fixed to year 2000 values. Three simulations were performed scaling

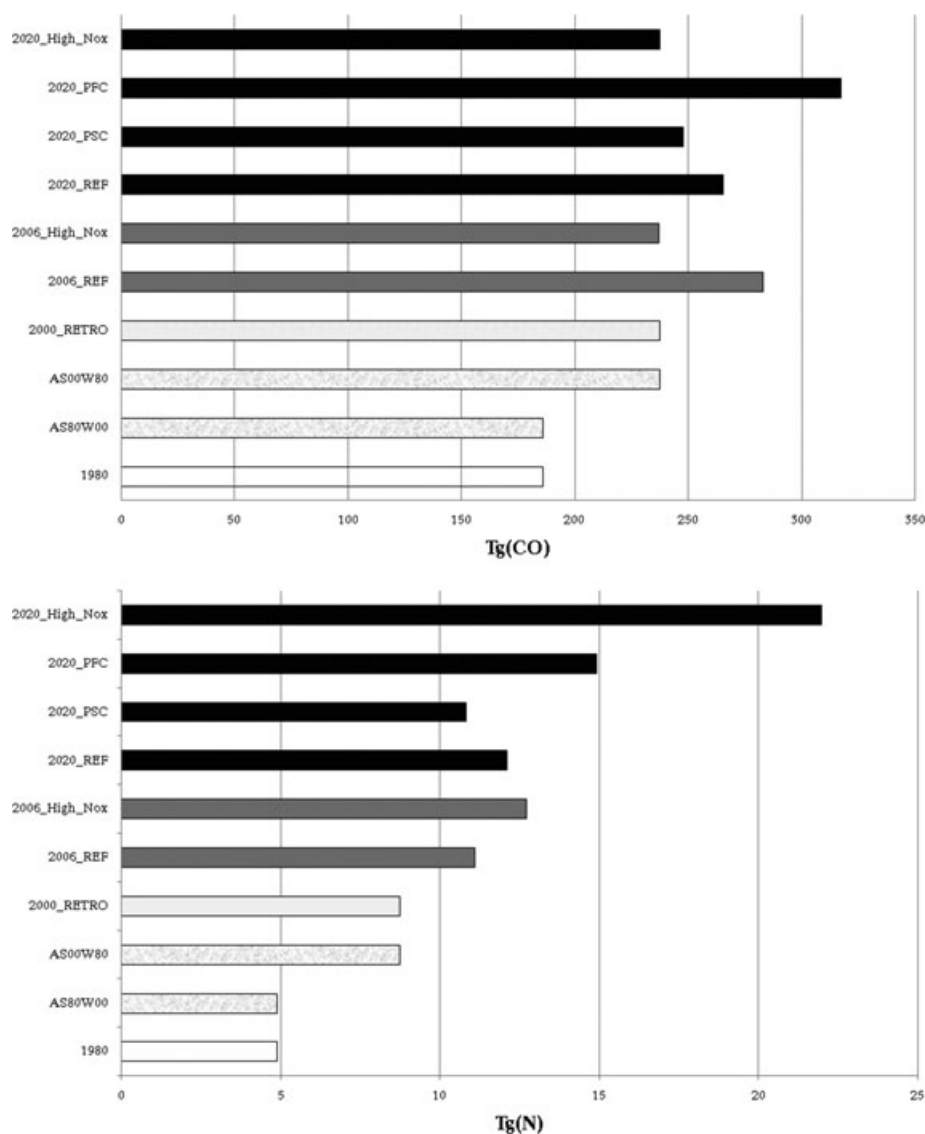


Fig. 1. Total anthropogenic CO (top) and NO_x (below) emissions in Southeast Asia for the simulations used in the calculations of OH and methane lifetime.

the 2000 Southeast Asian emissions with the 2000–2020 REAS trends (Ohara et al., 2007). The REAS 2020 projections consist of three scenarios for China where the development is regarded as most uncertain and one scenario (2020_REF) for the rest of Southeast Asia. In the 2020_PFC (Policy Failed Case) the emissions in China in 2020 are high due to continuation of the current energy structure and limited development of new technologies. The 2020_REF (Reference) is characterized by more efficient energy conservation, changes to cleaner technology and moderate switches to new energy and control technologies. In the 2020_PSC (Policy Succeed Case) scenario the emissions in China are low due to introduction of new energy and control technologies and initiatives on environmental policies. We also did a 2020_high_NO_x study where we assumed the same yearly

rate of emission increase for all components as from 2000 to 2006 in the 2006_high_NO_x study.

3. Results

The OH response to perturbations in NO_x emissions is non-linear dependent on the ambient NO_x concentration (Lelieveld et al., 2004). It is therefore important that the model realistically represents the NO_x distribution over our focus region. To fulfil this in polluted regions one needs both an accurate chemistry scheme in the model and also very importantly an emission inventory of high quality as input. For year 2000, we performed a comparison to GOME satellite retrieved tropospheric NO₂ columns over Southeast Asia (Richter et al., 2005) (Figs. 2

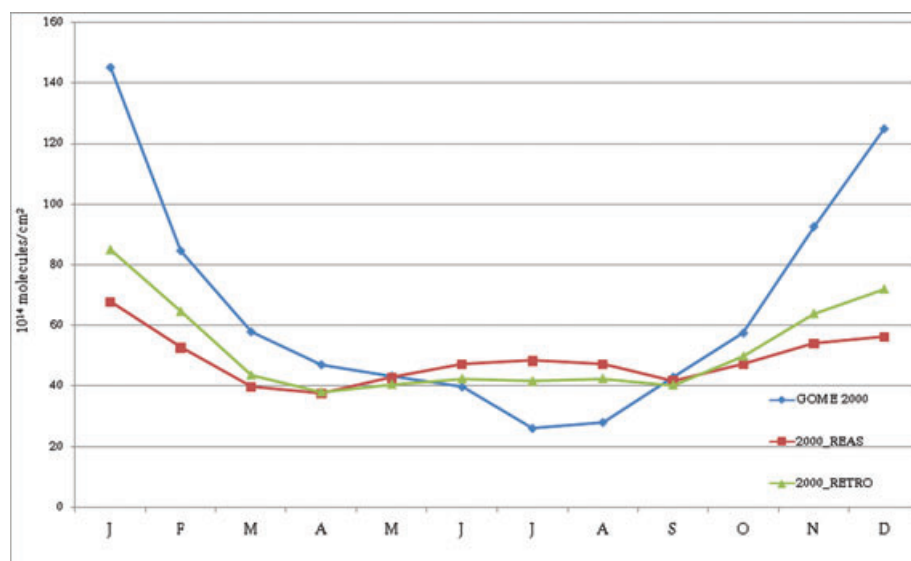


Fig. 2. Comparison year 2000 monthly average 10:30 localtime NO₂ column central eastern China (30–40° N, 110–123° E).

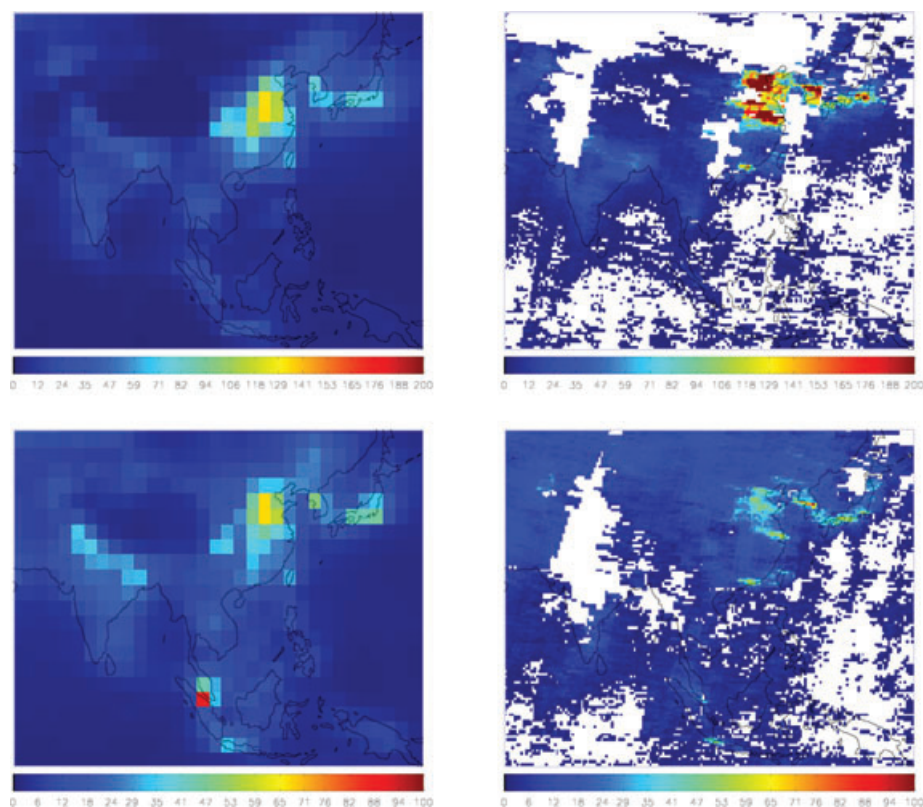


Fig. 3. Comparison of year 2000 monthly average (January upper row, July lower row) 10:30 localtime NO₂ columns over Southeast Asia. 2000_RETRO simulation (left-hand column) and the GOME 2000 (right-hand column).

and 3). In Fig. 2, the comparison over central eastern China is made using both the RETRO emission (2000_RETRO, Table 1) and the REAS inventory (2000_REAS) in Southeast Asia. For both inventories the fit to the satellite data is satisfactory. The correlation coefficient (r) is 0.82 the 2000_REAS simulation

and 0.96 for 2000_RETRO, while the normalized mean biases (nmb) are -26.1 and -20.9% , respectively. Our model results fail to fully reproduce the strength of the seasonal cycle in the NO₂ column but the Bremen retrieval used for this comparison shows somewhat stronger seasonality than the other GOME

retrievals (van Noije et al., 2006). As discussed by (Uno et al., 2007; Wang et al., 2007; Zhang et al., 2007) it might also be that current inventories for Southeast Asia not fully resolve the seasonality of emissions. In general, the correspondence seems to be better than for the comparison performed with several models in ACCENT (van Noije et al., 2006). The simulation using the RETRO emissions, 2000_RETRO ($r = 0.96$) capture the seasonality better than the study 2000_REAS ($r = 0.82$) using the REAS inventory. This is very likely because the RETRO inventory includes seasonal variation in the anthropogenic emissions. In our model the lacking seasonality in the REAS inventory result a mid-summer maximum in NO_2 column that seems unrealistic as this is not found by the satellite. In a recent study a model with the REAS inventory (strongly) underestimated the NO_2 columns over the same region (Uno et al., 2007). The spatial distributions for 2000_RETRO and GOME over Southeast Asia are compared in Fig. 3. In winter the model reproduces the structure and magnitudes retrieved from the GOME instrument in the most of the region. An exception is the most polluted regions where the model underestimates the satellite derived NO_2 columns. In summer the modelled NO_2 columns are slightly high over northeastern China and a little too low over the Korean peninsula. The large difference over Sumatra (red hotspot in figure of model column) suggests that the biomass burning emissions there might be overestimated. Though smearing out the signal due to coarse resolution the models shows realistic signals for cities like Singapore, Jakarta, Taipei and heavy populated and industrialised regions of Korea, Japan and China. The signal near Gungzhou and Hong Kong is weaker or more diffuse than the GOME signal.

The simulation of long-lived and secondary formed species was tested in TRACE-P (Wild et al., 2003; Hsu et al., 2004). To study this further we included a comparison to surface observations of CO , O_3 and HNO_3 from the WDCGG (<http://gaw.kishou.go.jp/wdcgg>) and EANET (<http://www.eanet.cc>) (Figs. 4–7). The role of CO on OH was referred to in the introduction. Key components for the OH budget for Asian outflow is also discussed by (Mao et al., 2008). Photolysis of

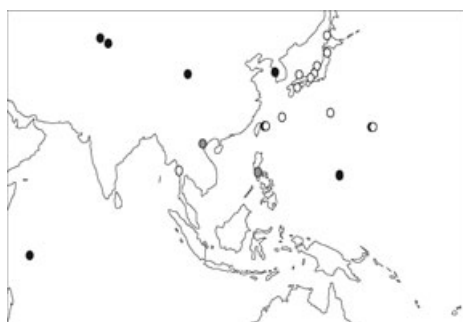


Fig. 4. Surface stations used for comparisons in Figs. 5–7. Black circles are CO stations, white are ozone stations and grey are HNO_3 stations.

O_3 and subsequent reaction with excited oxygen is an important source of OH . In order to get ozone correct and appropriate NO_y and HO_x cycling HNO_3 should also be simulated reasonably. We excluded urban stations from the comparison since it cannot be expected that the coarse resolution of the model resolve non-linear NO_x and ozone chemistry on scales of a few kilometres. The correspondence between the model and CO observations is remarkably good at oceanic stations influenced by Asian outflow with $r > 0.94$ and nmb of minus a few percent. The model for instance reproduces the high values at the Seychelles in the beginning of the year when the winter monsoon brings pollutant air-masses from the Asian continent. The CO concentrations at the polluted site Tae-ahn peninsula and continental stations at higher altitudes (Mount Waliguan and Kazakhstan) are slightly underestimated. Overall the mean correlation for the CO comparisons is 0.77 and the mean normalized mean bias -16.4% . There is a tendency that the modelled seasonal amplitude of CO and ozone is weaker than for the measurements at many stations. This could be another indication that even if we have seasonality in the preferred emission inventory it is not profound enough. The model also performs well for ozone over background oceanic stations. All observations over land areas except for one are over Japan and there the magnitude of simulated ozone is reasonable in most cases though the model is not capable of reproducing the seasonality at all stations (mean $r = 0.57$). It might be due to missing compliance between the coarse resolution of the model and different stations more or less situated in background, rural and urban influenced air masses. The nmb varies in sign for the different stations and the mean nmb of $+4.9\%$ only indicates that there is no systematic over- or underestimation by the model. The number of HNO_3 observations at non-urban sites available for year 2000 from the EANET network are sparse and far too few to be conclusive. The two comparisons we include in this study indicate resemblance in seasonality (mean $r = 0.58$) and magnitude but perhaps a slight underestimate by the model (mean nmb $= -29\%$).

We find that anthropogenic emission changes throughout the world resulted in small OH and methane lifetime changes from 1980 to 2000 (Fig. 8). This is in agreement with the findings in the RETRO project using the same emissions inventory but also including year-to-year changes in meteorology, biomass burning and stratospheric ozone (Szopa et al., 2007). The magnitude of the global changes (2000_RETRO versus 1980_RETRO) is also similar to the findings from (Dalsøren and Isaksen, 2006) for the period 1990–2001 using a different emission inventory. Inversions based on methylchloroform (Krol and Lelieveld, 2003; Bousquet et al., 2005; Prinn et al., 2005) and ^{14}CO (Manning et al., 2005) observations also found rather small trends in OH over the same period. In Table 2 the calculated OH changes for some recent studies are compared. Table 2 only indicates the range of changes and care should be taken in comparing the numbers directly since all studies show large interannual

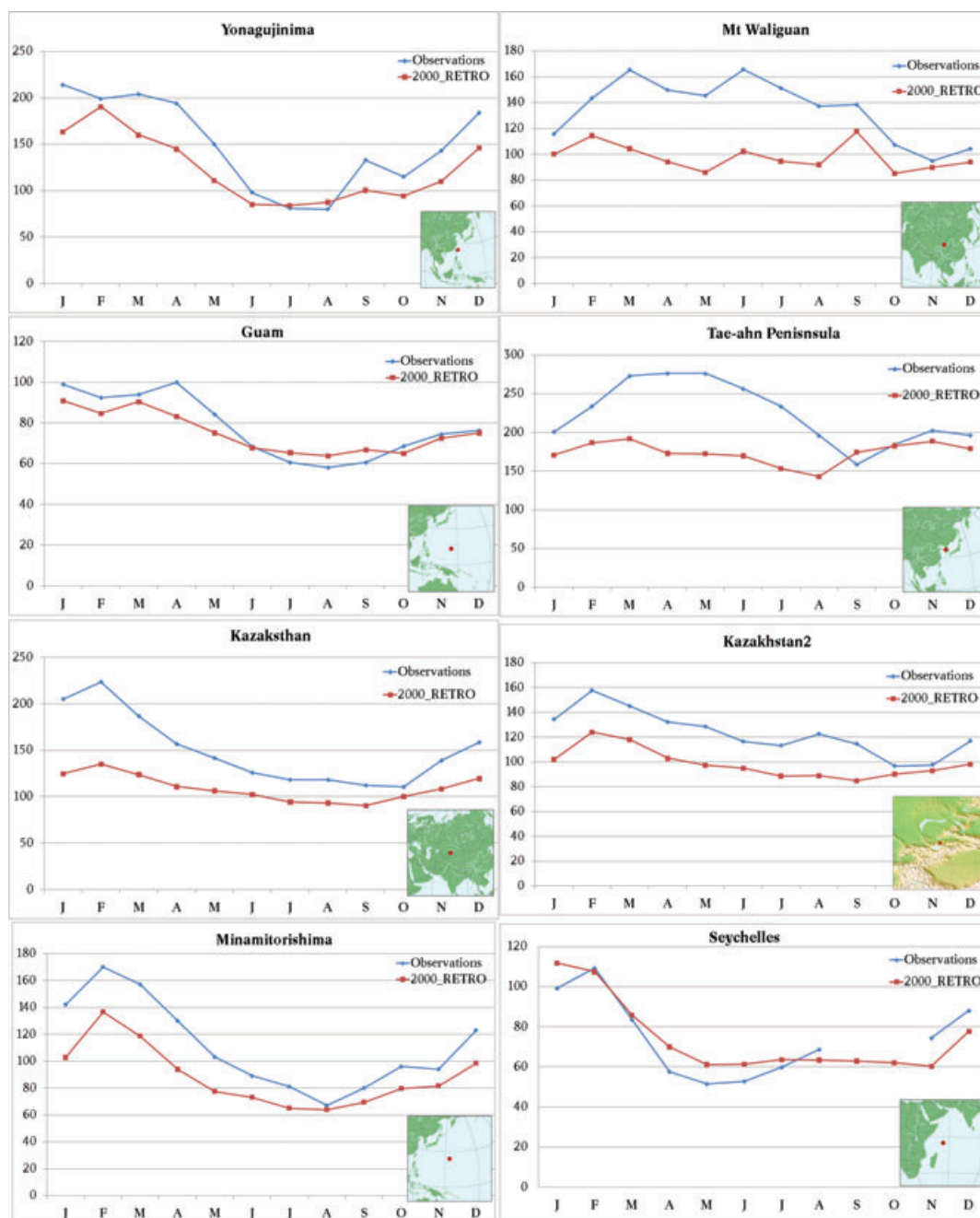


Fig. 5. Comparison between model results (2000_RETRO) and observations for year 2000 of CO for stations in or near Southeast Asia. Unit is ppbv.

variations and there are uncertainties and differences in calculation methods applied either for models (Lawrence et al., 2001), methylchloroform (Krol and Lelieveld, 2003) and ^{14}CO (Krol et al., 2008). The anthropogenic emission changes over Southeast Asia (AS00W80 run) resulted in a significant increase in global OH concentration and a reduction in methane lifetime. The overall effect on these indicators from anthropogenic emission changes in the rest of the world went in the opposite direction (simulation AS80W00). The reasons for different de-

velopment in various regions are discussed further in (Dalsøren and Isaksen, 2006) where the change in CO/NO_x emission ratio was found decisive for evolution of OH concentrations.

For the 2000–2006 period the two projections we use for Southeast Asian emissions give responses in opposite directions. In the standard 2006_REF scaling a slight negative OH change of -0.23% is found followed by a small increase in methane lifetime (Fig. 8). The more radical 2006_High_ NO_x scaling leads to changes of about $\pm 1\%$ (Fig. 8). The reason for the diverse

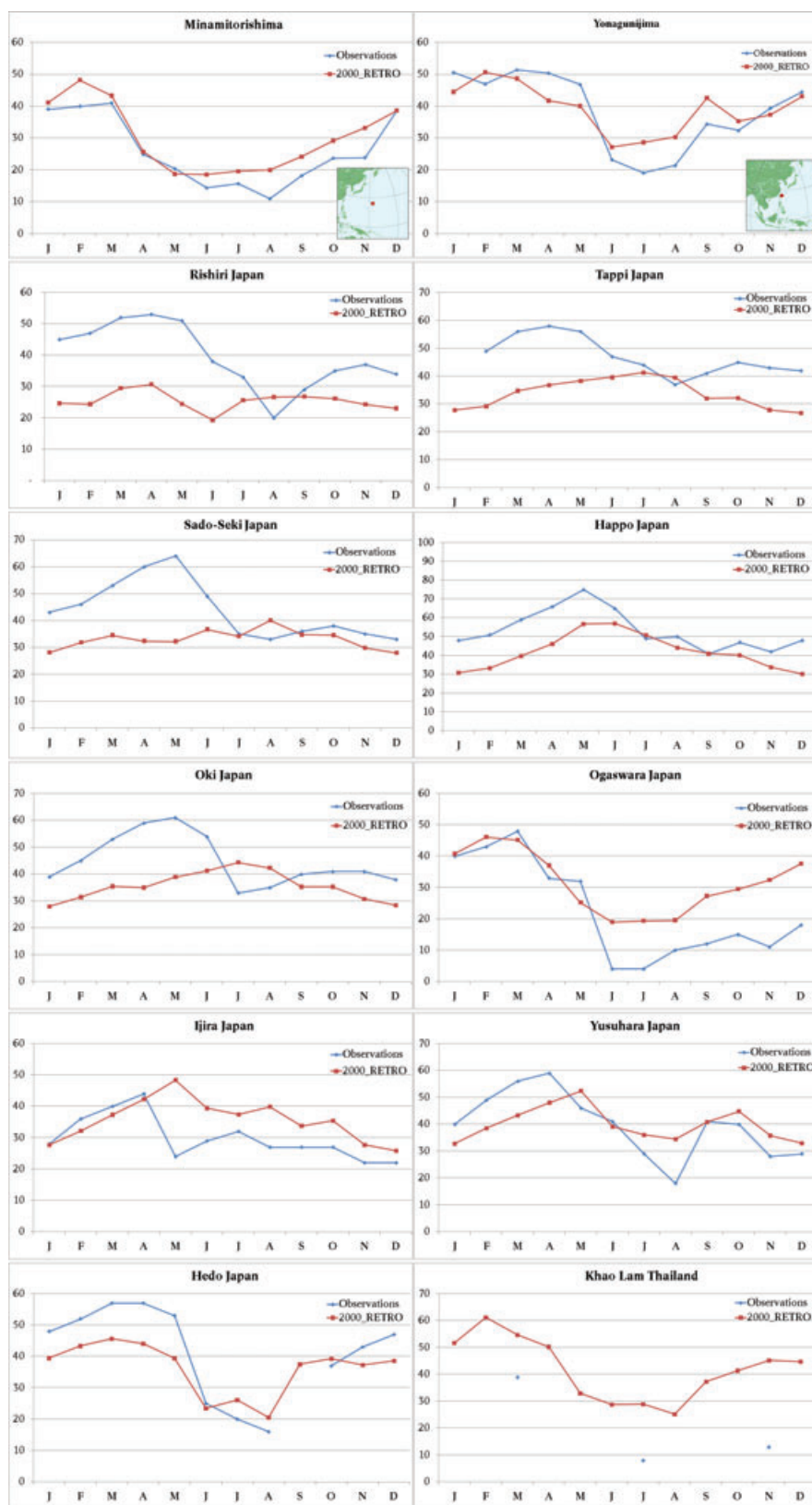


Fig. 6. Comparison between model results (2000_RETRO) and observations for year 2000 of ozone for stations in or near Southeast Asia. Unit is ppbv.

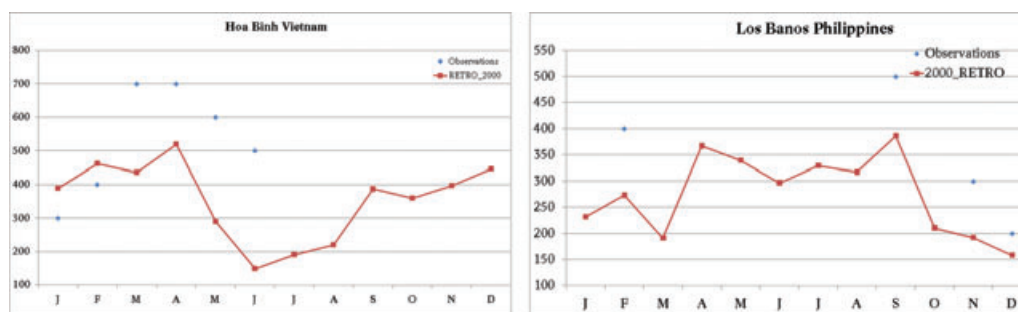


Fig. 7. Comparison between model results (2000_RETRO) and observations for year 2000 of HNO_3 for stations in or near Southeast Asia. Unit is pptv.

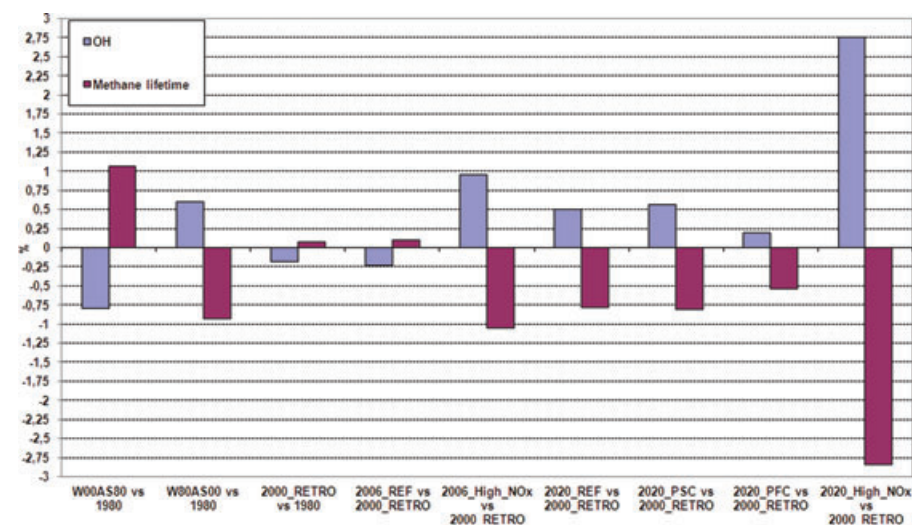


Fig. 8. Relative changes in global averaged OH and methane lifetime for the different simulations.

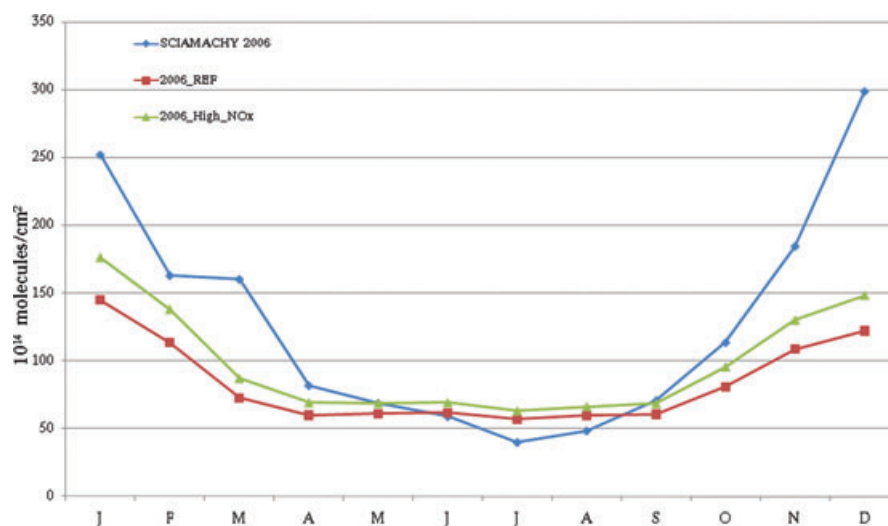
responses is related to the different assumptions on development of CO and NMVOC versus NO_x emissions, and in particular the CO/NO_x emission ratio. Even if the changes might be considered small they are much larger than the global changes over the longer time period 1980–2000. Several papers have been published about trends in satellite columns over central eastern China throughout the last decade (Richter et al., 2005; Akimoto et al., 2006; Uno et al., 2007; Zhang et al., 2007; Van Der A et al., 2008). The findings indicate dramatic increases in NO_2 . In Figs. 9 and 10 we compare the model derived NO_2 columns in the 2006_REF and the 2006_High_ NO_x with the estimated columns from SCIAMACHY data for 2006. The increase in the NO_2 column density from 2000 to 2006 over central eastern China is 95.3%. In 2006_REF the increase is 60.9% ($r = 0.91$, $\text{nmb} = -34.8\%$) whereas in the 2006_High_ NO_x the increase is 89.3% ($r = 0.92$, $\text{nmb} = -23.3\%$). It is interesting that the setup 2006_High_ NO_x best reproduces the trend (Fig. 9) and especially the large increase over China in wintertime (Fig. 10). This might be another indication that emission inventories underestimate recent NO_x emission increases in parts of Southeast Asia. This has been suggested in recent literature (referred to above and in the introduction). Though the

2006_High_ NO_x projection has simplifications also for other components (assuming no changes in CO and NMVOCs emissions) it should be realistic as an upper/lower estimate of the development of OH and methane lifetime due to Southeast Asian emissions.

For 2020 all applied emission scenarios lead to increases in OH and reductions in methane lifetime (Fig. 8). The high NO_x case departs from the others with its high increase in OH of 2.75% and a 2.85% decrease in methane lifetime. This change is similar to the global effect of all international ship emissions (Endresen et al., 2003; Dalsøren et al., 2007; Dalsøren et al., 2008), a source characterized by its large NO_x emissions, small CO emissions, and thereby large effect on OH and methane lifetime. The OH increase per year is slightly lower than for the high NO_x case from 2000 to 2006. This is explained by the fact that we see stronger non-linear chemical effects (OH reductions in highly NO_x polluted regions during wintertime) in the 2020 case. Since we tend to underestimate the year 2000 winter NO_2 columns over central eastern China derived from GOME we might slightly overestimate the OH increase after 2000. The OH increase in Southeast Asia is 4.7% in 2020_High_ NO_x , such a large change would have significant effect on the oxidation of

Table 2. Calculated OH changes for the last decades from recent studies

Study	Method	Period	Global OH change
Manning et al. (2005)	^{14}CO	1989–2003	No significant trend Southern Hemisphere (SH)
Bousquet et al. (2005)	Methylchloroform	1980–2000	$-0.7 \pm 0.2\%/ \text{year}$
Krol and Lelieveld (2003)	Methylchloroform	1978–2000	+12% 1978–1990 Slightly larger decline 1991–2000
Prinn et al. (2005)	Methylchloroform	1978–2003 2003–04 relative to 1979–1981	$0.2\% / \text{yr}$ (-0.4 to $+0.8$) -0.18% (-9 to $+13$)
Szopa et al. (2007)	Models	1960–2000	Small multidecadal changes
Dentener et al. (2003)	Model	1979–1993	$+0.24 \pm 0.06\% / \text{yr}$
		1979–1993 ^a	$-0.12 \pm 0.01\% / \text{yr}$
Dalsøren and Isaksen (2006)	Model	1990–2001 ^a	$+0.08\% / \text{yr}$, no significant trend SH ($+0.01\% / \text{yr}$), $+0.15\%$ NH
This study	Model	2000 relative to 1980 ^b	-0.18%

^aEffect of anthropogenic emission changes only^bEffect of anthropogenic emission changes only and no year to year variation biomass burning.Fig. 9. Comparison year 2006 monthly average 10:30 localtime NO_2 column central eastern China ($30\text{--}40^\circ\text{N}$, $110\text{--}123^\circ\text{E}$).

regional pollutants with short and intermediate lifetimes. Of the three scenarios adopted from the REAS inventory it is interestingly 2020_PSC (Policy Succeeded Case for China) that has the largest impact on OH and methane. Though the changes for the three REAS scenarios from 2000 to 2020 are less than $\pm 1\%$ this is similar to the effect of emission changes in the region from 1980 to 2000 and quite much larger than the impact (less than $\pm 0.2\%$) due to global emission changes from 1980 to 2000 (Fig. 8).

Even if the focus region is situated at low latitudes the absolute magnitude of the methane changes are not much larger

than for the OH perturbations. As mentioned in the introduction this is often the case due to the temperature dependency of the reaction between OH and methane (Dalsøren and Isaksen, 2006; Atkinson, 2007). The reason why we do not see such a strong dependency is likely that the largest emission changes within the region are taking place in China close to mid latitudes and also that during the summer monsoon air masses are effectively transported out of the boundary layer over much of the Asian continent. This moderates OH changes in the summer boundary layer where you have the highest temperatures.

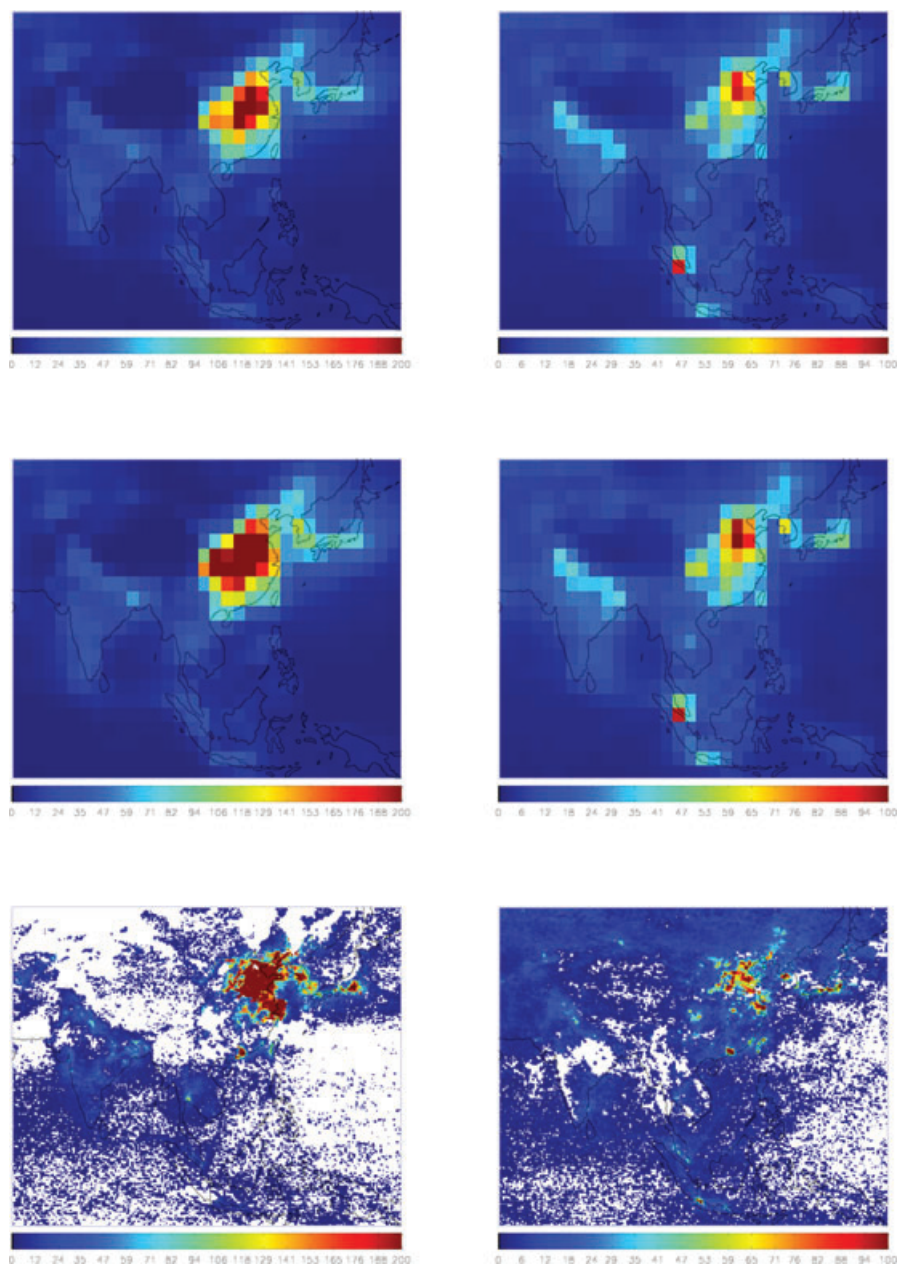


Fig. 10. Comparison of year 2006 monthly average (January left-hand column, July right-hand column) 10:30 localtime NO_2 columns over Southeast Asia. 2006_Ref simulation (upper row), 2006_High_ NO_x (mid row) and SCIAMACHY (lower row).

4. Discussion

Despite small changes in global average values, regional changes in OH may be significant as noted by (Dalsøren and Isaksen, 2006). Compensation due to different regional development or concurrent emission changes in key components might have occurred during the last decades (Wang and Jacob, 1998; Lelieveld et al., 2002). There is no guarantee that this will occur in the future. One region that could be a driver also for global changes due to recent and possibly future abrupt emission changes is

Southeast Asia. After year 2000 this study only focused on emission changes in Southeast Asia and their potential contribution to changes in future OH and methane. It should be noted that the contribution to some extent could be influenced by emission changes in other regions. Pollution from other source regions could be transported to Southeast Asia and perturb the chemistry within the region. Transport of PAN to Southeast Asia and decomposition to NO_2 near the surface would be one example. This could affect non-linear chemical processes and thereby change the contribution and effect of Southeast Asian emissions.

5. Conclusions

In agreement with former studies we find that global anthropogenic emission changes have resulted in small changes in OH and in methane lifetime from 1980 to 2000. The model studies indicate cancellation between substantial effects from emission changes in Southeast Asia and the overall impact of emission changes in the rest of the world. Comparison for 2000 with satellite measurements show that the model is able to reproduce retrieved tropospheric NO₂ columns over Southeast Asia. The strength or existence of seasonal variation in the emission inventory is found to be of importance. In a high emission case the 2006 NO_x emissions were fitted to the 2000–2006 increase in energy consumption in China. This case shows better agreement with the satellite retrieved 2000–2006 tropospheric NO₂ column trend over central eastern China than a reference case. The satellite comparison thereby highlights an issue noted in other studies: The question may be raised whether current emission inventories underestimate the recent increase in NO_x emissions in Southeast Asia. The development of global OH and methane due to emission changes in Southeast Asia after 2000 is also very dependent on the projected emissions and the balance between changes in NO_x and CO emissions. From 2000 to 2006 the sign of OH and methane lifetime changes are different in the reference case and the high NO_x emission case. As noted earlier, though the high NO_x emission case is an upper estimate of OH changes it is better than the reference scenario in reproducing measured NO₂ trends. From 2000 to 2020 the contributions from Southeast Asia are moderate in three scenarios but much larger than the small overall global emission impact from 1980 to 2000. In the 2020 high Southeast Asian NO_x emission case the impact on global OH and methane is large. It is of comparable absolute magnitude to large emission sectors like biomass burning (Dalsøren, 2001) and international ship traffic (Endresen et al., 2003; Dalsøren et al., 2007; Dalsøren et al., 2008). The OH increase in Southeast Asia is 4.7% in 2020_High_NO_x, such a large change would have significant effect on the oxidation of regional pollutants with short and intermediate lifetimes.

References

- Akimoto, H., Ohara, T., Kurokawa, J.-I. and Horii, N. 2006. Verification of energy consumption in China during 1996–2003 by using satellite observational data. *Atmos. Environ.* **40**, 7663–7667.
- ATA 2007. Air Transport Association. Annual Traffic and Ops: World Airlines. Available at: <http://www.airlines.org/economics/traffic/World+Airline+Traffic.htm>
- Atkinson, R. 2007. Gas-phase tropospheric chemistry of organic compounds: a review. *Atmos. Environ.* **41**, 200–240.
- Berglen, T. F., Berntsen, T. K., Isaksen, I. S. A. and Sundet, J. K. 2004. A global model of coupled sulfur/oxidant chemistry in the troposphere: the sulfur cycle. *J. Geophys. Res.* **109**, D19310, doi:10.1029/2003JD003948.
- Bousquet, P., Hauglustaine, D. A., Peylin, P., Carouge, C. and Ciais, P. 2005. Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform. *Atmos. Chem. Phys.* **5**, 2635–2656.
- Boersma, K. F., Eskes, H. J. and Brinksma E. J. 2004. Error analysis for tropospheric NO₂ retrieval from space. *J. Geophys. Res.* **109**, D04311, doi:10.1029/2003JD003962.
- Dalsøren, S. B. 2001. *3D CTM studies of Biomass Burning and Changes in Atmospheric Oxidation Capacity*. Scientiarum Thesis. Department of Geophysics, University of Oslo, Oslo, Norway, 185 pp.
- Dalsøren, S. B. and Isaksen, I. S. A. 2006. CTM study of changes in tropospheric hydroxyl distribution 1990–2001 and its impact on methane. *Geophys. Res. Lett.*, **33**, L23811, doi:10.1029/2006GL027295.
- Dalsøren, S. B., Endresen, Ø., Isaksen, I. S. A., Gravir, G. and Sørsgård, E. 2007. Environmental impacts of the expected increase in sea transportation, with a particular focus on oil and gas scenarios for Norway and northwest Russia. *J. Geophys. Res.* **112**, D02310, doi:10.1029/2005JD006927.
- Dalsøren, S. B., Eide, M. S., Endresen, Ø., Mjelde, A., Gravir, G. and co-authors. 2009. Update on emissions and environmental impacts from the international fleet of ships: the contribution from major ship types and ports. *Atmos. Chem. Phys.* **9**, 2171–2194.
- Dentener, F., Peters, W., Krol, M., van Weele, M., Bergamaschi, P. and co-authors. 2003. Interannual variability and trend of CH₄ lifetime as a measure for OH changes in the 1979–1993 time period. *J. Geophys. Res.* **108**(D15), 4442, doi:10.1029/2002JD002916.
- Ding, A. J., Wang, T., Thouret, V. C., Cammas, J.-P. and Nédélec, P. 2008. Tropospheric ozone climatology over Beijing: analysis of aircraft data from the MOZAIC program. *Atmos. Chem. Phys.* **8**, 1–13.
- Endresen, Ø., Sørsgård, E., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A. and co-authors. 2003. Emission from international sea transportation and environmental impact. *J. Geophys. Res.* **108**(D17), 4560, doi:10.1029/2002JD002898.
- Emmons, L. K., Edwards, D. P., Deeter, M. N., Gille, J. C., Campos, T. and co-authors. 2008. Measurements of pollution in the troposphere (MOPITT) validation through 2006. *Atmos. Chem. Phys. Discuss.* **8**, 18 091–18 109.
- Ghude, S. D., Fadnavis, S., Beig, G., Polade, S. D. and Van Der A, R. J. 2008. Detection of surface emission hot spots, trends, and seasonal cycle from satellite-retrieved NO₂ over India. *J. Geophys. Res.* **113**, D20305, doi:10.1029/2007JD009615.
- Gregory, D., Morcrette, J.-J., Jacob, C., Beljaars, A. C. M. and Stockdal, T. 2000. Revision of convection, radiation and cloud schemes in the ECMWF Integrated Forecast System. *Q. J. R. Meteorol. Soc.* **126**, 1685–1710.
- Hsu, J., Prather, M. J., Wild, O., Sundet, J. K., Isaksen, I. S. A. and co-authors. 2004. Are the TRACE-P measurements representative of the western Pacific during March 2001? *J. Geophys. Res.* **109**, D02314, doi:10.1029/2003JD004002.
- IATA. 2005. Carrier Tracker, Based on IATA Monthly International Statistics (MIS). Monthly Traffic Analysis. October 2005. Available at: <http://www.iata.org>.
- Isaksen, I. S. A., Zerefos, C., Kourtidis, K., Meleti, C., Dalsøren, S. B. and co-authors. 2005. Tropospheric ozone changes at unpolluted and semipolluted regions induced by stratospheric ozone changes. *J. Geophys. Res.* **110**, D02302, doi:10.1029/2004JD004618.
- Klimont, Z., Streets, D., Gupta, S., Cofala, J., Lixin, F. and Ichikawa, Y. 2002. Anthropogenic Emissions of Non-Methane Volatile Organic Compounds (NMVOC) in China. *Atmos. Environ.* **36**(8), 1309–1322.

- Krol, M. and Lelieveld, J. 2003. Can the variability in tropospheric OH be deduced from measurements of 1,1,1-trichloroethane (methyl chloroform)? *J. Geophys. Res.* **108**(D3), 4125, doi:10.1029/2002JD002423.
- Krol, M. C., Meirink, J. F., Bergamaschi, P., Mak, J. E., Lowe, D. and co-authors. 2008. What can ^{14}CO measurements tell us about OH? *Atmos. Chem. Phys.* **8**, 5033–5044.
- Lawrence, M. G., Jöckel, P. and von Kuhlmann, R., 2001. What does the global mean OH concentration tell us? *Atmos. Chem. Phys.* **1**, 37–49.
- Lelieveld, J., Peters, W., Dentener, F. J. and Krol, M. C. 2002. Stability of tropospheric hydroxyl chemistry. *J. Geophys. Res.* **107**(D23), 4715, doi:10.1029/2002JD002272.
- Lelieveld, J., Dentener, F. J., Peters, W. and Krol, M. C. 2004. On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere. *Atmos. Chem. Phys.* **4**, 2337–2344.
- Leue, C., Wenig, M., Wagner, T., Platt, U. and Jähne, B. 2001. Quantitative analysis of NO_x emissions from GOME satellite image sequences. *J. Geophys. Res.* **106**, 5493–5505.
- Levy, H. 1971. Normal atmosphere: large radical and formaldehyde concentrations predicted. *Science* **173**, 141–143.
- Lin, J., Zhou, N., Levine, M. and Friedly, D. 2008. Taking out 1 billion tons of CO_2 : the magic of China's 11th Five-Year Plan? *Energy Policy* **36**, 954–970.
- Manning, M. R., Lowe, D. C., Moss, R. C., Bodeker, G. E. and Allan, W. 2005. Short-term variations in the oxidizing power of the atmosphere. *Nature* **436**, 1001–1004.
- Mao, J., Ren, X., Brune, W. H., Olson, J. R., Crawford, J. H. and co-authors. 2008. Airborne measurement of OH reactivity during INTEX-B. *Atmos. Chem. Phys. Discuss.* **8**, 14 217–14 246.
- Martin, R. V., Chance, K., Jacob, D. J., Kurosu, T. P., Spurr, R. J. D. and co-authors. 2002. An improved retrieval of tropospheric nitrogen dioxide from GOME. *J. Geophys. Res.* **107**, 4437–445.
- OECD/IEA. 2007. World energy outlook 2007, Executive summary, China and India insights. International Energy Agency (IEA), Head of Communication and Information Office, 9 rue de la Fédération, 75739 Paris Cedex 15, France.
- Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K. and co-authors. 2007. An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmos. Chem. Phys.* **7**, 4419–4444.
- Prinn, R. G., Huang, J., Weiss, R. F., Cunnold, D. M., Fraser, P. J. and co-authors. 2005. Evidence for variability of atmospheric hydroxyl radicals over the past quarter century. *Geophys. Res. Lett.* **32**, L07809, doi:10.1029/2004GL022228.
- Raupach, M. R., Marland, G., Ciais, P., Quéré, C. L., Canadell, J. G. and co-authors. 2007. Global and regional drivers of accelerating CO_2 emissions. *PNAS* **104**, 10 288–10 293.
- Richter, A. and Burrows, J. P. 2002. Retrieval of Tropospheric NO_2 from GOME measurements. *Adv. Space Res.* **29**, 1673–1683.
- Richter, A., Burrows, J. P., Nüß, H., Granier, C. and Niemeier, U. 2005. Increase in tropospheric nitrogen dioxide over China observed from space. *Nature* **437**, 129–132.
- Schultz, M., Bolscher, M., van het, Pulles, T., Brand, R. and co-authors. 2007. RETRO report on emission data sets and methodologies for estimating emissions. Workpackage 1, Deliverable D1–6. EU-Contract No. EVK2-CT-2002–00170. Available at: http://retro.enes.org/reports/D1–6_final.pdf
- Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke and co-authors. 2008. Global wildland fire emissions from 1960 to 2000. *Global Biogeochem. Cycles* **22**, GB2002, doi:10.1029/2007GB003031.
- Sheehan, P. and Sun, F. 2006. Energy use and CO_2 emissions in China: retrospect and prospect. In: *Climate Change Project Working Paper Series*. Centre for Strategic Economic Studies, Victoria University, PO Box 14428, Melbourne VIC 8001, Australia.
- Streets, D. G., Zhang, Q., Wang, L., He, K., Hao, J. and co-authors. 2006. Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *J. Geophys. Res.* **111**, D14306, doi:10.1029/2006JD007118.
- Streets, D. G., Yu, C., Wu, Y., Chin, M., Zhao, Z. and co-authors. 2008. Aerosol trends over China, 1980–2000. *Atmos. Res.* **88**(2), 174–182, ISSN 0169–8095, doi:10.1016/j.atmosres.2007.10.016.
- Szopa, S., Schultz, M. G., Hauglustaine, D. A., Dalsøren, S., van Noije, T. and co-authors. 2007. Report on the reanalysis simulations. Deliverable D4.4 REanalysis of the TROpospheric chemical composition over the past 40 years: a long-term modelling study of tropospheric chemistry. Funded under the 5th EU framework programme. EU-contract EVK2-CT-2002–00170, 66 pp.
- Tanimoto, H., Sawa, Y., Yonemura, S., Yumimoto, K., Matsueda, H. and co-authors. 2008. Diagnosing recent CO emissions and ozone evolution in East Asia using coordinated surface observations, adjoint inverse modeling, and MOPITT satellite data. *Atmos. Chem. Phys.* **8**, 3867–3880.
- Thompson, A. M. 1992. The oxidizing capacity of the Earth's atmosphere: probable past and future changes. *Science* **256**, 1157–1165.
- Tsutsumi, Y., Mori, K., Ikegami, M., Tashiro, T. and Tsuboi, K. 2006. Long-term trends of greenhouse gases in regional and background events observed during 1998–2004 at Yonagunijima located to the east of the Asian continent. *Atmos. Environ.* **40**, 5868–5879.
- Uno, I., He, Y., Ohara, T., Yamaji, K., Kurokawa, J.-I. and co-authors. 2007. Systematic analysis of interannual and seasonal variations of model-simulated tropospheric NO_2 in Asia and comparison with GOME-satellite data. *Atmos. Chem. Phys.* **7**, 1671–1681.
- UNCTAD. 2006. United Nations Conference on Trade and Development. Review of Maritime Transport, 2006. Report by the UNCTAD secretariat. United Nations. New York and Geneva. UNCTAD/RMT/2006. United Nations publications. Sales No. E.06.II.D.7. ISBN 92–1–112699–1. ISSN 0566–7682.
- Van Der A, R. J., Peters, D. H. M. U., Eskes, H. J., Boersma, K. F., Van Roozendael, M. and co-authors. 2006. Detection of the trend and seasonal variation in tropospheric NO_2 over China. *J. Geophys. Res.* **111**, D12317, doi:10.1029/2005JD006594.
- Van Der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendael, M. and co-authors. 2008. Trends, seasonal variability and dominant NO_x source derived from a ten year record of NO_2 measured from space. *J. Geophys. Res.* **113**, D04302, doi:10.1029/2007JD009021.
- van Noije, T. P. C., Eskes, H. J., Dentener, F. J., Stevenson, D. S., Ellingsen, K. and co-authors. 2006. Multi-model ensemble simulations of tropospheric NO_2 compared with GOME retrievals for the year 2000. *Atmos. Chem. Phys.* **6**, 2943–2979.
- Wang, Y. H. and Jacob, D. J. 1998. Anthropogenic forcing on tropospheric ozone and OH since preindustrial times. *J. Geophys. Res.* **103**, 123–131.

- Wang, Y., McElroy, M. B., Martin, R. V., Streets, D. G., Zhang, Q. and co-authors. 2007. Seasonal variability of NO_x emissions over east China constrained by satellite observations: implications for combustion and microbial sources. *J. Geophys. Res.* **112**, D06301, doi:10.1029/2006JD007538.
- Wild, O., Sundet, J. K., Prather, M. J., Isaksen, I. S. A., Akimoto, H. and co-authors. 2003. Chemical transport model ozone simulations for spring 2001 over the western Pacific: comparisons with TRACE-P lidar, ozonesondes, and Total Ozone Mapping Spectrometer columns. *J. Geophys. Res.* **108**(D21), 8826, doi:10.1029/2002JD003283.
- Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter and co-authors. 2007. NO_x emission trends for China, 1995–2004: the view from the ground and the view from space. *J. Geophys. Res.* **112**, D22306, doi:10.1029/2007JD008684.
- Zhang, L., Jacob, D. J., Boersma, K. F., Jaffe, D. A., Olson, J. R. and co-authors. 2008. Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality in North America: an integrated analysis using satellite, aircraft, ozonesonde, and surface observations. *Atmos. Chem. Phys.* **8**, 6117–6136.