A regional model for surface ozone in Southeast Asia

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ABSTRACT

As part of the model intercomparison study MICS Asia II, the Swedish MATCH model was set up for Southeast and East Asia. In that study, the comprehensive photochemistry scheme of MATCH was used for the first time in Asia. The current work focuses on results of surface ozone from the MATCH model simulations falling outside the model intercomparison study. Model results of surface ozone concentrations for the entire year of 2001 were investigated and compared with measurements in Southeast Asia.

The model produced higher surface ozone concentrations than the observations at all of the non-remote stations investigated but underestimated during the dry season at remote locations. Modelled seasonal variation was similar to, but less pronounced than, the variation in the measurements. This study indicates that NO_x is the limiting precursor for ozone production in the model, while the fractionation in different species and total amount of non-methane volatile organic compounds (NMVOC) emissions are less important. Naturally emitted NMVOC, isoprene, is an important precursor of surface ozone at certain conditions, and a better inventory of these emissions is needed. Deposition velocities of ozone also have impact on surface concentrations. To improve the model performance, it is important to add a land use inventory with corresponding deposition velocities.

1. Introduction

Ozone is a secondary pollutant known to influence human health and agricultural efficiency. The main ozone precursors are nitrogen oxides (NO_x) and volatile organic compounds (VOCs). Emissions and atmospheric concentrations of these agents are increasing rapidly in Southeast Asia as a result of the fast growth of the transport sector and industrialization. In 2001, the transport sector contributed 37% of the NO_x emissions, while 27% came from power generation and 18% from industry (Streets et al., 2003). Industry, transportation and power generation by fossil fuel are important sources of NMVOCs, but biomass burning and domestic biofuel emit even higher amounts and are difficult to quantify and map.

A large intercomparison study of photochemical transport models for air pollutants, called MICS Asia II, was recently carried out (Carmichael et al., 2008). The aim of MICS Asia II was to gain a common understanding of model performance and uncertainties in Asia and to investigate the transport and deposition of sulphur and nitrogen compounds, ozone and aerosols in East Asia. The results of that study show that at least eight regional scale models have been applied to Southeast and East

Asia (Carmichael et al., 2008). One of these is the Swedish Multiscale Atmospheric Transport and Chemistry model (MATCH) used in the current study to investigate near-surface ozone in Southeast Asia. The model has been used in previous studies of ozone in Europe and Sweden (Langner et al., 2005; Solberg et al., 2005) and in Southeast Asia for studies of acidification (Engardt and Leong, 2001; Engardt et al., 2005; Siniarovina and Engardt, 2005).

The validation of modelled ozone in MICS Asia II points out that all regional models set-up have difficulties reproducing measured ozone concentrations and that more effort is required to explore the uncertainties and improve the model's abilities (Han et al., 2008). A way forward can be to nest urban scale models with better emission inventories into the regional models. An example of such urban scale modelling within this region is given by Zhang et al. (2005). In that study, two models are used to simulate photochemistry over the Bangkok metropolitan region, an area of 88 km × 72 km, during a 2-d period in 1997.

The monitoring of ozone in Southeast Asia is developing rapidly but for the modelled year only 52 stations were found, of which the main part was located in cities or nearby large emission sources in Thailand.

The current study deals with the difficulties of moving a regional photochemistry, transport and dispersion model from one part of the world, Europe, to another, Southeast Asia. As it is important to find out whether the model accurately

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represents reality, the main focus of the present paper is on comparisons between simulated and measured values, with a critical evaluation of resulting differences including sensitivity studies of uncertain model parameters.

2. Methods

2.1. Model description

This study developed and used a standard set-up for MATCH in Asia. To test the robustness of the set-up, several sensitivity studies were performed of which some are more thoroughly discussed.

The MATCH model is a three-dimensional Eulerian model developed at the Swedish Meteorological and Hydrological Institute (SMHI; Robertson et al., 1999). It was used in MICS Asia phase one to simulate sulphur transport and deposition (Carmichael et al., 2002).

The MATCH model can be used with a number of different chemistry, deposition and advection schemes. In this survey, thermal and photochemical gas phase chemistry based on Simpson et al. (1993) considers 70 species. The chemical scheme includes 110 thermal, 28 photochemical, 2 aqueous phase, 5 aerosol reactions and 4 gas phase, aqueous phase and aerosol equilibria. The formation of ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3) is modelled following Hov et al. (1994) with some modifications. The model includes methane, ethane, ethene, methanol, ethanol, formaldehyde, acetaldehyde, butanone, propene, n-butane, o-xylene, isoprene and their reaction products. These compounds are chosen to represent all different volatile organic molecules emitted to the atmosphere. They span the normal range of ozone creation potentials for most important organic pollutants (Pleijel et al., 1996).

The meteorological data were taken from ECMWF. They were updated every 6 h and then interpolated to 1-h resolution. These data showed very good agreement with measurements compared to eight other regional models in the MICS-Asia study (Carmichael et al., 2008). The correlation coefficient was above 0.65 for the wind direction and 0.8 for the wind speed. The relative humidity was most poorly reproduced, with a correlation coefficient of 0.4, while the temperature was almost exactly achieved by the model.

The resolution was $0.5° \times 0.5°$ in the horizontal direction, with 25 vertical layers reaching the top boundary at approximately 8 km. The full domain reaches from 13°S to 54°N and 75°E to 158°E, but the focus in this study is on a smaller part including most countries in Southeast Asia. The simulation period started on 1 February 2001 and ended 1 April 2002 to capture the seasonal variations and differences between 2001 and 2002.

Dry deposition velocities were derived as a function of stability and species and were given a diurnal variation over land. A sensitivity test was conducted, decreasing the dry deposition of ozone over land by half. The deposition is treated in the model with a land/sea dependency. The model included only one type of land use, however; for example, no distinctions were made between forest and crops.

2.2. Emission data and boundary conditions

Anthropogenic emissions of SO_x, NO_x, NH_3, CH_4, NMVOC and CO from Streets et al. (2003) were used with monthly variations. The anthropogenic NMVOCs were divided into 11 component classes. In a sensitivity test, splitting factors from previous work in Europe were used, while the base set-up of the model considered detailed information about the NMVOCs in Southeast Asia based on data provided by D. Streets, personal communication 2005 (Table 1). In the Asian case, the amount of NMVOCs from biomass burning is higher. As the uncertainty of total NMVOC is over 200% (Streets et al., 2003), two tests of how this influences the total ozone were completed. In the first case, the emitted NMVOC was reduced to 50% and in a second test increased to 200%. The spatial distribution of emissions remained unaltered in all cases.

The area emissions were released in the three lowest vertical layers of the model, 50% in layer 1, reaching from the surface to 20 m, 30% in layer 2, from 20 to 50 m and 20% in layer 3, from 50 to 100 m. Emissions from volcanoes and large point sources were released at heights of 1500 m and in layer 4 (100–200 m), respectively. A sensitivity test of the emission heights was done with all area emissions only in layer 1. The sensitivity of the temporal distribution of emissions was tested by implementing a roughly estimated diurnal cycle of NO_x and anthropogenic VOCs. The emissions doubled during 10 daytime hours, then diminished during 2 h to reach 0.2% of the average emission. This low emission was kept during the night and then increased again in the morning to reach the higher daytime value.

There were also natural emissions of isoprene according to GEIA (Guenther et al. 1995) that were released in the lowest model layer, with a diurnal cycle linked to sunset–sunrise. According to Karl et al. (2004), measurements indicate that the isoprene fluxes could be three to four times lower than originally reported by Guenther et al. (1995), for which reason our model was also tested with a three times lower isoprene emission.

Constant boundary values at the south, east, west, north and top boundaries were applied. As the boundaries for the model domain are at least 20 grid boxes from the area investigated here, no special sensitivity analysis of the boundary values was made at the south, east, west and north boundaries; however, the effect of the upper boundary on surface ozone was investigated.

3. Measurement data

The focus in this study is Southeast Asia. Though most of the monitoring stations used for evaluating the model performance are located in Thailand, there are also eight remote stations in other parts of Southeast Asia. Data from five institutions were used for comparison with modelled ozone:

Table 1. Translation of NMVOC species from Streets inventory and NMVOC species used in the model. Details about the two different divisions of NMVOCs, European and Southeast Asian set-up, investigated

Representative ozone-forming VOC in MATCH	Includes the following species in the Streets et al. (2003) inventory	Fraction of total NMVOC emission		Dominating source in Southeast Asia
		European set-up	Southeast Asian set-up	
Ethane	Ethane, acetylene	0.07	0.1	Domestic biofuels
n-butane	Butane, propane, pentanes, ketones other alkanes	0.37	0.25	Non-domestic (industry/power generation, fossil fuels)
o-xylene	Xylenes + benzene, toluene, other aromatics	0.25	0.17	Non-domestic
Ethene	Ethene	0.04	0.1	Biomass burning, domestic biofuels, industry/power
Propene	Propene, terminal alkenes, internal alkenes	0.04	0.11	Industry/power, domestic biofuels, biomass burning
Methanol	No emissions	0.01	0	No emissions
Ethanol	Other	0.14	0.2	Biomass burning
Formaldehyde	Formaldehyde	0.01	0.03	Biomass burning
Acetaldehyde	Other aldehydes	0	0.03	Biomass burning
Butanone	No emissions	0.03	0	No emissions Non-domestic
Non-reactive	Halocarbons	0.04	0.01	No emissions

(1) Hourly data on ozone and NO_x from 36 stations in Thailand were used as provided by the Bureau of Air Quality and Noise Management, Pollution Control Department (PCD) of the Ministry of Natural Resources and Environment, Thailand. The ozone was measured by API Photometric Ozone analyser model 400, which is the equivalent of the US EPA EQ0A-0992–087 method. These stations are all considered to be urban, near urban or near a special emission source, as this is where monitoring of surface ozone as a risk for health effects is done.

(2) Data from Differential Optical Absorption Spectroscopy (DOAS) measurements of ozone over Manila at the Philippines, provided by the Manila Observatory, were also used. This is also an urban station, and the comparison with model results is complicated by the location of the station close to the ocean.

(3) Data from passive samplers at six remote stations in Thailand provided by IVL, the Swedish Environmental Research Institute, and ERTC, the Environmental Research and Training Centre in Bangkok (Iverfeldt et al., 2003), were considered. The benefit of the latter data is that they are collected at remote stations while the disadvantage is the monthly temporal resolution, leaving questions about diurnal variation unanswered.

(4) Typical diurnal variation and annual average of ozone in 1996–1997 were reported in Pochanart et al. (2001).

(5) Annual median values of ozone from passive diffusive samplers at nine remote locations in Southeast Asia presented by Carmichael et al. (2003) were also used.

The stations are divided into nine groups with respect to their environment according to Table 2.

4. Model results and discussion

4.1. Annual mean concentration

The annual mean ozone concentration at 55 stations across Southeast Asia is presented in Table 2. On an annual basis, the model generally overestimates measured values. For non-remote stations, the modelled ozone for a single station was up to seven times higher, and the average difference for all non-remote stations was 14 ppbv. The modelled concentrations were more similar to the measured ones at remote locations, where the highest overestimation was less than three times the measured value, and the model produced lower concentrations at two stations, with differences of up to 30% of the measured concentrations.

The modelled high annual mean concentrations at non-remote stations in Table 2 can be an effect of all the stations being located in areas where there is a special need for measurements, such as in industrial and urban environments or nearby large point emission sources. Averaged emissions over the grid cell do not capture the local NO_x maxima. High emissions of NO_x lead local minima of ozone, and this can explain higher model predictions of ozone than what is measured. In reality, the high emissions give ozone production downwind of the emission source, and, for this reason, the model should underestimate the ozone at more remote locations, which is indeed seen at three remote stations.

The resolution in the model does not make it optimal for giving good values at non-remote stations as it is designed to

Table 2. Description and classification of the monitoring stations together with annual mean ozone concentration

Type	Ref	Station	Name	Latitude	Longitude	Annual mean concentration O_3 (ppbv)	
						Measurement	Model
Bangkok	PCD	1	Ratburana, Thailand	13.7	100.5	14	27
Urban	PCD	2	Junkasame, Thailand	13.8	100.6	7	34
Ambient	PCD	3	Ramkhamhaeng, Thailand	13.8	100.6	24	34
	PCD	4	National Housing Authority, Thailand	13.8	100.7	17	34
	PCD	5	Huai Khwang, Thailand	13.8	100.6	9	34
	PCD	6	None-tree Vitaya, Thailand	13.7	100.6	10	27
	PCD	7	Dept. of Energy Affairs, Thailand	13.8	100.5	20	34
	PCD	8	Singharatpitayakom, Thailand	13.7	100.5	18	27
Bangkok	PCD	9	Thonburi Highway Dist., Thailand	13.6	100.4	16	27
Urban	PCD	10	Thonburi Substation, Thailand	13.7	100.5	8	27
Roadside	PCD	11	Traffic Police Residence, Thailand	13.8	100.6	8	34
	PCD	12	Dindang Housing Authority, Thailand	13.8	100.6	5	34
Bangkok	PCD	13	Rangsit, Thailand	14.0	100.6	9	34
Suburban	PCD	14	Sukothai University, Thailand	13.9	100.6	14	34
Ambient	PCD	15	Sanamchan Nakhonpathom, Thailand	13.8	100.1	22	30
Cement	PCD	16	Saraburi, Thailand	14.5	100.9	13	33
Industrial	PCD	17	Kao Noy Saraburi, Thailand	14.5	100.9	19	33
Seaside	PCD	18	Ratchaburi Eng. Department, Thailand	13.4	99.9	19	27
Industrial	PCD	19	Samutsakhon, Thailand	13.5	100.3	16	27
Ambient	PCD	20	MaptaPut, Thailand	12.7	101.2	16	30
	PCD	21	Rayong, Thailand	12.7	101.3	20	29
	PCD	22	Rayong, Thailand	12.7	101.1	16	30
	PCD	23	Laem Chabang Chonburi, Thailand	13.1	100.9	17	26
	PCD	24	Siracha Chonburi, Thailand	13.2	100.9	17	26
	PCD	25	General Educationion Chonburi, Thailand	13.4	101.0	17	24
Coal Mine	PCD	26	Lampang, Thailand	18.3	99.5	14	27
Valley	PCD	27	Sob Pad, Thailand	18.3	99.8	12	28
	PCD	28	Ta See, Thailand	18.4	99.8	10	25
	PCD	29	Mae Moh, Thailand	18.3	99.7	17	27
Cities	PCD	30	Nakhonsawan, Thailand	15.7	100.1	24	27
Roadside	PCD	31	Suratthani, Thailand	9.2	99.3	12	23
	PCD	32	Phuket, Thailand	7.9	98.4	14	30
	PCD	33	Khonkaen, Thailand	16.4	102.8	14	33
	PCD	34	Nakhonratchasrima, Thailand	15.0	102.1	18	31
Northern	PCD	35	Chiang Mai, Thailand	18.8	99.0	20	29
	PCD	36	Chiang Mai, Thailand	18.8	99.0	13	29
Remote	CAR	37	Tanah Rata, Malaysia	4.0	101.0	16	25
	CAR	38	Lawa Mandau, Malaysia	6.0	116.0	17	34
	CAR	39	Bukit Kototabang, Indonesia	0.0	100.0	11	19
	CAR	40	Kalimantan, Indonesia	-2.0	114.0	10	12
	CAR	41	Mt St Thomas, Philippines	16.0	120.0	26	26
	PHIL	42	Metro Manila	14.5	121.0	28[a]	33[a]
	CAR	43	Chiang Mai, Thailand	20.0	99.0	14	28
	CAR	44	Nakhon Sri Thammarat, Thailand	8.0	100.0	9	25
	CAR	45	Luang, Prabang. Laos	20.0	102.0	11	23
	CAR	46	Savvanaketh, Laos	17.0	106.0	15	27
	IVE	47	Nationel forest park Phatoob, Thailand	18.9	100.7	13[b]	28[b]
	IVE	48	Phumipol dam, Thailand	17.2	99.0	18[b]	24[b]
	IVE	49	Nam-Phung dam, Thailand	17.0	104.0	26[b]	30[b]

Table 2. Continued

Type	Ref	Station	Name	Latitude	Longitude	Annual mean concentration O$_3$ (ppbv)	
						Measurement	Model
	IVE	50	Skaeret Research Station, Thailand	15.0	102.0	25[b]	29[b]
	IVE	51	Pathumthani, Thailand	14.0	101.0	24[b]	35[b]
	IVE	52	Srinakarin, Thailand	14.0	99.0	27[b]	22[b]
	IVE	53	Tung Khaye nationel forest park, Thai	8.0	100.0	13[b]	26[b]
	PON	54	Inthanon, Thailand	19.0	98.0	27	23
	PON	55	Srinakarin, Thailand	14.0	99.0	28	21

PCD—Pollution Control Department.
CAR—Carmichael et al. (2003).
IVE—Iverfeldt et al. (2003).
PON—Ponchanart et al. (2000).
[a] Not annual mean but mean of September, October, November, January, February, March.
[b] Not annual mean but mean of June 2001–March 2002.

produce mean values for 0.5° × 0.5° grid cells, the resolution of the emission inventory and the driving meteorology.

The modelled NO$_x$ concentration was for the non-remote locations lower than the measured values. Typically model values were less than 10 ppbv while the monitoring concentrations were between 10 and 80 ppbv. For two remote stations with measurement data, both the measured and modelled NO$_x$ levels were low (station Tanah Rata model 2.5 ppbv and measurement 1 ppbv, station Khao Lam Dam model less than 0.5 ppbv and measurement 2 ppbv). The modelled concentration of NMVOCs was also low. Compared to measurements in Bangkok, Zhang and Oanh (2002) report 2.5 and 1.5 ppmC for January and August while the model estimate is only 0.16 and 0.06 ppmC, respectively. The model can not reproduce concentrations from measurements in heavily polluted urban environments, where such high concentrations are found. The explanation of the low-modelled precursor concentrations can be that the grid size mean of NO$_x$ and NMVOCs favours O$_3$ production, compared to the more diverse concentrations in reality.

4.2. Seasonal variation

The monthly mean near-surface ozone concentrations for three months, April, August and December, are presented in Fig. 1, together with monthly accumulated precipitation predicted by the model. April is the beginning of the rainy season in the north while August is in the middle of the rainy season for most of Southeast Asia except southern Indonesia, which has its high peak precipitation in December when the northern part of the domain is dry. Figure 2 shows the averaged seasonal variations for all Thai stations from PCD for February 2001 to March 2002, and Fig. 3 shows the seasonal variations for six remote stations with monthly data available for the modelling period. At remote stations, the modelled ozone is higher than measurements during the wet season. To the contrary, during the dry season, the modelled ozone does not reach the measured values at four

of six stations. The model seems to have less seasonal variation than reality.

The seasonal variation reported in Figs. 2 and 3 has a characteristic low for both the model and measurements between May and October and a period of higher concentrations from November to April. The low concentration period coincides with the rainy season that has greater wet deposition of precursors and southwesterly monsoon with transport from the ocean, while November to April is the dry season with high solar radiation and northeasterly monsoon. The modelled values have their maxima in November while the measured values have their maxima a few months later, in January or February.

4.3. Daily and diurnal variation

Data with hourly temporal resolution were available only at non-remote stations in Thailand. How well the model did reproduce the day-by-day and hour-by-hour variations differed a lot from station-to-station and from month-to-month. The span of the correlations of the hourly values for each month, representing how well the diurnal cycle was simulated, and the correlation of the diurnal means for the entire period, is illustrated in Fig. 4. Station 15, a Bangkok suburban location, is chosen to represent a station where the modelled series is following the measured values well, while station 21, in the seaside industrial group, is chosen as an example of a station where the correlation is poor. Time-series of diurnal means for eight weeks at these two stations is presented in Fig. 5, and the diurnal mean variation for each month at the two chosen stations is seen in Fig. 6. Correlations with DOAS data from the Philippines were not as good as for the Thai stations. By studying time-series from several locations in the model (without measurements), it was found that there is no or small diurnal variation over the ocean, while the diurnal cycle is much more evident at insular locations and on the mainland. The reason for small diurnal variation over ocean is that the deposition velocity of ozone to water is small
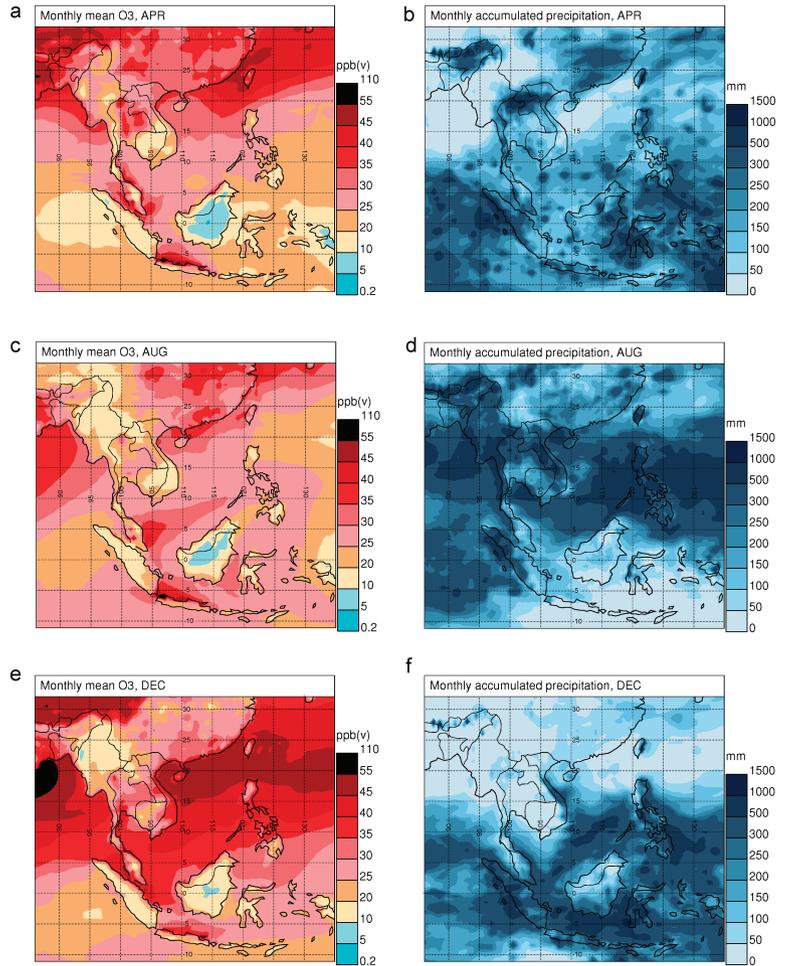
Fig. 1. Modelled monthly mean near-surface ozone concentration and precipitation for 3 months, April, August and December 2001.

and constant and ocean emissions of precursors is lower. This implies that the coastal grid cells, containing mostly water, are given an incorrect decreased diurnal variation of ozone. This can be seen in, for example, Metro Manila and most stations within the group Seaside Industrial Ambient. In remote locations on mainland Thailand, the diurnal pattern is least pronounced in the rainy season from June to August, similar to the urban locations (Fig. 6A). Comparing field campaign data from Mae Moh, Thailand (Matsuda et al., 2005), with results from January to April 2002, modelled diurnal maxima occur at the same time as measurement maxima, and the modelled mean has the same magnitude as the measurements. This is also true for the measured concentrations from the nearby PCD station, station 29, Mae Moh, for these dry months.

The overestimation of the modelled ozone is often high during nighttime. The maximum daytime values are not always overpredicted, as are the nighttime concentrations. Typically modelled nighttime ozone is 10–30 ppbv while measured concentrations are less than 10 ppbv at most stations. Modelled and measured daily maxima are 20–70 ppbv. The average ozone concentrations produced by the model would correspond much more closely to
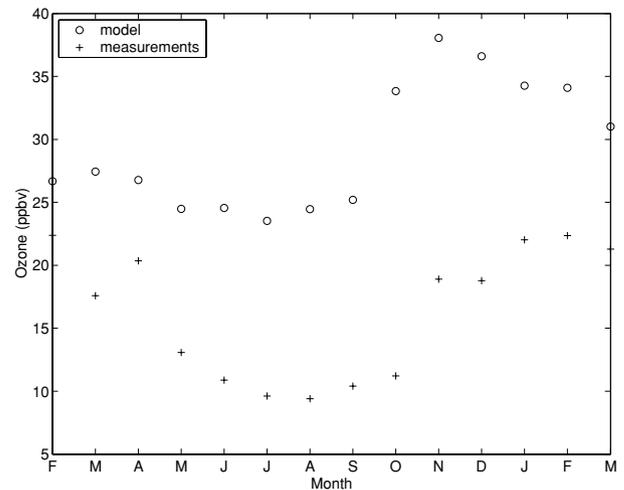

Fig. 2. Monthly means of ozone averaged over 36 non-remote stations from February 2001 to March 2002.
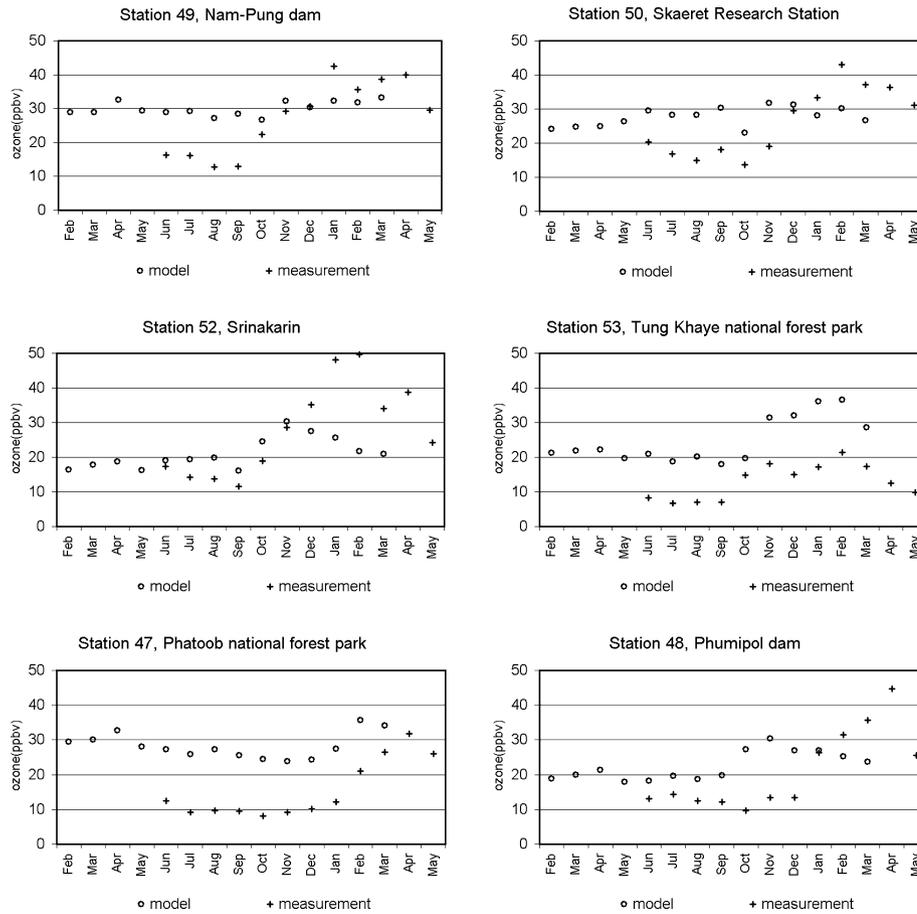
Fig. 3. Seasonal variation of ozone at remote stations in Thailand. Measured data from Iverfeldt et al. (2003).
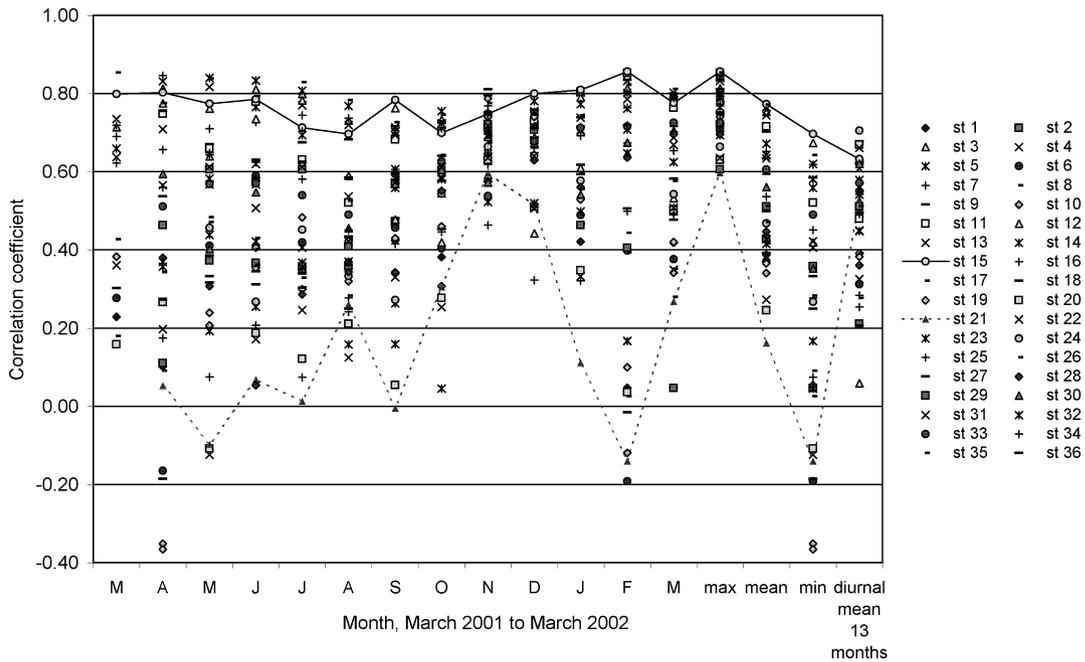

Fig. 4. Correlation coefficients between the hourly measured and modelled concentrations. Max, mean and min refer to the highest, average and lowest monthly correlation coefficients, respectively.
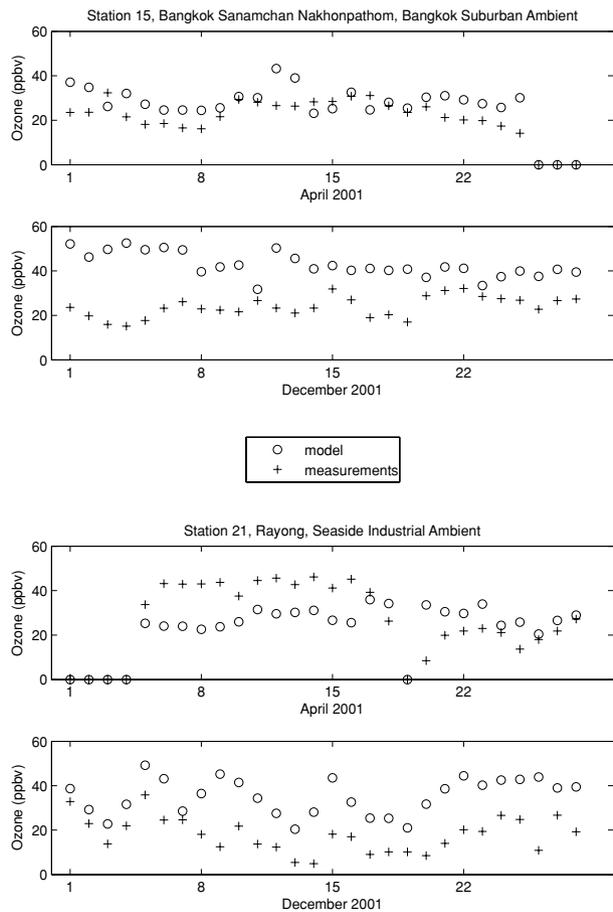
Fig. 5. Diurnal mean of measured and modelled ozone at two different stations, one suburban, in Bangkok, and one seaside industrial, in Rayon, during eight weeks.

the measurements if the lower nighttime concentrations were better reproduced.

4.4. Sensitivity tests

4.4.1. Deposition velocity. To test the robustness with regard to different deposition velocities of ozone, the ozone deposition velocity in the model was decreased by half. The maximum change in ozone concentration at a remote location was 8 ppbv, corresponding to a 30% higher value for the lower deposition velocity simulation. At most remote locations, the change in concentration was between 10 and 30%, while the change over the ocean was always less than 5% and for non-remote locations less than 10 ppbv.

4.4.2. Emissions. The emission inventory is an important part of the modelling study. Compared to three other inventories, the Asian inventory, REAS (Ohara et al., 2006, the global EDGAR 3.2 (http://www.mnp.nl/edgar/model/v32ft2000edgar/) and the global IIASA (http://www.iiasa.ac.at/rains/global_emiss/global_emiss1.html), the emission inventory

used (Streets et al., 2003) is found to have lower emissions (Ohara et al., 2007). For example, the NO_x emissions in Southeast Asia are at least 20% lower than the three other inventories (REAS 3770 kt yr^{-1}, Streets et al. 3058, EDGAR 3913 and IIASA 3944). In the case of CO, it is 40% lower over Southeast Asia (REAS 54514 kt yr^{-1}, Streets et al. 34045, EDGAR 42606 and IIASA 39800). These differences are not constant over the domain; for example, NO_x for Thailand is 1086 kt/y in Streets et al. (2003), while the REAS inventory reports only 592 kt/y. Some of this difference can be due to the treatment of international aviation, shipping and open biomass burning. The newer REAS inventory (Ohara et al., 2006) does not yet include open biomass burning, which is difficult to estimate for this region.

The difference in absolute ozone concentration due to speciation of VOC emissions over the entire region is smaller than 4 ppbv for both the annual and the monthly mean values. The European differentiation between NMVOCs emissions results in less surface ozone, where ozone is high and greater where ozone is low. Close to emission sources, the Southeast Asian set-up gives 0–1 ppbv higher ozone concentrations and in more remote areas a maximum of 3 ppbv lower. A comparison of the two schemes with respect to ozone-forming potential (Carter, 2003) indicates that the total maximal incremental reactivity (MIR) is higher in the Asian division. The uncertainty in NMVOC emissions estimates is greater than 200% (Streets et al., 2003), indicating that the total emission of NMVOCs is a more important factor that might have a great influence on our results. However, the results of the model 50, and 200% NMVOC emissions do not change the annual mean ozone concentration by more than −2 ppbv and +4 ppbv, respectively. The decrease in ozone was uniform over the domain when the NMVOC emissions were decreased, while in the latter case the increase near large emission regions such as Bangkok, Chiang Mai, Singapore and Jakarta was somewhat more distinct, but did not exceed 4 ppbv annually or 10 ppbv monthly.

The vertical emission pattern was tested with all area emissions in the lowest model layer, but the annual modelled ozone changed less than 4 ppbv in any grid cell.

The inclusion of a diurnal variation of emissions of NO_x and VOCs increased the annual mean concentrations of ozone over inland polluted areas as much as 20 ppbv, while the change over ocean was small, a decrease that in a few grid boxes exceeded 2 ppbv. Close to areas with high emissions, where ozone in measurements has a strong diurnal cycle, the modelled ozone actually increased both during day and night when applying a diurnal variation of NO_x and VOC emissions. This means that the ozone is even more overestimated compared to urban measurements when a diurnal variation of its precursors was included.

Figure 7 shows the changes in surface ozone when the natural isoprene emissions are reduced by a factor of three, following the findings of Karl et al. (2004). The highest increase in near-surface ozone occurs at Borneo, mainland Malaysia, and
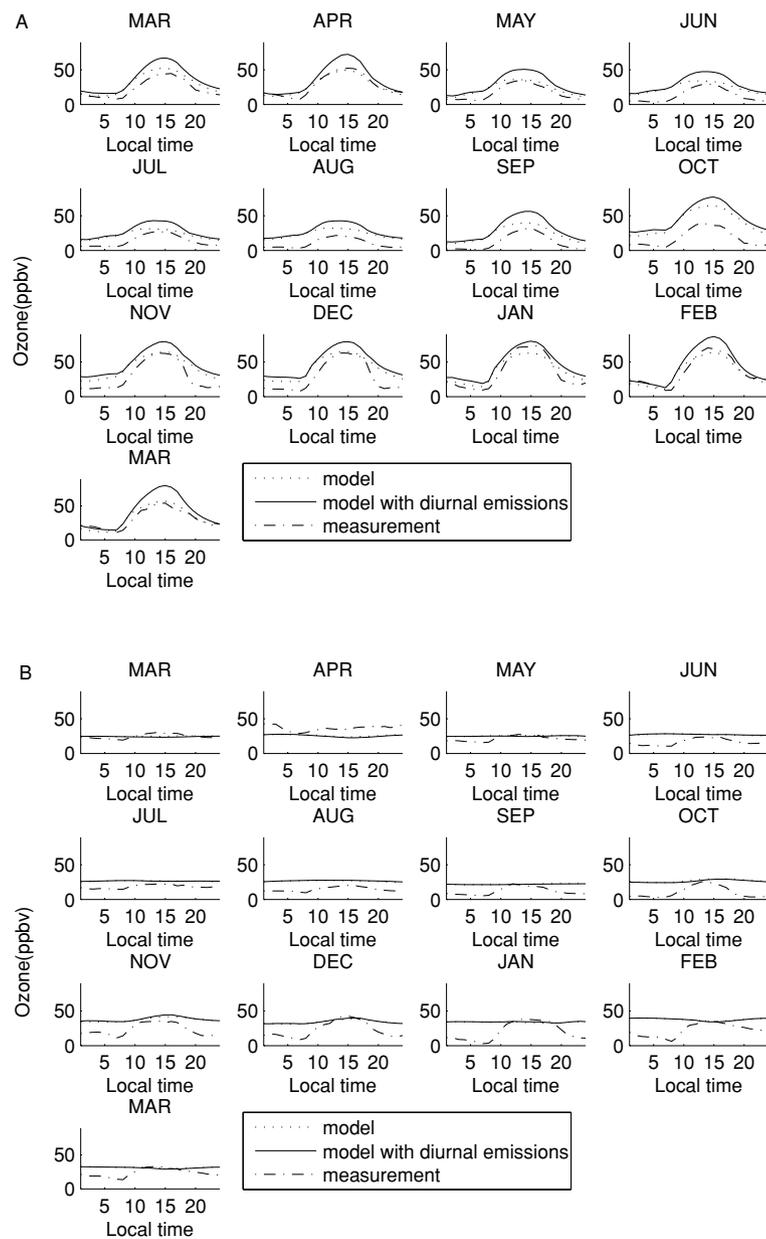
Fig. 6. Mean diurnal variation of O$_3$ concentration for each month of the simulation period. (A) Sanamchan Nakhonpathom, a Bangkok suburban station. This station showed the best correlation between modelled and measured concentrations. (B) Rayong, in the group Seaside Industrial Ambient, showed the poorest correlation between modelled and measured concentrations. Model results for two different set-ups are shown together with observed diurnal variation.

Sumatra, while the greatest decrease is found near and around Chiang Mai, Kuala Lumpur, Singapore, Jakarta and Telukbetung. Where there is little anthropogenic NO$_x$ emission, more isoprene has a negative influence on the ozone production; accordingly, the ozone increases when some isoprene is removed at Borneo, Sumatra and the less industrialized regions of mainland Malaysia. Where there are large anthropogenic NO$_x$ emissions, the reduced isoprene also reduces ozone concentrations. As the concentration of isoprene in the model became unreasonably high, at some locations more than a 75 ppbv monthly mean when isoprene was used according to GEIA (Guenther et al., 1995). As isoprene is an important precursor of ozone, it is urgent to attain reasonable emissions of it. A model for biogenic

emissions, such as the Model of Exchange of Gases between the Atmosphere and Nature, (MEGAN; Guenther and Wiedinmyer, 2004), could probably improve our model results. Another way to achieve better isoprene concentrations in the model would be to add isoprene deposition. Isodorov et al. (2007) report that the "isoprene absorption by underlying surface can be 2–25% of phytogenous emission of this hydrocarbon" .

4.4.3. Boundary concentrations. Two profiles of ozone concentration using different upper boundary conditions of ozone are presented in Fig. 8. The surface ozone over land seems to be affected little by the changed upper boundary. In the entire domain, except for very high altitude locations, the maximum change in monthly surface ozone over land was 6 ppbv, and the
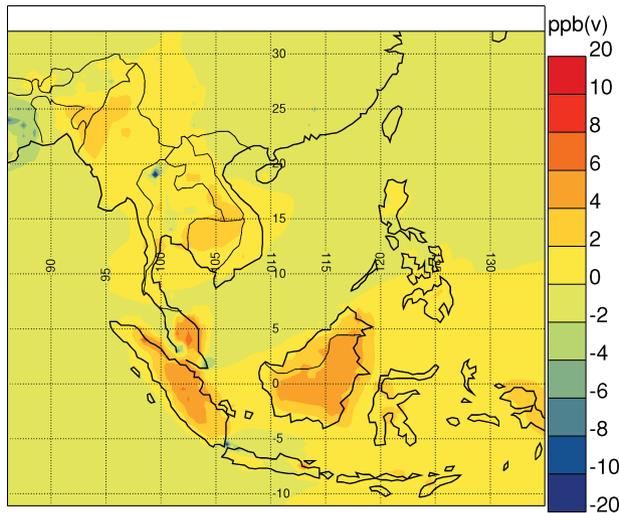
Fig. 7. Difference in annual mean ozone concentration from model set-up with 1/3 of isoprene emissions and model set-up with 100% isoprene emissions.
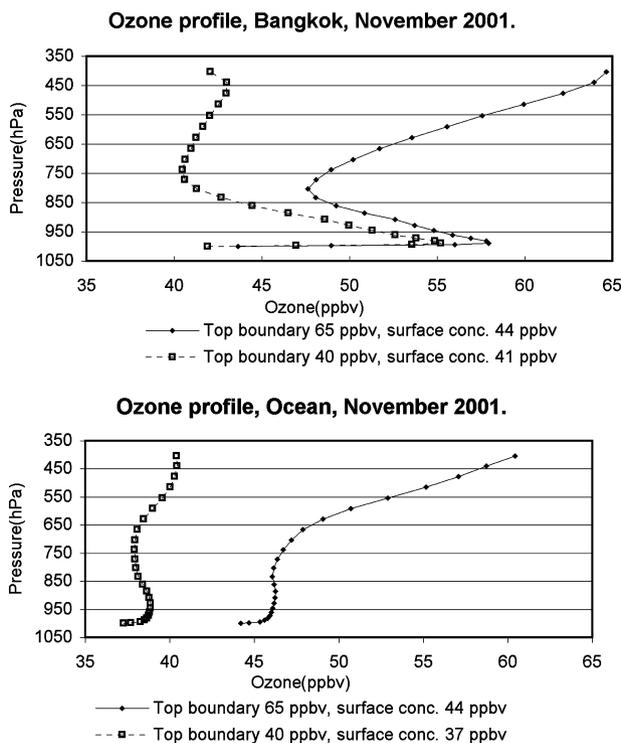

Fig. 8. Modelled vertical ozone profiles showing the dependence of surface ozone on upper boundary values. (A) Bangkok (13.75°N, 100.5°E), (B) ocean (15°N, 115°E).

change was most often below 2 ppbv when the top boundary was changed by 25 ppbv. In contrast, the ozone concentration over the ocean is influenced more by the top boundary. Figure 8 illustrates a location and month where the difference in surface ozone due to the top boundary over the ocean was evident.

5. Conclusions

The present study deals with modelling of near-surface ozone in Southeast Asia and was aimed at simulating 1 yr at the beginning of the current century for which we had access to meteorological data and a recent emission inventory. For this year, it was possible to find only a limited amount of measurement data for comparisons. Nevertheless, the results are promising and can be used as a basis for further modelling studies in the area, hopefully with more measurement data for comparison.

The model produces higher concentrations of ozone but considerably lower NO_x levels than measured concentrations.

Sensitivity tests of NO_x and VOC emissions indicate that NO_x is a limiting factor in the ozone chemistry formulation used in the model. The NO_x seems to be removed too fast by ozone production reactions, leading to an overestimation of O_3 and underestimation of NO_x concentrations. This can be an effect of the grid size used in the model; as the NO_x is emitted, it is immediately spread in the entire grid cell, resulting in concentrations favourable for ozone production. Even though different NMVOCs have different ozone-forming potentials and reactions paths, varying the relative abundance of them showed a minor impact on ozone formation. The modelled ozone distribution for Southeast Asia is characterized by high concentrations near industrial areas such as Bangkok, Kuala Lumpur, Singapore and Jakarta. Nesting of urban scale models of these areas and refined emission estimates could improve the model results. Ozone concentrations over the ocean are low when the monsoon is southwesterly and high when the northeasterly monsoon transports pollutants from the continent.

Even though, the most urgent air quality problems are found on the local scale in the Asian mega cities, there is a great need of improving modelling on the regional scale to understand the total system and to possibly trace less evident effects of anthropogenic emissions.

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