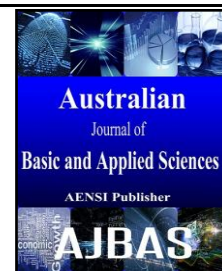




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### Climate Change Impacts on Hydroxyl Radical (OH) from the CiTtyCAT Tropospheric Chemistry Model Simulations

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#### ABSTRACT

Hydroxyl radical, OH at the tropospheric level plays an important role as a cleansing agent in the atmosphere for atmospheric greenhouse and trace gases. Hydroxyl radical also prevents the enhancement of these gases. Without OH, the Earth's atmosphere chemical composition will be very different and might be harmful to living beings. In this study, the authors have explored the impact of regional climate change to the tropospheric OH in urban, rural and remote areas in Malaysia. This paper aims to develop a climate change scenario for A2 emissions over Malaysia using RCM PRECIS model and used the climate output as input to CiTtyCAT, a tropospheric chemistry model. In the 21<sup>st</sup> century, the surface temperature and precipitation are found to increase both during the wet season (winter season) and dry season (summer season). In response to climate change relative to the baseline OH concentration, tropospheric OH concentration has been found highly variable in the study locations over Malaysia. These projections were observed in both seasons. This study has suggested that tropospheric OH sensitivity is not only to climate change but other factors in response OH concentration variability as well.

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#### INTRODUCTION

Oxidation is needed for the removal of greenhouse gases (GHG) from the atmosphere and prevents enhancement of these gases. In the atmosphere, GHGs are oxidized and converted to forms that can be removed either by dry or wet deposition (Fuglestedt, 1995). The oxidizing capacity of the atmosphere is determined by the concentrations of the reactive oxidants in the atmosphere which include hydroxyl radical (OH), ozone (O<sub>3</sub>), nitrate radicals (NO<sub>3</sub>), excited state atomic oxygen (O(<sup>1</sup>D)), hydroperoxyl radicals (HO<sub>2</sub>), organic peroxy radicals (RO<sub>2</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), ground state atomic oxygen (O(<sup>3</sup>P)) and organic peroxides (ROOH). In the troposphere, the oxidation is heavily depends on the level of hydroxyl radical (OH) (Levy, 1971; Morgenstern *et al.*, 2013) and its geographical distribution (Levy, 1971). Other species are also contributing in atmospheric oxidation processes but they are less important (Fuglestedt, 1995).

Tropospheric hydroxyl radical plays an important role as a removal mechanism for atmospheric trace gases, through oxidation of

atmospheric pollutants such as nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO) and non-methane volatile organic compounds (NMVOCs), and removes ozone depleting substances such as hydrofluorocarbons (HFCs) (DeMore *et al.*, 1996). The radical OH is also known to participate in the formation of atmospheric aerosols such as sulfate, nitrate and secondary organics (e.g. Koch *et al.*, 2006). In the troposphere, the chemistry of OH is strongly linked with ozone, where the production of OH is initiated by the photolysis of ozone at wavelengths smaller than 320 nm (R1), where it produces excited oxygen atoms, O(<sup>1</sup>D) that can attack water vapor to produce the hydroxyl free radical (OH) (R2) (Atkinson *et al.*, 2004).



In environment with high levels of ozone, shortwave radiation and high humidity shall favor the production of OH. Ozone as OH precursor also depends on emissions of its precursors and on climatic conditions; shortwave radiation is affected by overhead absorption by ozone, scattering and

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absorption by clouds and aerosols, and reflection from the Earth's surface (Madronich, 1987; Thompson, 1992; Voulgarakis *et al.*, 2009a); and water vapour abundances are largely determined by temperature. A growth of ozone implies an increase of OH concentration but increasing in emission of CH<sub>4</sub>, CO and NMHC could decrease the tropospheric OH concentration over large region. Depending on geographical distribution, high variability of tropospheric OH was observed in a number of studies. Region with enhancement of NO<sub>x</sub> and O<sub>3</sub> will tend to increase OH concentration while enhancement of CO and CH<sub>4</sub> will decrease OH concentration in the troposphere (Wang and Jacob, 1998). Reduction of OH concentration due to the enhancement of CH<sub>4</sub> was also observed and found to play important role in affecting the tropospheric oxidizing capacity in comparison with changes in stratospheric ozone and climate changes (Morgenstern *et al.*, 2013).

The effect of climate change on tropospheric OH concentration is relatively smaller than the CH<sub>4</sub> and highly uncertain (Naik *et al.*, 2013). A number of studies on climate-chemistry interaction using the climate-chemistry models (CCMs) have demonstrated the feedback of this interaction on tropospheric hydroxyl radical, OH variation (Thompson, 1992; Johnson *et al.*, 1999; Murazaki and Hess, 2006; Doherty *et al.*, 2013). By using a Lagrangian chemistry-transport model (STOCHEM) with prediction between years 1990 - 2075 and climate response alone of temperature changes between 2.5 and 3.5°C with fixed emission forcing, the global OH was increased by 14% (Johnson *et al.*, 1999). In another study by using MOZART (Model of Ozone and Other Related Chemical Tracers), the tropospheric OH concentration was increased by 3.5% during summer season with the increased of both surface temperature and water vapor (Murazaki and Hess, 2006). Meanwhile, with the increase of water vapor alone of 19%, the OH concentration was increased by 6% in the end of the century (Doherty *et al.*, 2013). In understanding the spatial variability of OH, considering the dependency on a variety of physical and chemical processes in the atmosphere is somewhat challenging. For this reason, modeling tools have been widely used to investigate the spatial variability of OH and its driver.

In this paper, investigation was focused on tropospheric OH spatial distribution variability in response to climate change over Malaysia region. This study has selected A2 emission scenario for the regional climate model and the climate data output was used as input to the tropospheric chemistry model to investigate the impact of climate change on tropospheric OH concentration in urban, suburban, and remote areas in Malaysia.

### **Methodology:**

#### **Regional climate model:**

The domain for the regional climate study is the Malaysian region (comprises of Peninsular Malaysia and East Malaysia) and covering parts of Indonesia (Kalimatan, Borneo) with coordinate of N 0°00' to N 8°00' and E 100°00' to E 120°00'. The regional climate model, PRECIS (Providing Regional Climate for Impact Studies) version 1.9.3 (Jones *et al.*, 2004), was used to simulate the future climate (2090 – 2100) of the region based on A2 emission scenario. The A2 scenario describes a very heterogeneous world (Thompson *et al.*, 1989). It has been described as a self-reliance and preservation of local identities with fertility patterns across regions that converge very slowly, resulting in a continuously increasing global population. Economic development is primarily seen as regionally-oriented and per capita economic growth and technological change are more fragmented and slower than in others scenarios. Boundary data/model input ECHAM4 was used in this study. An ECHAM4 climate model has been developed from the ECMWF atmospheric model (European Centre for Medium Range Weather Forecast) (Roegner *et al.*, 1996) as an evolutionary series of the original from the spectral weather prediction model (Simmons *et al.*, 1989).

#### **Chemistry model:**

The climate output generated from the regional climate model was used as input to the tropospheric chemistry CiTTyCAT model (Cambridge Tropospheric Trajectory Model of Chemistry and Transport). The CiTTyCAT model is a box model which follows a 3D Lagrangian air parcel trajectory (Evans *et al.*, 2000). The model was used to simulate the tropospheric ozone in a number of specific locations throughout the study region. A box model is a simple and excellent tool that can be used to perform studies of chemical processes without interference from dynamical factors. The model framework consists of three main aspects (Wild, 1996): first, is a chemical scheme, which consists of the kinetic parameters for the chemical species of interest, production and loss terms for each of the chemical species; second, is the photolysis scheme which considers the effects of solar radiation on chemical species, mainly on the production of free radicals that play important roles in atmospheric chemistry; and third, is the physical scheme, which describes the non-chemical sources and sinks for chemical species such as emissions sources from surface, and loss terms due to dry and wet deposition. A detailed description of the model can also be found in a number of studies (Wild, 1996; Emmerson *et al.*, 2004; Sentian, 2009; Pugh *et al.*, 2012).

The existing chemical scheme was improved by adding the halocarbon chemical scheme. A total of 42 halocarbon species, 87 bimolecular reactions and 9 termolecular reactions were added into the model.

The model was used to investigate changes in tropospheric chemistry under specific conditions by picking up emissions, performing chemical reactions, and depositing some of the products of the reactions on the earth's surface. In this study, six locations were selected with different background status to investigate the tropospheric ozone response to projected climate change. The locations were as follows: Danum Valley, (Sabah Borneo: N 05°00' E 117°49.8'); Kota Kinabalu, (Sabah Borneo: N 05°53.6' E 116°02.6'); Kapit (Sarawak Borneo: N 02°00.4' E 112°56.2'); Sg. Petani, (Kedah-Peninsular Malaysia: N 05°37.9' E 100°28.2'); and Kuala Lumpur (Peninsular Malaysia: N 03°06.4' E 101°43.1'). The model was set to run for an 8-day model run at a 300-second time step.

## RESULTS AND DISCUSSION

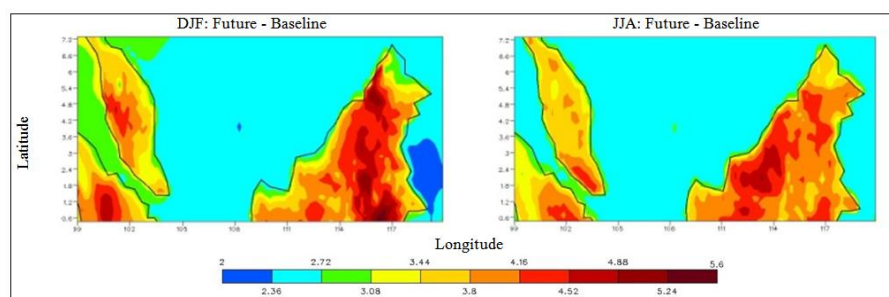
### *Regional climate change scenario in Malaysia:*

The projected surface temperature under A2 emission scenario over Malaysia region was about 31°C during winter season (Dec-Jan-Feb; DJF) and 32°C during summer season (Jun-Jul-Aug; JJA). Relative to the baseline scenario, the projected surface temperature increased by 3°C (10%) in both seasons (Table 2). The highest change of surface temperature over the domain was observed in Southwest of Sabah and some areas of Sarawak and Kalimantan, Indonesia. The changes of surface temperature were relatively higher in land areas compared to sea areas of the Malaysia domain (Fig. 1). In term of total precipitation, the projected precipitation in both seasons was found to increase by about 0.2 mm/day (12%) during winter season (DJF) and 0.3 mm/day (22%) during summer season (JJA) (Table 1). The highest changes in precipitation over the Malaysia region were observed during the summer season (JJA) in some areas of Kalimantan, Indonesia.

### *Response of climate change on tropospheric OH:*

In response to the regional climate change, the variability of future tropospheric OH concentration was found in the lowest OH concentration in remote and rural areas (Danum Valley and Kapit) and highest over urban areas of Sg. Petani during January and Kota Kinabalu during July. Danum Valley and Kapit, both in remote/rural areas and relatively unpolluted regions, have shown a decrease in future tropospheric OH concentration of between 0.03 and  $0.21 \times 10^6$  molecule  $\text{cm}^{-1}$  (21.4 – 51.9%) in both seasons in response to the climate changes in the range. Another study by Thompson (1992) by using 1D model has found that there was decrease of tropospheric OH by 0.3% per year in the 21<sup>st</sup> century relative to year 1985. Meanwhile, in more urban environment such as Kuala Lumpur, Sg. Petani and Kota Kinabalu, OH concentration changes were found variable.

During wet season (DJF), the tropospheric OH concentration was increased by  $0.06 \times 10^6$  molecule  $\text{cm}^{-1}$  (Kuala Lumpur) and  $0.1 \times 10^6$  molecule  $\text{cm}^{-1}$  (Sg. Petani). Meanwhile, during summer season (JJA) the increase of tropospheric OH was only found in Sg. Petani with changes of  $0.71 \times 10^6$  molecule  $\text{cm}^{-1}$  (14. %). The increase of the OH concentration in the future was found to be in agreement of other investigations on the impact of climate change on tropospheric OH (Johnson *et al.*, 1999; Murazaki and Hess, 2006; Doherty *et al.*, 2013). In the same study (Murazaki and Hess, 2006; Doherty *et al.*, 2013), the increase of surface temperature and precipitation can lead to increase in OH concentration. However, the changes of the OH concentration in the future due to the climate change are highly uncertain (Naik *et al.*, 2013). Other factors such as depletion of stratospheric ozone, global cloud cover, ultraviolet radiation, and the increase of GHGs emission may affects the OH concentration in the troposphere (Thompson *et al.*, 1992; Morgenstern *et al.*, 2013).



**Fig. 1:** Surface temperature ( $^{\circ}\text{C}$ ) changes: Future A2 – Baseline A2 (Note: DJF-Dec-Jan-Feb; JJA-Jun-Jul-Aug).

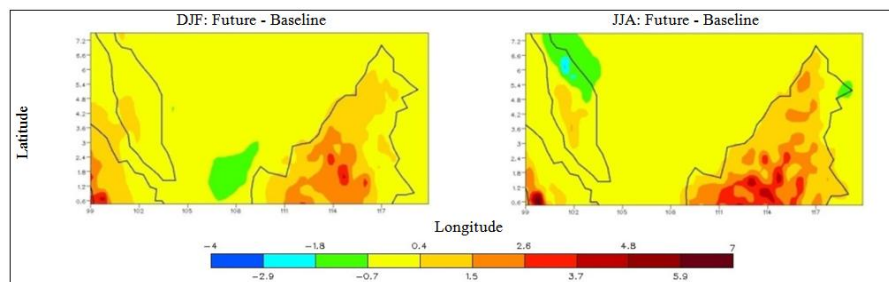
### *Conclusions:*

Climate change is prominent under the A2 scenario over the Malaysian region, where warmer atmospheric conditions and more precipitation were projected, as surface temperature and total

precipitation increased more than 10% and 12% respectively in both seasons. Under warmer and wet atmospheric conditions, the impact on future tropospheric OH concentration over Malaysia was

high variable. These results also suggest that, apart from climate change, the OH concentration variability of the region is another factor that equally

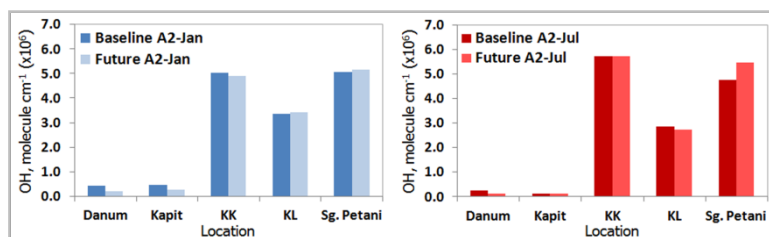
important to consider, such as future anthropogenic and GHGs emissions, depletion of stratospheric ozone and global cloud cover.



**Fig. 2:** Precipitation changes (mm/day): Future A2 – Baseline A2 (Note: DJF-Dec-Jan-Feb; JJA-Jun-Jul-Aug).

**Table 1:** Climate changes in the 21<sup>st</sup> century under A2 emission scenario.

| Parameter                           | Season       |              |
|-------------------------------------|--------------|--------------|
|                                     | DJF (Wet)    | JJA (Dry)    |
| Temperature, °C                     | 3.05 (10.8%) | 3.04 (10.6%) |
| Precipitation, mm day <sup>-1</sup> | 0.17 (11.5%) | 0.29 (21.8%) |



**Fig. 3:** Changes of tropospheric ozone in response to climate changes during wet (January) season and dry (July) season.

**Table 2:** Changes and percentages of changes of OH concentrations (molecule cm<sup>-1</sup>) in end of the century relative to baseline during January.

| Locations                                 | January (Wet Season)          |                             |                  |
|-------------------------------------------|-------------------------------|-----------------------------|------------------|
|                                           | Baseline A2 ( $\times 10^6$ ) | Future A2 ( $\times 10^6$ ) | Differences      |
| Danum<br>(N 05°00' E117°49.8')            | 0.42                          | 0.21                        | -0.21<br>(50.0%) |
| Kapit<br>(N 02°00.4' E 112°56.2')         | 0.46                          | 0.26                        | -0.20<br>(43.5%) |
| Kota Kinabalu<br>(N 05°53.6' E 116°02.6') | 5.03                          | 4.89                        | -0.14<br>(2.8%)  |
| Kuala Lumpur<br>(N 03°06.4' E 101°43.1')  | 3.36                          | 3.42                        | 0.06<br>(0.2%)   |
| Sg. Petani<br>(N 05°37.9' E 100°28.2')    | 5.07                          | 5.17                        | 0.1<br>(2.0%)    |

**Table 3:** Changes and percentages of changes of OH concentrations (molecule cm<sup>-1</sup>) in end of the century relative to baseline during July.

| Locations                                 | July (Dry Season)             |                             |                  |
|-------------------------------------------|-------------------------------|-----------------------------|------------------|
|                                           | Baseline A2 ( $\times 10^6$ ) | Future A2 ( $\times 10^6$ ) | Differences      |
| Danum<br>(N 05°00' E117°49.8')            | 0.27                          | 0.13                        | -0.14<br>(51.9%) |
| Kapit<br>(N 02°00.4' E 112°56.2')         | 0.14                          | 0.11                        | -0.03<br>(21.4%) |
| Kota Kinabalu<br>(N 05°53.6' E 116°02.6') | 5.73                          | 5.72                        | -0.01<br>(0.2%)  |
| Kuala Lumpur<br>(N 03°06.4' E 101°43.1')  | 2.87                          | 2.73                        | -0.14<br>(4.9%)  |
| Sg. Petani<br>(N 05°37.9' E 100°28.2')    | 4.76                          | 5.47                        | 0.71<br>(14.9%)  |

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