Assessment of aerosol direct effects on PM$_{2.5}$ and O$_3$ air quality in Continental Southeast Asia

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Abstract. An online coupled modeling system composed of Weather Research and Forecasting (WRF) model and Community Multiscale Air Quality (CMAQ) model was applied to assess aerosol direct effects on air quality with the focus on fine particulate matter (PM$_{2.5}$) and ozone (O$_3$) in Continental Southeast Asia where has suffered significant air pollution recently due to rapid development. The results showed that, in four focused countries including Cambodia, Laos, Thailand, and Vietnam, the direct effects moderately decreased shortwave radiation, temperature, planetary boundary layer (PBL) height, and wind speed by -9.08%, -0.44°C, -10.27%, and -2.21% in dry season, and -2.37%, -0.04°C, -2.05%, and -0.57% in wet season, respectively. Consequently, PM$_{2.5}$ concentration was found to increase by +10.51% in dry season and +1.44% in wet season. O$_3$ concentration was decreased by -2.76% in dry season while slightly increased by +0.56% in wet season. The increasing effect of aerosols on PM$_{2.5}$ concentration was caused by the more stable atmospheric condition. The increase or decrease in O$_3$ concentration depended on the responses of atmospheric dynamics as well as photolysis rates of photochemical reactions to direct effects.

1. Introduction

Environmental management cannot keep pace with continuously growths of economic and population at a rapid rate, thus it results in significant air pollution in Southeast Asia. Air quality in Continental Southeast Asia varies widely over space and time. In rural areas, biomass burning is the most dominant source of air pollutants followed by vehicle emission, whereas, in urban areas, vehicle and industrial emissions are the most important sources. During the dry season, from November to April, agricultural biomass burning and forest fire frequently cause severe particulate pollution not only in the local areas but also across the whole region and beyond [1]. Adverse air pollution has undesirable effects on human health and ecosystems. The most severe health effects can be attributed to fine particulate matter (PM$_{2.5}$) and ozone (O$_3$) [2]. Hence reduction of O$_3$ and PM$_{2.5}$ concentrations has become one of the key objectives of air quality control in the region.

In order to understand sources, transport, and chemical transformation of air pollutants for further air quality control, scientists have widely used air quality models. The air quality model traditionally used is called offline model which is decoupled treatments of meteorology and chemistry. Recently, online model which allows to facilitate feedbacks between chemistry and meteorology [3] has been developed rapidly and considered as a very promising way for future atmospheric simulation. In line with this trend, United States Environmental Protection Agency (US EPA) coupled the Weather Research and Forecasting (WRF) model and the Community Multiscale Air Quality (CMAQ) model to create a two-way online coupled modeling system. The essence of this interaction is focused on the direct radiative effects of scattering and absorbing aerosol in the troposphere that in turn affect
radiation calculations in the meteorological model, WRF [4]. This development not only helps to more realistically simulate the atmosphere but also gives a chance in investigating aerosol direct effects on the air quality. Aerosol direct effects can cause increase or decrease in air pollutant concentrations through change in meteorological condition. Assessment of aerosol impacts is thus important for supporting the development of pollution control strategies. In addition, this topic is interesting since aerosol direct effects on air quality is far from being well understood in Continental Southeast Asia. Therefore, this paper aimed to evaluate the impacts of aerosol direct effects on meteorology and air quality in Continental Southeast Asia targeted on four countries, namely Cambodia, Laos, Thailand and Vietnam. Focused pollutants are PM$_{2.5}$ and O$_3$. To fulfill the objectives, WRF and CMAQ models were coupled in both one-way and two-way online for the whole historical year 2014.

2. Methodology

2.1. Modelling framework and simulation design

Modeling domain together with locations of observation sites is presented in Figure 1. The focused domain covered almost part of continental region of Southeast Asia. Four countries was targeted including Cambodia, Laos, Thailand and Vietnam. The domain had a horizontal resolution of 24 km x 24 km, and 98 grid cells in each direction.

The online coupled WRF-CMAQ modeling system was applied for air quality simulations in the study region. The datasets used for the modeling system included geological data from USGS (United States Geological Survey); RTG_SST_HR (real time global high resolution sea surface temperature), and FNL (final analysis) meteorological data which were provided by the United States National Centers for Environmental Prediction (NCEP); chemical initial and boundary condition data produced by the global chemistry transport model, MOZART (Model Ozone and Related Chemical Tracer); HTAP (Hemispheric Transport of Air Pollution), FINN (Fire Inventory from National Center for Atmospheric Research), and Aerocom (Aerosol Comparisons between Observations and Models) emission databases, and biogenic volatile organic compounds (BVOCs) emission data outputted from MEGAN (Model of Emissions of Gases and Aerosols from Nature).

![Figure 1. Modeling domain and locations of observation sites.](image)

To capture aerosol direct effects, two simulations were designed: one-way online simulation (without feedback from CMAQ to WRF) and two-way online simulation (with feedback from CMAQ to WRF) for entire historical year 2014. Since the feedback in two-way simulation was implemented to represent aerosol direct effects, their impacts on meteorology and air quality were calculated as the differences between the results of two-way online simulation and one-way online simulation, and the contribution percentages of direct effects were calculated as the ratio of these differences to the one-way simulation results.

2.2. Model evaluation

Model performance was evaluated by statistical calculation of model results against observations. Hourly observed radiation, temperature, wind speed, pressure, and relative humidity were gathered from 7 stations located in Vietnam. Hourly observed PM$_{2.5}$ and O$_3$ concentrations were gathered from 10 stations, 6 stations located in Vietnam, and 4 stations located in Thailand (Figure 1). Beside traditional statistical metrics using correlation coefficient (R), mean bias (B$_{MB}$), mean absolute gross error (E$_{MAGE}$), this study was also followed the evaluation method proposed by Yu et al. (2006) using
normalized mean bias factor ($B_{NMBF}$) and normalized mean absolute error factor ($E_{NMAEF}$) [5].

3. Results and discussion

3.1. Model performance against observation

In general, the coupled WRF-CMAQ model showed a good performance. For meteorological simulation, modeled temperature, radiation, humidity, and pressure were highly consistent with observations as shown by R values (Table 1). The model overestimated the observation by the factor of 1.43 (two-way online simulation) for radiation. No significant overestimation or underestimation was found for temperature, humidity and pressure. For wind speed, its correlation with observation was lower than those of the other variables and the model overestimated the observation by the factor of 1.74 (two-way online simulation). For air quality simulation, the correlation coefficients for $PM_{2.5}$ and $O_3$ were acceptable. However, overestimations were found in model calculation for the two pollutants. The model overestimated $PM_{2.5}$ concentration by the factor of 1.26 (two-way online simulation) and $O_3$ concentration by the factor of 1.57 (two-way online simulation).

Table 1. Statistical evaluation for model performance.

| Parameter       | n    | $\bar{Y}$ | Run type | $\bar{M}$ | R    | $B_{MB}$ | $E_{MAGE}$ | $B_{NMBF}$ | $E_{NMAEF}$ |
|-----------------|------|-----------|----------|-----------|------|----------|------------|------------|-------------|
| Temperature ($^\circ$C) | 42471 | 25.59     | One-way  | 24.35     | 0.92 | -1.24    | 2.07       |            |             |
| Radiation (W/m$^2$)     | 42471 | 111.15    | One-way  | 174.61    | 0.76 | 63.46    | 94.66      | 0.57       | 0.85        |
| Humidity (%)            | 42471 | 79.46     | One-way  | 80.00     | 0.62 | 0.54     | 9.51       | 0.01       | 0.12        |
| Pressure (mBar)          | 42471 | 1009.48   | One-way  | 1006.99   | 0.88 | -2.49    | 3.79       | 0.00       | 0.00        |
| Wind speed (m/s)        | 2920  | 1.39      | One-way  | 2.50      | 0.48 | 1.12     | 1.40       | 0.81       | 1.01        |
| $PM_{2.5}$ ($\mu g/m^3$) | 59259 | 29.82     | One-way  | 35.40     | 0.57 | 5.58     | 20.81      | 0.19       | 0.70        |
| $O_3$ (ppb)             | 50246 | 21.57     | One-way  | 34.24     | 0.54 | 12.67    | 17.23      | 0.59       | 0.80        |

3.2. Meteorology response to aerosol direct effects

Under the aerosol direct effects, ground-level shortwave radiation, temperature, PBL height, and wind speed showed decreasing trends of mean values (Figure 2). The reduction in these meteorological variables were -5.99%, -0.24°C, -7.02%, and -1.43% on average, and in the range of -15.38% to -1.31%, -0.67°C to +0.05°C, -21.03% to +0.67%, and -6.96% to +1.69%, respectively, for the whole year in four focused countries. In dry season (from November to April), these contribution were -9.08%, -0.44°C, -10.27%, and -2.21% on average and in the range of -25.87% to -0.76%, -1.25°C to +0.05°C, -36.80% to +2.84%, and -11.89% to +2.86%, respectively. In wet season (from May to October), they were -2.37%, -0.04°C, -2.05%, and -0.57% on average and in the range of -8.36% to +2.52%, -0.25°C to +0.12°C, -9.66% to +3.64%, and -3.44% to +1.23%, respectively. These results showed that the direct effects were larger in dry season than those in wet season. The reason was that $PM_{2.5}$ concentration in dry season was higher than that in wet season. Among four countries, Northern Laos suffered the highest effects and Cambodia suffered the lowest effects from aerosols, there had no much difference in the effects between Thailand and Vietnam.
3.3. **PM$_{2.5}$ response to aerosol direct effects**

The spatial distributions of contribution percentages of aerosol direct effects to ground-level PM$_{2.5}$ concentration are shown in Figure 3. Over four focused countries, aerosols mostly produced increasing effects on PM$_{2.5}$ concentrations with contribution percentage of +8.56% on average and in the range from -4.75% to +44.74% for the whole year. In both dry season and wet season, PM$_{2.5}$ concentrations were increased with the percentage of +10.51% on average and in the range from -5.50% to +52.08%, and +1.44% on average and in the range from -5.15% to +9.82%, respectively. Similarly to meteorology, the effects on PM$_{2.5}$ were larger in dry season than those in wet season because of lower PM$_{2.5}$ loadings in wet season. The contribution percentages were high in Northern Laos in dry season where was found to have high PM$_{2.5}$ concentration. In contrast with other countries, in Cambodia where was found to have low PM$_{2.5}$ concentration, contribution percentage was negative with the
value of -0.05% in wet season. The average contribution percentages in Thailand and Vietnam were quite similar. The results indicated that the effects on PM$_{2.5}$ concentration were highly associated with PM$_{2.5}$ concentration level; the effects were large at high PM$_{2.5}$ periods and locations.

![Figure 3](image)

**Figure 3.** Spatial distributions of contribution percentages of aerosol direct effects to ground-level PM2.5 concentration.

|                  | Radiation, % | Temperature, °C | PBL height, % | Wind speed, % |
|------------------|--------------|-----------------|---------------|--------------|
| Whole year       | PM$_{2.5}$, %| -0.74           | -0.76         | -0.81        | -0.68        |
| Dry season       | PM$_{2.5}$, %| -0.72           | -0.79         | -0.81        | -0.69        |
| Wet season       | PM$_{2.5}$, %| -0.32           | -0.41         | -0.37        | -0.24        |

The increase in PM$_{2.5}$ concentration could be explained as followed. Shortwave radiation is scattered by aerosols, thus shortwave radiation arriving the ground surface is decreased, and then temperature near the ground surface is decreased. On the other hand, temperature in the atmosphere at high level is raised as aerosols absorbed shortwave radiation and heat is released. Thus, temperature inversion is enhanced that leads to increase in the atmospheric stability and decrease in PBL height. Therefore, the vertical dispersion of pollutants is reduced. Besides, the horizontal transportation is also reduced because of reducing in wind speed. As a result, PM$_{2.5}$ accumulates more and its concentration is increased. This explanation was confirmed by high negative correlations between the responses of PM$_{2.5}$ concentration and the responses of shortwave radiation, temperature, PBL height, and wind speed (Table 2). Note that, the correlation coefficients in wet season were low because in this season, PM$_{2.5}$ concentration was low, thus the direct effects were not significant, so that, the relationship could not be clearly observed.

**3.4. O$_3$ response to aerosol direct effects**

The spatial distributions of contribution percentages of aerosol direct effects to ground-level O$_3$ concentration are shown in Figure 4. Aerosol direct effects produced a slightly decreasing effect on O$_3$ concentration with the percentage of -1.51% on average and in the range from -3.86% to +1.47% for the whole year in four focused countries. In dry season, O$_3$ concentration was decreased with the percentage of -2.76% on average and in the range from -7.27% to +1.75% while, in wet season, O$_3$ concentration was slightly increased with percentage of +0.56% on average and in the range from -1.85% to +3.38%. There had not much difference in contribution percentages among four countries.
In order to explain the response of O$_3$ concentration to aerosol direct effects, both atmospheric dynamics as well as photolysis rates of photochemical reactions should be taken into account. First, change in atmospheric dynamics due to direct effects, more stabilized atmosphere and shallower PBL height as mentioned above, leads to increase in NO$_x$ concentration. In case of VOC-limited regime, the increase in NO$_x$ concentration enhances titration by NO as in equation (3) then causing a decrease in O$_3$ concentration. But in case of NO$_x$-limited regime, the increase in NO$_x$ concentration facilitates O$_3$ formation as in equations (1) and (2) then causing an increase in O$_3$ concentration. Second, reducing in photolysis rate due to the decrease in surface shortwave radiation caused by direct effects reduces the generation of O$_3$ as in equation (1) then causing a decrease in O$_3$ concentration [6]. Therefore, the overall response of O$_3$ concentration to direct effects results from the competition of two opposing effects, a decreasing effect and an increasing effect. In this study, it was found that, in four focused countries, the mean ratio of non-methane hydrocarbon to nitrogen oxides (NMHC/NO$_x$) in wet season, 71.35 (ppbC/ppb), was higher than that in dry season, 33.85 (ppbC/ppb). In addition, in wet season, the direct effects on shortwave radiation were less than those in dry season (Figure 2) that led to less reduction in photolysis rate compared to dry season. Thus, the increasing effect was slightly larger in wet season while the decreasing effect was larger in dry season.

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\begin{align*}
\text{NO}_2 + \text{hv} & \rightarrow \text{NO} + \text{O} \\
\text{O} + \text{O}_2 & \rightarrow \text{O}_3 \\
\text{NO} + \text{O}_3 & \rightarrow \text{NO}_2 + \text{O}_2
\end{align*}
\]

(1)  
(2)  
(3)

4. Conclusion
The coupled meteorology and atmospheric chemistry WRF-CMAQ model together with utilized data, in overall, was able to reproduce the observation acceptably. Aerosol direct effects were recognizable in the region, particularly, under high PM$_{2.5}$ pollution conditions. Dry season suffered larger impacts than wet season. Regarding meteorology, aerosol produced decreasing effects on shortwave radiation, temperature, PBL height, and wind speed. In term of air quality, the inclusion of aerosol direct effects resulted in an increase in PM$_{2.5}$ concentration and a decrease in O$_3$ concentration for the whole year. The effects on O$_3$ concentration were not significantly compared with those on PM$_{2.5}$ concentration. In closing, this application of the online coupled WRF-CMAQ modeling system demonstrated the model performance and presented a first using this system to study aerosol direct effects in the region. The results of this study can provide valuable information for developing effective pollution control strategies in the regions.

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