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## **Evaluation of the current state of modelled ozone, PM and deposition fields in the Pearl River Delta, China**

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### **Introduction**

The Pearl River Delta (PRD) region is one of the most developed regions in southern China. It is made up of three megacities regions (i.e., Guangzhou, Shenzhen and Hong Kong) and numerous small cities. It accounts for about 4% of the total population in China and contributes about 19% of China's gross domestic product in recent years. The large amount of energy consumption caused by rapid economic growth and intense human activity in PRD region gives rise to great pollutant emission and finally causes air quality deterioration. Based on recent air quality studies, we can generally draw a conclusion that it is a mixture of primary and secondary pollutions in PRD region (Chan and Yao, 2008). A recent series of field campaigns in the PRD region, namely PRIDE-PRD, organized by Peking University has helped us to gain insight and achieve a more specific understanding of air pollution and radiative process in this region (Zhang et al., 2008). An important conclusion, which can be drawn from these campaigns, is that the major pollutants, such as ozone (O<sub>3</sub>) and particulate matter (PM) in the PRD region often co-exist as a regional problem.

O<sub>3</sub>, as a secondary pollutant, has drawn intensive attention from many researchers. When studying O<sub>3</sub>, the short-time O<sub>3</sub> episodes are often concerned. From the previous studies in the PRD region, we have obtained some results on the meteorological conditions associated with O<sub>3</sub> episodes (Ding et al., 2004; Huang et al., 2005), the chemical sensitivities of O<sub>3</sub> to precursors (Wang et al., 2005; Zhang et al., 2008; Wang et al., 2010) and the relative importance of different atmospheric processes to ozone concentrations (Wang et al., 2010; Lam et al., 2005). Relatively fewer studies have aimed at explaining long-term ozone budget and transport characteristics and chemical production rate in the PRD region. On the other hand, as another critical pollutant in this region, PM has not been studied intensively in the PRD region, except at Hong Kong. The key issues associated with PM over PRD include the concentration of carbonaceous species, the formation of secondary particulate matters, the hygroscopic properties of particulate matters and the optical properties of particulate matters (Cheng et al., 2006; Hagler et al., 2006; Huang et al., 2006; Zhang et al., 2008).

The previous studies on O<sub>3</sub> or PM in PRD often focused on a few episodic days or a certain typical pollution seasons. In this report, we summarize the modelling results on O<sub>3</sub> and PM for a whole year to discern the seasonal variations and inter-annual variations of air pollution in PRD.

## Modeling method

We use CMAQ (version 4.5.1) with the State Air Pollution Research Centre version 99 (SAPRC-99) chemical mechanism to simulate the distribution of O<sub>3</sub> and PM<sub>10</sub> and the deposition fields of sulphate and nitrate in the Pearl River Delta region in south China during the whole year of 2006. The modelling results of Jan, Apr, Jul and Oct are shown to represent the seasonal variation.

The modelling domain with a horizontal grid spacing of 36km covers the entire China (Figure 1a). The grid has 13 vertical layers from the surface to an altitude of 17km above the ground, with 7 layers below 1000m and the surface layer of about 19m. The results covering the PRD region are extracted to represent the current state of air quality in PRD.

The fifth-generation Pennsylvania State University/National Centre for Atmospheric Research Mesoscale Model (MM5 version 3.7) is used to simulate the meteorological fields to drive CMAQ. The Intex-B emission inventory (Zhang et al., 2009) with 0.5°×0.5° resolution is used for the modelling domain as anthropogenic emission and the non-agricultural open fire emission is provided by Peking University (Song et al., 2009). As for the biogenic NO<sub>x</sub> and VOC, the Biogenic Emissions Inventory System version 3.09 (BEIS 3.09, Vukovich and Pierce, 2002) with a Chinese plantation survey dataset (Pearl River Delta environmental protection planning committee, 2006) is used to estimate the emission amounts.

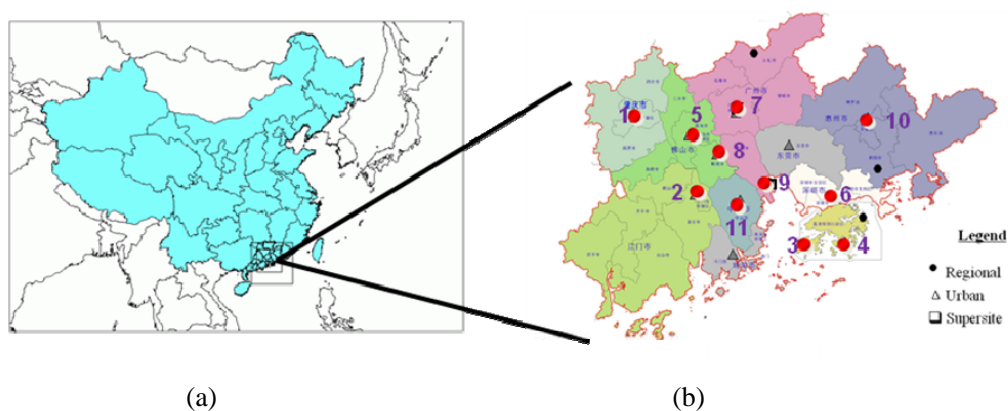


Figure 1. (a) the 36 km domain for CMAQ simulation; (b) the 11 sites which are marked by red dots are used to evaluate the model performances: 1-Chengzhong, 2-Donghu, 3-TungCung, 4-TseunWan, 5-Huijingcheng, 6-Liyuan, 7- Luhu, 8-Shunde, 9-Wanqingsha, 10-Xiapu, 11-Zimaling.

Figure 1b shows the location of monitoring sites in PRD, the observational data in these 11 sites are used to evaluate the model performances with three statistical measures, i.e., correlation coefficient (COR), normalized mean bias (NMB) and normalized mean error (NME).

### Evaluation and seasonal variations of modelled O<sub>3</sub> and PM

The average time series of the observations and modelling results at the above 11 sites for the pollutants O<sub>3</sub> and PM<sub>10</sub> are presented in Figure 2 to examine the overall model performances. As for O<sub>3</sub>, the CMAQ model successfully captured the day-to-day variation for the year 2006 with a COR of 0.76 (see Figure 2a). It can also be seen that for seasonal variation, there are significant spring peak, autumn peak and summer trough for O<sub>3</sub> in PRD. In spring, CMAQ shows the worst performance with a remarkable overprediction of O<sub>3</sub>, especially in March. As for PM<sub>10</sub>, Figure 2b clearly demonstrates that the CMAQ model successfully captured the day-to-day variation of PM<sub>10</sub> for the year 2006 with a COR of 0.72. For the whole year, the average concentration of PM<sub>10</sub> is underestimated by 21% with a maximum bias in spring. The CMAQ model successfully captured the values of PM<sub>10</sub> in winter and autumn, which were the most polluted seasons.

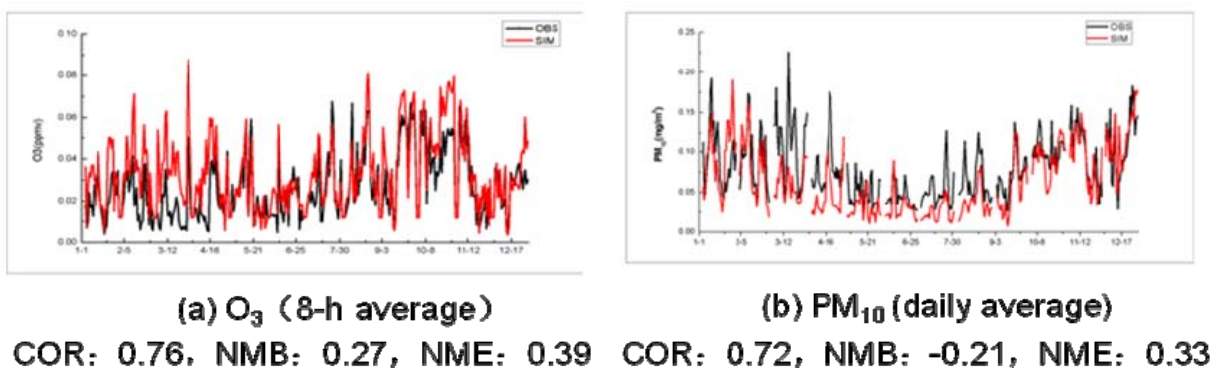


Figure 2. Time series of (a) 8-h O<sub>3</sub> and (b) daily PM<sub>10</sub> averaged at the 11 PRD sites for the whole year 2006

When taking seasonal variation into consideration, we take December, January and February as winter season (DJF), March, April, May as spring (MAM), June, July, August as summer (JJA) and September, October and November as autumn (SON).

Figure 3 presents the seasonal variation of surface O<sub>3</sub> concentration in the PRD region. It can be seen that in all seasons, there exist lower concentrations in the central PRD region than in its surrounding areas. In winter, most of the PRD region shows low O<sub>3</sub> due to low temperature and weak solar radiation. In spring, first O<sub>3</sub> peak in a year occurs due to increasing temperature and

strengthening radiation intensity. In summer, the O<sub>3</sub> presents a summer trough along the invasion pathway of the East Asia Monsoon because of the great monsoon precipitation caused by the inflow of clean marine air masses. In autumn, relatively dry weather and great downward flow cause the highest O<sub>3</sub> concentration in a year.

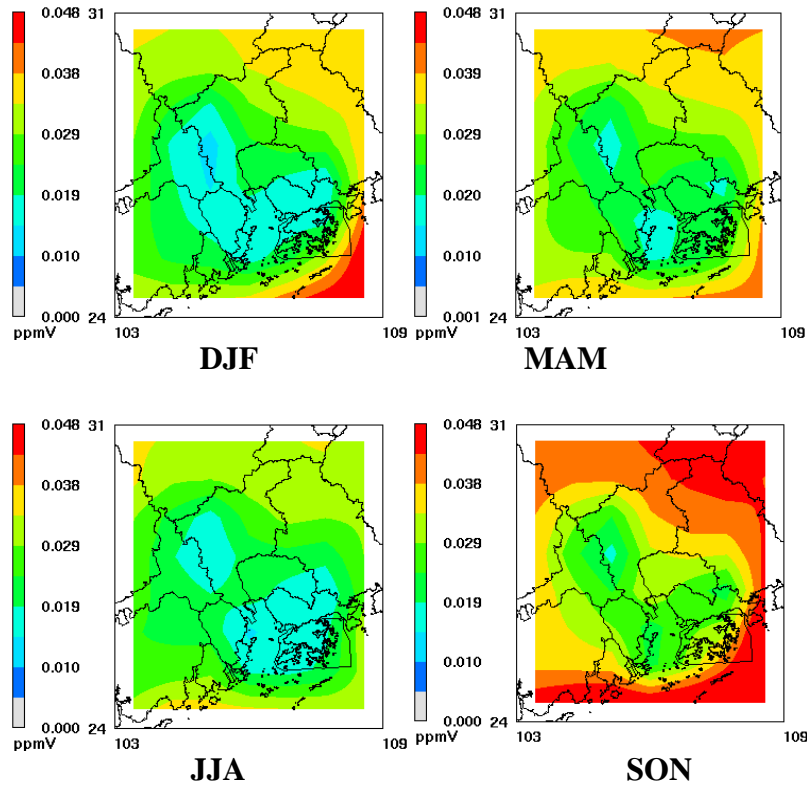


Figure 3 Seasonal variation and spatial distribution of surface O<sub>3</sub> concentration in the PRD region

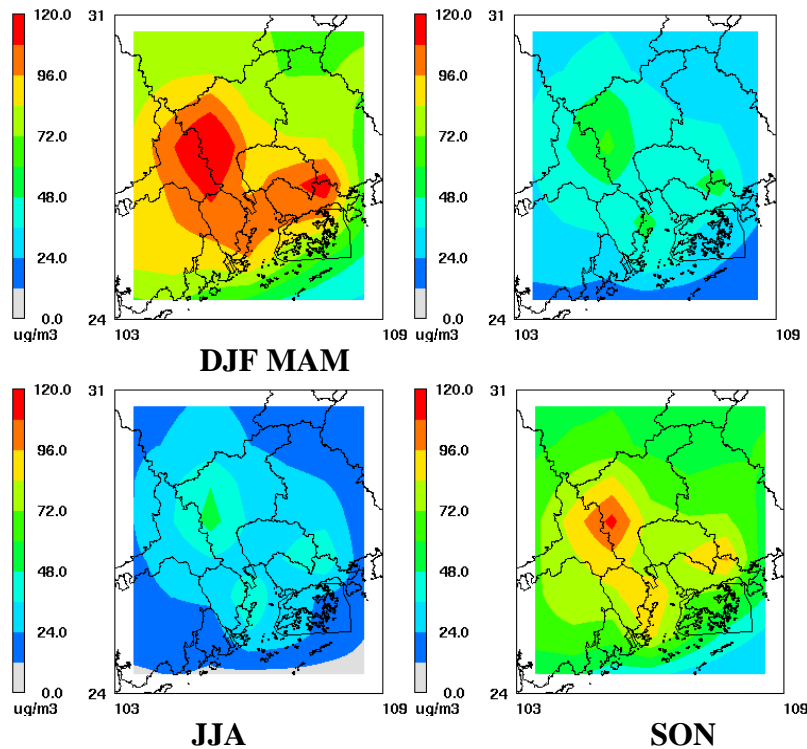


Figure 4. Seasonal variation and spatial distribution of surface PM<sub>10</sub> concentration in the PRD region

Figure 4 presents the seasonal variations of surface  $PM_{10}$  concentration in PRD region. It can be seen that in PRD region, the distribution of  $PM_{10}$  shows great seasonal variation and in contrary to  $O_3$ , there exist higher concentrations in the central PRD region than in its surrounding areas. In winter, the  $PM_{10}$  concentrations are at the highest level due to greater emission from anthropogenic sources, relatively low PBL and not enough precipitation to wash out the particulate matters. In spring, with the advent of the rainy season, the  $PM_{10}$  concentration declines, but we can also find relatively high levels in the highly emitted areas such as Guangzhou and Foshan. In summer, the invasion of East Asia monsoon causes the inflow of clean marine air masses together with much precipitation, resulting in the lowest  $PM_{10}$  concentration throughout the year. In autumn, the relatively less rainfall and low wind speed favour the accumulation of particulate matters, and as a result, the  $PM_{10}$  concentration stays at a high level as well.

## Seasonal variation of deposition fields in PRD

### SO<sub>2</sub>

Figure 5 presents the season variation of SO<sub>2</sub> dry deposition field in the PRD region. The results show that the seasonal variation of dry deposition fields of SO<sub>2</sub> is not very significant except that in winter. Higher dry deposition could be found along the border of Guangzhou and Foshan. In winter, high dry deposition occurs in the Pearl River Estuary region and over the sea, which might be attributed to the strong north winds resulting in relatively high SO<sub>2</sub> concentration in southern PRD areas.

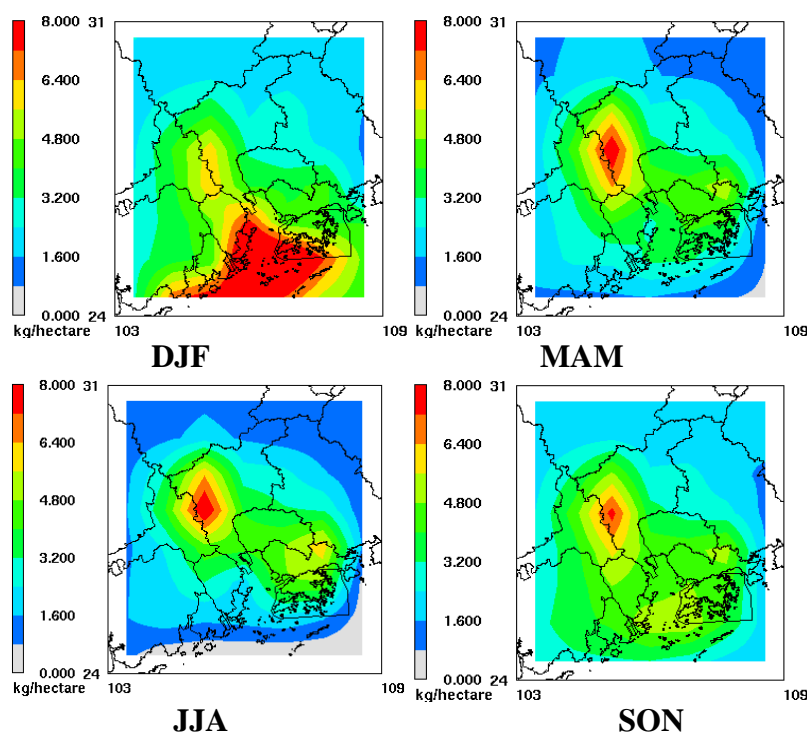


Figure5. Seasonal variation of SO<sub>2</sub> dry deposition in PRD

Figure 6 presents the seasonal variation of SO<sub>2</sub> wet deposition in PRD. The results show spring/summer maximum and winter/autumn minimum of SO<sub>2</sub> wet deposition field. The intensity of SO<sub>2</sub> wet deposition correlates well with its ambient concentration and the amount of rainfall. In winter, weak wet deposition is due to relatively low rainfall in despite of high ambient SO<sub>2</sub> levels. In spring, the highest wet deposition is found due to increasing rainfall. In summer, though the rainfall intensity is strong, the SO<sub>2</sub> levels are lower than those in the spring time. As a result, the wet deposition values are lower in summer than those in spring. In autumn, this dry season finally causes low wet deposition intensity.

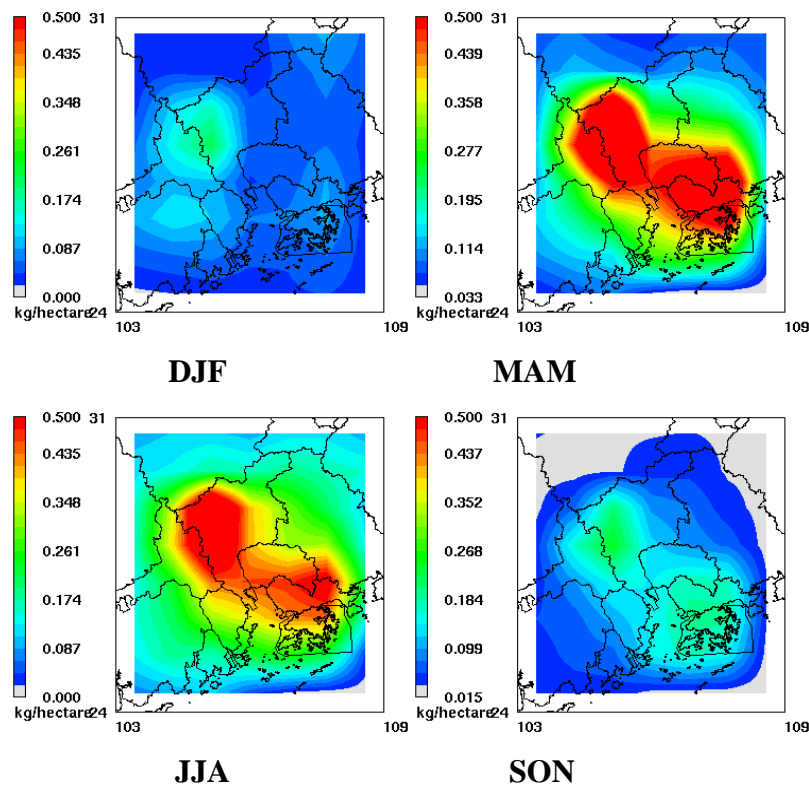


Figure6. Seasonal variation of SO<sub>2</sub> wet deposition in PRD

## NO<sub>2</sub>

Figure 7 presents the seasonal variation of NO<sub>2</sub> deposition, which is dominated by dry deposition. The seasons with most intensified deposition of NO<sub>2</sub> are summer and autumn. The deposition field correlates with the ambient concentration and the downward dry deposition velocity. In winter, low temperature and relatively weak turbulence result in long residential time of NO<sub>2</sub> in the ambient air, therefore, relatively weak deposition is obtained. In summer, the concentration of NO<sub>2</sub> is relatively low but greater turbulence lead to shorter residential time of NO<sub>2</sub> in the air, resulting in higher deposition. In autumn, higher NO<sub>2</sub> concentration and great downward flow cause maximum deposition of NO<sub>2</sub> throughout the year.



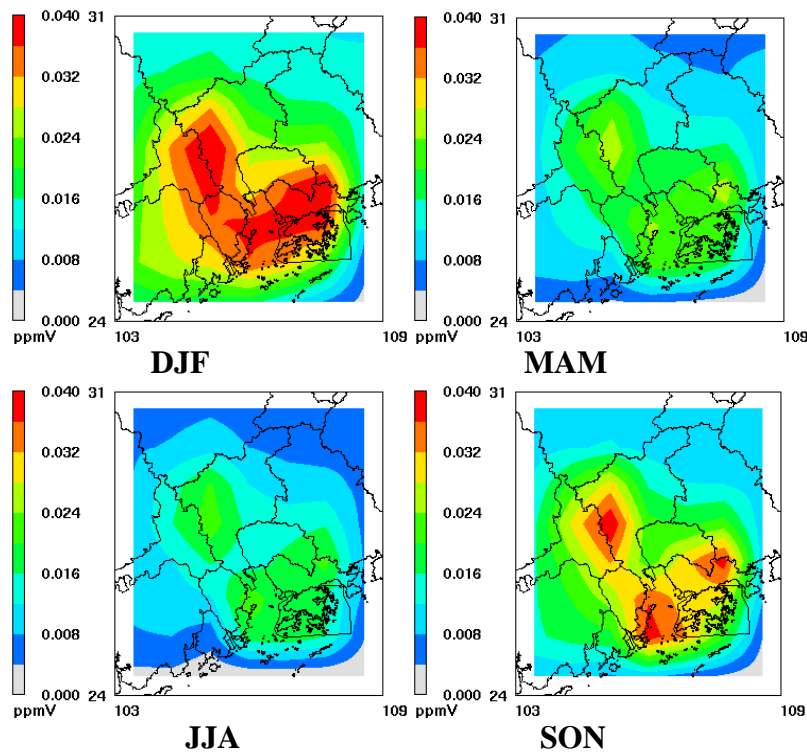


Figure7. Seasonal variation of NO<sub>2</sub> deposition fields in PRD

### O<sub>3</sub>

Figure 8 presents the seasonal variation of O<sub>3</sub> deposition field in PRD. Due to the wet deposition flux is of several magnitudes of orders smaller than the dry deposition field, only the total deposition fields are showed. In winter, relatively low O<sub>3</sub> concentration and relatively long O<sub>3</sub> residential time cause the lowest deposition in the year. In spring, the increasing deposition flux is found due to higher O<sub>3</sub> concentration and short residential time. In summer, although Asia monsoons give rise to the lowest O<sub>3</sub> concentration, strong turbulence and the shortest residential time result in the highest deposition in one year. In autumn, the highest O<sub>3</sub> concentration and the significant downward flow result in a higher deposition field.

### PM<sub>10</sub>

Figure 9 is the season variation of PM<sub>10</sub> dry deposition field in PRD. It shows no significant seasonal variations. High values are often found in Guangzhou, Foshan and Hong Kong area. This pattern is similar to that of SO<sub>2</sub> dry deposition in the PRD region.

Figure10 presents the season variation of PM<sub>10</sub> wet deposition field in PRD. It shows significant seasonal variations. The ambient concentration and the precipitation amount or the cloud pro-

cess would affect the wet deposition flux greatly. Lower wet deposition flux in winter is due to little rainfall though the ambient  $PM_{10}$  concentration is high. In spring, the increasing wet deposition is mainly due to the increasing rainfall. In summer, wet marine air gives rise to high precipitation, which causes the highest wet deposition in the year. In autumn, though decreasing rainfall happens, the high ambient concentration also results in high wet deposition field. Compared to dry deposition, wet deposition is a more effective way to clean the particulate matter in the atmosphere.

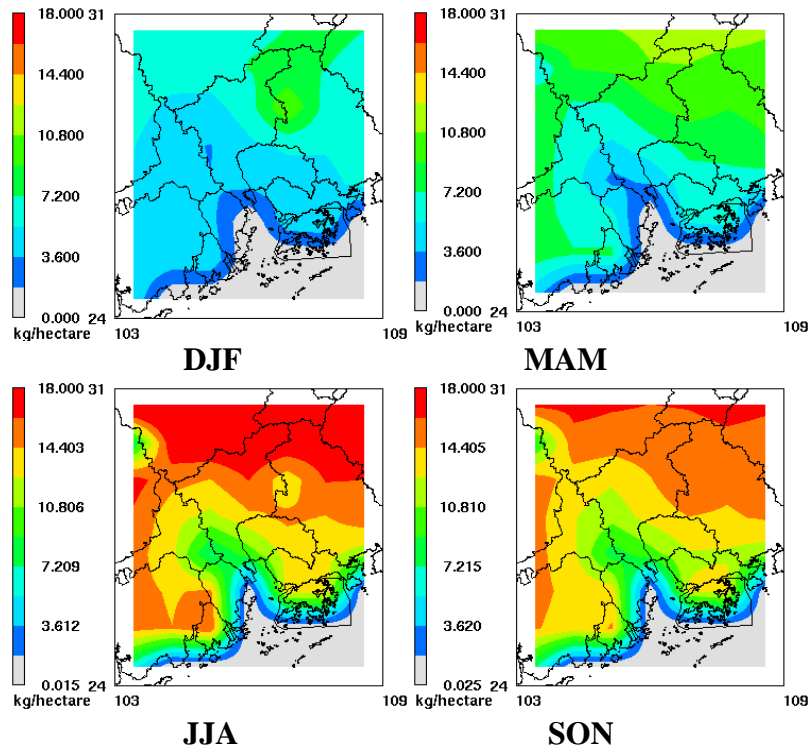


Figure 8. Seasonal variation of  $O_3$  deposition fields in PRD

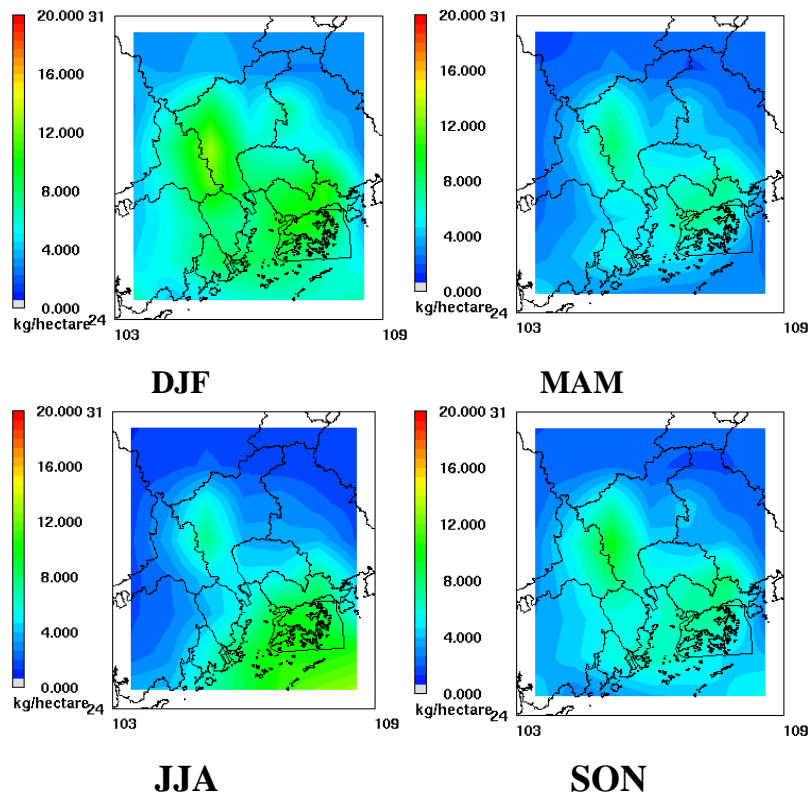


Figure 9. Seasonal variation of PM<sub>10</sub> dry deposition flux in PRD

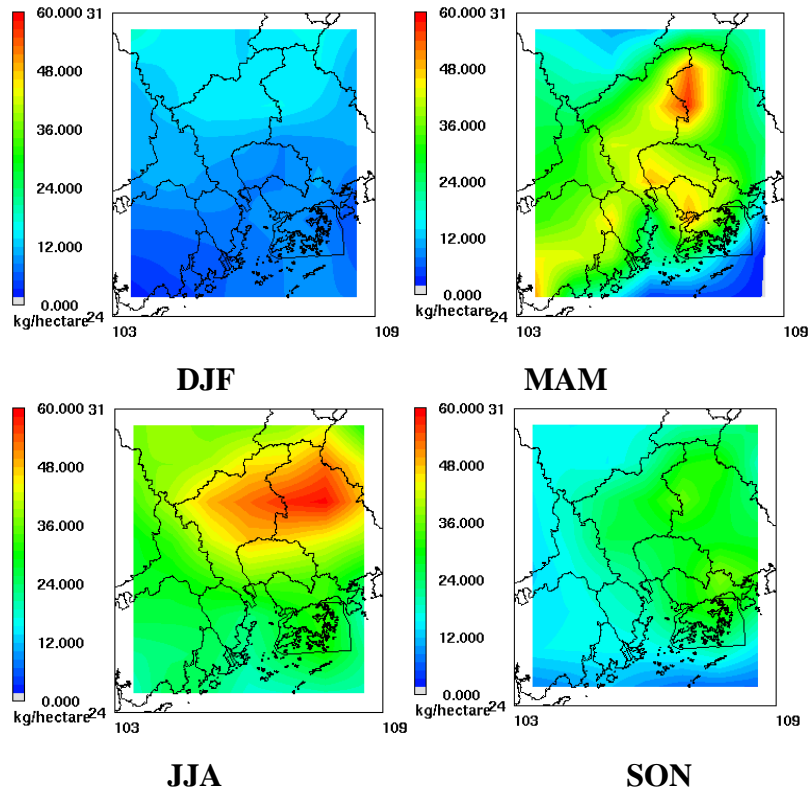


Figure10. Seasonal variation of PM<sub>10</sub> wet deposition flux in PRD

We also examined the seasonal variation of the dry deposition fields of sulphate and nitrate in PRD. The dry deposition of sulfate shows less significant seasonal variation except in July. High temperature and high water vapors are favourable to the formation of sulfate, so a high sulfate value

can be expected in July. However, more sulfate is removed from the atmosphere by wet deposition, so the dry deposition flux remains low in July. In other months, higher dry deposition can be found in Shenzhen and Hong Kong, which may imply that the atmospheric environment there can accelerate the formation of sulfate. As for nitrate, more significant seasonal variation of dry deposition can be found. Higher dry deposition is obtained in January and October than that in July and October.

Compared to dry deposition, wet deposition is a more effective way to remove the particulate matters from the atmosphere, with higher fluxes in all months. The wet deposition shows significant seasonal variation. It is in a great correlation with the particulate matter concentrations and the amount of precipitation. In April and July, both sulfate and nitrate show great wet deposition flux in PRD due to more rainfall. In January, low precipitation causes less wet deposition. October has also fewer rainy days as in January over the PRD region; however, the ambient particulate matter shows high concentrations in October, resulting in higher wet deposition than that in January.

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