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The regional budget of O_3 mass and concentration variations within the atmospheric boundary layer using the CMAQ model: An example from the Pearl River Delta, China

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Tropospheric O_3 pollution notably contributes to the deterioration of air quality in many metropolitan regions, resulting in detrimental effects on human health and ecosystem. Due to the moderate atmospheric lifetimes of O_3, horizontal transport, exchange between atmospheric boundary layer (ABL) and free troposphere (FT), and chemical process within the ABL all potentially play important roles in regional O_3 pollution. In this study, we developed a post-calculation tool to quantify the hourly contributions of these processes to the regional budget of O_3 mass and concentration variations within the ABL based on the modelling results of the Community Multiscale Air Quality (CMAQ) model. The new features of this tool include: (1) the contributions of ABL-FT exchange on O_3 pollution can be quantified; (2) horizontally, the targeted region can be freely defined by users and vertically, the volumes are non-fixed owing to the diurnal variations of ABL; and (3) the budgets of O_3 mass and concentration variations are separately calculated and analysed. The Pearl River Delta (PRD) region, located in the South China and faced with severe O_3 pollution, was selected as the target region in this study. Results show that the variations of total O_3 mass within the ABL of the PRD were controlled by ABL-FT exchange, that is, the increase (decrease) of O_3 mass in the morning (afternoon) was driven by O_3 inflow (outflow) through ABL-FT exchange. By contrast, it was the chemical process that drove the variations of regional-mean O_3 concentrations. Except that ABL-FT exchange contributed to the rise of O_3 concentrations in several hours after sunrise, O_3 transport did not lead to the notable variation of O_3 concentration in the remaining hours of the day. Combining source apportionment methods, we found that outside O_3 (including O_3 produced by emissions within the East and Central China and background O_3) entered the PRD mainly through ABL-FT exchange. For chemical process, local sources played a major part, but the contributions of outside emissions cannot be neglected, suggesting the contributions of precursor transport. The effects of typhoon periphery, the weather system most related to O_3 pollution in the PRD, were also examined by comparing the budget results on O_3 pollution days with and without the occurrence of typhoons. The usage of this tool will help to comprehensively understand the influence of transport and chemical process in O_3 pollution on

the regional scale, which is crucial for effective and strategic O_3 control.

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