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Supplement of

Rethinking the role of transport and photochemistry in regional ozone pollution: insights from ozone concentration and mass budgets

Kun Qu et al.

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Three texts, nine figures and two tables are included in this Supporting Information for the paper entitled “Rethinking the role of transport and photochemistry in regional ozone pollution: Insights from ozone mass and concentration budgets”.

**Texts:**
- Text S1 describes the detailed processes of O\textsubscript{3} budget calculations in this study.
- Text S2 compares the equations of O\textsubscript{3} concentration budget calculations used in this study with these in 1-D models (Eq. (1) in the manuscript).
- Text S3 presents the results of model validation of atmospheric boundary layer (ABL) height, wind and O\textsubscript{3} mixing profiles based on the IAGOS dataset.

**Figures:**
- Figure S1 indicates two calculation paths for the regional O\textsubscript{3} concentration budget within an hour.
- Figure S2 shows the comparison between IAGOS and modelling atmospheric boundary layer height in Hong Kong during the daytime of Oct. 2015.
- Figure S3 compares IAGOS and modelling wind roses in three height ranges (0-1 km, 1-2 km and 2-5 km) in Hong Kong during the two representative months.
- Figure S4 compares IAGOS and CMAQ modelling vertical profiles of O\textsubscript{3} mixing ratios in Hong Kong during the two representative months.
- Figure S5 presents the spatial distributions of 18 sites of the Guangdong-Hong Kong-Macao Pearl River Delta (PRD) Regional Air Quality Monitoring Network.
- Figure S6 compared the mean diurnal changes of O\textsubscript{3} concentrations in the PRD from three sources: observational near-ground O\textsubscript{3} concentrations, modelled near-ground O\textsubscript{3} concentrations and ABL-mean O\textsubscript{3} concentrations.
- Figure S7 displays the spatial distributions of mean contributions of vertical exchange through the ABL top due to advection perpendicular to the ABL top and its slope (ABLex-A) to O\textsubscript{3} mass changes in the morning and afternoon of two representative months.
- Figure S8 is the flow diagram of the O\textsubscript{3} budget calculation processes.
- Figure S9 is the flow diagram of the O\textsubscript{3} budget calculation in Step I (or the post-processing tool flux\textunderscore 4d\textunderscore cal).

**Tables:**
- Table S1 gives more detailed information on the O\textsubscript{3} polluted days of the PRD in the two representative months.
- Table S2 lists the formulas in the O\textsubscript{3} budget calculations, the parameters used and their corresponding source files in the flux\textunderscore 4d\textunderscore cal tool.
Text S1. Detailed processes of O\textsubscript{3} budget calculations

As the flow diagram shown in Fig. S8, there are two steps in the calculations of O\textsubscript{3} budgets based on the WRF-CMAQ modelling results:

1) **Step I: Quantifications of process contributions to O\textsubscript{3} mass and volume changes**

The post-processing tool `flux_4d_cal` was developed using FORTRAN90 for this step. For all grid columns in d02 except for these adjacent to the boundaries of the modelling domain, the following contents are calculated by the tool:

- Hourly contributions of horizontal transport to O\textsubscript{3} mass changes within the ABL, including these in the x- and y-directions;
- Hourly contributions of vertical exchange due to the changes of ABL height (ABLex-H) to O\textsubscript{3} mass changes within the ABL;
- Hourly contributions of vertical exchange due to advection perpendicular to the ABL top and its slope (ABLex-A) to O\textsubscript{3} mass changes within the ABL, including these in the x-, y- and z-directions;
- Hourly contributions of other processes (gas-phase chemistry, cloud process and dry deposition) to O\textsubscript{3} mass changes within the ABL;
- Hourly transported air volumes by each transport process;
- Total O\textsubscript{3} masses within the ABL at both the start and end of each hour;
- ABL heights at the starting and end hours.

All of the above values can be found in the NetCDF (nc) output files, and they are used in the Step II calculations.

To finish the calculations of Step I, several input files are needed:

- Meteorological files processed by the MCIP module in CMAQ from the WRF outputs, which include the METCRO2D (meteorological parameters in the 2-D space), METCRO3D (meteorological parameters in the 3-D space) and MERDOT3D (wind speeds in the 3-D space) files;
- Pollutant concentration output files (CONC files) modelled by CMAQ, where hourly instantaneous O\textsubscript{3} concentrations are stored;
- Process Analysis (PA) output files modelled by CMAQ, where the hourly, nested contributions of gas-phase chemistry, cloud process and dry deposition to O\textsubscript{3} concentration are stored.

For most of the files used here, the setting of spatial domains and times should be consistent; otherwise, the calculations would not be performed or generate wrong results. Additionally, users should provide the resolution of the modelling domain and the indexes of contributions by three non-transport O\textsubscript{3} processes in the PA files for further calculations.
The flow chart of the calculation in $flux_{\ 4d\ _cal}$ is shown in Fig. S9. The calculation formulas for the grid column $(i, j)$, parameters used and their source files are summarized in Table S2. There are four loops in the calculations, which are the loops of x-, y-grids, time steps and vertical layers. We assume that there are 60 time-steps within an hour, and parameters at each time step can be interpolated linearly by their values at the starting and end hours. The hourly contribution of non-transport processes to O$_3$ in a grid cell is divided equally to these within each time step. For every layer within the ABL, contributions to O$_3$ mass changes and volumes related to horizontal transport and non-transport processes are calculated and summed up to derive the total contributions in the ABL column. For layers where the ABL top is located, besides these aforementioned parameters, contributions to O$_3$ mass changes and volumes related to vertical exchange (ABLex-H and ABLex-A) are also calculated. Besides, total O$_3$ masses within the ABL at the start and end of each hour are also calculated, and ABL heights at the starting and end hours can be read from the METCRO2D files.

The height of night-time stable ABL can be severely underestimated by normally used ABL parameterization, especially when the Richardson number is used (Dai et al., 2014). To reduce the influence of imprecise ABL heights in the O$_3$ budget calculations, here, we set the lowest ABL height limit as 350 m for all hours, which is an approximate value close to the values reported by night-time observations in summer or autumn in the Pearl River Delta (PRD) (Chan et al., 2006; Fan et al., 2011; He et al., 2021; Song et al., 2021). The results of the budget conservation examination (Fig. 3 in the manuscript) also suggest that the choice of this value is acceptable. Further studies are surely needed to determine this value better. However, we focus on the causes of daytime ozone pollution; thus, night-time budgets do not notably influence the conclusions of this study.

2) Step II: Regional O$_3$ budget calculations and conservation examinations

This step aims to: 1) calculate the hourly O$_3$ mass and concentration budgets within the ABL of the user-defined regions and 2) check whether the conservation between the changes of O$_3$ masses/concentrations modelled by CMAQ and the net contributions of O$_3$-related processes calculated above can be achieved. Besides the nc file generated in Step I, the definition of the grids in user-defined targeted regions, including the border grids and non-border grids, should also be provided by the users. Any software with basic data analysis and nc-file processing functions (Python, MATLAB, R, etc.) can be applied for this step.

The contents in this step include:

- Calculation of hourly contributions of horizontal transport to O$_3$ mass changes through each user-defined border grid. For horizontal transport through one type of border, its contributions in every interfaces between the border grids of this type and the outside regions, in both x- and y-directions, are summed up as the total contributions of the process in the regional-level O$_3$ mass budget.
• Calculation of hourly contributions of vertical exchange through the ABL top and other non-transport processes to O₃ mass changes. For one process, its contributions in all grids within the targeted regions are summed up as the total contributions of the process in the regional-level O₃ mass budget.

• Calculation of the hourly O₃ concentration budget (the contributions of O₃-related processes to the hourly variations of mean O₃ concentrations over the ABL of the targeted region) based on the contributions of O₃-related processes to O₃ mass changes and the volumes or volumes changes linked to the processes.

More details on the calculation of the O₃ concentration budget are introduced as follows. As displayed in Fig. S1, within an hour, the mean O₃ concentration within the ABL of the targeted region changes from c₀ to c₁. During daytime, O₃ mass and ABL volume both change notably, making it difficult to quantify the contributions to O₃ concentration variations by various O₃-related processes. To simplify the calculation, two calculation paths (shown as the red arrow lines in Fig. S1; c₁ and c₂ are the reference O₃ concentrations separately for two calculation paths) are used in the calculations, assuming that only O₃ mass or ABL volume change in each step of two paths. For the path “c₀ → c₁ → c₁”, the first step is the ABL volume change step, with O₃ concentration change described as:

$$c_{r1} - c_0 = c_0 \times \left( \frac{\sum H_0}{\sum H_1} - 1 \right) \tag{S1}$$

where H₀ and H₁ are the ABL heights at the starting and end hours. It is counted as part of the contributions by ABLEex-H. The second step is the O₃ concentration change step, with O₃ concentration change described as:

$$c_1 - c_{r1} = \frac{\sum (F_{htrans} - c_{r1} \times \Delta V_{htrans})}{L^2 \times \sum H_1} + \frac{\sum (F_{ABLex-A} - c_{r1} \times \Delta V_{ABLex-A})}{L^2 \times \sum H_1} + \frac{F_{ABLex-H}}{L^2 \times \sum H_1}$$

$$+ \frac{F_{chem}}{L^2 \times \sum H_1} + \frac{F_{cloud}}{L^2 \times \sum H_1} + \frac{F_{ddep}}{L^2 \times \sum H_1} \tag{S2}$$

where F_{htrans}, F_{ABLex-A}, F_{ABLex-H}, F_{chem}, F_{cloud} and F_{ddep} indicate the contributions of horizontal transport, ABLEex-A, ABLEex-H, gas-phase chemistry, cloud process and dry deposition, respectively, to O₃ mass changes; ΔV_{htrans} and ΔV_{ABLex-A} are the volumes of transported air parcels attributed to horizontal transport and ABLEex-A, respectively, within an hour; L denotes the length of the grid cell, or the horizontal resolution of the model. The six terms on the right-hand side of Eq. (S2) are separately classified as the contributions of horizontal transport, ABLEex-A, ABLEex-H, gas-phase chemistry, cloud process and dry deposition in the O₃ concentration budgets. Note that the contributions of ABLEex-H are separately calculated in two steps, and they are summed up as the total contribution of ABLEex-H in the O₃ concentration budget. Similarly, for the path “c₀ → c₂ → c₁”, the changes in O₃ concentration in two steps can be described as:

$$c_{r2} - c_0 = \frac{\sum (F_{htrans} - c_0 \times \Delta V_{htrans})}{L^2 \times \sum H_0} + \frac{\sum (F_{ABLex-A} - c_0 \times \Delta V_{ABLex-A})}{L^2 \times \sum H_0} + \frac{F_{ABLex-H}}{L^2 \times \sum H_0}$$

$$+ \frac{F_{chem}}{L^2 \times \sum H_0} + \frac{F_{cloud}}{L^2 \times \sum H_0} + \frac{F_{ddep}}{L^2 \times \sum H_0} \tag{S3}$$
\[ c_1 - c_{r2} = c_{r2} \times \left( \frac{\sum H_0}{\sum H_1} - 1 \right) \] (S4)

The contributions of various O₃-related processes are classified correspondingly. The final result of individual contribution is estimated as the average value of the contributions calculated based on two calculation paths.
Text S2. Comparisons of the O\textsubscript{3} concentration budget calculations between this study and 1-D models

When the region column in the Chemical Transport Models (CTMs) is thin enough to resemble a line, transport contributions in the O\textsubscript{3} concentration budget calculations based on the CTMs results (Eqs. (6-7) in the manuscript) are expected to have the same forms as those in 1-D models, or Eqs. (1) and (3) in the manuscript:

\[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} = -\bar{u} \frac{\partial \langle c_{O_3} \rangle}{\partial x} - \bar{v} \frac{\partial \langle c_{O_3} \rangle}{\partial y} - \frac{\partial \overline{c_{O_3} w'}}{\partial z} + S(O_3) \]  

\[ - \frac{\partial \overline{c_{O_3} w'}}{\partial z} = \frac{\Delta c_{O_3}}{H} \frac{\partial H}{\partial t} + \frac{\Delta c_{O_3}}{H} \left( u_h \frac{\partial H}{\partial x} + v_h \frac{\partial H}{\partial y} - w_h \right) \]  

where $\langle c_{O_3} \rangle$ is the mean O\textsubscript{3} concentration over the ABL of the studied region; $\bar{u}$ and $\bar{v}$ are the mean horizontal wind speeds in the x- and y-direction; $S(O_3)$ is the total contribution of non-transport processes to O\textsubscript{3} mass changes; $\Delta c_{O_3}$ is the difference of O\textsubscript{3} concentrations above and within the ABL; $H$ is the ABL height; $u_h$, $v_h$ and $w_h$ are the ABL-top wind speeds in the x, y and z-direction, respectively. Thus, we can use it to check the validity of the O\textsubscript{3} concentration budget calculations in this study.

Here the contributions of horizontal transport to the variations of $\langle c_{O_3} \rangle$ can be described as (Eq. (6) in the manuscript):

\[ \left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{htrans} = F_{htrans} + \langle c_{O_3} \rangle (V - dV) \cdot \frac{dV}{V} - \langle c \rangle = \frac{F_{htrans} - \langle c_{O_3} \rangle dV}{V} \]  

(S5)

where $F_{htrans}$ is the contributions of horizontal transport to O\textsubscript{3} mass changes; $V$ is the original volume of the PRD grids below the ABL; $dV$ is the volume of transported parcels. Assume that the length of the region in the x-direction is $dx$, thus,

\[ V = S \, dx \]  

(S6)

where $S$ is the area of the interface. As calculated in the O\textsubscript{3} mass budget, in the unit time,

\[ F_{htrans} = \langle c_{O_3} \rangle_{trans} uL \]  

(S7)

\[ dV = \bar{u} S \]  

(S8)

where $\langle c_{O_3} \rangle_{trans}$ is the O\textsubscript{3} concentration in the transported air parcels, and. Therefore, from Eqs. (S5)-(S8), we can get:

\[ \left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{htrans} = \bar{u} \frac{\langle c_{O_3} \rangle_{trans} - \langle c_{O_3} \rangle}{dx} = -\bar{u} \frac{d\langle c_{O_3} \rangle}{dx} \]  

(S9)

which is the same as the first term on the right side of Eq. (1) in the manuscript. Similarly, the contribution of horizontal transport in the y-direction can be expressed as the second term on the right side of Eq. (1) in the manuscript.

For ABLE\textsubscript{H}, its contributions when $V$ is much higher than $dV$ (this assumption can be normally met when the period is short) are:

\[ \left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{ABLE-H} = F_{ABLE-H} + \langle c_{O_3} \rangle (V + dV) - \langle c_{O_3} \rangle = \frac{F_{ABLE-H} - \langle c_{O_3} \rangle dV}{V} \]  

(S10)
where $F_{\text{ABLex-H}}$ is the O$_3$ mass change attributed to ABLex-H. In the unit time,

$$F_{\text{ABLex-H}} = c_h \frac{\partial H}{\partial t} L^2$$  \hspace{1cm} (S11)

$$dV = \frac{\partial H}{\partial t} L^2$$  \hspace{1cm} (S12)

$$V = HL^2$$  \hspace{1cm} (S13)

where $c_h$ is the O$_3$ concentration above the ABL; $L$ is the width of the grid cell (equal to the horizontal resolution of the model). Therefore, from Eqs. (S10)-(S13),

$$\left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{\text{ABLex-H}} = c_h - \langle c_{O_3} \rangle \frac{\partial H}{\partial t} = \frac{\Delta c_{O_3}}{H} \frac{\partial H}{\partial t}$$  \hspace{1cm} (S14)

where $\Delta c_{O_3}$ is the difference of O$_3$ concentrations above and within the ABL ($c_h - \langle c_{O_3} \rangle$). Apparently, it is the same as the first term on the right side of Eq. (3) in the manuscript.

For ABLex-A,

$$\left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{\text{ABLex-A}} = \frac{F_{\text{ABLex-A}} + \langle c_{O_3} \rangle (V - dV)}{V} - \langle c_{O_3} \rangle = \frac{F_{\text{ABLex-A}} - \langle c_{O_3} \rangle dV}{V}$$  \hspace{1cm} (S15)

$F_{\text{ABLex-M}}$ is the contributions of ABLex-A to O$_3$ mass change. In the unit time,

$$F_{\text{ABLex-A}} = c_h \left( u_h \frac{\partial H}{\partial x} + v_h \frac{\partial H}{\partial y} - w_h \right) L^2$$  \hspace{1cm} (S16)

$$dV = \left( u_h \frac{\partial H}{\partial x} + v_h \frac{\partial H}{\partial y} - w_h \right) L^2$$  \hspace{1cm} (S17)

$$V = HL^2$$  \hspace{1cm} (S18)

Therefore, from Eq. (S15-18),

$$\left[ \frac{\partial \langle c_{O_3} \rangle}{\partial t} \right]_{\text{ABLex-A}} = \frac{c_h - \langle c_{O_3} \rangle}{H} \left( u_h \frac{\partial H}{\partial x} + v_h \frac{\partial H}{\partial y} - w_h \right) = \frac{\Delta c_{O_3}}{H} \left( u_h \frac{\partial H}{\partial x} + v_h \frac{\partial H}{\partial y} - w_h \right)$$  \hspace{1cm} (S19)

It is the same as the second term on the right side of Eq. (3) in the manuscript.

Based on the above discussion, for the contributions of all transport processes considered in the O$_3$ budget calculations, the above formulas (Eqs. (S9), (S14) and (S19)) are the same as those used in 1-D models (Janssen and Pozzer, 2015; Vilà-Guerau de Arellano et al., 2015), suggesting their applicability in the quantification of the O$_3$ concentration budget using CTMs modelling results.
Text S3. Model validation of ABL height, wind and O₃ mixing ratio profiles based on the IAGOS dataset

IAGOS (In-service Aircraft of a Global Observing System; https://www.iagos.org) is a global aircraft-based observing system, where state-of-the-art instruments deployed in aircraft are used to measure reactive gases, greenhouse gases, aerosol and clouds in the troposphere and lower stratosphere (Petzold et al., 2016). Meteorological parameters, including air temperature, wind speed and direction, are also provided by IAGOS. When the aircraft climb up or descend, these measurements are suitable for obtaining the vertical profiles of parameters with high resolutions, which provides valuable observational datasets for the model validation in the vertical direction.

To ensure reasonable quantifications of the O₃ budgets, the IAGOS dataset in two representative months in Hong Kong (located in the south PRD) was used to evaluate the modelling performance of WRF-CMAQ in this study. We focused on comparing parameters within the height range of 0-5 km. Since observational data is often missing in some height ranges and the vertical resolution of modelling results is relatively low, for the temperatures, wind speeds and directions at different heights, we calculated their mean observational and modelling values within every 500 m height range (i.e., 0-500 m, 500-1000 m, etc.) as the data used in the comparisons. The detailed evaluations are introduced as follows:

(1) Atmospheric boundary layer (ABL) heights:
ABL height is an important parameter in the O₃ budget calculations — it is used in the quantification of the contributions from all O₃-related processes. Therefore, a good modelling performance of ABL height is important for accurately analyzing O₃ budgets. In this study, the observational ABL heights were determined using the profiles of potential temperature (θ) in IAGOS, and they are defined as the heights where the lapse rate of θ (\(\partial \theta / \partial z\), the rate of θ changing over height change) reaches its maximum values (Dai et al., 2014). Since there are limited profiles available in July 2016 and night-time ABL heights are hard to be determined accurately, we only evaluated the modelling performance of ABL heights during the daytime (6:00-18:00 Local Time (LT)) of Oct. 2015. As shown in Fig. S2, the mean bias (MB) between modelling and observational ABL heights in Hong Kong is only -1.1 m, and a good correlation between ABL heights from two datasets (\(R = 0.76\)) suggests that the mean diurnal cycles of ABL during daytime can be modelled well. Though the modelling performance of ABL heights is satisfying based on the IAGOS dataset in Hong Kong, more comprehensive comparisons based on three-dimensional observations with higher spatiotemporal resolutions and coverages are still required for more accurate O₃ budget estimates in future studies.

(2) Wind profiles:
Figure S3 shows the IAGOS and modelling wind roses within the height ranges of 0-1000 m, 1000-2000 m and 2000-5000 m. Both datasets indicate that higher wind speeds can be generally found at higher altitudes. In autumn, WRF overestimates wind speed below 1000 m by 0.6 m/s (16%) but underestimates it above 1000 m. In summer, the biases between wind speeds
in the two datasets are relatively smaller, especially at lower heights (< 2000 m). Both datasets show similar prevailing wind directions at different height ranges and seasons. Thus, the modelling performance of wind speeds and directions in the vertical direction is acceptable.

(3) O3 mixing ratio profiles:
The comparisons between observational and modelling profiles of the O3 mixing ratio are displayed in Fig. S4. Few O3 profiles were available in July 2016, and the useable ones were mostly measured during clean periods. Thus, the comparison was mainly based on the results in Oct. 2015 (the number of IAGOS O3 profiles available for the comparisons is 41). Both datasets show that the O3 mixing ratio decreases with height in Hong Kong. Below the height of 1000 m, the observational and modelling O3 mixing ratios are 71.4 ppbv and 75.8 ppbv, respectively. Within the height range of 1000-2000 m, the O3 mixing ratio is overestimated by 26%. High O3 levels during Oct. 13-24 and relatively low O3 levels in other periods can be found in both datasets, suggesting that the development, maintenance and dissipation of O3 pollution in this month were modelled well. Therefore, the performance of O3 profile modelling can also meet the requirement of O3 budget calculations.
**Figure S1.** Two calculation paths for the regional-level O₃ concentration budget within an hour. $m_\text{pollutant}$ indicates the total mass of pollutants in the atmospheric boundary layer (ABL) of the studied region; V is the volume of the ABL of the targeted region; L is the length of the grids (equal to the horizontal resolution of the model); H is the ABL heights; $t₀$ and $t₁$ are the starting and end hour, respectively; $c₀$ and $c₁$ are the concentrations of pollutants in $t₀$ and $t₁$, respectively; $c_r₁$ and $c_r₂$ are the reference concentrations of pollutants for two calculation paths.

**Figure S2.** Comparisons between IAGOS and modelling atmospheric boundary layer height in Hong Kong during the daytime of Oct. 2015. n, the number of the available data pairs for the comparison; MB, mean bias; R, correlation factor.
Figure S3. Comparisons between IAGOS and modelling wind roses in Hong Kong in (a) Oct. 2015 and (b) July 2016. Results within the height range of 0-1000 m, 1000-2000 m, and 2000-5000 m were separately displayed.
Figure S4. Comparisons between IAGOS and CMAQ modelling vertical profiles of O₃ mixing ratios (ppb) in Hong Kong in (a) Oct. 2015 and (b) July 2016. The heights of the atmospheric boundary layer (ABL) modelled by WRF in two representative months are shown as solid black lines.

Figure S6. Mean diurnal change of the hourly variations of observational, modelling mean near-ground O\textsubscript{3} concentrations in 18 sites of the Guangdong-Hong Kong-Macao regional monitoring network and modelling mean O\textsubscript{3} concentration over the atmospheric boundary layer (ABL) of the Pearl River Delta on the polluted days of autumn (Oct. 2015) and summer (July 2016). “mod” and “obs” are short for models and observations, respectively; R, correlation factor.

Figure S7. The spatial distributions of contributions of ABLex-A to O\textsubscript{3} mass changes on the polluted days of Oct. 2015. (a-b) Contributions through vertical advection; (c-d) contributions through horizontal advection. (a,c) The mean results during the morning hours (6:00-14:00 LT); (b,d) the mean results during the afternoon hours (14:00-19:00 LT).
Figure S8. Flow diagram of the O₃ budget calculation processes. ABL, atmospheric boundary layer; ABLex-H, vertical exchange through the ABL top due to the changes of ABL height; ABLex-A, vertical exchange through the ABL top due to advection perpendicular to the ABL top and its slope.

Figure S9. Flow diagram of the O₃ budget calculation in Step I (or the post-processing tool flux_4d_cal). NCOL, NROW and NLAY indicate the number of columns, rows and vertical layers in the modelling domain. ABL, atmospheric boundary layer. METCRO2D, 2-dimensional meteorological outputs from the MCIP module in CMAQ; METCRO3D, 3-dimensional meteorological outputs from the MCIP module in CMAQ; METDOT3D, 3-dimensional wind fields outputs from the MCIP module in CMAQ; CONC, 3-dimensional outputs of pollutant concentrations from CMAQ; PA, 3-dimensional outputs of hourly contributions by non-transport processes to O₃ from CMAQ.
Table S1. Information on the O₃ polluted days of the Pearl River Delta (PRD) in Oct. 2015 and July 2016. MDA1, the maximum 1-hr O₃ concentrations; MDA8, the maximum 8-hr average O₃ concentrations.

<table>
<thead>
<tr>
<th>Dates</th>
<th>Influencing Weather Systems</th>
<th>O₃ concentrations in the PRD (the maximum values in nine municipals of the PRD, released by the China National Environmental Monitoring Centre; µg/m³)</th>
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<td></td>
<td></td>
<td>MDA1</td>
</tr>
<tr>
<td>Oct.13, 2015</td>
<td></td>
<td>201</td>
</tr>
<tr>
<td>Oct.14, 2015</td>
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<td>301</td>
</tr>
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<td>July 22, 2016</td>
<td>Subtropical High</td>
<td>211</td>
</tr>
<tr>
<td>July 23, 2016</td>
<td></td>
<td>223</td>
</tr>
<tr>
<td>July 24, 2016</td>
<td></td>
<td>265</td>
</tr>
<tr>
<td>July 25, 2016</td>
<td>Subtropical High</td>
<td>334</td>
</tr>
<tr>
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<td></td>
<td>235</td>
</tr>
<tr>
<td>July 29, 2016</td>
<td></td>
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</tr>
<tr>
<td>July 30, 2016</td>
<td>Typhoon Nida</td>
<td>268</td>
</tr>
<tr>
<td>July 31, 2016</td>
<td></td>
<td>385</td>
</tr>
<tr>
<td>Source of Parameters</td>
<td>Parameters used</td>
<td>Formulas in the calculation of process contributions to mass changes for the grid cell</td>
</tr>
<tr>
<td>----------------------</td>
<td>----------------</td>
<td>----------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Other process</td>
<td>Other process</td>
<td>Formula: $\sum_{i} p_{i} \Delta v_{i}$, where $p_{i}$ are the process contributions and $\Delta v_{i}$ are the mass changes.</td>
</tr>
</tbody>
</table>

**Table 2.** Formulas in the calculations of process contributions to mass changes.
References (of the Supplement)


