Analysis of seasonal ozone budget and spring ozone latitudinal gradient variation in the boundary layer of the Asia-Pacific region

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HIGHLIGHTS
- Seasonal variation and latitudinal gradient of O3 in AP are reproduced in MOZART-4.
- Budget analysis confirms the dominant factor controlling BL O3 seasonal variation.
- Quantitative contributions to O3 latitude gradient are fixed by tagged method.

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ABSTRACT
The ozone (O3) budget in the boundary layer of the Asia-Pacific region (AP) was studied from 2001 to 2007 using the output of Model of Ozone and Related chemical Tracers, version 4 (MOZART-4). The model-simulated O3 data agree well with observed values. O3 budget analysis using the model output confirms that the dominant factor controlling seasonal variation of O3 differs by region. Photochemistry was found to play a critical role over Japan, the Korean Peninsula and Eastern China. Over the northwestern Pacific Ocean, advective flux was found to drive the seasonal variation of O3 concentrations. The large latitudinal gradient in O3 with a maximum of 52 ppbv over the marine boundary layer around 35°N during the spring was mainly due to chemistry; meanwhile, advection was found to weaken the gradient. The contribution of stratospheric O3 was ranked second (20%) to the local contribution (25%) in Japan and the Korean Peninsula near 35°N. The rate of O3 export from China’s boundary layer was the highest (approximately 30%) in low latitudes and decreased with increasing latitude, while the contribution of North America and Europe increased with increasing latitude, from 10% in lower latitudes to 24% in higher latitudes.

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1. Introduction

Tropospheric Ozone (O3) is affected by transport from the O3-rich stratosphere, photochemical production following oxidation of CO and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NOx) and by removal via photolysis, reaction with radicals, and deposition in the Earth’s surface (Wild, 2007). It is considered to be an air pollutant that causes damage to human health and natural ecosystems. O3 also acts as a direct greenhouse gas (Solomon et al., 2007), and it has been reported that tropospheric O3 levels observed over East Asia have been rising over the past decades, likely as a consequence of increasing emissions of NOx from Asia (Naja and Akimoto, 2004; Wang et al., 2009). The high O3 concentration over East Asia not only impacts the regional air quality but also increases the background O3 in the entire Northern Hemisphere due to regional and intercontinental transport, especially during the spring (Akimoto et al., 1996; Liu et al., 2003; Zhang et al., 2003).

Observations have shown that O3 reaches its maximum during spring and then drops to a minimum in the summer over the Asia-Pacific region and remote mid-latitude areas in the Northern Hemisphere (Wild and Akimoto, 2001; Pochanart et al., 1999, 2002, 2003; Yamaji et al., 2006, 2008). Moreover, a summer peak of O3 has been observed at some inland locations of East Asia (Zhu et al.,...
2004; Ding et al., 2008). Based on the present understanding, the spring O3 maximum could have resulted from comprehensive impacts of stratospheric intrusions (Carmichael et al., 1998; Langford, 1999; Levy et al., 1985), regional photochemical production (Penkett and Brice, 1986; Penkett et al., 1993), and intercontinental transport (Pochanart et al., 2002; Wild and Akimoto, 2001). While Kim and Lee (2010) noted that the stratospheric input was not the main source for the spring O3 maximum over East Asia and the Pacific Ocean, Zhang et al. (2012) suggested that the stratosphere contributed 5 ± 3 ppbv (12%) of the surface O3 over South China in spring based on model simulation. The summer minimum of O3 has been attributed to the incursion of Asian summer monsoon with lower-level air pollutants from the Northwest Pacific Ocean (Chan et al., 1998; Zbinden et al., 2006). Similar results were also reported for the South Asian monsoon region of India (David and Nair, 2011). The peak time of the spring maximum was shifted by a few months at different locations from early spring to early summer over the Asia-Pacific region (Chan et al., 1998; He et al., 2008), which may be caused by the intercontinental transport from the Asia Continent (Tanimoto et al., 2005) and the seasonal variation of stratosphere and troposphere exchange (STE).

Although many studies have given comprehensive discussion regarding the seasonal cycles of boundary layer O3 and the possible reasons for the peak values over East Asia, an unambiguous conclusion of the mechanisms that lead to the formation of O3 seasonal variation has not been drawn, and the spring maximum of O3 is still a fundamental issue in tropospheric chemistry research. The present study focuses on the impacts of physical and chemical processes on the seasonal variation and spring latitudinal gradient of boundary layer O3 over the Asia-Pacific region using output values from the 3D CTM, MOZART-4 (Model of Ozone and Related chemical Tracers, version 4). Recently, O3 budget has been intensively investigated using model outputs (Emmons et al., 2003; Li et al., 2007; Wild, 2007), and tagged tracer approaches have been used to discuss the roles of transport from various selected regions around the globe on O3 distribution (Wang et al., 1998a, 1998b, 1998c; Grewe, 2004; Sudo and Akimoto, 2007; Emmons et al., 2012). This study combines the quantitative analysis of the O3 budget with two tagged approaches, which were implemented with MOZART-4, to identify the main sources of the spring O3-latitude gradient. Lastly, the relative importance of physical and chemical processes on the O3 level is evaluated.

2. Data and model output

2.1. Data

Some of the monthly observational O3 data were taken from eight regional stations of the Acid Deposition Monitoring Network in East Asia (EANET) from 2001 to 2007: Cheju, Rishiri, Tappi, Sado-Seki, Oki, Yusuhara, Hedo, and Ogasawara. The descriptions of these sites can be found at http://www.eanet.asia/site/index.html. Others are from the WMO-World Data Centre for Greenhouse Gases (WDCGG, http://ds.data.jma.go.jp/gmd/wdcgg/) from 2001 to 2007, which includes the four sites of Ryori, Tae-Ahn Peninsula, Yonagunijima, and Minamitorishima. The monthly data for Beijing, Tianjin, Mt. Tai, Mt. Huang, and Hong Kong are from Zong et al. (2007), Yao et al. (2011), Li et al. (2007), and Wang et al. (2009), respectively. The O3 data at Lin’an are from Yang et al. (2008). The location information of all sites is provided in Figs. 1 and 2.

2.2. Model

MOZART is a global tropospheric CTM that has been extensively evaluated by comparing results with observations from ozonesonde, aircraft, and surface monitoring stations. The output data

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**Fig. 1.** Comparison of modeled O3 (ppbv) (dashed line) and observations (solid line) with their mean biases at selected sites over Japan and the Korean Peninsula during 2001–2007. All of the monthly data were calculated based on the mean of seven years’ data. R is the correlation coefficient (same below). The degree of freedom is 82, and the data passed the t-test at the 95% confidence level (p-value < 0.05).
have been shown to simulate the concentrations of tropospheric O₃ and its precursors reasonably well (Horowitz et al., 2003; Pfister et al., 2008a, 2008b; Emmons et al., 2010). In this study, we used MOZART-4, which is described and evaluated in great detail in Emmons et al. (2010). MOZART-4 was run with the standard chemical mechanism (see Emmons et al., 2010 for details) in this study. The Synoz (synthetic ozone) scheme of McLinden et al. (2000) was used as an upper boundary condition for O₃ flux in the stratosphere, yielding a cross-tropopause O₃ flux of 500 Tg/yr. This was driven by the NCEP/NCAR reanalysis meteorological field, having a horizontal resolution of approximately 2.8° x 2.8°, with 28 vertical levels from the surface to approximate 2 hPa. The initial condition and emission were based on NCAR Community Data Portal, which was introduced by Emmons et al. (2010). The model was run with a time step of 20 min from 1 Jan 2000 to 31 Dec 2007, and the first year was discarded as spin-up. It is noted that the modeled results used for analyses are based on the mean values in the boundary layer (the six lowest-lower layers in the model, surface to ~2 km) and that only the surface values were used for model evaluation.

2.3. Tagged experiments

Wu et al. (2009) showed that estimates of O₃ contributions are significantly different when estimated by removing all of the emissions in a source region, in comparison to making small perturbations in the emissions, which is due to the nonlinearity of O₃ photochemistry. To some extent, the tagged methods can decrease the error caused by nonlinearity. In this study, two tagged experiments (listed in Table 1) were performed to quantify the source of the latitudinal gradient in O₃ during the spring. To isolate the contributions from individual source regions, we used a tagged tracer method, which was introduced by Sudo and Akimoto (2007). The tagged method treats a chemical species emitted or chemically produced in a certain region as a separate tracer and calculates its transport, chemical transform and surface deposition. We classify O₃ by 10 source regions as shown in Table 1. The O₃S is an O₃ tracer of stratospheric origin (Sudo and Akimoto, 2007). Strato is the O₃S contribution rate. The rest of the air column is classified as REST.

Another tagged experiment quantified the source contribution from Eastern China. This experiment tagged NO emissions and the resulting products, following them to the production of O₃, as used by Emmons et al. (2012). The technique added synthetic tracers to the chemical mechanism that do not modify the original chemistry but make use of the mixing ratios and loss rates of the full, standard chemistry. Emmons et al. (2012) presented the details of the mechanism and illustrated the additive equalities of the technique and comparisons to other attribution techniques. A test was also shown by Emmons et al. (2012) to illustrate that the tagging method is sound, and it has been implemented without any errors. The simulations of tagged experiments were run from 1 Jan 2000 to 31 Dec 2007 with the first year as a spin-up simulation.

3. Results and discussion

3.1. Validation of simulated O₃

Figs. 1 and 2 show the comparisons of the modeled monthly O₃ with observations over Japan, the Korean Peninsula and Eastern China. Seasonal variations of O₃ were reproduced by the model. The discrepancies in summer O₃ may result from the significant effects of clouds and wet processes. The model also properly reproduces the different variability of O₃ at six sites over Eastern China (Fig. 2). Peak O₃ appears in May in Tianjin, Lin’an and Mt. Huang; in June in Beijing and Mt. Tai; and in autumn in Hong Kong. The O₃ values over Southeastern China (represented by Lin’an, Mt. Huang and

![Fig. 2. Comparison of the modeled O₃ (ppbv; dashed line) and observations (solid line) at selected sites over Eastern China. The degree of freedom is 10, and the data passed the t-test at the 95% confidence level (p value < 0.05).](image)
Hong Kong) are characterized by a double-peak pattern in the late spring and autumn and have a single peak in late spring or early summer in the north part of Eastern China (Tianjin and Beijing). The correlation coefficient (R) between the simulated and observed results ranges from approximately 0.61 to 0.96 over Japan and the Korean Peninsula and from 0.73 to 0.94 for the sites over Eastern China. All correlations passed the t-test under the 95% confidence level. The high correlation of 0.96 at four oceanic sites is primarily attributed to their remote locations with less direct impact of continental emissions. Most land sites at mid latitude may be affected by local anthropogenic emissions and complex topography, which may also be responsible for the relatively low correlation for mid-latitude land sites.

3.2. Seasonal cycles and budget of boundary-layer O₃ over the Asia-Pacific region

Based on the seasonal O₃ patterns discussed in Section 3.1, the NO emission rate in East Asia, and the classification of monitoring sites (He et al., 2008), we define four regions over the Asia-Pacific as shown in Fig. 3. Region I (18°–28°N, 122°–135°E) has a high correlation coefficient of 0.96 between observed and simulated O₃ and has a less direct effect of continental emissions. Region II (28°–46°N, 122°–144°E), including Japan and the Korean Peninsula, has correlation coefficients from 0.61 to 0.85 and is affected by local anthropogenic emissions. Region III (33°–43°N, 112°–122°E) and Region IV (22°–33°N, 112°–122°E) are located in Eastern China (divided by the latitude of 33°N) and have relatively great emissions of anthropogenic pollutants. The four regions are all affected by the Asian monsoon. We define two regions in Eastern China for the differences in pollutant emission rates and climate (Ding and Chan, 2005).

In the proceeding section, the controlling factors of O₃ concentration (transport, net photochemical production and deposition) in the four regions are discussed. Here, every term affecting the O₃ budget is evaluated quantitatively from the surface to approximately 2 km (boundary layer) in each region. The rate of change of O₃ can be expressed as:

\[
\frac{dO_3}{dt} = \text{Chem} + \text{Adv} + \text{Con} + \text{Dif} - \text{Dep}
\]

where Chem represents the net chemical production; Adv, Con and Dif are the transport fluxes of horizontal and vertical advection, convection and diffusion, respectively; and Dep is the dry deposition rate. Fig. 4 shows each of these components of the O₃ budget for the four regions: Net chemistry, Advection, Convection, Diffusion, and Dry deposition.

As shown in Fig. 4, O₃ has a February–March maximum and a summer minimum in the boundary layer over the study regions. This is similar to the results of Danielsen (1968) and Langford (1999). The seasonal variability of O₃ is inconsistent with that of O₃, except in Region I where the peaks of O₃ and O₃ (30% of O₃) are in the same period (early spring). This result implies that the seasonal cycle of O₃ is partly due to stratospheric downward transport, especially in Region I during the spring.

In Region I, O₃ exhibits an early-spring maximum (approximately 44 ppbv) and a summer minimum (approximately 19 ppbv). The net chemistry, advection, vertical transport, and deposition are relatively small compared to those of the other regions. The net monthly rates of changes are obviously negative from February to August (net-dO₃/dt in Fig. 4), in agreement with the successively decreasing tendency of O₃. Net chemistry shows a loss of O₃, and the rate of loss reaches its maximum in the spring. Advection, convection and diffusion have positive values with distinct seasonal variations, indicating that Region I is an O₃ inflow region. Because of the subtropical high, southeasterly wind prevails during the summer (Fig. 5b). The lower O₃ mixing ratios in the upwind remote ocean lead to weaker advection in summer in Region I. Weaker dry deposition results from lower local O₃ mixing ratios in summer. From the above discussions, we can see that the inflow resulting from advection, convection and diffusion is the main source of O₃ in Region I, while chemistry is the sink. Advection and stratospheric contribution may play critical roles in the seasonal cycle of O₃ due to their similar seasonal variations.

In Region II, O₃ shows a seasonal pattern with two peaks at 50.6 and 42.6 ppbv in spring and autumn, respectively. Net chemistry is positive throughout the year and has two peaks of 80 and 110 ppbv/month in the spring and autumn, respectively. Negative advection exists during the whole year, and positive diffusion only exists in summer, which reveals that Region II is the O₃ outflow region. As such, the main factor causing O₃ seasonal cycle in Region II is photochemistry.

In Region III, O₃ has a summer maximum (60.8 ppbv) and a late fall–early winter minimum (39.9 ppbv). Net chemistry, advection and deposition are relatively large. Net chemistry reaches a maximum of 280 ppbv/month in summer, with a minimum of

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**Fig. 3.** Average emission rate (μg/m²/s) during 2001–2007 of (a) NO (including anthropic, biomass burning and soil emissions) and (b) CO at the surface in spring used in the model simulations (shaded). The four regions for the budget analyses in (a) are indicated by blue boxes. The solid blue line in (b) indicates the diagnostic transect. The black boxes in (b) mark two of the 10 tagged regions. The locations of monitoring sites are indicated by solid dots. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
5 ppbv/month in December. Northeastern China has less precipitation and is relatively drier than Region II and Region IV in the summer (Yang et al., 2003), resulting in a summer maximum of photochemistry in Region III due to stronger solar radiation and lower humidity. Another important factor causing the summer maximum of chemistry is the high level of O3 precursors. Advection has a negative value (approximately -105 ppbv/month) in summer due to the outflow of polluted air from Region III and the inflow of clean air from the upwind region. Thus, the O3 seasonal cycle in Region III mainly results from photochemistry.

In Region IV, the double-peak pattern of O3 is distinctly distinguished from that in Region I where O3 is low from March to November. The first peak (53 ppbv) occurs in spring. After spring, the O3 mixing ratio begins to decrease until July, when a minimum of 39 ppbv occurs. In autumn, there is a second peak, which is slightly lower than the springtime peak. The net chemistry presents

Fig. 4. Monthly O3 mixing ratio (units: ppbv), O3S (units: ppbv) and budget (units: ppbv/month) of MOZART-4 over the four regions. Advection, Convection, Diffusion, Net chemistry, and Dry deposition represent the fluxes of horizontal and vertical advection, convection, diffusion, the net photochemical production, and dry deposition, respectively. O3S is the stratospheric O3 tracer in the boundary layer. Net-dO3/dt represents the net monthly rate of change in the O3 mixing ratio between the adjacent two months (units: ppbv/month). R is the correlation coefficient between simulated and observed results at individual sites in the corresponding regions.

Fig. 5. Seven-year averaged wind vector (vector, unit: m/s) and geopotential height (contour, unit: m) on 850 hPa in April and July.

(a) Apr. (b) Jul.
two peaks in the spring and autumn, similar to the seasonal variation of O₃. Region IV is the O₃ outflow region due to the net negative effect of advection, convection and diffusion during the whole year. In summer, it is located at the bottom part of subtropical high, and the southeasterly wind prevails (Fig. 5), leading to the transport of oceanic air masses with low levels of O₃ and O₃ precursors to Region IV, as inferred from advection. However, low levels of O₃ precursors and more cloudy or rainy days (Yang et al., 2003) give weaker photochemistry during the summer. Thus, the seasonal variation of O₃ is principally affected by chemistry, and the summer minimum in Region IV results from weak chemical production and strong outflow.

3.3 Reasons for the spring latitudinal O₃ gradient

Tanimoto et al. (2005) indicated that exchanges between continental and maritime air masses driven by the Asian monsoon play a central role in producing O₃-latitudeal in homogeneity over East Asia, coupled with regional photochemistry. The authors also estimated that the regional build-up of O₃ over Japan is from anthropogenic emissions in Eastern China and the Korean Peninsula. As discussed in Section 3.2, the O₃-latitudeal gradient crosses Region I and Region II where the controlling factors of O₃ concentration differ significantly. What is the central role controlling the O₃-latitudeal gradient? What are the implications of Chinese pollution export to the latitudeal gradient? The following experiments quantified the impact of the physical and chemical processes on the O₃-latitudeal gradient by analyzing O₃ budgets and the source of O₃ using tagged experiments.

Fig. 6a shows that the spring O₃ concentration increases with latitude, reaching a maximum (52 ppbv) around 35°N, and then decreases from 35° to 45°N, which is in agreement with the result of Tanimoto et al. (2005). The stratospheric tracer is weakest at low latitudes (approximately 5 ppbv) and continues to increase until 32°N (to approximately 10 ppbv). It is less than one-third of the total mean O₃ mixing ratio, which is in accordance with the results of Monks (2000). This finding indicates that the stratospheric input is not the dominant reason leading to the O₃-latitudeal gradient. Fig. 6b shows that convection has little impact on the O₃-latitudeal gradient for its small change with latitude, except at 35°N. Positive roles of net chemistry and diffusion and negative advection in the relatively low latitudes (20°–24°N) reveal that low latitude area is the net O₃ outflow and that the spring maximum at 20°–24°N is mainly due to net chemistry. The area between 24° and 28°N is the O₃ inflow region due to the positive roles of advection and diffusion. The net chemistry increases with latitude from 28°N to its peak at 35°N. The advection, convection and diffusion terms become negative (outflow of O₃), and advection plays a main role in O₃ outflow. At 42°N, the effect is reversed. It is concluded that the spring O₃-latitudeal gradient is mainly due to net chemistry, and advection makes the gradient weaker.

The results of the tagged experiments are shown in Fig. 7. The stratospheric contribution rate of O₃ (Strato) increases with latitude and accounts for 20% of the total at 35°N. The contribution rate of O₃ in the boundary layer over Japan and the Korean Peninsula (JK-BL) increases with latitude and reaches a peak (approximately 25%) at 35°N. The rate of O₃ export from China’s boundary layer (CN-BL) is the highest in low latitudes, approximately 35%, and then decreases with latitude. It is less than 15% at 35°N. Although each contribution rate of O₃ from the boundary layer of North America and Europe and from the entire free troposphere is relatively low, together they contribute roughly 20% of the total O₃ at 35°N. At a relatively higher latitude, the contribution reaches 25%. The rate of REST is relatively low at 35°N, at only 14%. As such, the key contributor is CN-BL (30%) south of 32°N, and Strato and JK-BL are ranked second and third. Around 35°N, the contributions for the high-O₃ values are arranged in a decreasing order, except for REST, as follows: JK-BL (local, 25%), Strato (20%), CN-BL (15%), ER-BL (7%), ER-HT (6%), NA-BL (5%), NA-HT (4%), CN-HT (3%), and JK-HT(2%). North of 38°N, JK-BL is still the key contributor, but it slightly decreases with latitude while ER-BL, NA-BL and REST increase.

O₃ can be produced further downstream from the emission regions of its precursors. The export of China’s emissions leads to secondary O₃ production at mid-latitudes. Jaegle et al. (2003) found that the influence of secondary O₃ production increased O₃ by 2–4 ppbv on average in the Northeast Pacific for the 0–6 km column using GEOS-CHEM. The precursors of O₃ over China and Japan are higher than those in the Northeast Pacific (Fig. 3a and b). Because most of the atmosphere is NOₓ-limited, we use a tagging NO tracer to identify the contribution of NO and its resulting products in Eastern China to O₃ over Japan, which was used by Emmons et al. (2012). The simulation result shows that the
contribution of NO and its resulting products in Eastern China decrease with latitude and account for 7.8 ppbv (15.4%) at 35° N (not shown), which is consistent with the tagged experiment in Fig. 7.

4. Conclusions

Generally, the monthly mean O3 mixing ratio shows a maximum over the Asia-Pacific region during early spring to early summer, as reported by previous studies. Four regions are defined according to the seasonal cycle patterns to identify the impacts of physical and chemical processes based on quantitative analysis of the O3 budget using 2001–2007 simulations of MOZART-4. Two tagged numerical experiments were performed to quantify the relative importance of physical and chemical processes on the reasons for the O3-latitudinal gradient in the spring. The main results are summarized as follows:

1. The model simulation reasonably reproduced seasonal variabilities of O3 over the Asia-Pacific region, with a particularly excellent correlation at the low-latitude oceanic site.

2. Over the Northwest Pacific Ocean, the boundary-layer O3 shows a maximum during late winter and early spring and a minimum in summer, resulting from advection effects. Over Japan, the Korean Peninsula and Southeastern China, the seasonal pattern of O3 is dominated by two peaks in the spring and fall and by a summer trough in the net chemistry. The O3 seasonal pattern in Northeastern China has a single summer peak, due to the strong photochemical production in the high level of NOx precursors under the dry weather and strong solar radiation.

3. The large latitudinal gradient in O3 with a maximum around 35° N in spring is mainly due to net chemistry, whereas advection weakens the O3 gradient. The contribution of stratospheric O3 is ranked second (20%) to the local contribution of the boundary layer of Japan and the Korean Peninsula (25%) near 35° N. The rate of O3 export from China’s boundary layer is the highest at lower latitudes, approximately 30%, and decreases with latitude at 15% at 35° N. The contribution of North America and Europe increases with latitude and reaches 22% at 35° N.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.06.006.

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