Mechanism of Formation of the Ozone Valley over the Tibetan Plateau in Summer—Transport and Chemical Process of Ozone

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(Received February 4, 2002; revised September 7, 2002)

ABSTRACT

With the 3D chemical transport model OSLO CTM2, the valley of total column ozone over the Tibetan Plateau in summer is reproduced. The results show that when the ozone valley occurs and develops, the transport process plays the main part in the ozone reduction, but the chemical process partly compensates for the transport process. In the dynamic transport process of ozone, the horizontal transport process plays the main part in the ozone reduction in May, but brings about the ozone increase in June and July. The vertical advective process gradually takes the main role in the ozone reduction in June and July. The effect of convective activities rises gradually so that this effect cannot be overlooked in July, as its magnitude is comparable to that of the net changes. The effect of the gaseous chemical process brings about ozone increases which are more than the net changes sometimes, so the chemical effect is also important.

Key words: Tibetan Plateau, ozone valley, dynamic transport process, chemical process

1. Introduction

By analyzing TOMS data, Zhou et al. (1995) found that there is a low value center of total column ozone over the Tibetan Plateau in summertime. As there are convergent upward flows over the Tibetan Plateau in summer under the influence of the South Asia High, they thought that the convergent upward flows would cause the ozone valley over the Tibetan Plateau, and it is a pathway through which the materials in the lower troposphere are transported into the stratosphere. Zou (1996) researched the seasonal variation and trend of the ozone over the Tibetan Plateau with TOMS data. The scientists from NASA (1995, private communication) and Zou and Gao (1997) analyzed SAGE I and II data respectively to find that the ozone valley over the Tibetan Plateau in summer happens at 15–20 km altitude. Fu et al. (1997) simulated the ozone valley over the Tibetan Plateau with a 2D chemical model, whose results illustrated that the low value center of total column ozone over the Tibetan Plateau is mostly attributed to the dynamic and thermodynamic effects of the Tibetan Plateau, and the effect of chemical processes can be overlooked. Liu and Li (2001) found that the Tibetan plateau is one of three regions of the rapidest decrease of total ozone in the area at the same latitude by using TOMS data (1979–1992), and the ozone valley would be deepened.

In summer, there is a low value center of total ozone over the Tibetan Plateau, and the deepened ozone valley makes the ozone layer above 450 000 km^2 of ground become thinner (Liu and Li, 2001), which would cause an increase of UV radiation at the surface over the Tibetan Plateau. The increase of UV radiation significantly influences the human beings and ecosystem of the Tibetan Plateau. Therefore, Chinese scientists are paying more attention to the ozone changes over the Tibetan Plateau. Because the 2D model has the limitation that it cannot exhibit longitudinal effects. Fu et al. indicated only the dynamic and thermodynamic effects of the Tibetan Plateau, and the chemical effect. There are some problems that should be studied further, such as the role of the dynamic and chemical processes. Hence, we use a 3D chemical model to simulate the ozone valley over the Tibetan Plateau, and analyze the role of each process.

2. Description of the model

The OSLO CTM2 is an off-line chemical transport/tracer model (CTM), which uses precalculated transport and physical fields to simulate chemical

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turnover and distribution in the atmosphere. The model is valid for the global troposphere, and three-dimensional with the model domain reaching from the ground up to 10 hPa for the current data set. The model horizontal resolution is determined by the input data. The data set used in this study is ECMWF forecast data with the T21 grid (5.625° × 5.625°) in 1996. The vertical resolution of the model is determined by the input data and we use 19 levels from the surface up to 10 hPa.

Advection is handled by means of the second order moment (Prather, 1986). Convection is based on the Tiedtke mass flux scheme (Tiedtke, 1989), in which vertical transport of species is determined by the surplus/deficit of mass flux in a column. The chemical scheme is dealt with by the QSSA approach (Hesstvedt et al., 1978; Berntsen and Isaksen, 1997). Photodissociation is performed on-line following Wild et al. (2000). Natural emissions are based upon GEIA and EDGAR, and anthropogenic emissions are from Müller (1992) (see Table 1), in which subsonic aircraft NO_x emission is referenced as in the IPCC year 2000 scenario. Deposition is treated according to Wesley (1989), and the boundary layer according to the Holtslag K-profile scheme (Holtslag et al., 1990). Dry deposition velocities in the model are given in Table 2. The influence of stratospheric ozone is a fixed ozone flux of 450 Tg per year. The model involves 48 species, such as: O_x, NO_y, OH_x, and NMHCs, etc. 85 reactions and 16 photolysis reactions. The split operator method is adopted to solve,

$$\begin{cases} \dfrac{\partial \phi}{\partial t} = \left(\dfrac{\partial \phi}{\partial t}\right)_{\text{dyn.}} + \left(\dfrac{\partial \phi}{\partial t}\right)_{\text{chem.}}, \\ \dfrac{\partial \phi}{\partial t}_{\text{dyn.}} = \left(\dfrac{\partial \phi}{\partial t}\right)_{\text{adv.}} + \left(\dfrac{\partial \phi}{\partial t}\right)_{\text{conv.}} + \left(\dfrac{\partial \phi}{\partial t}\right)_{\text{B.L.}}, \end{cases} \tag{1}$$

where ϕ represents trace gas concentration; t, time; subscript dyn., the dynamic process; chem., the chemical process; adv., the advective process; conv., the convective process; and B. L., the effect of the boundary layer. Detailed introductions of OSLO CTM2 are in Sundet (1997), Jonson et al. (2001), and Kraabol (2000). The model is run for 15 months and the results of the last 4 months are analyzed.

3. Results and analysis

The ozone valley is mainly the ozone reduction at 15–20 km high that the Tibetan Plateau in summer produces. The OSLO CTM2 domain is from the ground to 10 hPa, therefore, the model is able to simulate the seasonal variation of the ozone valley over the Tibetan Plateau. Figure 1a shows modeled total ozone in July, and Fig. 1b exhibits observed total ozone from TOMS data in July. By comparing these two figures, it can be seen that the modeled total ozone is less than the TOMS data. This is because the model vertical height is limited. As ozone above 10 hPa is approximately longitudinally uniform, the modeled total ozone can basically represent the distribution and seasonal characteristics of the real total ozone. But there are some differences between these two distributions, such as the TOMS data showing that the high center of total ozone in the Northern Hemisphere is located in middle latitudes, however, this characteristic is missing from the model. The observation (Fig. 1b) indicates that there are two low centers of total ozone over the Tibetan Plateau and Rock Mountains, respectively. From Fig. 1a, it is found that the model is capable of reproducing the two low centers of total ozone. Figure 2a displays the difference between 90°E and the longitudinally averaged modeled total ozone. Being similar to Fig. 2a, Fig. 2b depicts the difference between 90°E and the longitudinally averaged TOMS total ozone. The comparison of these two figures shows that the modeled valley of total ozone over the Tibetan Plateau is in excellent agreement with the TOMS observation, but the modeled intensity of the ozone valley is less than that of the TOMS data, namely, the modeled difference of total ozone is 25 DU, but the observed difference is 30 DU.

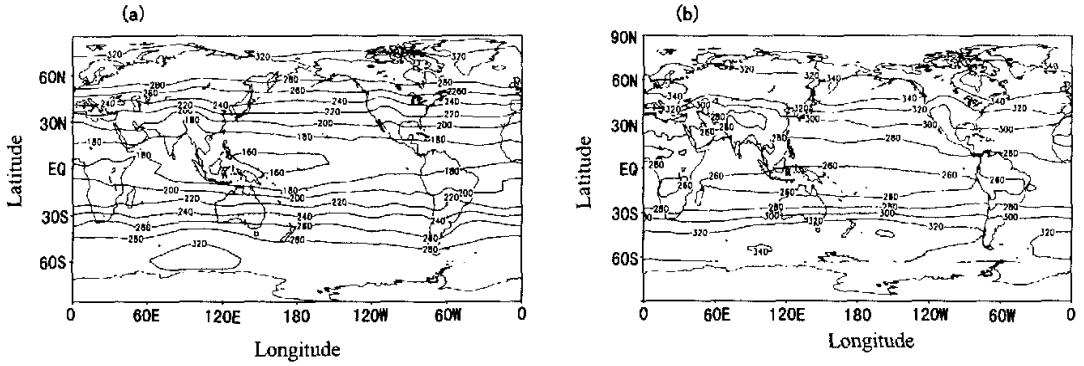

Fig. 1. Distribution of total column ozone in July, (a) model results, (b) TOMS data, units: DU.

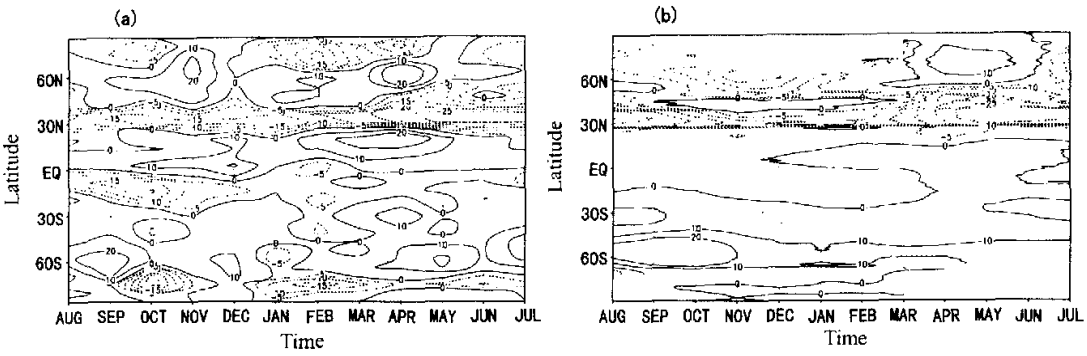

Fig. 2. Difference of total column ozone between 90°E and zonal mean, (a) model results, (b) TOMS data, units: DU.

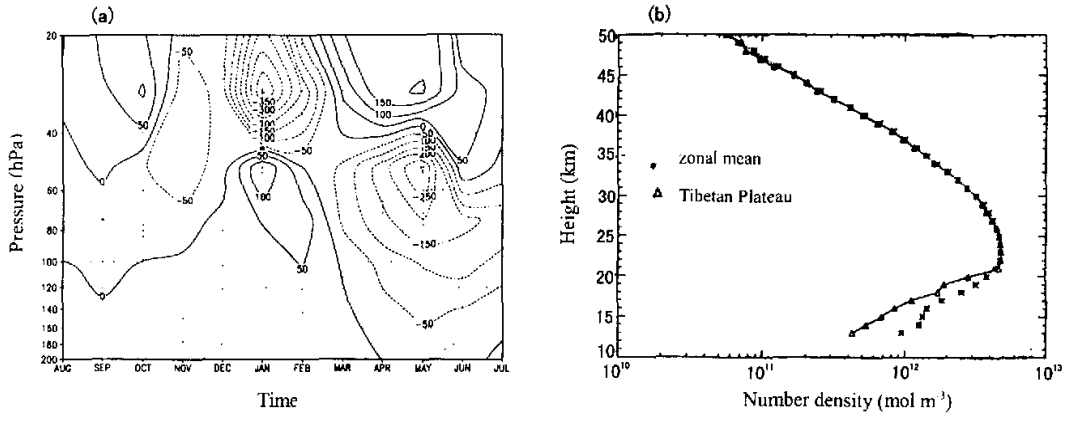

Fig. 3. (a A height–time section of the ozone difference between grid point (31°N, 90°E) over the Tibet an Plateau and the zonal mean (units: ppdv). (b) Ozone profile on the Tibetan Plateau and the zonal mean from SAGE data (from NASA) on 15 May 1986 (units: mol m^{-3}).

The above results indicate that the model is able to simulate the seasonal variation of the ozone valley over the Tibetan Plateau in summer. Figure 3a shows the modeled height–time variation of the difference between the ozone of the Tibetan an Plateau (31°N, 90°E) and the longitudinal average, and Fig. 3b depicts the difference between the ozone of the Tibetan Plateau and the longitudinal average from SAGE data. The observation indicates that the ozone valley over the Tibet an Plateau in summer mainly exists at 15 22 km altitude. The model results reproduce very well that the production of the ozone valley over the Tibetan Plateau in summer is primarily ascribed to the ozone reduction from 120 hPa to 40 hPa (equivalent to 14–24 km altitude).

The above comparison of modeled and observed results sufficiently illuminates that the model is able to reproduce the formation and variation of the ozone valley over the Tibetan Plateau in summer. This is the basis of the further analysis of every process. In order to analyze the effects of transport and chemical process on the formation of the ozone valley over the Tibetan Plateau, the grid point (31°N, 90°E, 100 hPa) over the Tibetan Plateau is selected for the analysis of ozone budgets. Generally, a statistic of the material budget is the sum of every process from one hour to another hour. But, when the changes of trace concentration are analyzed, the average concentration is usually used, such as in the above results, monthly averaged concentrations are used. Therefore, the analyses of budgets are not the same as the analyses of concentration. In order to be consistent with average concentration, a method is used as follows,

$$\begin{cases} x_1 = x_0 + \sum\limits_j p_{j1}\,, \\ x_2 = x_1 + \sum\limits_j p_{j2} = x_0 + \sum\limits_j (p_{j1} + p_{j2})\,, \\ x_i = x_{i-1} + \sum\limits_j p_{ji} = x_0 + \sum\limits_j (p_{j1} + p_{j2} + \cdots + p_{ji})\,, \\ x_m = x_{m-1} + \sum\limits_j p_{jm} = x_0 + \sum\limits_j (p_{j1} + p_{j2} + \cdots + p_{jm})\,, \\ \bar{x} = x_0 + \sum\limits_j (p_{j1} + \dfrac{m-1}{m} p_{j2} + \dfrac{m-2}{m} p_{j3} + \cdots + \dfrac{1}{m} p_{jm})\,, \end{cases} \tag{2}$$

where x_i represents trace concentration at the ith hour; p_{ji}, the effects of the jth process at the ith hour; and $\bar{x}$, the average concentration. The sum in the brackets is the effect of the jth process on the average concentration. Compared with the effect of every process, the reason for changes in average concentration is explored. From Eq.(2), it is known that the changes of average concentration of different periods which started at the same hour are ascribed to changes of the effect of every process. It is meaningless how big the effect of a process in a given period is, but the changes of average concentration and the difference of the effect of every process are meaningful. That is to say, the changes of average concentration are caused by the difference of the effect of every process. Table 1 shows the changes of average concentration, the effect of every process, and the differences of every process between different periods.

From Table 1, it is found that the average concentrations in the four periods decrease gradually, which means that the monthly average concentration deceases gradually from April to July. The results in Table 1 illustrate that the effects of every process in different periods are different. Compared with the results of April and April–May, it can be seen that the ozone average concentration decreases from 241.3 in April to 214.1 ppbv in April–May, and the ozone reduction is attributable to horizonal and vertical transport, in which the effect of horizonal transport (−25.1 ppbv) is higher than that of vertical advection (13.3 ppbv), but the effect of convective activites is small (−0.2 ppbv). In the horizontal transport, the effect of latitudinal transport reduces ozone (−753.0 ppbv), but that of longitudinal transport increases ozone (717.9 ppbv). The magnitude of latitudinal and longitudinal transport is much bigger than that of net horizontal and vertical transport. The dynamic transport makes ozone decrease by 38.6 ppbv; but the effect of the chemical process leads ozone to increase 14.8 ppbv, which is opposite to that of the dynamic process. The sum of all the effects is an ozone reduction of 23.2 ppbv. Because the magnitude of the effect of the chemical process is equivalent to that of the net effect, the chemical effect cannot be neglected.

Similar to the changes between April and April May, the average concentration (199.3 ppbv) in April–June is less than in April–May. The table indicates that the ozone reduction in June is mostly ascribable to the vertical advection (119.7 ppbv), and the effect of convective activities is small (−1.6 ppbv). Meanwhile, the horizontal transport and chemical process result in ozone increases of 83.2 and 23.4 ppbv, respectively. That is to say, the horizontal transport which leads to ozone decrease in May makes ozone increase in June. The changes of the horizontal transport are because the ozone increases induced by the longitudinal transport are more than the ozone reduction resulting from the latitudinal transport. The sum of ozone changes is an ozone reduction of 15.6 ppbv. Like the ozone changes in April–June, the effect of every process on ozone becomes stronger in July than in June. The horizontal transport and chemical process make ozone increase 149.7 and 43.0 ppbv, respectively. The vertical advection and convective activities lead ozone to decrease 184.6 and 7.4 ppbv. While the effect of every process on ozone significantly intensifies the net effect on ozone becomes weaker by 9.9 ppbv. The effect of convective activities is equal to that of the net effect on the order of magnitude, so the effect of the convective activities cannot be neglected.

From the above analyses of April–May, April June, and April–July, it is found that the effects of the longitudinal and latitudinal transport gradually become smaller, especially latitudinal transport. Contrarily, the effects of vertical advection, convective activities, and the chemical process increasingly intensify. In June and July, the effect of the vertical advection becomes the main factor of the changes of the ozone valley over the Tibetan Plateau. On the whole, the effect of the dynamic process on the ozone valley over the Tibetan Plateau is reviewed as follows. In May, what is the early formation of the ozone valley over the Tibetan Plateau, the horizontal transport is the main part of the dynamic process that results in the ozone valley over the Tibetan Plateau. However, in June and July, the vertical advection becomes the main part of the dynamic process.

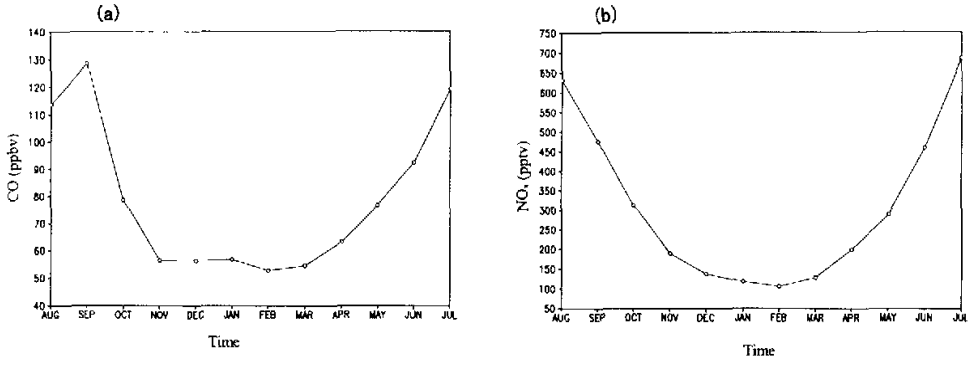

Fig. 4. Seasonal variation of (a) CO (units: ppdv) and (b) NO_x (units: ppdv) at (31°N, 90°E, 100 hPa).

Table 1. Budget of Ozone (units: ppbv)

	Apr. 1 to Apr. 30	Apr. 1 to May 31	Apr. 1 to Jun. 30	Apr. 1 to Jun. 31
Avg. Conc.	241.333	214.098(-27.2)	199.341(-14.8)	189.590(-9.8)
Lon. Transport	1480.288	2208.212(717.9)	2648.866(440.6)	2864.468(211.6)
Lat. Transport	1440.558	2193.613(-753.0)	2551.049(-357.4)	2616.951(-65.9)
Horiz. Transport	39.730	14.599(-25.1)	97.817(83.2)	247.517(149.7)
Vert. Adv.	38.525	25.192(-13.3)	-94.464(-119.7)	-288.096(183.6)
convective	0.0	-0..169(-0.169)	1.759(-1.6)	-9.164(-7.4)
dynamic	78.255	39.622(-38.633)	1.594(38.032)	-49.743(-51.337)
chemical	6.560	21.363(14.8)	44.774(23.4)	87.732(43.0)
sum	80.377	57.140(23.2)	41.539(-15.6)	31.649(-9.9)

*Notes: Avg. Conc. represents average concentration in a period; Lon. Transport, longitudinal transport; Lat. Transport, latitudinal transport; Horiz. Transport, horizontal transport (the sum of Lon. Trans. and Lat. Trans.); Vert. Adv., vertical advection; convective, convective activities; dynamic, dynamic process (the sum of Horiz. Transport, Vert. Adv., and convective); chemical, Chemical process. The brackets show the difference between the current and previous period.

The magnitude of the effect of the chemical process on changes of ozone concentration is sometimes more than the net ozone effects, so the chemical effect is important. Results of some models indicate that the production of tropospheric ozone depends on NO_x concentration to a considerable degree. In the boundary layer, because of high NO_x concentration, the production of ozone is most efficient in the troposphere. In the middle troposphere, the depletion of ozone is more than the production of ozone due to the lowest NO_x concentration. In the upper troposphere, the chemical process produces ozone under higher NO_x concentration (Bertsen and Isaksen, 1997; Flaøt and Hov, 1996). Figures 4a and 4b show the seasonal variation of CO and NO_x at the grid point (31°N, 90°E, 100 hPa), which increase gradually from April to July. Because of the increase of NO_x and ozone precursors such as CO etc. the chemical production of ozone is enhanced gradually. Therefore, the enhancement of the chemical production of ozone can be attributed to the increase of NO_x and CO concentration.

4. Summary and discussion

From the above analyses it can been seen that when the ozone valley is produced and developed the transport process of ozone plays the main role, and the chemical process partly compensates for the ozone reduction caused by the transport process. In the dynamic transport process of ozone, the horizontal transport process plays the chief role in the ozone reduction in May, but brings about an ozone increase in June and July. The vertical advective process gradually becomes the main role in the ozone reduction in June and July. The effects of convective activities rise little by little so that this effect cannot be overlooked in July as its magnitude is comparable to that of the

net changes. The effect of the gaseous chemical process brings about an ozone increase that is sometimes more than the net changes, so the chemical effect is also important. The variations of the dynamic transport process are closely associated with the variation of the circulation over the Tibetan Plateau and its surroundings. The chief characteristic of the summer circulation is that the South Asia High is over the Tibetan Plateau and its surroundings. In May, the center of the South Asia High moves from the Philippine Sea to the Zhongnan Peninsula (21°N, 101°E), the intensity of the center increases a little, whose closed contour line at 100 hPa is 16640 gpm. When the South Asia High moves over the Tibetan Plateau in June, its center is situated at (29°N, 86°E), whose intensity significantly increases, with the closed contour line of the center at 100 hPa of 16760 gpm. In July, the South Asia High continues to move westward, with center situated over the Iran Plateau (32°N, 60°E), and at the same time, the intensity of the center is further strengthen, with the closed contour line of the center at 100 hPa of 16840 gpm (Zhu et al., 1980). With the movement of the South Asia High, the flow field over the Tibetan Plateau and its surroundings changes accordingly. In May, the airflow from the lower troposphere of the southern, northern, and eastern Tibetan Plateau converges into the middle and upper troposphere over the Tibetan Plateau. In June, a strong updraft also occurs in the western Tibetan Plateau. The updraft from the surroundings of the Tibetan Plateau becomes the strongest in July (Bian et al., 1997). Integrated with the analyses of the model results and weather, we further depict the dynamic and chemical processes in the formation and development of the ozone valley over the Tibetan Plateau. In May (that is the early period of formation of the ozone valley over the Tibetan Plateau), the formation of the ozone valley over the Tibetan Plateau is primarily caused by the South Asia High moving northwestward to bring ozone of low concentration from low latitudes. Then, while the South Asia High moves over the Tibetan Plateau and its intensity is strengthened, ozone of low concentration from the lower troposphere around the Tibetan Plateau is transported into the upper troposphere and lower stratosphere over the Tibetan Plateau. Consequently, vertical advection plays the main role in the formation of the ozone valley over the Tibetan Plateau, but the horizontal transport partly compensates for the effect of the vertical advection. At same time, the effect of the chemical process plays an important role and cannot be overlooked.

Acknowledgments. The authors wish to thank Prof. I. S. A. Isaksen and Dr. J. K. Sundat from the University of Oslo for their help.

REFERENCES

Bertsen, T., and I. S. A. Isaksen, 1997: A global 3D chemical transport model for the troposphere, 1: Model description and CO and ozone results. *J. Geophys. Res.*, **102**, 21239–21280.

Bian J. C., Li W. L., and Zhou X. J., 1997: Analysis of the seasonal variation feature of the winds structure over Tibetan Plateau and its surroundings. *Ozone Changes Over China and Its Influences on Climate and Environment*, Zhou X. J., Ed., China Meteorological Press, Beijing, 257–273.

Flatøy, F., and Ø. Hov, 1996: Three-dimensional model studies of the effect of NO_x emission from aircraft in the upper troposphere over Europe and North Atlantic. *J. Geophys. Res.*, **101**, 1401–1422.

Fu C., Li W. L., and Zhou X. J., 1997: Numerical simulation of the formation of ozone valley over Tibetan Plateau in summer, its surroundings. *Ozone Changes Over China and Its Influences on Climate and Environment*, Zhou X. J., Ed., China Meteorological Press, Beijing, 274–285.

Hesstvedt, E., Ø. Hov, and I. S. A. Isaksen, 1978: Quasi steady-state approximation in air pollution modelling: Comparison of two numerical schemes for oxidant prediction. *Int. Journal of Chem. Kinetics*, **X**, 971–994.

Holtslag, A. A. M., E. I. F. DrBruijn, and H. -L. Pan, 1990: A high resolution air mass transformation model for short-range weather forecasting. *Mon. Wea. Rev.*, **118**, 1561–1575.

Jonson, J. E., J. K. Sundet, and L. Tarrason, 2001: Model calculations of present and future levels of ozone and ozone precursors with a global and regional model. *Atmos. Environ.*, **35**, 525–535.

Kraabol, A. G., 2000: Impact of NO_x emissions from subsonic aircraft on the chemical composition of the atmosphere: Model calculation. Ph.D. thesis, Dept. of Geophysics, University of Oslo, Norway, 117pp.

Müller, J., 1992: Geographical distribution and seasonal variation of surface emissions and deposition velocities of atmospheric trace gases. *J. Geophys. Res.*, **97**, 3787–3804.

Liu Y., and Li W. L., 2001: Deepening of ozone valley over the Tibetan Plateau and its possible influences. *Acta Meteorologica Sinica*, **59**, 97–106. (in Chinese)

Prather, M. J., 1986: Numerical advection by conservation of second-order moments. *J. Geophys. Res.*, **91**, 6671–6681.

Sundet, J. K., 1997: Model studies with a 3D global CTM using ECMWF data. Ph.D. thesis, Dept. of Geophysics, University of Oslo, Norway, 101pp.

Tiedtke, M., 1989: A comprehensive mass flux scheme for cumulus parameterisation on large scale models. *Mon. Wea. Rev.*, **117**, 1779–1800.

Wesley, M. L., 1989: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmos. Environ.*, **23**, 1293–1304.

Wild, O., X. Zhu, and M. J. Prather, 2000: Fast-J: Accurate simulation of in- and below-cloud photolysis in tropospheric chemical models. *J. Atmos. Chem.*, **37**, 245–282.

Zhou X. J., Luo C., Li W. L., and Shi J. E., 1995: Ozone changes over China and low center over Tibetan Plateau. *Chinese Science Bulletin*, **4**, 1396–1398.

Zhu F. K., Lu L. H., Chen X. J., and Zhao W., 1980: *South Asia High*, Science in China Press.
Zou H., 1996: Seasonal variation and trends of TOMS ozone over Tibet. *Geophys. Res. Lett.*, **23**, 1029–1032.
Zou H., and Gao Y. Q., 1997: Vertical ozone profile over Tibet using SAGE I and II data. *Advances in Atmospheric Sciences.*, **14**, 505–512.

青藏高原夏季臭氧低谷形成的机理

--臭氧输送和化学过程

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摘 要

利用三维化学输送模式（OSLO CTM2）模拟青藏高原夏季臭氧低谷。结果表明：在青藏高原夏季臭氧低谷的形成和变化过程中，动力输送过程起着最主要作用，化学过程部分补偿了输送过程引起的臭氧减少。在动力输送过程中，水平输送在5月份是造成臭氧减少的主要原因，可在6月和7月却变为使臭氧增加；垂直平流的作用不断增强，在6月和7月成为臭氧减少的主要因素；对流输送的作用在7月份大幅增加，其引起的臭氧减少可以与净的变化相比，其作用也不可忽视。气相的化学过程引起的臭氧增加的量值有时超过了臭氧的净变化的大小，因此它也起着重要作用。

关键词：青藏高原，臭氧低谷，动力输送过程，化学过程