ADVANCES IN ATMOSPHERIC SCIENCES, VOL. 20, NO. 4, 2003, PP. 565–574

Impact of Aircraft NO_x Emission on NO_x and Ozone over China

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(Received March 26, 2002; revised April 3, 2003)

ABSTRACT

A three-dimensional global chemistry transport model (OSLO CTM2) is used to investigate the impact of subsonic aircraft NO_x emission on NO_x and ozone over China in terms of a year 2000 scenario of subsonic aircraft NO_x emission. The results show that subsonic aircraft NO_x emission significantly affects northern China, which makes NO_x at 250 hPa increase by about 50 pptv with the highest percentage of 60% in January, and leading to an ozone increase of 8 ppbv with 5% relative change in April. The NO_x increase is mainly attributed to the transport process, but ozone increase is produced by the chemical process. The NO_x increases by less than 10 pptv by virtue of subsonic aircraft NO_x emission over China, and ozone changes less than 0.4 ppbv. When subsonic aircraft NO_x emission over China is doubled, its influence is still relatively small.

Key words: aircraft emission, NO_x, ozone

1. Introduction

It has been shown that emissions from aircraft have an influence on the trace components of the global atmosphere and thereby the climate (IPCC, 1999; Brasseur et al., 1998). The total radiative forcing from aircraft emissions in 1992 is about 3.5% of the total radiative forcing caused by all anthropogenic activities. Aircraft emission of NO_x, water vapour, sulphur oxides, soot, and carbon dioxide can directly or indirectly enhance the level of so-called greenhouse gases, increase the particle loading, and alter cloudiness in the atmosphere. The greatest contributions are estimated to be from the increase in ozone, carbon dioxide, and contrails (IPCC, 1999). Ozone is a greenhouse gas, and its changes in the middle and upper troposphere have the strongest impact on the radiative forcing of ozone (Wang and Sze, 1980; Lacis et al., 1990; Hansen et al., 1997). The aircraft emission of NO_x has the potential to increase ozone in the upper troposphere. Aircraft emissions of NO_x for 1992 have been found to increase the ozone concentration in the atmosphere by up to 6% in northern mid-latitudes at cruise altitude (IPCC, 1999). Based on a future aircraft scenario, this change is expected to increase to 13% by the year 2050 (IPCC, 1999).

With the rapid economic development and opening to the world of China, Chinese aviation over the mainland has been developing rapidly also. Chinese aviation has increased 25 times more in 1997 compared with 1978, whose rapidest growth rate is about 20% per year from 1989 to 1995, with 7.5% from 1996 to 1997 (CAAC, 1998). Chinese aviation has been developing so rapidly that we need to pay more attention to the effect of Chinese aviation. How does aircraft emission of NO_x influence NO_x and ozone over China? How does emission of NO_x from subsonic aircraft in China influence global NO_x and ozone? We will explore these questions in this paper.

2. Description of model

OSLO CTM2 is an off-line chemical transport/tracer model (CTM; Developed by the Department of Geophysics, University of Oslo), which uses pre-calculated transport and physical fields to simulate chemical turnover and distribution in the atmosphere. The model is valid for the global troposphere, and is 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 a T21 ($5.625^\circ \times 5.625^\circ$)

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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 Müeller (1992) (see Table 1). According to NASA–92 inventory, subsonic aircraft NO_x emission is scaled up to 0.7 Tg (N) yr^{-1} to represent emissions for the year 2000. The yearly averaged subsonic aircraft NO_x emission is interpolated into the model grids, but because the grid height varies with the seasons, the emission in the grid also varies, especially, in high latitudes. Deposition is treated through the use of 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. Influence of stratospheric ozone is a fixed ozone flux of 450 Tg per year.

Table 1. Overview of natural and anthropogenic emissions in OSLO-CTM2

NO_x	(Tg yr^{-1})
Fossil fuels	31.7
Biofuels	1.3
Biomass burning	7.1
Aircraft	0.7
Lightning	5.0
Soils	5.6
Stratosphere	∼0.5
TOTAL	51.9
CO	(Tg yr^{-1})
Fossil fuels	650
Biomass burning	700
Oceans	50
Vegetation	150
TOTAL	1550
NMHC	(Tg(C) yr^{-1})
Isoprene	220
Acetone	30
Fossil fuels	124
Biomass burning	32
TOTAL	406

Methane concentration is fixed to 1700 ppbv in the Southern Hemisphere and 1790 ppbv in the Northern Hemisphere.

Table 2. 1 m dry deposition velocities (V_0) used in the model

			V_0 (cm s^{-1})
Species	Land	Sea	Ice and snow
O_3	0.6	0.1	0.05
NO_2	0.1	0.05	0.02
HNO_3	4.0	1.0	0.05
PAN	0.2	0	0.0
H_2O_2	1.0	1.0	0.05
CO	0.03	0	0

The model is able to simulate the seasonal variation of ozone and CO observed at a number of stations both in the Northern and Southern Hemispheres (Sundet, 1997; Jonson et al., 2001; Kraabol, 2000). The estimated transport of ozone in the upper troposphere and lower stratosphere is in good agreement with observations within the measurement of ozone and water vapor by airbuses in the service aircraft (MOZAIC) program (Marenco et al., 1998).

3. Experiments and results

Four experiments are carried out as follows: The first one (A) is a reference case, which includes global subsonic aircraft NO_x emission; the second (B) excludes the subsonic aircraft emission. The third (C) is the same as the reference case except that subsonic aircraft NO_x emission over China is excluded; The fourth (D) is similar to Case C, but in which subsonic aircraft NO_x emission over China is doubled. These experiments run for 15 months, and the results of the last 12 months are analyzed.

Figure 1a shows the NO_x distribution between 300 and 190 hPa in summer obtained by the measurements of NO_xAR from spring 1995 to spring 1996 (taken from IPCC, 1999), from which it can be seen that there are three high value centers in the Northern Hemisphere. These high centers are located in the southeastern U.S. (40°N, 80°W), Middle East (40°N, 40°E), and Russia and Mongolia (60°N, 80°E), with 400–450, 300–350, and 300–350 pptv, respectively. In comparing the observations and the model results (Fig. 1b) it is found that the modeled concentrations of NO_x at 250 hPa over the southeastern U.S. and Middle East are high values with 350 and 300 pptv, which are lower than the observation. The simulated results basically reproduce the NO_x distribution and concentrations in the upper troposphere, but the model does not reflect the high center over Russia and Mongolia. Because NO_x distribution and concentration in the upper troposphers is influenced by various processes, such as the exchanges between stratosphere and troposphere, lightning, aircraft NO_x emission, and convective activities, the observations during a period do not accurately represent the characteristics of the long-term average condition.

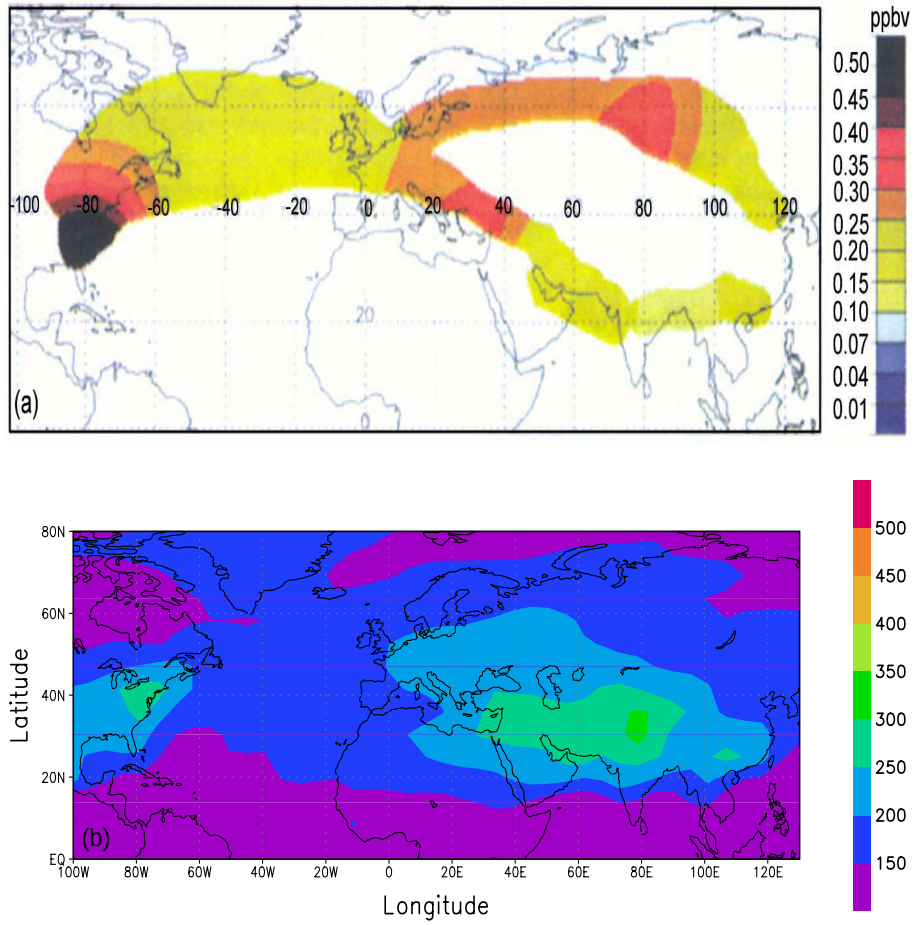

Fig. 1. NO_x distribution in the upper troposphere. (a) NO_x concentration field in the altitude regions between 300 and 190 hPa in summer obtained by the measurements of NO_xAR from spring 1995 to spring 1996 (taken from IPCC, 1999). (b) Simulated NO_x distribution at 250 hPa in summer.

Table 3. Comparison of NO_x observations and model results

	NO_x Observation	model
In winter		
Over NAFC from 9 to 12 km (40°–65°N, 10°–40°W)	96–111 pptv	80–100 pptv
Over eastern U. S. (35°–60°N, 50°–80°W) (Brunner et al., 1998)	124–155 pptv	80–100 pptv
In summer		
over North Atlantic from 10.5 to 11.5 km (Ziereis et al., 1999)	120–230 pptv	150–210 pptv
over North Atlantic	132–142 pptv	
over eastern U. S. (Brunner et al., 1998)	200–361 pptv	210–270 pptv
In autumn eastern U. S., NAFC, and Europe from 10.5 to 11.5 km (Ziereis et al., 1999)	90–140 pptv	100–140 pptv
Northern China (NO_xAR, IPCC, 1999)	150–200 pptv	150–180 pptv
Southern China (NO_xAR, IPCC, 1999)	150–200 pptv	150–180 pptv

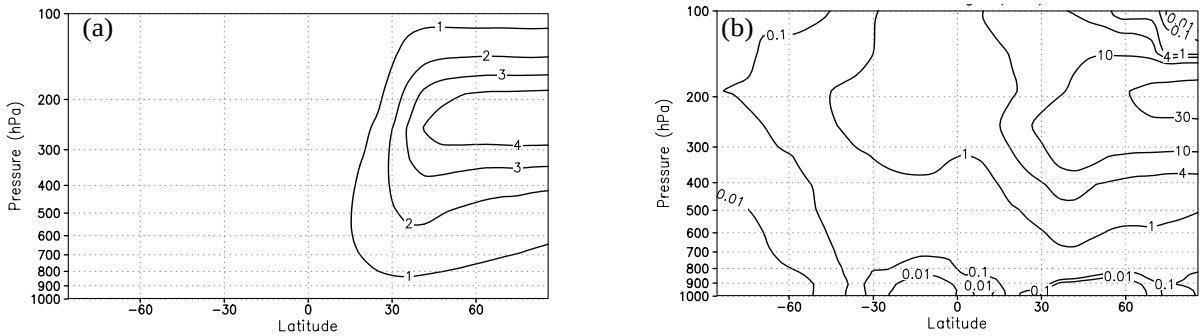

Fig. 2. (a) Simulated annual zonal average increase of ozone, and (b) zonal mean NO_x changes in July.

Table 3 reviews some of the observations and model results in the reference case, which indicates that the model results coincide well with the observations while the model results in winter are lower than the observation to some extent, especially in the eastern U. S. In general the model has the capability to simulate NO_x seasonal variation at subsonic aircraft cruise height.

As many processes affect the atmospheric traces in the upper troposphere, the changes of the traces are quite complicated. In the IPCC report (1999), it is found that the results from six models are not absolutely uniform, especially in the distribution and size of ozone and NO_x increases due to aircraft emission. The common conclusion is that NO_x concentration is obviously raised, and ozone increases by virtue of aircraft NO_x emission. Under the condition of the 2015 inventory, the maxima of annual zonal average increase of ozone from the six models range from 7 ppbv to 11 ppv; and under the condition of 1992 aircraft NO_x emission, the analogues are from 4.4 ppbv to 12.7 ppv (IPCC, 1999). Meanwhile, an assessment prepared for the European Commission (Brasseur et al., 1998) concluded that the current fleet of commercial aircraft should have increased NO_x abundances by 50–100 pptv near 200 hPa, with a corresponding increase in ozone concentrations of 5–9 ppbv (i.e., 4%–8%), during summertime. Figure 2 exhibits the modeled annual zonal average changes of ozone and NO_x changes in July due to aircraft NO_x emission under the condition of the 2000 aircraft NO_x inventory. The highest ozone change is 5 ppbv, while the maximum of NO_x changes is about 50 pptv. The above results indicate that the model is capable of simulating well the changes of ozone and NO_x under the effects of aircraft NO_x emission.

Figure 3 displays the NO_x increase at 250 hPa caused by aircraft NO_x emission (the difference between Case A and B). Model results suggest that the highest NO_x increases are more than 80 pptv in January, and the analogue in July is only 55 pptv in the eastern U.S. and West Europe. Because lightning and convective activities make NO_x in the upper troposphere increase in summer, the effect of aircraft NO_x emission is lower in summer than in winter (25% vs 80%). Schlager et al. (1999) observed NO_x enhancement of 50–150 pptv inside the main aircraft corridor of the North Atlantic in September and October 1997, and concluded that this was due mainly to aircraft emission based on simulation and observation. From the figure, it is found that the modeled NO_x increases of about 60–80 ppbv are in good agreement with the observation in October. When the NO_x increases due to aircraft NO_x emission mainly happen in 30°–60°N, NO_x content in northern China (including Xinjiang, Inner Mongolia, and northeast China) also significantly changes under this effect. The NO_x increases in northern China are 40–55 pptv in winter, spring, and autumn with 40%–60% relative change, but the counterpart in summer is only about 10 pptv with 10% relative change.

As subsonic aircraft NO_x emission may affect climate through changing ozone, more attention is paid to the effect of subsonic aircraft NO_x emissions. Figure 4 shows simulated monthly mean ozone increase at 250 hPa caused by aircraft NO_x emission. Model results indicate that the ozone increases dominantly occur from 40°N to the Arctic, and are the highest in spring with 9 ppbv maximum, lowest in autumn with 3 ppbv maximum. In winter, ozone increase chiefly emerges in 30°–60°N, and is transported into the Arctic. Comparing Figs. 3 and 4, it is found that the locations of the highest NO_x increase are different from the ozone counterparts, with the maximum of ozone change occurring downwind of the NO_x increase. Therefore, ozone increases of 6–8 ppbv are produced in northern China with 5% relative change in spring, but they are lowest in summer and autumn with a maximum of about 2.5 ppbv.

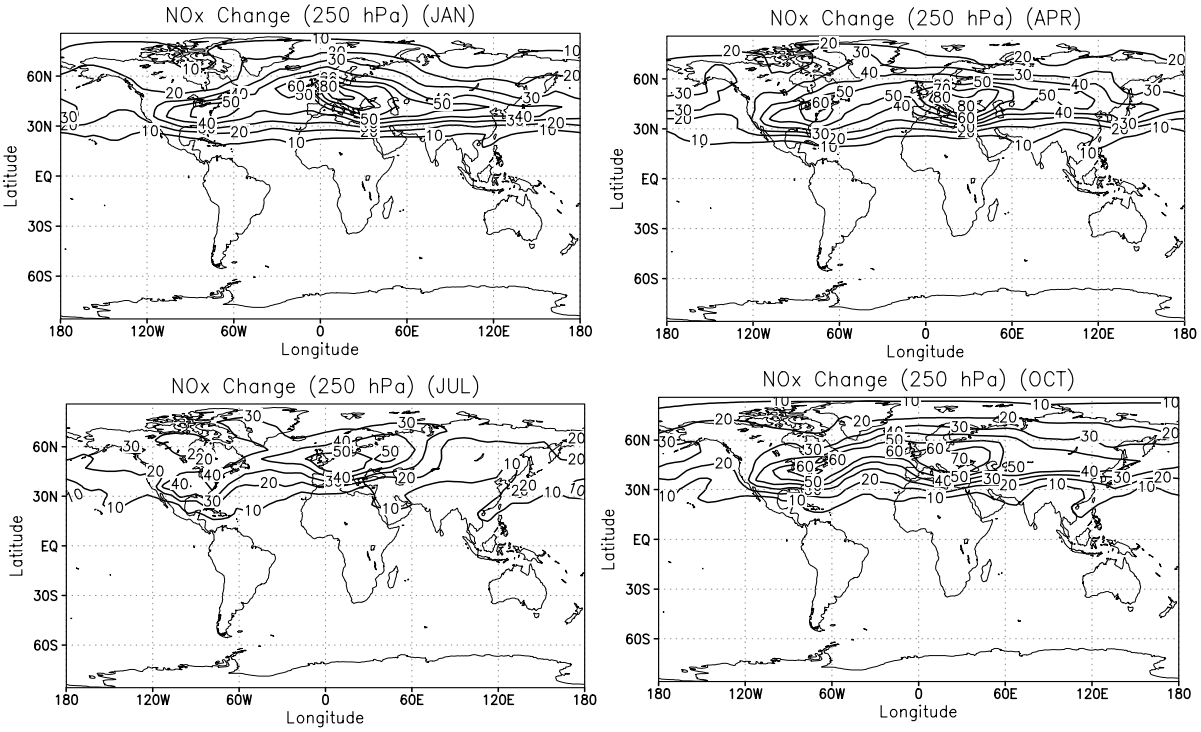

Fig. 3. The NO_x change at 250 hPa due to aircraft NO_x emission (units: pptv).

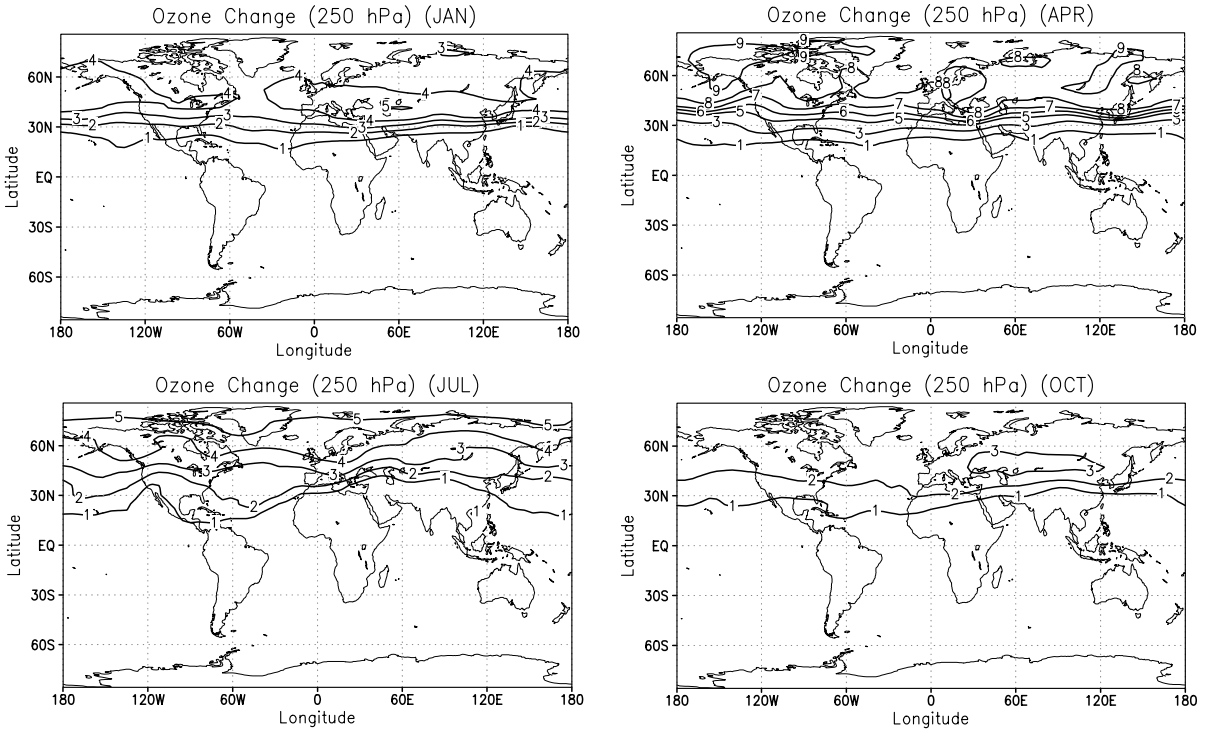

Fig. 4. The ozone change at 250 hPa due to aircraft NO_x emission (units: ppbv).

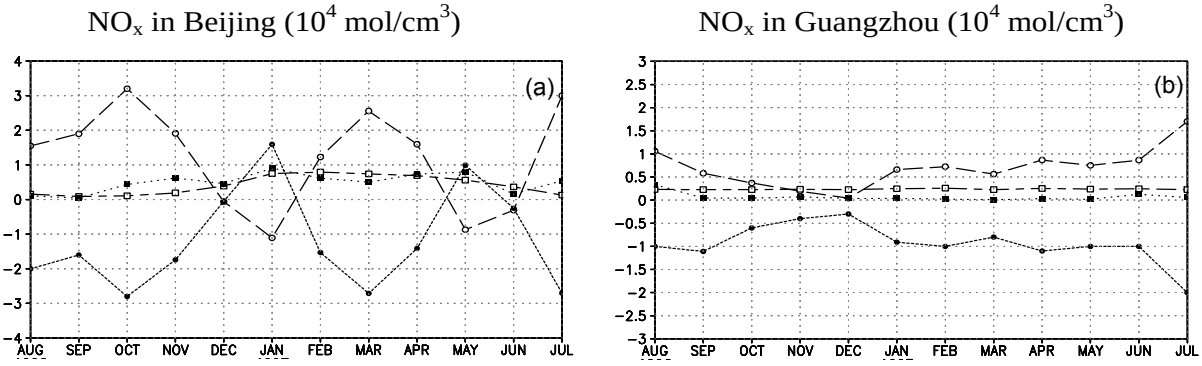

Fig. 5. The changes of NO_x budgets in (a) Beijing and (b) Guangzhou; open circles: transport, solid circles: chemical loss, open squares: local emission, solid squares: sum.

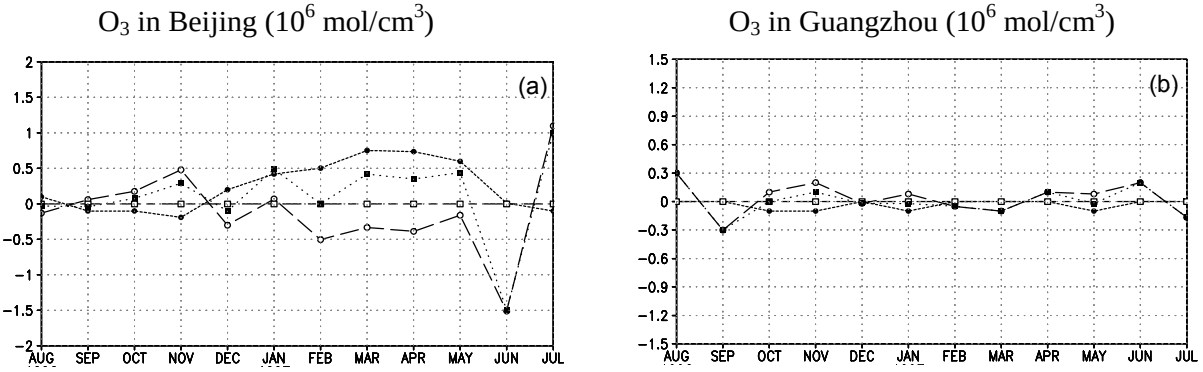

Fig. 6. The changes of ozone budgets in (a) Beijing and (b) Guangzhou; open circles: transport, solid circles: chemical loss, open squares: local emission, solid squares: sum.

Ozone in the upper troposphere is influenced by various processes, such as the exchange between stratosphere and troposphere, convective activities, advection, and chemical process. The effect of chemical process on ozone is under control of the radiation, temperature, and ozone concentration, etc. In spring, ozone increases from 40°N to the Arctic and the ratio of NO_2 to NO_x changes from 90% in winter to 45% in spring in the Arctic. This indicates that the efficiency of ozone production rises under the effect of sufficient sunlight. In summer, the atmospheric circulation transports trace gases in low-middle latitudes into high latitudes and the Arctic, such as NO_x, etc. Meanwhile, under the influence of strong sun radiation in the Arctic, ozone is produced efficiently, and chemical loss of ozone is the lowest in the year because of low ozone concentration in summer over the Arctic. Therefore, the ozone increase in Arctic is significant in spring and summer.

In order to comprehend deeply the model results, the budgets of NO_x and ozone at 200 hPa in Beijing and Guangzhou are analyzed. The processes that affect trace concentration consist of transport, chemical process, and local emission. Figure 5 depicts differences of the effect of each process between Case A (reference) and Case B in the NO_x budget. The result shows that in Beijing the NO_x transport is the main NO_x source during most of the year, and chemical loss nearly balances the sum of transports. In Guangzhou, the transport and chemical process play important roles in the NO_x budget, namely, the chemical loss counteracts the increase of the transport and emission. Figure 6, which highlights ozone budgets, is similar to Fig. 5. In winter and spring, the production of the chemical process in Beijing is the main factor in ozone increase, and the transport process causes ozone decrease; during summer and fall, the chemical process reduces the ozone a little, the transport process brings more ozone. In Guangzhou, the chemical process exhausts ozone, the ozone increase fully results from the

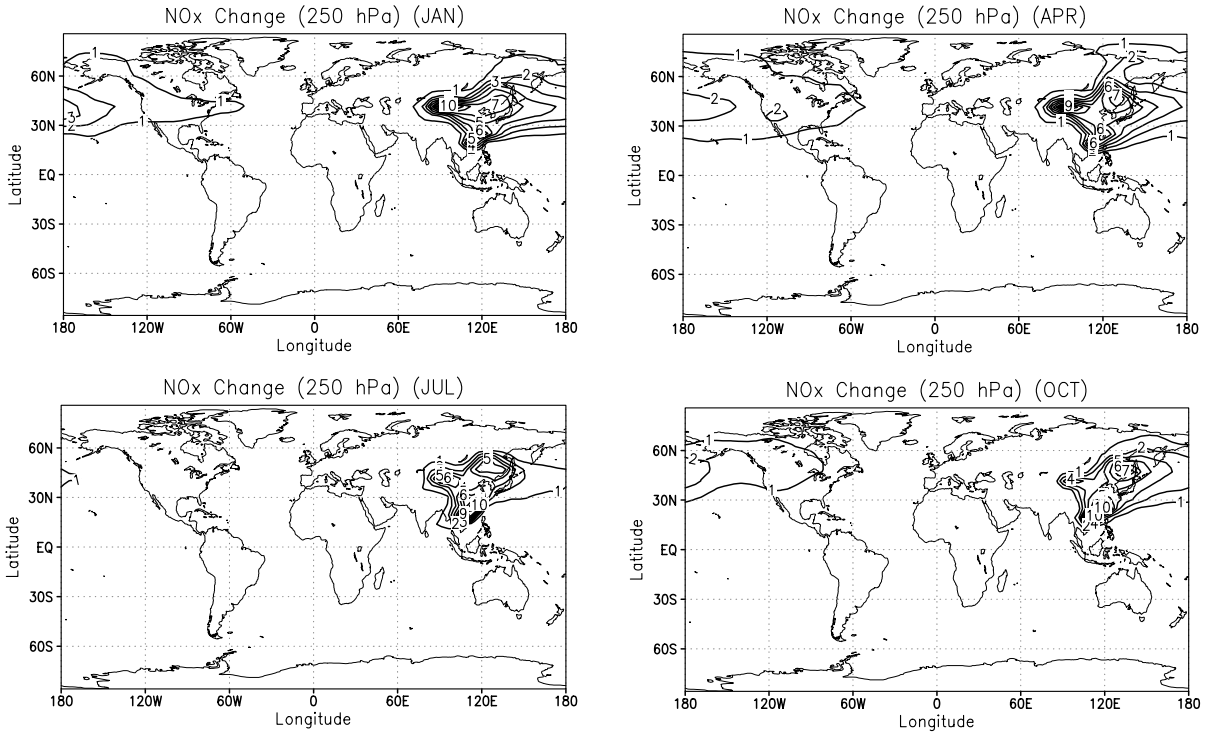

Fig. 7. The NO_x change at 250 hPa due to aircraft NO_x emission over China (units: pptv).

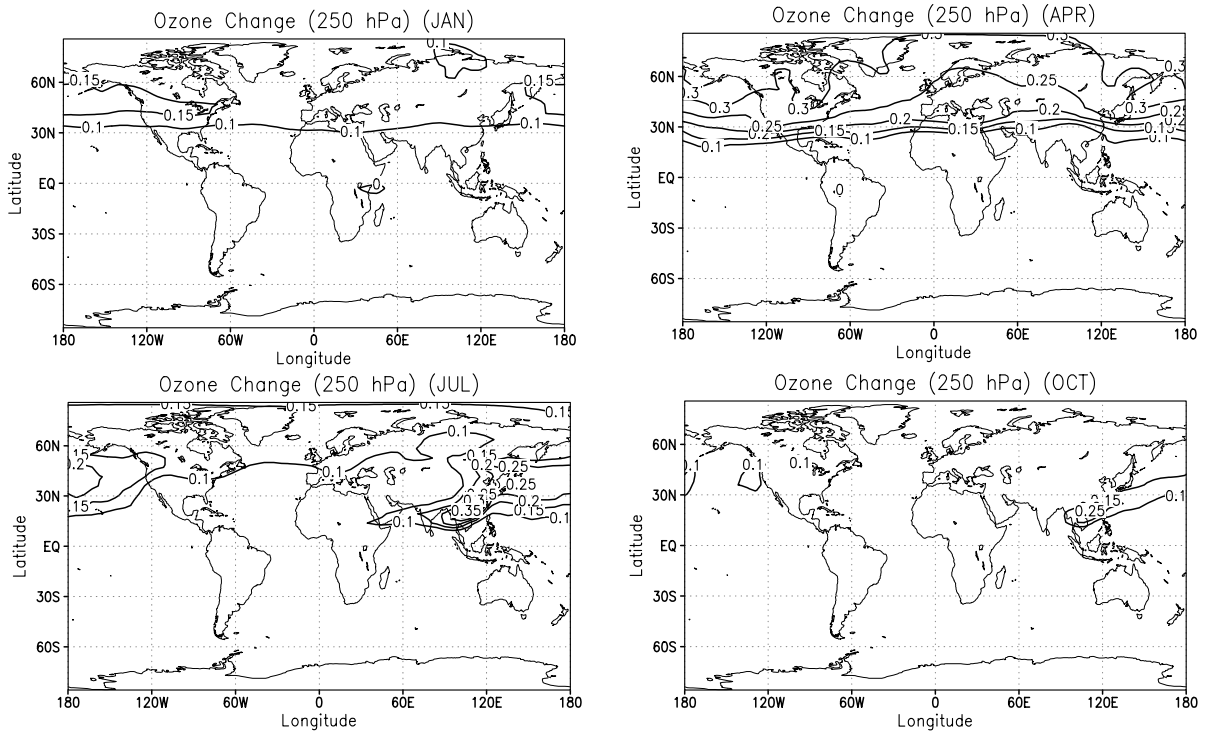

Fig. 8. The ozone change at 250 hPa due to aircraft NO_x emission over China (units: ppbv).

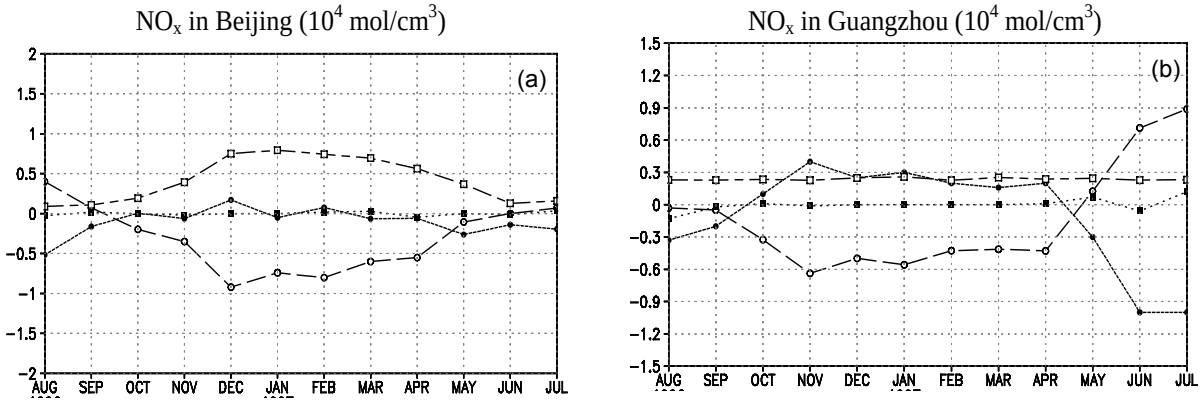

Fig. 9. The changes of NO_x budgets in (a) Beijing and (b) Guangzhou; open circles: transport, solid circles: chemical loss, open squares: local emission, solid squares: sum.

transport process. These results show that NO_x increase in China induced by aircraft emission mainly results from upward transportation; the ozone enhancement in northern China is produced by the local chemical process; in southern China, the chemical process depletes ozone.

The comparison of Case A and C can also confirm further the above results about NO_x and ozone budgets. The difference between Case A and C represents the influence of aircraft NO_x emission over China on NO_x and ozone (Figs. 7 and 8). From Fig. 7, it can be seen that seasonal variation of NO_x increase is little, and its maximum is only 10 pptv; there are three centers of NO_x increase, which are situated in the southeast part of China, the northeast part of China, and the Xinjiang Uygur Autonomous Region, respectively. In contrast to the results of global aircraft emission, it is found that the contribution of local aircraft NO_x emission to NO_x increase in northern China is almost one fifth of the total NO_x enhancements. Figure 8 shows ozone increase resulting from aircraft NO_x emission over China, whose maximum is only 0.35 ppbv in spring and minimum is 0.15 ppbv in fall. These ozone arguments are much less than those caused by global aircraft NO_x emission. The NO_x budgets (Fig. 9) show that during winter and spring, aircraft NO_x emission makes NO_x content increase, and transport process cause it to decrease; the effect of the chemical process in Beijing is small, but the effect in Guangzhou makes NO_x increase which is about equal to local aircraft NO_x emission. In summer, the chemical process reduces NO_x. Meanwhile, ozone change between Case A and C is mostly similar to ozone change between Case A and B, but is less. Under the influence of local aircraft NO_x emission, the chemical process in summer results in ozone increase while the transport process causes ozone decrease. Because the net effect of each process is relatively small, ozone changes little.

Based on the statistics of the CAAC, the growth rate of Chinese aviation is about 7%–8% per year in 1997. Chinese aviation transportation will double by 2010 if it continues to grow at this rate. When aircraft NO_x emission over China is doubled, NO_x and ozone arguments also rise in linear manner. The maximum of NO_x increase is about 20 pptv, and the maximum of ozone increase is about 0.7 ppbv in spring.

4. Summary

The above results show that the NO_x increase of 50 pptv from aircraft NO_x emission is 60% of total NO_x at 250 hPa in winter over northern China; and ozone increases by 5% at 250 hPa with 8 pptv in spring due to aircraft emission. But NO_x increase caused by local aircraft NO_x emission over China is no more than 10 pptv, and ozone increase is less than 0.4 ppbv. In northern China, NO_x increase is mainly from the transport process, and ozone increase is produced by the chemical process. Even if aircraft NO_x emission over China is doubled, the influence of aircraft NO_x emission over China will still be limited.

In addition, we pay more attention to the fact that individual model results are not fully the same as the others; the difference is mainly in seasonal variation of ozone enhancement. For example, OSLO-CTM2, OSLO-CTM1 model (Berntsen and Isaksen, 1999), and the STOCHEM model (Stevenson et al., 1997) have maxima in spring; IMAGES model (Brasseur et al., 1996) and CTMK model (Wauben et al., 1997) have maxima in summer; ECHAM3/CHEM model (Dameris et al., 1998) even has maxima in winter. Kraabol (2000) gave an explanation of ozone max-

ima in spring, saying that during spring the radiation is strong enough to cause an efficient photochemical ozone production due to the extra NO_x added from aircraft. The chemical backgroud conditions in the upper troposphere with low NO_x levels because of little convective activity (i.e., small lightning emission and low transport from the PBL) and high NMHC concentration accumulated during the winter, give an efficient ozone production (cf. Fig. 1 in Brasseur et al., 1996). Whether ozone maxima happen in winter and spring or summer, the ozone over northern China is remarkably influenced by aircraft NO_x emission, but the maxima in summer is relatively smaller than the other two cases.

Acknowledgments. This work was supported by the National Natural Science Foundation of China under Grant No.49805008 and the Young Grant of the Chinese Academy of Meteorological Sciences. This work also received support from the Research Council of Norway.

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飞机排放的NO_x对中国地区NO_x和臭氧的影响

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摘 要

根据2000年亚音速飞机排放的NO_x，利用三维全球化学输送模式(OSLOCTM2)研究亚音速飞机排放的NO_x对中国地区NO_x和臭氧的影响。模式结果表明：亚音速飞机排放的NO_x明显地影响中国北方地区，在1月份，250 hPa 高度NO_x的浓度增加大约50 pptv，最大的相对变化为60%；在4月份，250 hPa 高度臭氧增加8 ppbv，相对变化为5%。NO_x的增加主要是由于输送过程引起的，但臭氧的增加则是化学过程生成的结果。由于中国地区亚音速飞机排放的NO_x造成的NO_x的增加不超过10 pptv，而且臭氧增加小于0.4 ppbv。即使中国地区亚音速飞机排放的NO_x 增加一倍，这个影响仍然比较小。

关键词：飞机排放，NO_x，臭氧