

Intercontinental Transport and Chemical Transformation of Ozone and Its Precursors from East Asia

Oliver WILD and Hajime AKIMOTO

Frontier Research System for Global Change, Yokohama 236-0001, Japan

Abstract—The influence of anthropogenic trace gas emissions on the global troposphere is controlled by both chemical and dynamical processes, and the interaction between them controls any long-range impacts. We use the FRSGC-UCI 3-D chemical transport model to quantify the effects of anthropogenic emissions from the developing countries of East Asia on neighbouring continents and on the global troposphere. We show how the coupling of chemistry and meteorology leads to the largest changes in ozone over North America and Europe in spring and autumn, and demonstrate that future increases in precursor emissions may be expected to have significant effects on background levels of ozone at a hemispheric scale.

INTRODUCTION

Ozone in the troposphere is important both as a climate gas (e.g., Granier and Shine, 1999) and as the major source of the principal atmospheric oxidizing agent, the hydroxyl radical, OH (e.g., Chameides and Walker, 1973). Comparison of recent surface ozone measurements with those from the end of last century indicate that concentrations have increased significantly, a change which is largely attributable to increases in anthropogenic emissions of precursors (Volz and Kley, 1988). Ozone formation over the major heavily-populated industrial regions of the Northern Hemisphere has largely been studied in a regional context (e.g., Kotamarthi and Carmichael, 1990); however, these regions lie in a similar latitude band, and their pollution problems may therefore be readily coupled by atmospheric transport processes. The effects of East Asian emissions on ozone concentrations in the northwestern U.S. have been detected in measurement campaigns (Jaffe *et al.*, 1999) and corroborated by model studies (Berntsen *et al.*, 1999). In this paper, we quantify the production and export of ozone from the developing countries of East Asia and the magnitude of the subsequent ozone changes far downwind. We investigate how the chemical and dynamical processes controlling these ozone changes interact through the year, with the aim of improving our understanding of how anthropogenic changes may affect tropospheric composition.

These studies have been performed using the Frontier Research System for Global Change University of California, Irvine (FRSGC-UCI) Chemical Transport

Model (CTM) described in earlier work (Wild and Prather, 2000). The model is run using a year of meteorological data from the GISS II' GCM (Rind and Lerner, 1996) at $4^\circ \times 5^\circ$ resolution; the resolution has been degraded to $8^\circ \times 10^\circ$ to reduce computational costs, and the data is cycled for multiple-year runs. The model includes a detailed tropospheric chemistry scheme, including hydrocarbon oxidation, using the ASAD package (Carver *et al.*, 1997), and photolysis rates are calculated with the Fast-J scheme (Wild *et al.*, 2000) including a full treatment of scattering in clouds. The effects of stratospheric chemistry are simulated by applying first-order rates of change above a tracer-diagnosed tropopause (McLinden *et al.*, 2000). Emissions are principally taken from the EDGAR dataset (Olivier *et al.*, 1996), with additional NO sources from lightning; wet and dry deposition are based on surface vegetation type, precipitation and convective activity.

EFFECTS OF EAST ASIAN EMISSIONS

To determine the effects of anthropogenic emissions on regional and global ozone budgets, we increase the emissions of NO_x, CO and NMHC from surface fossil fuel sources in East Asia by 10%. This perturbation is small enough to avoid significant non-linear chemical influences on O₃ formation, but large enough to produce a measurable global response. It provides a reasonably realistic short-term scenario for the developing countries of the East Asian region, where NO_x emissions are currently thought to be increasing at 4–5% per year (van Aardenne *et al.*, 1999). In order to focus on these developing regions, we do not increase emissions over Japan.

We find that the additional emissions lead to an increase in net ozone production of 2.57 Tg/yr in the boundary layer over Asia; 77% of the additional ozone is transported out of the region, and the remaining 23% (0.58 Tg) is deposited. The ozone production efficiency of NO_x, defined as the number of molecules of ozone created per molecule of NO_x oxidised, averages 3.4 and ranges from 1.7 in January to 6.4 in July. Globally, the increase in net ozone production is 1.65 Tg/yr, and consequently the global troposphere outside the Asian region accounts for additional destruction of 0.92 Tg/yr. However, this is localised in the lower troposphere, where destruction of transported ozone dominates additional formation from transported precursors; in the upper troposphere the reverse is true, and additional production dominates. Between 400 and 150 hPa (about 6.4–13 km), additional ozone formation is 1.53 Tg/yr, while additional transport from the boundary layer is 0.65 Tg/yr; in-situ chemical processes therefore account for 70% of the additional pollution ozone at these altitudes.

LONG-RANGE IMPACTS FROM EAST ASIA

We demonstrate the extent of the atmospheric effects of these emission increases by considering the mean changes in ozone by season at the surface

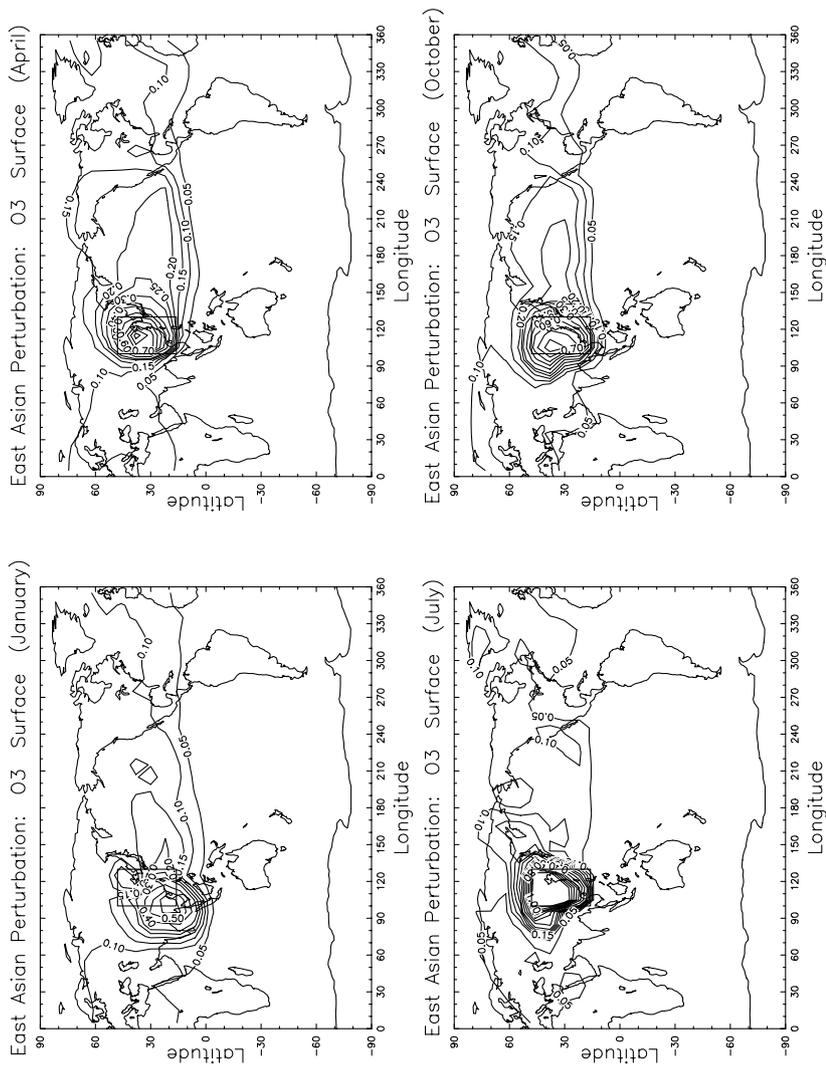


Fig. 1. Perturbation in monthly mean ozone (in ppbv) at the surface for January, April, July and October caused by 10% increased fossil fuel/industrial emissions over East Asia.

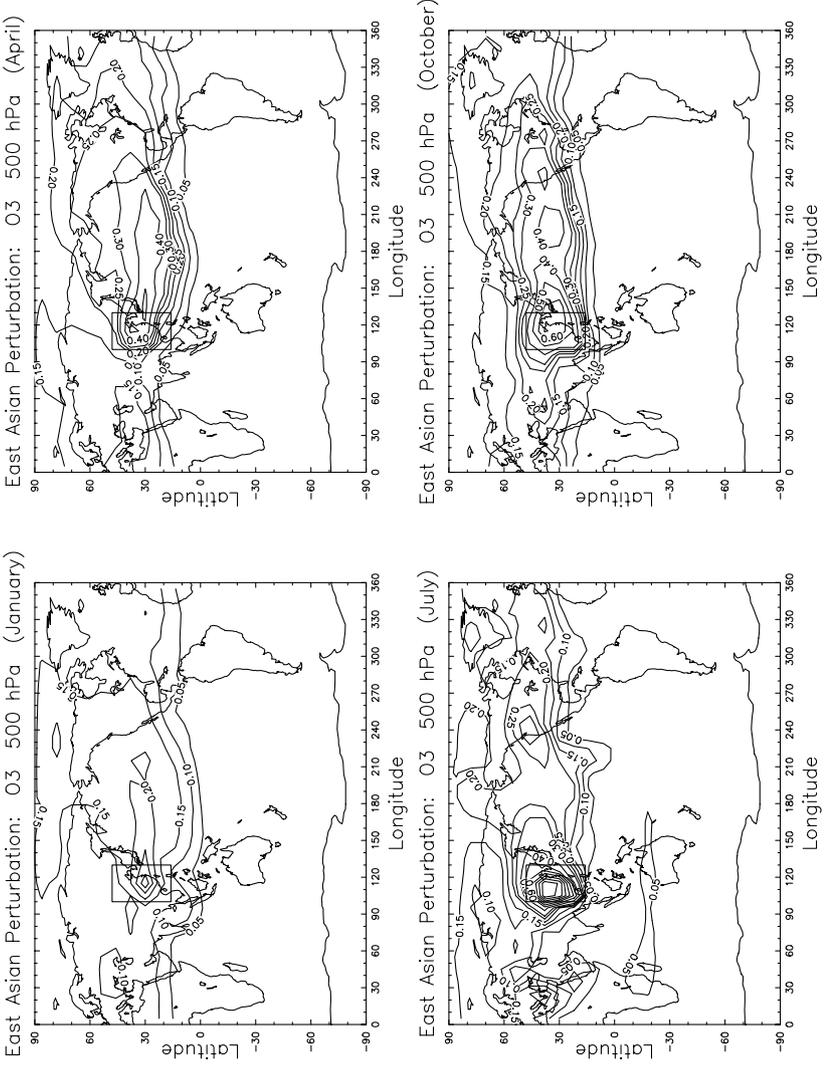


Fig. 2. Perturbation in monthly mean ozone (in ppbv) at 500 hPa for January, April, July and October.

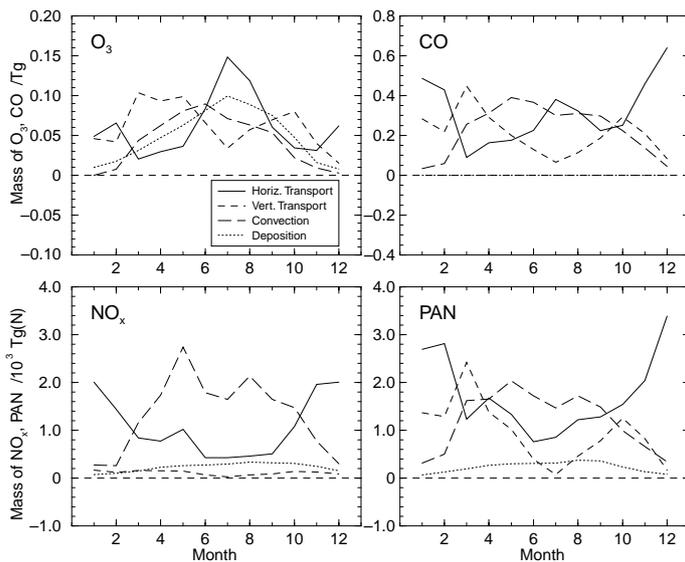


Fig. 3. Monthly mass fluxes of O_3 , CO, NO_x and PAN, formed from the increased emissions, through the physical processes controlling removal or export of each species from the continental boundary layer.

(Fig. 1) and in the mid-troposphere at 500 hPa (about 4.8 km, Fig. 2). In the winter, when chemical production is relatively slow, increases in boundary layer ozone over the emission region are small, close to zero in northern industrial regions. The main export from the boundary layer is south-westerly, towards the Indian subcontinent; vertical transport is limited, and enhancements over the Pacific Ocean are therefore small. In the spring, boundary layer production is much greater, and significant enhancements are seen throughout the emission region. Vertical transport is more efficient, and rapid horizontal transport in the mid- and upper troposphere, coupled with further ozone production, leads to relatively large enhancements over the Pacific and as far as the west coast of North America in the lower and mid-troposphere.

In summer, ozone production is greatest, and the regional boundary layer sees the largest enhancements; however, more stagnant meteorological conditions and shorter chemical lifetimes mean that while concentrations at 500 hPa are greatly enhanced over the region, the downwind effects are significantly smaller than in spring, particularly at lower levels. In the autumn, chemical production is reduced and chemical lifetimes are longer, and hence the situation is similar to the spring, although horizontal transport in the mid-troposphere is less strong, and precursor concentrations are rather lower, so that enhancements over the Pacific are smaller.

MECHANISMS FOR EXPORT

We illustrate the main mechanisms for export by considering changes in the mass flux of species out of the boundary layer over Asia due to the 10% emissions increase, see Fig. 3. We consider deposition, horizontal advection, deep convection and vertical transport associated with pressure systems, such as the frontal lifting described by Stohl and Trickl (1999). While convective activity is greatest in the summer, non-convective vertical transport peaks in spring and autumn; in summer and winter, high pressure systems over the continent lead to greater export of pollution by horizontal advection than by vertical transport. This effect is exaggerated for ozone as boundary layer enhancements are much greater in summer than winter, and for PAN, where the reverse is true. Despite the different seasonality, convective and non-convective lifting make similar contributions to the total annual vertical transport of long-lived species from the boundary layer; for short-lived species such as NO_x , convection is strongly dominant, as the timescale for convective transport is rather shorter than for other removal mechanisms. Total vertical transport of PAN and CO peaks in March, whilst that of O_3 and NO_x peaks in May. Chemical production of ozone above the boundary layer is most dependent on exported NO_x , and hence is more sensitive to convective than non-convective lifting, peaking in May.

INTERCONTINENTAL IMPACTS

To demonstrate the intercontinental impacts of the increased emissions, we show the perturbations in monthly mean ozone over East Asia (16–48°N, 100–130°E), the United States (24–48°N, 120–70°W) and Europe (32–64°N, 10°W–30°E) at the surface, 650 hPa (about 3 km) and 200 hPa (about 11 km), in Fig. 4. These perturbations reflect the seasonal variations shown in Fig. 1; the spatial variance demonstrates the variability in the monthly means over each region. Ozone is most greatly enhanced in the boundary layer over East Asia, as expected, but enhancements in the upper troposphere over the region are also large, and peak in August when the coupled effects of vertical transport and in-situ chemical production are greatest. The downwind impacts are greatest in spring and autumn, and are concentrated in the upper troposphere; surface enhancements are controlled principally by subsidence of these upper-tropospheric perturbations. Over the Pacific, mid- and upper-tropospheric transport is most rapid in spring and autumn at these latitudes; in summer the perturbations over the U.S. are smallest due to slower, more northerly transport, shorter chemical lifetimes, and more influence from air masses from the south. Over Europe the flow is more uniform, and southerly flow less prevalent in summer; although upper tropospheric enhancements are similar to those over the U.S., surface and mid-tropospheric enhancements are smaller.

The regional mean perturbations at the surface range from 0.04–0.16 ppbv over the U.S., and are only slightly smaller, 0.04–0.13 ppbv over Europe. Individual surface sites experience pollution episodes reaching a maximum of 0.31 ppbv at Boulder (40°N, –105°E) and 0.18 ppbv at Hohenpeissenberg (48°N,

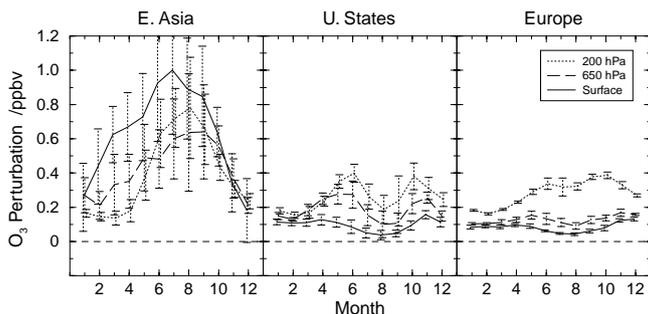


Fig. 4. Differences in monthly mean ozone mixing ratios (in ppbv) at the surface, 650 hPa and 200 hPa over East Asia, the U.S., and Europe from a 10% increase in emissions over Asia. Error bars indicate the spatial variance of monthly means within each region.

11°E). For a three-fold increase in Asian emissions, as expected between 1985 and 2010, Jacob *et al.* (1999) estimate monthly mean ozone perturbations of 2–6 ppbv in the western U.S. and 1–3 ppbv in the eastern U.S., peaking between April and June. Assuming a linear scaling from our 10% increased emissions, we find perturbations over the whole region of 0.8–3.2 ppbv, with a peak in October–November in addition to the spring peak. We also suggest that the perturbations over Europe will be almost as large, at 0.8–2.6 ppbv, and show a similar seasonality.

CONCLUSIONS

Increased emissions of oxidant precursors over East Asia leads to considerable extra ozone formation over the region, and significant enhancements at a hemispheric scale. In spring and autumn, efficient vertical transport out of the boundary layer, rapid horizontal transport in the upper troposphere, and significant chemical formation combine to give the greatest ozone enhancements downwind. While it has already been shown that increases in Asian emissions may be large enough to affect air quality, and hence human health and crop yields, over the U.S. (Jacob *et al.*, 1999), we demonstrate that the effects over Europe are only marginally smaller.

In addition, there is a direct impact on the oxidising capacity of the troposphere; while the East Asian region sees additional ozone formation, peaking in the boundary layer in summer, there is increased net destruction outside the region, as additional ozone formation from exported precursors is rather less than the additional destruction of directly-exported ozone. This may have direct implications for greenhouse gases such as methane and the HFCs, which have chemical lifetimes controlled by tropospheric OH. We also demonstrate that, while direct lifting of ozone formed in the continental boundary layer is important, chemical formation in the upper troposphere accounts for about 70% of the additional ozone from pollution sources at these altitudes.

REFERENCES

- Berntsen, T. K., S. Karlsdottir, and D. A. Jaffe, Influence of Asian emissions on the composition of air reaching the North Western United States, *Geophys. Res. Lett.*, **26**, 2171–2174, 1999.
- Carver, G. D., P. D. Brown, and O. Wild, The ASAD atmospheric chemistry integration package and chemical reaction database, *Computer Physics Communications*, **105**, 197–215, 1997.
- Chameides, W. L. and J. C. G. Walker, A photochemical theory for tropospheric ozone, *J. Geophys. Res.*, **78**, 8751–8760, 1973.
- Granier, C. and K. P. Shine, Climate effects of ozone and halocarbon changes, in Scientific Assessment of Ozone Depletion: 1998, WMO Global Ozone Research and Monitoring Project, Report 44, 1999.
- Jacob, D. J., J. A. Logan, and P. P. Murti, Effect of rising Asian emissions on surface ozone in the United States, *Geophys. Res. Lett.*, **26**, 2175–2178, 1999.
- Jaffe, D. *et al.*, Transport of Asian air pollution to North America, *Geophys. Res. Lett.*, **26**, 711–714, 1999.
- Kotamarthi, V. R. and G. R. Carmichael, The long range transport of pollutants in the Pacific Rim region, *Atmos. Env.*, **24**, 1521–1534, 1990.
- McLinden, C. A., S. Olsen, B. Hannegan, O. Wild, M. J. Prather, and J. Sundet, Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, *J. Geophys. Res.*, **105**, 14,653–14,665, 2000.
- Olivier, J. G. J. *et al.*, Description of EDGAR Version 2.0, RIVM/TNO report 771060 002, RIVM, Bilthoven, December 1996.
- Rind, D. and J. Lerner, Use of on-line tracers as a diagnostic tool in general circulation model development: 1. Horizontal and vertical transport in the troposphere, *J. Geophys. Res.*, **101**, 12,667–12,683, 1996.
- Stohl, A. and T. Trickl, A textbook example of long-range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe, *J. Geophys. Res.*, **104**, 30,445–30,462, 1999.
- van Aardenne, J. A., G. R. Carmichael, H. Levy, D. Streets, and L. Hordijk, Anthropogenic NO_x emissions in Asia in the period 1990–2020, *Atmos. Env.*, **33**, 633–646, 1999.
- Volz, A. and D. Kley, Evaluation of the Montsouris series of ozone measurements made in the nineteenth century, *Nature*, **322**, 240–242, 1988.
- Wild, O. and M. J. Prather, Excitation of the primary tropospheric chemical mode in a global 3-D model, *J. Geophys. Res.*, **105**, 24,647–24,660, 2000.
- Wild, O., X. Zhu, and M. J. Prather, Fast-J: Accurate simulation of in- and below-cloud photolysis in Global Chemical Models, *J. Atmos. Chem.*, **37**, 245–282, 2000.

O. Wild (e-mail: oliver@jamstec.go.jp) and H. Akimoto