Impacts of East Asia on the Tropospheric Ozone Budget

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Abstract. We present ozone budgets over the three major northern hemisphere industrialized regions calculated using a 3-D chemical transport model. For each region we estimate the magnitudes of the production and export of ozone and its precursors, and show how these vary through the year. For East Asia, we investigate the degree to which increased emissions affect the other regions, and demonstrate that in most situations the global atmosphere away from the continental boundary layer acts as a net chemical sink for ozone formed from anthropogenic precursor emissions.

Introduction

The export of ozone from the major northern hemisphere industrialized population centres of North America, Europe and East Asia is thought to make a significant contribution to ozone concentrations in the troposphere. The magnitude of the summertime export from North America, first estimated by *Parrish et al.* [1993], has been assessed using 3-D CTMs [*Jacob et al.*, 1993] and extended to seasonal effects [*Liang et al.*, 1998], and similar estimates have been made for Europe and East Asia. In this paper we present exports from all three regions calculated with a global model, focusing on the magnitude of the impact of each region on the global troposphere, and assessing the downwind influences. In particular, we focus on the rapidly-developing East Asian region and the seasonality of its effects on North America and Europe.

Modelling Studies

We use the UCI global tropospheric CTM from the University of California, Irvine, running off-line with a year of winds from the GISS GCM. The model has a resolution of $4°\times5°$ in the horizontal, and 9 vertical levels from the surface to 10 hPa; for the calculations presented here the resolution has been degraded to $8°\times10°$ to reduce computational requirements. The model includes a detailed treatment of tropospheric chemistry including the oxidation of representative hydrocarbons in addition to HO_x, O_x, NO_x and CH_4 chemistry [*Wild and Prather*, 2000]. Photolysis rates are calculated online with the Fast-J scheme [*Wild et al.*, 2000] including full treatment of scattering in clouds. Emissions are based on 1990 data from EDGAR v.2 [*Olivier et al.*, 1996], with additional emissions for NO from lightning and aircraft sources, and with emissions of isoprene from *Guenther et al.* [1995] reduced to 220 Tg(C)/yr. For these studies, the flux of O_3 from the stratosphere is 475 Tg/yr, surface deposition removes 704 Tg/yr, and there is net chemical production of 229 Tg/yr.

Continental Budgets

The annual budgets for ozone and NO_x in the boundary layer over the main industrialized regions are shown in Table 1. The net O_3 production includes chemical formation from precursors from all sources; the net export term is the difference between transport of chemically-produced O_3 out of the region and transport of free-tropospheric O_3 in. Each region is a net exporter of O_3; in addition, about 20% of emitted nitrogen is exported from the boundary layer as NO_x, most of which is lifted into the free troposphere and leads to further O_3 production above the region. Total production over each region between the surface and 150 hPa (about 13 km) is indicated at the bottom of the table.

Table 1. Annual Regional O_3 and NO_x Budgets

	U.States	Europe	E.Asia
Boundary Layer O_3 (Tg/yr)			
Net O_3 production	60.5	30.7	49.1
O_3 deposition	49.1	27.2	34.5
Net O_3 export	11.4	3.4	14.7
Boundary Layer N (Tg(N)/yr)			
NO_x emissions	7.68	5.79	4.67
NO_x+PAN export	1.41	1.24	1.14
Emitted N exported as NO_x	18%	21%	24%
Total Regional O_3 (Tg/yr)			
Net O_3 production	81.5	49.1	74.7
Net O_3 export	32.3	21.9	40.2

Annual exports from the U.S. boundary layer (11 Tg(O_3), 1.4 Tg(N)) are similar to those derived by *Liang et al.*, [1998] (6 Tg(O_3), 1.4 Tg(N)). Exports from the European boundary layer are smaller in magnitude due to lower production efficiencies and wintertime titration; exports from East Asia are larger due to greater vertical transport.

Anthropogenic Emissions

The effects of anthropogenic emissions are investigated by applying a 10% perturbation to emissions of NO_x, CO and NMHCs from these sources in each region in turn. The differences in the budgets are shown in Table 2. The annual mean production efficiency (net number of molecules of O_3 formed per molecule of NO_x oxidized) is largest over East Asia, reflecting the lower NO_x emission intensities. In each region, most (66–77%) of the additional O_3 formed is exported. Global net O_3 production is also elevated, but for the U.S. and East Asia this is less than the additional boundary layer production, and hence we find that regions outside the perturbed boundary layer act as a net sink of O_3. Although chemical production is enhanced globally due to exported NO_x, chemical loss of O_3 is also enhanced due to the additional O_3 exported; in the upper troposphere, the former effect dominates globally, but in the mid- and lower troposphere the additional loss dominates. For Europe, in contrast to the other regions, reduced chemical O_3 formation in the boundary layer from lower ozone production efficiencies and wintertime titration, and greater deposition downwind

over the Eurasian continent lead to reduced chemical O_3 removal, and net production dominates.

Table 2. Production and Fate of Anthropogenic O_3 From 10% Increased Emissions in Each Region

	U.States	Europe	E.Asia
Boundary Layer O_3 (Tg/yr)			
Net O_3 production	2.48	1.30	2.57
O_3 deposition	0.85	0.36	0.58
Net O_3 export	1.63	0.94	1.99
Fraction O_3 exported	66%	72%	77%
O_3 production efficiency	1.24	0.92	2.74
Global O_3 (Tg/yr)			
Net O_3 production	2.13	2.00	1.65
O_3 deposition	2.04	1.91	1.48
Additional Burden	0.09	0.09	0.17

East Asia

For increased East Asian emissions, boundary layer O_3 production peaks in the summer, when temperatures are highest and insolation greatest; deposition and export follow this peak. Export of NO_x from the entire region (up to 150 hPa) is greatest in the winter, but boundary layer exports remain relatively constant due to increased convection in summertime. In the mid-troposphere net O_3 production is enhanced in spring and autumn, but very little in summer; this represents a trade-off between greater summertime O_3 production and greater loss of O_3 transported from the boundary layer. In the upper troposphere production is enhanced in the summer when convection of NO_x from the boundary layer is greatest. These regions contribute an extra 0.86 Tg(O_3)/yr, 25% of the total additional O_3 production over the region.

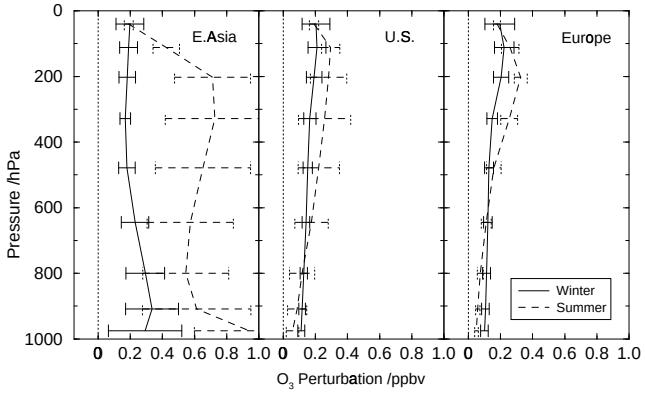

Figure 1. Profiles of ozone enhancement (in ppbv) over each region in winter (DJF) and summer (JJA) for 10% increased emissions over East Asia. Error bars show the variance of grid-box monthly means within each region.

O_3 mixing ratios are enhanced at all levels, but the absolute increases are greatest in the mid- and upper troposphere, see Figure 1. During the winter, the increases are modest and relatively uniform in altitude; in the summer Asian regional O_3 production is greater, and the variability in the enhancements over different parts of the region is rather greater over the emission region and downwind over North America. Upper tropospheric production is enhanced globally; greater deposition over the continents erodes the enhancement at the surface, and leads to steeper tropospheric gradients.

Consequences of Increased Emissions

An increase of 10% in the 1990 anthropogenic emissions for East Asia leads to an enhancement of 70–190 pptv in mean surface O_3 mixing ratios over the U.S. Assuming a linear trend, this is similar to the 1–6 ppbv enhancement calculated by *Jacob et al.* [1999] for a 200% enhancement. The enhancement is greatest in spring and autumn, when transport across the Pacific is quickest, and a minimum in summer. Mean surface O_3 mixing ratios over Europe are enhanced by 35–150 pptv, only slightly less than those over the Eastern U.S. For reference, East Asian emissions are currently increasing at about 5%/yr [*van Aardenne et al.*, 1999].

Outside the polluted boundary layer, the principal enhancements occur in the upper troposphere. The radiative forcing effects of tropospheric ozone are a maximum in this region, and hence it is likely that increases in anthropogenic emissions may lead to significant climate effects through increases in O_3.

We note that increased anthropogenic emissions may lead to greater net O_3 loss in the lower and mid-troposphere away from the continental boundary layer. This effect may be reduced for European emissions, where deposition downwind plays a greater role, but may increase the importance of the Atlantic and Pacific troposphere as chemical sinks of O_3.

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