

Effect of rising Asian emissions on surface ozone in the United States

Daniel J. Jacob, Jennifer A. Logan and Prashant P. Murti

Division of Engineering and Applied Science, and Department of Earth and Planetary Sciences, Harvard University

Abstract. The effect of increasing fossil fuel combustion in eastern Asia on surface O₃ air pollution in the United States is examined with a global three-dimensional tropospheric chemistry model. Tripling of Asian anthropogenic emissions from 1985 to 2010 is expected to increase monthly mean O₃ concentrations by 2–6 ppbv in the western United States and by 1–3 ppbv in the eastern United States, the maximum effect being in April–June. This increase would more than offset the benefits of 25% domestic reductions in anthropogenic emissions of NO_x and hydrocarbons in the western United States. Asian influence may be less under the stagnant conditions leading to violations of the U.S. air quality standard. Nevertheless, our results suggest that a global perspective is necessary when designing a strategy to meet regional O₃ air quality objectives.

1. Introduction

The U.S. national air quality standard for O₃ concentrations in surface air was revised in 1997 from 120 ppbv (1-hour average, not to be exceeded more than once per year) to 80 ppbv (8-hour average, not to be exceeded more than three times per year). Nonattainment areas under the old standard were mainly urban and suburban, or downwind of large metropolitan centers [EPA, 1996a]. The new standard expands the geographical scale of the regulatory problem. Saylor *et al.* [1998] find that many rural areas of the United States that were in compliance with the old standard will likely fail the new standard.

As the O₃ air quality standard decreases, better understanding of the O₃ background becomes increasingly important in the design of air pollution control strategies. Background O₃ concentrations in surface air over the United States, as derived from measurements at clean sites or from the intercept of correlations between O₃ and NO_y (total reactive nitrogen oxides), are in the range 25–55 ppbv [Logan, 1989; Altshuler and Lefohn, 1996; Hirsch *et al.*, 1996]. This background can be largely attributed to transport from outside the United States, and it probably includes a major anthropogenic component. Model simulations show that with zero NO_x emissions in North America, and emissions elsewhere at present-day levels, long-range transport from outside the continent would maintain 20–40 ppbv O₃ in surface air over the United States in summer [Liang *et al.*, 1998]. The lifetime of O₃ in the free troposphere is sufficiently long that pollution from Europe, Asia, and North America can circumnavigate the globe and elevate O₃ throughout north-

ern midlatitudes. Measurements in the free troposphere at mountain sites over Europe show O₃ concentrations rising from 10 ppbv in 1890 to 20 ppbv in 1930 to 50 ppbv today [Marenco *et al.*, 1994].

We present here a modeling investigation of the effect of rising Asian emissions on O₃ concentrations in surface air over the United States. Energy consumption in China, India, and most of the rest of Asia increased by 5% yr⁻¹ over the period 1970–1995, and this rate of growth is expected to continue at least until 2015 [United States Department of Energy, 1997]. Fossil fuel combustion is the main source of energy in Asia and proceeds with minimal emission controls except in Japan. We may therefore expect NO_x and hydrocarbon emissions to grow in parallel to fossil fuel combustion. In contrast, little change is expected in U.S. emissions of NO_x over the next decade [EPA, 1996b]. An emission inventory for eastern Asia (east of Afghanistan) gives a fossil fuel combustion source of NO_x of 4.2 Tg N yr⁻¹ in 1985 [Kato and Akimoto, 1992], as compared to 6.3 Tg N yr⁻¹ in the United States [EPA, 1996b]. Kato and Akimoto [1992] report a 5.5% yr⁻¹ growth of NO_x emissions in eastern Asia outside Japan from 1975 to 1987. Assuming a 5% yr⁻¹ growth outside Japan from 1985 to 2010, we expect the east Asian source of NO_x in 2010 to amount to three times the 1985 Asian source and twice the U.S. source. Even a small contribution of this increasing Asian source to surface O₃ concentrations in the United States would make it more difficult for the United States to meet its air quality standard.

2. Model

Our analysis uses the Harvard-GISS global three-dimensional model of tropospheric chemistry. This model uses a 1-year archive of meteorological fields from a general circulation model (GCM) developed at the Goddard Institute for Space Studies (GISS) [Hansen *et al.*, 1983]. The horizontal resolution is 4°×5°. There are nine sigma levels in the vertical; the three lowest levels extend approximately to 0.5, 1.2, and 2.7 km above the surface. Meteorological information is updated every 4 hours. The model is too coarse to resolve urban plumes, but it shows some ability at simulating regional O₃ pollution episodes in the eastern United States [Jacob *et al.*, 1993].

We use here the latest version of the model described by Horowitz and Jacob [1999] which includes 80 species (24 chemical tracers) to describe tropospheric O₃-NO_x-hydrocarbon chemistry. The chemical mechanism is integrated with a fast Gear solver [Jacobson and Turco, 1994]. The model includes comprehensive inventories of anthropogenic and natural emissions. Emissions from fossil fuel combustion are from the 1985 Global Emission Inventory Activity (GEIA) inventory for NO_x [Benkovitz *et al.*, 1996]

Copyright 1999 by the American Geophysical Union.

Paper number 1999GL900450.
0094-8276/99/1999GL900450\$05.00

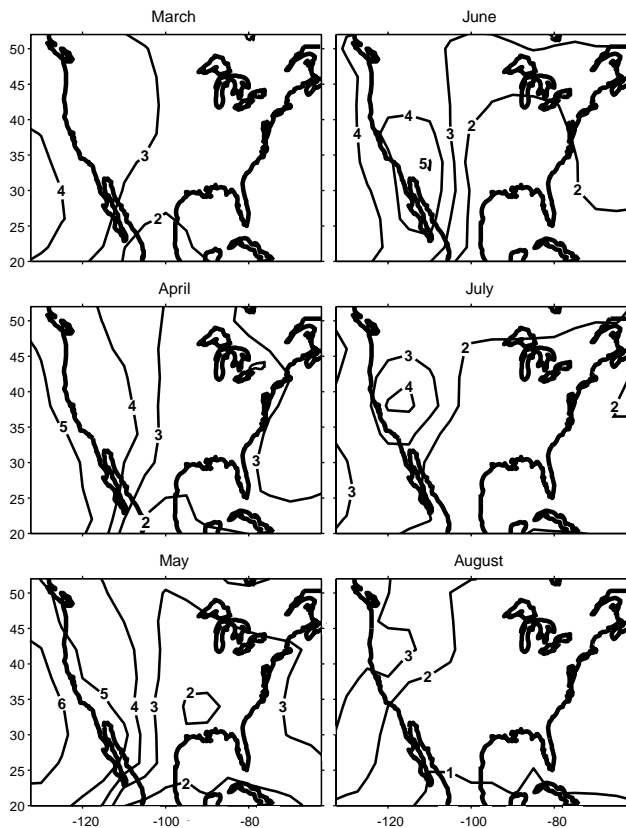


Figure 1. Increase of O_3 concentrations (ppbv) in surface air over the United States caused by tripling of anthropogenic emissions in eastern Asia from 1985 to 2010. Values are model monthly means for March–August.

and the Piccot *et al.* [1992] inventory for hydrocarbons. The GEIA inventory for eastern Asia is that of Kato and Akimoto [1992].

Global evaluation of the model with observations for O_3 and its precursors was presented by Wang *et al.* [1998]. The model reproduces well the general climatology of tropospheric ozone determined from ozone-sonde observations. A more specific evaluation with surface observations over the United States was presented by Horowitz *et al.* [1998] for summer and by Liang *et al.* [1998] for other seasons. The model reproduces observed monthly mean concentrations of O_3 at non-urban U.S. sites usually to within 10 ppbv. The largest discrepancy is in the south-central United States in summer where the model overestimates O_3 by more than 20 ppbv. This overestimate is due to a northeastward displacement of the Bermuda High in the GCM relative to its climatological position.

In this work, we conduct a sensitivity simulation for the year 2010 where the fossil fuel combustion source from Asia east of $60^\circ E$ (from Afghanistan eastward) is tripled from 1985 levels, while emissions from North America and Europe are held at 1985 levels. We also conduct sensitivity simulations where anthropogenic emissions of NO_x and non-methane hydrocarbons (NMHCs) in the United States are reduced by 25% from 1985 levels, with Asian emissions either at 1985 levels or at 2010 levels. We conduct a further sensitivity simulation with zero anthropogenic emissions in eastern Asia and 1985 levels elsewhere (zero case). All simulations are conducted for a period of 15 months from June 1

(year 1) to August 31 (year 2) using the same meteorological archive. The first 9 months of simulation are for initialization. We report results for March through August of year 2.

3. Results and discussion

Figure 1 shows the increases in monthly mean O_3 concentrations over the United States in March–August caused by tripling of anthropogenic emissions in eastern Asia from 1985 to 2010. Results are for the lowest model layer (0–500 m above the surface). The maximum increase is 5–6 ppbv over California in May. The increases are larger in the western United States (2–6 ppbv) than in the east (1–3 ppbv), reflecting the prevailing flow in the lower troposphere during spring and summer. The western United States are usually under the influence of westerly flow from the Pacific, while the eastern United States are usually under the influence of southerly flow from the Gulf of Mexico. Summer maxima of Asian influence over the western deserts (Figure 1) are due to strong dry convection entraining Asian pollution from aloft. Comparison of the 2010 case to the simulation with no anthropogenic Asian emissions (zero case) shows similar geographical patterns of Asian enhancement but doubled in magnitude. Tripling of Asian emissions from 1985 to 2010 thus doubles the magnitude of the Asian enhancement over the United States.

Subsidence of Asian O_3 transported in the free troposphere is the principal mechanism for Asian influence on surface O_3 over the United States in the model. Subsidence of Asian NO_x and PAN followed by O_3 production in the U.S. boundary layer is far less important. We find that increasing Asian emissions cause a slight decrease of NO_x concentrations in surface air over the United States because the higher O_3 concentrations lead to a shorter chemical lifetime of NO_x . Ozone concentrations in the free troposphere (700–300 hPa) over the United States and the extra tropical North Pacific increase by 5–10 ppbv as Asian emissions triple from 1985 to 2010 (Figure 2).

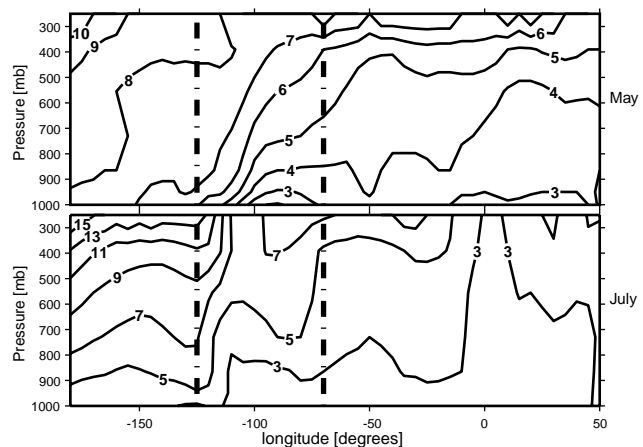


Figure 2. Increase of O_3 concentrations (ppbv) at $34^\circ N$ caused by tripling of anthropogenic emissions in eastern Asia from 1985 to 2010. Values are model monthly means for May and July and are shown as a function of longitude (degrees) and pressure. The dashed lines are the boundaries of the United States.

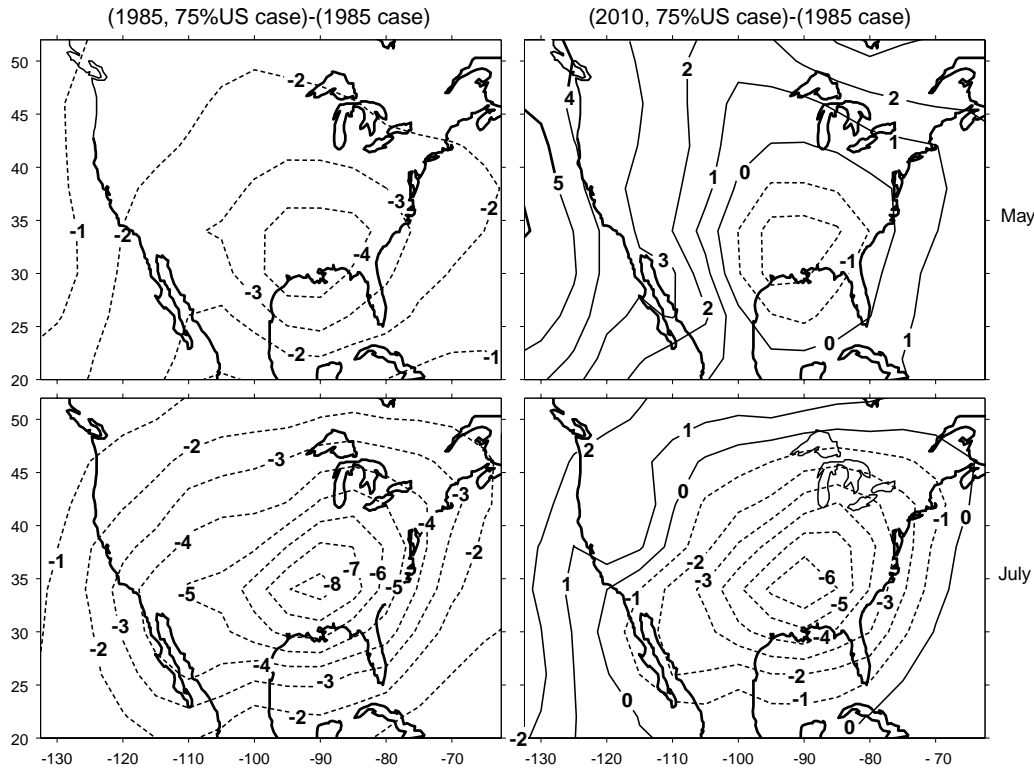


Figure 3. Changes of O_3 concentrations (ppbv) in surface air over the United States relative to 1985 values when 25% domestic reductions in anthropogenic NO_x and nonmethane hydrocarbon (NMHC) emissions are implemented, with Asian emissions at 1985 levels (left panels) and at 2010 levels (right panels). Values are model monthly means for May (top) and July (bottom).

Asian pollution influence on surface air in the United States in the model is maximum in April–June (Figure 1). In summer, deep monsoonal convection over eastern Asia and relatively weak westerlies shift the Asian pollution plume to higher altitudes and limit its long-range transport (Figure 2). Westerly transport of Asian air across the North Pacific is known to be strongest in the spring [Merrill, 1989; Savoie *et al.*, 1989]. Measurements by Jaffe *et al.* [1999] on the northwest coast of the United States in March–April reveal events of Asian pollution characterized by high CO and hydrocarbons; O_3 was not enhanced in these events, but more recent aircraft observations at the same location in spring show high O_3 layers associated with Asian pollution [D. Jaffe, personal communication, 1999].

Our model features little day-to-day variability (only a few ppbv) in the Asian pollution enhancement of O_3 concentrations over the United States. This result reflects an erroneous constancy of the GCM air flow in the free troposphere over the North Pacific. In a previous simulation of ^{222}Rn over the North Pacific, we found that the GISS GCM provides a good representation of the climatological mean westerly flow and its seasonal variation but has almost no day-to-day variability in the free troposphere [Balkanski *et al.*, 1992]. In contrast, aircraft observations of ^{222}Rn over the North Pacific show a large variability of Asian influence [Andreae *et al.*, 1988; Kritz *et al.*, 1990; Jaffe *et al.*, 1999].

The magnitude of the Asian pollution influence on surface O_3 over the United States must be considered from the perspective of the strongly nonlinear response of O_3 concentrations to changes in precursor emissions [Liu *et al.*, 1987;

Sillman *et al.*, 1990]. Model studies for the United States suggest that reducing domestic anthropogenic NO_x emissions by 50% would decrease mean summertime O_3 concentrations in surface air by typically 10–15 ppbv, while reducing anthropogenic NMHC emissions by 50% would have less than a 5 ppbv effect [McKeen *et al.*, 1991; Liang *et al.*, 1998]. In our standard model simulation with Asian emissions at 1985 levels, we find that a 25% reduction of anthropogenic NO_x and NMHC emissions in the United States decreases O_3 concentrations by 2–4 ppbv in May and by 2–8 ppbv in July over most of the country (Figure 3, left panels). Results from a simulation with the same 25% domestic reductions but with Asian emissions at 2010 levels are shown in the right panels of Figure 3. We find that in May the rise in Asian emissions from 1985 to 2010 more than offsets the benefits of 25% reductions in domestic emissions over the western half of the United States, and negates much of the effect of reduced emissions in the eastern half. The pattern is similar but weaker in summer.

Our model analysis thus suggests that increasing fossil fuel combustion in eastern Asia over the next decades will cause significant degradation of mean air quality in the western United States, with less effect in the east. From the perspective of meeting the air quality standard, it is critical to determine how the Asian influence correlates on a day-to-day basis with the stagnant weather conditions conducive to O_3 pollution episodes. One might expect a negative correlation [Jacob *et al.*, 1993] since pollution episodes are usually associated with strong subsidence inversions suppressing transport from the free troposphere. The insufficient

weather variability in the GISS GCM does not allow us to examine this issue here. We plan to examine it in the future with a simulation driven by assimilated meteorological observations.

We have assumed that emissions of O₃ precursors in eastern Asia from 1985 to 2010 would grow in parallel to fossil fuel combustion. This assumption is simplistic, as the mix of anthropogenic sources will evolve. Degradation of public health and agriculture [Chameides *et al.*, 1999] may eventually spur air pollution control measures in Asia. In our simulation for 2010 we find monthly mean surface O₃ concentrations over eastern Asia in the range 50–80 ppbv (spring-summer), comparable to values in the eastern United States at present. Recent O₃ measurements in China reported by Chameides *et al.* [1999] indicate episodes exceeding 100 ppbv, and model calculations reported in that same study suggest that episodes of up to 180 ppbv might be expected if emissions double.

Acknowledgments. This research was supported by the Electric Power Research Institute (EPRI) and by the NASA Atmospheric Chemistry Modeling and Analysis Program (ACMAP). We thank Pradeep Saxena for encouraging us to apply our model to this problem. This paper is dedicated to the memory of Geraldine M. Gardner.

References

- Altshuller, A. P. and A. S. Lefohn, Background ozone in the planetary boundary layer over the United States, *J. Air Waste Management Assoc.*, *46*, 134–145, 1996.
- Andreae, M. O., H. Berresheim, T. W. Andreae, M. A. Kritz, T. S. Bates, and J. T. Merrill, Vertical distribution of dimethylsulfide, sulfur dioxide, aerosol ions, and radon over the Northeast Pacific Ocean, *J. Atmos. Chem.*, *6*, 149–173, 1988.
- Balkanski, Y. J., D. J. Jacob, R. Arimoto, and M. A. Kritz, Long-range transport of radon-222 over the North Pacific Ocean: Implications for continental influence, *J. Atmos. Chem.*, *14*, 353–374, 1992.
- Benkovitz, C. M., M. T. Schultz, J. Pacyna, L. Tarrason, J. Dignon, E. C. Voldner, P. A. Spiro, J. A. Logan, and T. E. Graedel, Global gridded inventories for anthropogenic emissions of sulfur and nitrogen, *J. Geophys. Res.*, *101*, 29,239–29,253, 1996.
- Chameides, W. L., L. Xingsheng, T. Xiaoyan, Z. Xiuji, L. Chao, C. S. Kiang, J. St. John, R. D. Saylor, S. C. Liu, K. S. Lam, T. Wang, and F. Giorgi, Is ozone pollution affecting crop yields in China?, *Geophys. Res. Lett.*, *26*, 867–870, 1999.
- Environmental Protection Agency (EPA), National Air Quality and Emissions Trends Report, 1995, *EPA-454/R-96-005*, U.S. EPA, Research Triangle Park, NC 27111, 1996a.
- Environmental Protection Agency (EPA), National Air Pollutant Emissions Trends, 1900–1995, *EPA-454/R-96-007*, U.S. EPA, Research Triangle Park, NC 27111, 1996b.
- Hansen, J., G. Russell, D. Rind, P. Stone, A. Lacis, S. Lebedeff, R. Ruedy, and L. Travis, Efficient three-dimensional global models for climate studies: Models I and II, *Mon. Weath. Rev.*, *111* 609–662, 1983.
- Hirsch, A.I., J. W. Munger, D. J. Jacob, L. W. Horowitz, and A. H. Goldstein, Seasonal variation of the ozone production efficiency per unit NO_x at Harvard Forest, Massachusetts, *J. Geophys. Res.*, *101*, 12,659–12,666, 1996.
- Horowitz, L. W., and D. J. Jacob, Global impact of fossil fuel combustion on atmospheric NO_x, *J. Geophys. Res.*, in press, 1999.
- Horowitz, L. W., J. Liang, G. M. Gardner, and D. J. Jacob, Export of reactive nitrogen from North America during summertime, *J. Geophys. Res.*, *103*, 13,451–13,476, 1998.
- Jacob, D. J., J. A. Logan, G. M. Gardner, R. M. Yevich, C. M. Spivakovsky, S. C. Wofsy, S. Sillman, and M. J. Prather, Factors regulating ozone over the United States and its export to the global atmosphere, *J. Geophys. Res.*, *98*, 14817–14826, 1993.
- Jacobson, M. Z., and R. P. Turco, SMVGEAR: a sparse-matrix, vectorized Gear code for atmospheric models, *Atmos. Environ.*, *28*, 273–284, 1994.
- Jaffe, D., and 12 others, Transport of Asian air pollution to North America, *Geophys. Res. Lett.*, *26*, 711–714, 1999.
- Kato, N. and H. Akimoto, Anthropogenic emissions of SO₂ and NO_x in Asia: Emissions inventory, *Atmos. Environ.* *26A*, 2997–3017, 1992.
- Kritz, M. A., J.-C. LeRouley, and E. F. Danielsen, The China Clipper — fast advective transport of radon-rich air from the Asian boundary layer to the upper troposphere near California, *Tellus*, *42B*, 46–61, 1990.
- Liang, J., L. W. Horowitz, D. J. Jacob, Y. Wang, A. M. Fiore, J. A. Logan, G. M. Gardner, and J. W. Munger, Seasonal variations of reactive nitrogen species and ozone over the United States, and export fluxes to the global atmosphere, *J. Geophys. Res.*, *103*, 13,435–13,450, 1998.
- Liu, S. C., M. Trainer, F. C. Fehsenfeld, D. D. Parrish, E. J. Williams, D. W. Fahey, G. Hubler., and P. C. Murphy, Ozone production in the rural troposphere and implications for regional and global ozone production, *J. Geophys. Res.*, *92*, 4191–4207, 1987.
- Logan, J. A., Ozone in rural areas of the United States, *J. Geophys. Res.*, *94*, 8511–8532, 1989.
- Marengo, A., H. Gouget, P. Nedelec, and J.-P. Pages, Evidence of a long-term increase in tropospheric ozone from Pic du Midi data series: Consequences: Positive radiative forcing, *J. Geophys. Res.*, *99*, 16,617–16,632, 1994.
- McKeen, S. A., E.-Y. Hsie, and S. C. Liu, A study of the dependence of rural ozone on ozone precursors in the Eastern United States, *J. Geophys. Res.*, *96*, 15,377–15,394, 1991.
- Merrill, J. T., Atmospheric long-range transport to the Pacific Ocean, *Chemical Oceanography*, *10*, 15–50, 1989.
- Piccot, S. D., J. J. Watson, and J. W. Jones, A global inventory of volatile organic compound emissions from anthropogenic sources, *J. Geophys. Res.*, *97*, 9897–9912, 1992.
- Savoie, D. L., J. M. Prospero, and E. S. Saltzman, Nitrate, non-seasalt sulfate and methanesulfonate over the Pacific Ocean, *Chemical Oceanography*, *10*, 220–251, 1989.
- Saylor, R. D., W. L. Chameides, and E. B. Cowling, Implications of the new ozone National Ambient Air Quality Standards for compliance in rural areas, *J. Geophys. Res.*, *103*, 31,137–31,141, 1998.
- Sillman, S., J. A. Logan, and S. C. Wofsy, The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, *J. Geophys. Res.*, *95*, 1837–1852, 1990.
- United States Department of Energy (DOE) International Energy Outlook (IEO), 1997, Energy Information Administration, U.S. DOE, 1997. Available from <http://www.eia.doe.gov/oiaf/ieo97>, 1997.
- Wang, Y., J. A. Logan, and D. J. Jacob, Global simulation of tropospheric O₃-NO_x-hydrocarbon chemistry. 2. Model evaluation and global ozone budget, *J. Geophys. Res.*, *103*, 10,727–10,756, 1998b.

D. J. Jacob (djj@io.harvard.edu), J. A. Logan, P. P. Murti, Pierce Hall, 29 Oxford St., Cambridge, MA 02138.

(Received March 9, 1999; revised May 4, 1999; accepted May 10, 1999.)