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Past and future changes in global tropospheric ozone: Impact on radiative forcing

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Abstract. Calculations by a global three-dimensional chemical transport model of the atmosphere suggest that increased surface emissions of chemical compounds caused by industrial activities at mid-latitudes in the northern hemisphere and by biomass burning in the tropics since the middle of the 19th century have produced an increase in the abundance of tropospheric ozone along with a reduction in the oxidizing capacity of the atmosphere (globally averaged OH concentration reduced by 17% and methane lifetime enhanced by 1.5 years). These perturbations in tropospheric ozone result in a change in annually averaged radiative forcing of 0.37 W m^{-2} (0.62 W m^{-2} in the northern hemisphere during the summer months). Future changes (1990–2050) in tropospheric ozone associated with population increase and economic development (primarily in developing countries) are expected to be largest in the tropics, specifically in South and Southeast Asia. Further changes in the oxidizing capacity of the atmosphere could be small if the abundance of tropospheric water vapor increases as a result of anticipated climate change.

1. Introduction

The release into the atmosphere of chemical compounds, including methane, nonmethane hydrocarbons, carbon monoxide, and nitrogen oxides, produced by biomass burning (mostly in the tropics) and fossil-fuel combustion (mostly at mid-latitudes in the northern hemisphere) is believed to have caused the increase in tropospheric ozone concentrations observed at several locations in the northern hemisphere [*Wege et al.*, 1989; *Kley et al.*, 1994; *Staehelin and Schmid*, 1991; *Volz-Thomas*, 1992]. Because of the limited number of ozone sonde stations that have operated continuously over long periods of time, trends in tropospheric ozone are poorly documented, especially in the tropics and the southern hemisphere; limited data suggest, however, that surface ozone amounts over the European continent were typically 10–20 ppbv in the early 1900s (i.e., substantially lower than currently observed 20–30 ppbv wintertime values and 40–60 ppbv summertime values) [*Volz and Kley*, 1988; *Marenco et al.*, 1994].

2. The IMAGES Model

To investigate past and future evolutions of tropospheric ozone and its precursors, we use a three-dimensional chemical-transport model called IMAGES (described in detail and evaluated by *Müller and Brasseur* [1995]), which provides the global distribution and budget of the key chemical compounds in the troposphere for specified surface emissions and atmospheric dynamical fields. The model extends from the surface to 50 mbar (approximately 20 km altitude). It includes 60 chemical compounds (including 7 primary nonmethane hydrocarbons and their degradation products including peroxyacetyl nitrate and acetone) and 150 chemical and photochemical reactions. The spatial resolution is 5° in longitude and latitude, with 25 levels in the vertical. The model does not allow for feedback of the chemistry on the dynamics (which is the same for all model runs).

3. Past Ozone Changes

Before discussing the expected chemical composition of the future global troposphere, we first consider changes that have occurred since pre-industrial times. Pre-industrial conditions (assumed to occur in 1850) are simulated by removing all fossil-fuel related sources (including aircraft emissions), by reducing biomass burning emissions by an arbitrary factor of 3, and by assuming a background methane mixing ratio of 800 ppbv [*Houghton et al.*, 1990]. Stratospheric ozone and temperature are fixed at their 1850 level and remain unchanged for all model runs. Parameters describing the transport of chemical compounds are assumed to be the same as in 1990. Calculations performed with IMAGES suggest that, between 1850 and 1990, the zonally averaged summertime ozone amount at mid-latitudes in the northern hemisphere has increased by about 80% (15 ppbv) in the boundary layer, and by 40–70% (20–40 ppbv) in the free troposphere (Plate 1a). Increases in the upper troposphere (200 mbar) are approximately 10 ppbv (40%) in the tropics and 35 ppbv (20%) at 45°N. Changes in the tropospheric ozone-column abundance (not shown) are typically 20 Dobson units (DU) over the eastern United States and Europe, 15 DU over Southeast Asia, and 5 DU at mid-latitudes in the southern hemisphere. Differences in surface ozone concentrations between 1850 and 1990 (Plate 1b) range between 100 and 150% (20–40 ppbv) during summertime in Europe, the Far East, and most of the United States. These changes in boundary-layer ozone should be regarded as lower limits because they do not account for the changes in soil NO emissions associated with the increasing use of fertilizers (assumed in the model to be 0.5 Tg N/yr in 1990) at mid-northern latitudes. Over the 1850–1990 period, the mass-weighted OH concentration has decreased by 17%, corresponding to an increase of 1.5 years in the global lifetime of methane.

As ozone interacts with both shortwave (solar) and longwave (terrestrial) radiation, variations in its atmospheric abundance are expected to affect the climate system [*Lacis et al.*, 1990]. The global radiative forcing (defined as the variation in the net radiative flux at the tropopause) resulting from tropospheric ozone changes during the 1850–1990 period is estimated to be 0.43 W m^{-2} during June to August (0.14 W m^{-2} for shortwave and 0.29 W m^{-2} for longwave radiation) and 0.31 W m^{-2} (0.10 W m^{-2} for shortwave and 0.21 W m^{-2} for longwave radiation) during the December-February period (cloudy sky conditions using the cloud distribution derived from NCAR CCM-2, see *Kiehl and*

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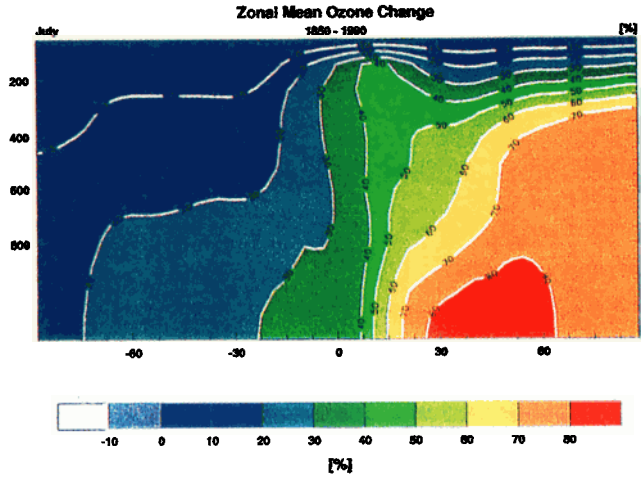

Plate 1(a). Change (percent) in the zonally averaged ozone concentration from 1850 to 1990 calculated as a function of latitude and height for July conditions.

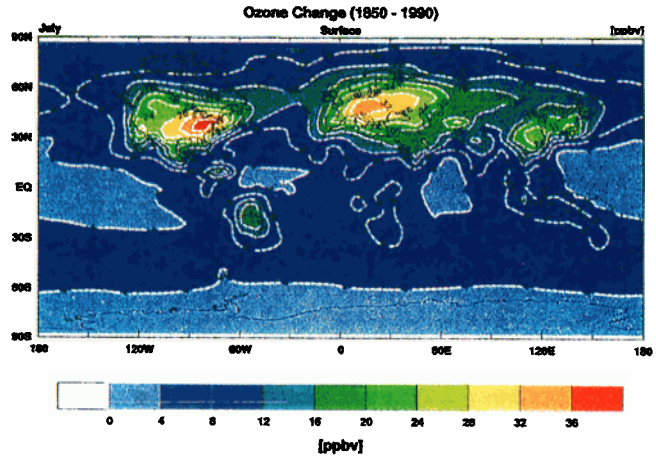

Plate 1(b). Change (ppbv) in the surface ozone mixing ratio from 1850 to 1990 calculated for July conditions.

Briegleb, 1993). These values represent a significant fraction of the total change in radiative forcing (2.4 W m^{-2}) estimated for the same period of time [*Houghton et al.*, 1996] and are within the range of model estimates of *Hauglustaine et al.* [1994]; *Chalita et al.* [1996]; *Lelieveld and van Dorland* [1995]; *Roelofs et al.* [1997]; *Forster et al.* [1996]; *Berntsen et al.* [1997]; *van Dorland et al.* [1997]. Details about the seasonal and hemispheric differences in the radiative forcing appear in Table 1. The mean forcing reaches 0.62 W m^{-2} in the northern hemisphere during summer, with local values larger than 0.8 W m^{-2} over the southeastern United States, the North Atlantic ocean, Eastern Europe and the Sahara (not shown).

The IMAGES model suggests that, in October when biomass burning in the tropics is most intense, the tropospheric ozone column abundance over the burning areas of South America and Africa, as well as in the South Atlantic, are 10–15 DU higher than if biomass burning were ignored. The magnitude of this ozone enhancement is similar to the variation in the tropospheric-ozone column observed by *Fishman and Brackett* [1997] between the burning and non-burning seasons, but is somewhat lower than the 20–25 DU contrast reported by *Hudson and Thompson* [1998]. This enhancement produces a radiative forcing of about 0.7 W m^{-2} in the tropical Atlantic, consistent with the recent estimate of *Portmann et al.* [1997].

4. Future Ozone Changes

Estimates of future changes in the ozone abundance and in the related climate forcing must be based on given scenarios for the emissions of ozone precursors, which are highly uncertain because they depend on assumed economic growth, technological advances, and responses to recent (e.g., Kyoto Agreement) and future regulatory measures. In this study, rather than considering several scenarios covering a large number of possibilities, we adopt the IS92a scenario of IPCC [*Houghton et al.*, 1992], which is regarded as a "medium case" for the year 2050. The increase factors for NO_x (2.1), CO (1.7), and anthropogenic nonmethane hydrocarbons (2.4) relative to 1990 are assumed to have the same geographical distribution as the increase factor for CO_2, with a proportionality factor determined to match the global emission in 2050 quoted by IPCC (scenario IS92a). As a result, the emissions of ozone precursors increase dramatically (factor 3 to 7) in most regions of the developing world (with the largest enhancement in South East Asia and Africa), but change only slightly in the developed world (including the former Soviet Union.). The methane mixing ratio in 2050 is assumed to be 2.7 ppmv [*Houghton et al.*, 1996]. In addition, consistent with the IS92a scenario, biomass-burning emissions are assumed to increase uniformly by 20% and future aircraft emissions (and their geographical distributions) are taken from *IPCC* [1998] and based on the NASA scenario.

Table 1. Net radiative forcing (W m^{-2}) (longwave and shortwave) associated with tropospheric ozone changes since the pre-industrial era, and between 1990 and 2050 (cloudy sky).

	Annual	DJF	MAM	JJA	SON
			1850–1990		
Global	0.37	0.31	0.37	0.43	0.39
NH	0.48	0.34	0.50	0.62	0.48
SH	0.27	0.29	0.24	0.23	0.31
			1990–2050		
Global	0.26	0.22	0.24	0.31	0.29
NH	0.34	0.25	0.32	0.44	0.38
SH	0.18	0.19	0.17	0.18	0.20

The change in the zonal-mean ozone amounts for the 1990–2050 period, as derived by the IMAGES model, appears in Plate 2a. The largest *relative* changes (more than 30% or 4 ppbv at the surface) are predicted to occur throughout the tropical troposphere in response to rapidly growing economies in Asia and in other developing countries, and to the assumed increase in biomass burning. In contrast, the largest *absolute* changes in the ozone amount (approximately 35 ppbv or 25%) are located at 300 mb in the northern mid- and high latitudes, and result from in-situ NO_x emissions by aircraft and transport from anthropogenic surface sources. In this upper tropospheric region of the atmosphere, the abundance of NO_x is expected to increase by more than 100 pptv between 1990 and 2050. Plate 2b shows that corresponding changes in boundary-layer ozone are larger than 50% (10–20 ppbv) over the populated areas of Asia and central America, and that enhanced ozone abundances (more than 40–50% increases) are predicted in the rapidly developing regions of the southern hemisphere (South Africa and eastern Brazil) during summertime (January). In the upper troposphere (250 mbar, see Plate 3), ozone increases typically by 20–30 ppbv (20–30%) in the northern hemisphere (40–60 ppbv when compared to the pre-industrial era). More than half of this enhancement results from the assumed increase in NO_x emissions associated with aircraft operations. The tropospheric ozone column is typically enhanced

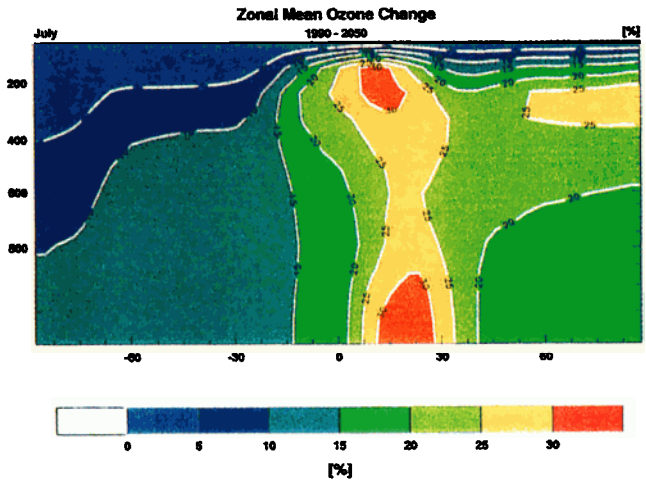

Plate 2(a). Change (percent) in the zonally averaged ozone concentration for the 1990–2050 period calculated as a function of latitude and height on the basis of the IPCC IS92a scenario.

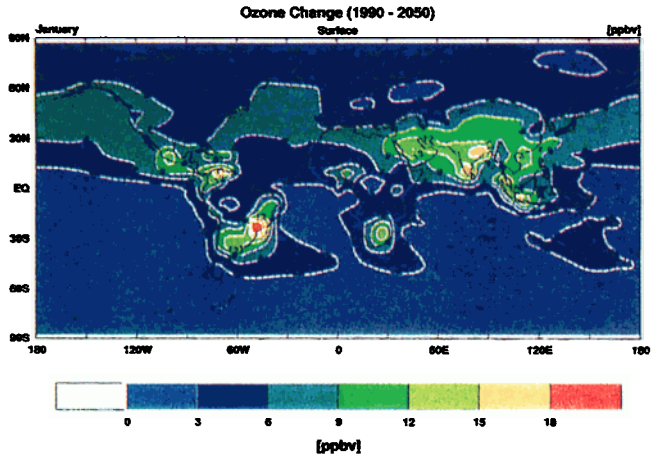

Plate 2(b). Change (ppbv) in the surface ozone mixing ratio for the 1990–2050 period calculated for January conditions.

by 5 DU in the tropics with local values reaching 10–15 DU over South and Southeast Asia. The globally averaged change in ozone radiative forcing calculated for the 1990–2050 period is 0.31 W m^{-2} (0.21 W m^{-2} for longwave and 0.10 W m^{-2} for shortwave effects) for the June-August period, and 0.22 W m^{-2} for the December-February period. The global mean OH density decreases by 5.5% relative to year 1990 and the global photochemical lifetime of methane increases from 8.6 years in 1990 to 9.1 years in 2050.

The future evolution of tropospheric ozone abundance could be modified by changes in tropospheric temperature and specific humidity associated with expected climate changes. To estimate the importance of this effect (upper limit), we applied to the zonal-mean temperature and water vapor fields as specified in IMAGES perturbations produced by the NCAR Community Climate Model coupled to a slab ocean [*Kiehl et al.*, 1998] for a doubling in the atmospheric CO_2 abundance. (The global annual-mean surface-temperature change is 2.05°C.) The temperature increase of a few degrees does not significantly affect the calculated ozone changes in year 2050. The increase in the water vapor concentration, however, which reaches 50% in the upper troposphere (tropics) and is of the order of 20% at 5 km altitude (all latitudes), leads to enhanced OH and HO_2 concentrations (globally 7% with 10% at 5 km, not shown), and to enhanced net ozone destruction. As a result, the ozone increase predicted for year 2050 is somewhat reduced (e.g., from 30 to 25% in the tropical upper troposphere) when this "climate" effect is taken into account in the model. In spite of significant perturbations expected in the future chemical composition of the troposphere, the mass-weighted mean OH concentration calculated by the model increases only by 2.2% between 1990 and 2050, if the abundance of water vapor increases (tending to increase the supply of OH) as a result of global climate change. The oxidizing capacity of the atmosphere, may therefore remain relatively unchanged in the next 5 decades. Other potential climate feedbacks, which remain poorly quantified and hence are not considered here (e.g., changes in cloudiness, increasing production of nitrogen oxides and ozone resulting from enhanced lightning activity in a warmer climate), could further affect the chemical composition of the global troposphere in the future [*Sinha and Toumi*, 1997]. Future ozone levels could also be affected by aerosol-induced changes in photolysis rates [*Dickerson et al.*, 1997] and in the NO_x photochemistry [*Dentener* et al., 1996].

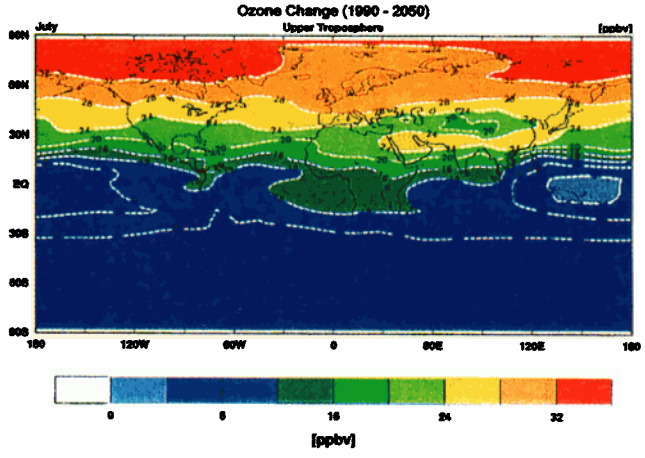

Plate 3. Change (ppbv) in the ozone concentration in the upper troposphere (250 mbar) predicted for the 1990–2050 period (July conditions).

5. Conclusions

Enhanced biomass burning in the tropics and fossil-fuel consumption at mid-latitudes in the northern hemisphere have substantially modified the global ozone budget in the troposphere since the pre-industrial era, and produced significant impacts on climate forcing. Based on suggested scenarios for population growth and energy consumption, the IMAGES model suggests that future changes in tropospheric ozone should be largest in the tropics, especially in Asia. However, based on the IPCC/IS92a scenario, the magnitude of globally averaged predicted ozone changes for the 1990–2050 period should be somewhat smaller than changes that have occurred since the pre-industrial era. The uncertainty in the ozone budget in the tropics, and specifically in the role of biomass burning, needs to be carefully addressed, because the sensitivity of climate to ozone changes is largest in these regions.

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