

Title: Examine the seasonal variability of ozone mixing ratios and budgets in the tropical southern Pacific

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Abstract.

Significant seasonal variations in the concentrations of ozone, NO_x, nonmethane hydrocarbons and other trace gases were observed in the southern tropical Pacific during the PEM-TROPICS-A (PT-A, Sep/Oct 1996) and PEM-TROPICS-B (PT-B, Mar/Apr 1999). Making these data sets more useful would require development of the relationship of these limited data sets to the seasonal and climatic variability of the atmosphere. Here we employ a three-dimensional global scale chemical transport model to generate the chemical climatology of troposphere and then evaluate the ozone budget in the southern tropical Pacific Ocean. We also identify key control factors in the model and compare model results with measurements. This process evaluates the effectiveness of the model in representing transport barriers, such as the Inter-Tropical Convergence Zone (ITCZ) and South Pacific Convergence zone (SPCZ) and the effects of biomass burning in South America and Africa on ozone in the middle of the Pacific Ocean. The model has considerable skill in reproducing the observed ozone profiles, including the seasonal variability. Transport of ozone from the west to the model mid-troposphere in the central Pacific during the PT-A period caused a net increase in the ozone burden. A net outflow of ozone from the region to the east reduced the ozone burden during the PT-B period. For the upper troposphere the horizontal advective flux inputs into the central Pacific show a similar trend. The input from the north was found to be much larger during the PT-B period than during the PT-A period. The *in situ* production and loss calculated in the 3-D model were in approximate agreement with values derived from measurements. The gradients in the model across the ITCZ and SPCZ are also compared with the measurements.

Introduction

Ozone is a product of atmospheric chemistry and also is a useful indicator as a result of the emissions of trace gases in to the troposphere from anthropogenic and natural emissions. The impact of anthropogenic emissions on the chemistry of the atmosphere has been evaluated in terms of ozone (NARSTO, 1996). It was shown that ozone is not only a key component for the smog, but also a significant and pervasive constituent in the chemistry of the troposphere. The primary net source of ozone in to the troposphere is the subsidence of stratospheric air and its primary net sink is the physical removal at the surface of the earth by dry deposition. The gross production and loss terms due to chemical reactions in the atmosphere are larger than these net production and loss terms by factors of 4 to 5, though the net increase in ozone due to the chemical process is expected to be much smaller than stratospheric import (WMO, 1996).

The impact of anthropogenic emissions on the chemistry for the different regions of the atmosphere is currently under evaluation based on limited data sets collected in the recent past and modeling activity. The main focus of the NASA PEM (Pacific Exploratory Mission) is to provide the atmospheric conditions in selected remote locations in the Pacific using aircraft platform based measurements, and corroborating with limited ground level data sets. Data collected during these measurement campaigns provide a snapshot of the atmospheric conditions prevailing in these regions during the

limited time periods (typically 1 to 2 months). Developing relationship of these limited data sets to seasonal and climatic variability of the atmosphere would be necessary. This could be done by either performing extensive comparison of these data sets with few available long-term monitoring for limited number of species, such as ozone and carbon monoxide, or using global scale models for generating chemical climatology. Here we employ the latter approach to evaluate the ozone budgets in the southern tropical Pacific Ocean and identify the key control factors in the model and compare it with the measurements. PT-A and PT-B field measurement experiments conducted by NASA under the GTE program offer such an opportunity by providing us with a database of intensive measurements of ozone and its precursors in the tropical southern Pacific during two different periods of the year. PT-A was conducted during August-October of 1996 in what would be the spring period in the Southern Hemisphere and was followed by PT-B conducted during the fall period of March-April 1999.

Experiment

In this study, we use the MOZART 3-D model for three separate runs. A one-year run with mixing ratios, fluxes and chemical tendency terms saved once a day was initially carried out. Results from this run were used to initialize two additional runs: one for the August-September period and the second for the March-April period in accordance with PT-A and PT-B time. Results from these runs were saved at a much higher frequency at once every hour for comparison with the PT-A and PT-B measurements.

The box-photochemical model was used to calculate the photochemical production and loss tendencies of ozone with constraints on radiation and trace gas mixing ratios measured from the PT-A and PT-B data sets. The one-minute averaged merged data files generated by the Harvard research group were used to make the calculations. Below is a detailed description of the results from these calculations.

Results

Figure 1 shows the modeled and measured profiles of ozone at Fiji in the southern tropics. A typical data points from Fiji for SHADOZ would include several launches in a month for 2 to 5 years. In general, the model is in good agreement with the data for both spring and fall seasons. MOZART is slightly under-estimating mid-tropospheric ozone during the months of March/April in Fiji.

Figure 2 is the plot for ozone mixing ratio from MOZART and measured during PT-B (Fig 2a) and PT-A (Fig.2b) periods as a function of latitude and altitude. If there are several data points in a particular altitude-latitude bin, the average of these values is used for plotting. In Fig 2a, the results from the model calculation is able to reproduce the low ozone values throughout the depth of the troposphere in the tropical Southern Hemisphere; however, several pockets of high ozone in the data set at 10-12 km are not captured in the model. These pockets of high ozone represent episodes of long-range transport of ozone from East Asia to the northern tropical Pacific and possibly from South Africa or Southern America to the extra tropical regions of the South Pacific (Maloney *et al.*, 2000). Whereas in Fig 2b for PT-A period, the model result shows only a slight elevation in ozone mixing ratios in the 25 to 15 S latitudes as compared to the significant elevations in the observed data set. It could be the reason of biomass burning

in the southern tip of Africa which is assumed to be the major source for the elevated ozone (Fuelberg *et al.*, 1999). A similar analysis was performed for CO as an indicator of biomass/anthropogenic influence on the air masses. In general, there is an excellent agreement between the model calculated and measured CO for this period.

Advective, convective, diffusive fluxes across each of the grid boundaries were calculated and saved once every hour for the limited in time calculations for the March/April and August/September model calculations. The mass change in each of the grid cells due to processes in the model, such as due to chemical production and chemical loss were also saved as one hour averages for these calculations. The fluxes derived from the model were analyzed for the central Pacific zone and defined as follows. The central Pacific region (CP) extends from 165 E to 120 W longitudes and from 30 N to 30 S latitude. The boxes were further divided in the vertical plane in to the lower tropospheric box extending from 0 to 2 km, mid tropospheric box extending from 2 to 7 km and upper tropospheric box extending from 7 to 12 km. Table 1 lists the model calculated fluxes averaged over a month in the central Pacific region for both PT-A and PT-B periods. Overall, model is able to catch the pattern of general circulation in this region, such as the lower tropospheric box (surface layer) ozone fluxes are directed from east to west, whereas the mid-tropospheric box (MT) and upper tropospheric box (UT) the flux direction is from east to west in alignment with the prevailing wind directions of subtropical jet. The diagram also shows ozone subsidence in this region for the vertical fluxes. As a result, there is a net ozone increase in CP mainly due to zonal transport mechanism.

Figure 3 presents the results of odd oxygen (O_x) production/loss term from the box-model and the MOZART 3D model for PT-B period as a function of altitude and latitude for the Southern Hemisphere. Overall, the 3D model has the same tendency as the box model results for most of the altitude and latitude bins. However, the model is much less likely to produce ozone in the upper troposphere (8-12 km) than the data from the box model.

Figure 4 shows the flight 16 data from PT-A and model calculated comparison for SPCZ analysis. Basically, model is able to distinguish a well-defined SPCZ as compared to the observation. And the stratospheric intrusion at around 20 degree south from Lidar measurement is also captured in the model.

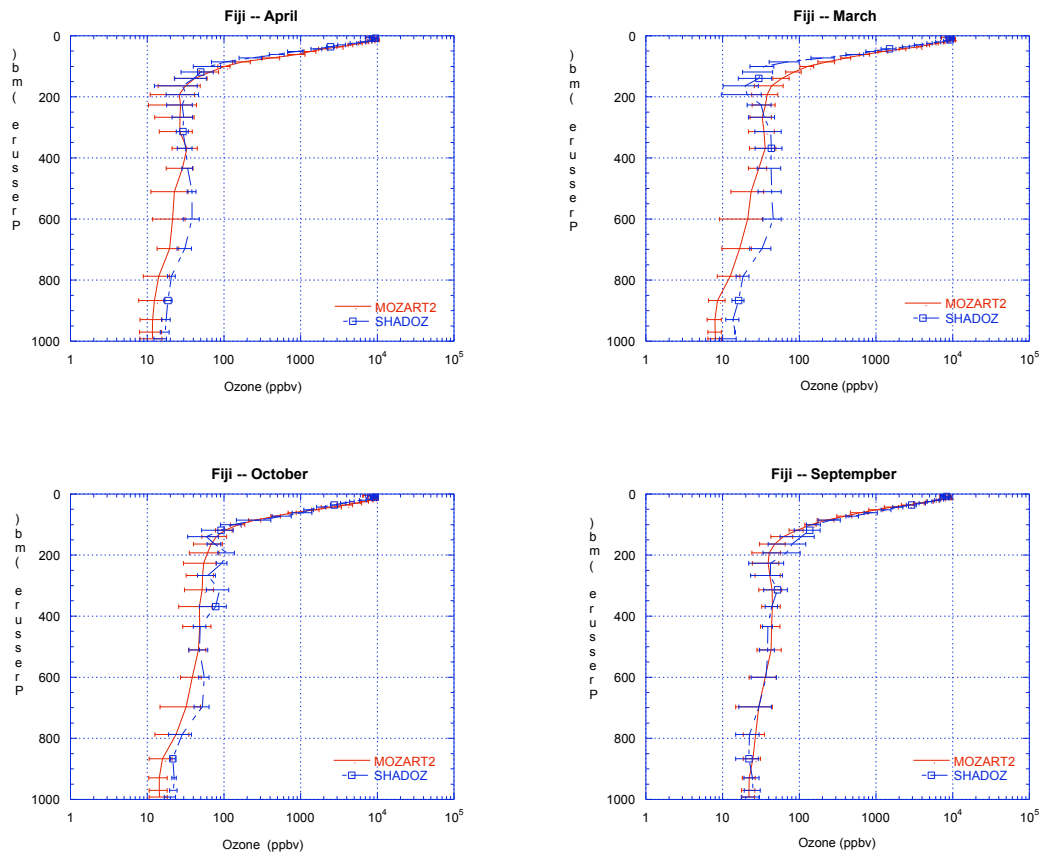


Figure 1. SHADOZ ozone sonde profiles, compared with MOZART (v2) calculations for Fiji.

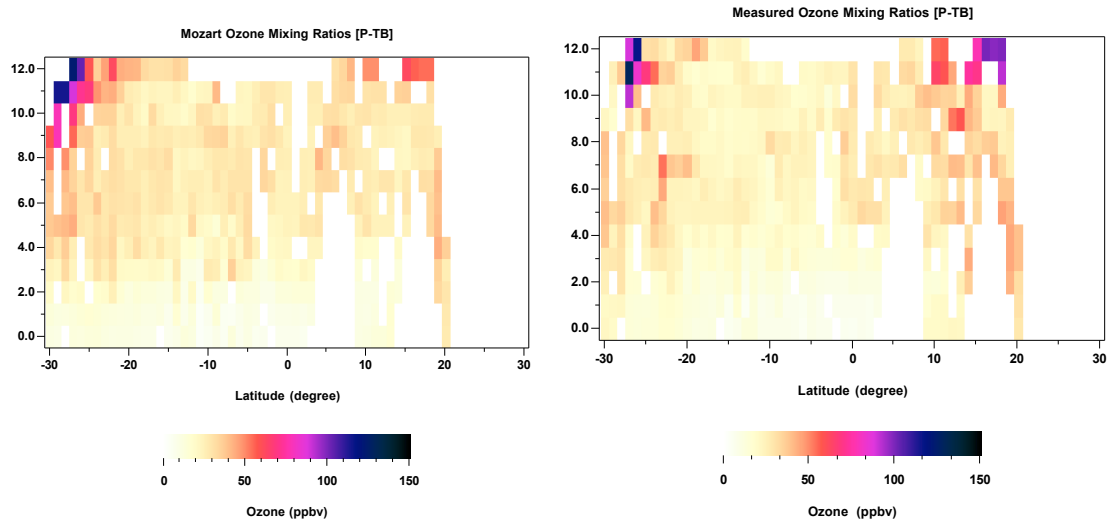


Figure 2a. MOZART calculated ozone (average, left panel) for March and April and sampled at the same locations from DC 8 flights during PT-B (right panel)

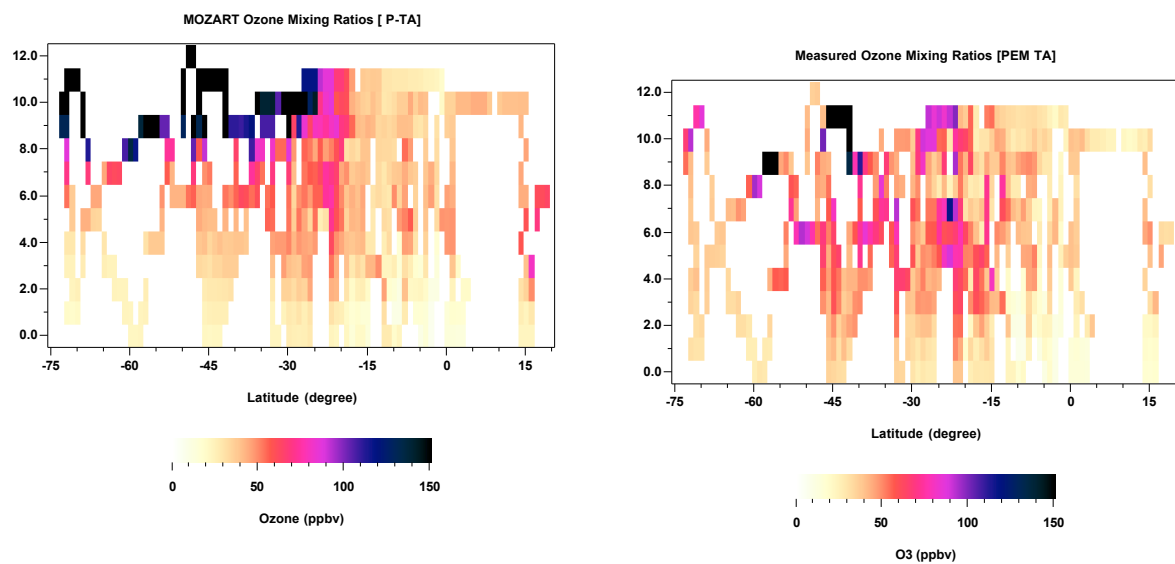


Figure 2b. MOZART calculated ozone (average, left panel) for September and sampled at the same locations from DC 8 flights during PT-A (right panel)

Table 1: Central Pacific Region (165E~135W, 30N~30S) Fluxes Analysis

Monthly Averaged Fluxes (kg/sec)		Directional Fluxes						Processes.				
		West	East	South	North	Bottom	Top	Adv	Depos	Conv.	Diff	Chem.
PEM-Tropics A (Sep.)	UT	22,470	20,340	-2,402	-1,896	-1,718	-972	877.6	216.9	-1,268	4.329	917.3
	MT	6,486	4,144	-1,456	-2,424	-2,589	-1,718	2,440	174	-1,443	-2.824	-1,262
	LT	-3,059	-5,060	-28.52	-754.5	0	-2,589	5,316	75.78	2,715	-1,287	-6,584
PEM-Tropics B (Mar.)	UT	18,840	27,650	-1,550	-10,510	-654.9	-1,221	710.2	301.4	-1,906	6.303	650
	MT	4,551	5,678	-694	-3,484	-571.6	-654.9	1,746	157.2	-62.86	-1.879	-1,554
	LT	-2,674	-4,219	-69.02	-1,465	0	-571.6	3,512	95.56	1,972	-1,029	-4,167
Net Fluxes		Latitudinal		Longitudinal		Vertical		% change = (B-A)/A x100%		E-W	N-S	Vert.
PEM-Tropics A (Sep.)	UT	2,130		-506		-746		UT		-514%	1871%	176%
	MT	2,342		968		-871		MT		-148%	188%	110%
	LT	2,001		726		2,589		LT		-22.8%	92%	-78%
PEM-Tropics B (Mar.)	UT	-8,810		8,960		566.1		UT		-514%	1871%	176%
	MT	-1,127		2,790		83.3		MT		-148%	188%	110%
	LT	1,545		1,396		571.6		LT		-22.8%	92%	-78%

Note:

UT: Upper tropospheric Box (7~12 km), MT: Middle tropospheric Box (2~7 km), LT: Lower tropospheric Box (0~2 km)

Adv: Advective Flux

Depos.: Depositional Flux

Conv.: Convective Flux

Diff.: Diffusive Flux

Chem.: Chemistry Rate

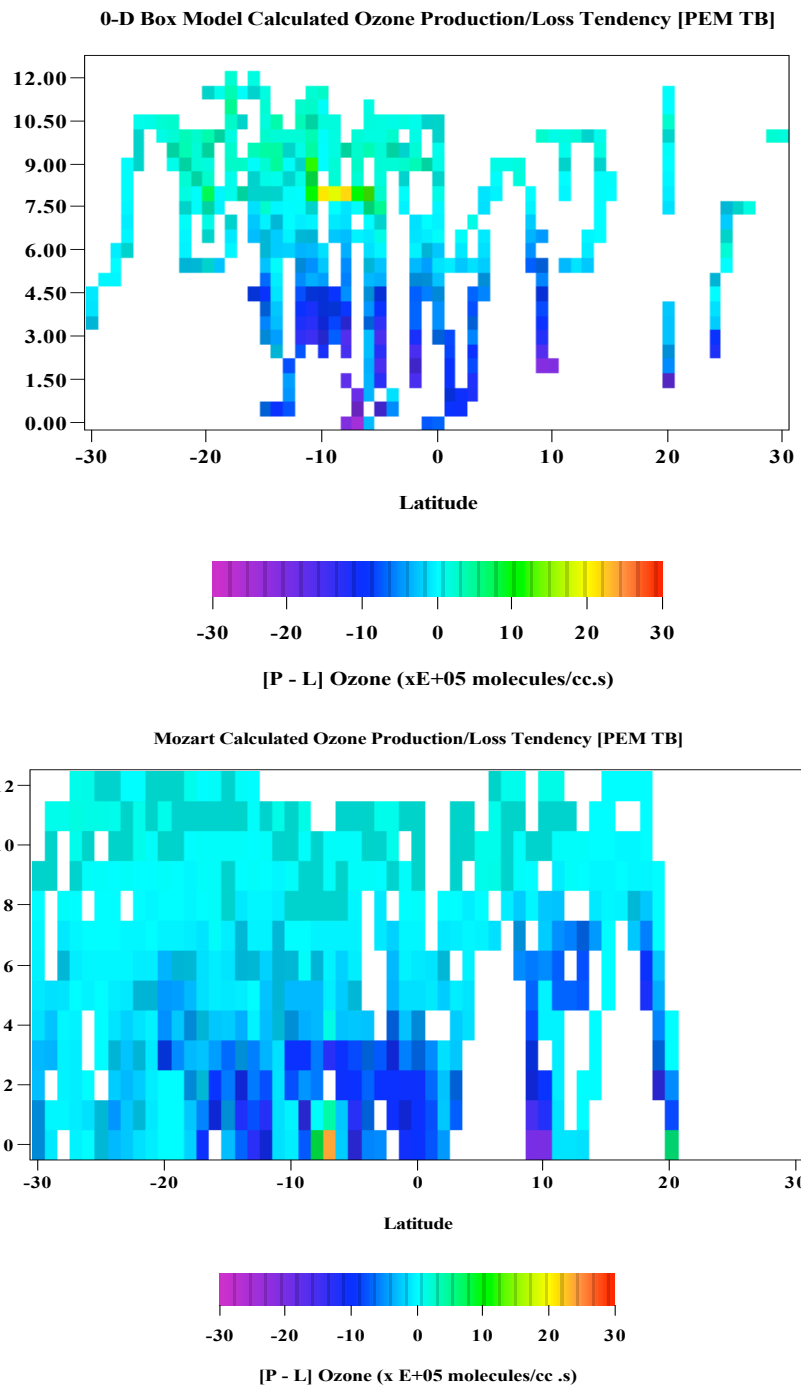


Figure 3 . 0D box model (upper panel) and MOZART(lower panel) calculated ozone production/loss tendency for the period of PT-B

Figure 4: Ozone profile from Flight 16 in PT-A (upper panel) and MOZART calculation (lower panel)

