Indonesian wildfires of 1997: Impact on tropospheric chemistry

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[1] The Indonesian wildfires of 1997 released large amounts of trace gases and aerosols (e.g., \sim 130 Tg of carbon monoxide (CO)) from September to November. Using the GEOS-CHEM model of tropospheric chemistry and transport, we conducted a study of this burning event, including sensitivity simulations, to estimate the impacts of the trace gas and aerosol emissions on tropospheric chemistry. The emissions from the fires were estimated using the Total Ozone Mapping Spectrometer (TOMS) Aerosol Index (AI) data product and fire-count data from the Along Track Scanning Radiometer (ATSR) World Fire Atlas as surrogates for biomass burning. The model captures most of the daily variations of CO and ozone (O_3) observed in the region affected by the pollution. Export of the pollution from the Indonesian region was primarily in the prevailing easterlies in the free troposphere to the tropical Indian Ocean where the bulk of the pollution lay between 20°N to 20°S from September to November. The model's tropospheric CO and O₃ columns were elevated by more than 50% and 10%, respectively, over the tropical Indian Ocean in a simulation with emissions from the fires relative to a simulation without the emissions. Another important export pathway was to the tropical and subtropical South Pacific Ocean in the southern subtropical jet. A more episodic pathway occurred to the tropics and subtropics of the North Pacific Ocean. By December, the tropospheric CO column from the fires had mixed zonally and somewhat symmetrically about the equator impacting both the Northern and Southern Hemisphere similarly. The model's CO column was elevated by 10-20% from 30°N to 45°S in December, by 5-10% poleward of 45° S, and by less than 5% poleward of 45° N. The relative impact of the fires was lower in the Northern Hemisphere, as the background CO column is typically higher there. The fires decreased the concentration of the hydroxyl radical (OH) in the model by more than 20% over much of the tropical Indian Ocean through consumption by CO, heterogeneous loss of odd-oxygen radicals (HO_x) on black (BC) and organic carbon (OC) aerosols, and reduction of UV light by the aerosols. The net direct, shortwave radiative forcing at the top of the atmosphere of OC and BC aerosols from the fires was relatively small, as their forcings were similar, but of opposite signs. The net forcing at the surface, however, was large, about -10 W m^{-2} over most of the tropical Indian Ocean and as low as -150 W m^{-2} over the burning regions in Indonesia, indicating that aerosols from the fires significantly perturbed the tropical radiative budget. The calculated forcing of O_3 was minor relative to those of BC and OC aerosols. INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution-urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere-constituent transport and chemistry; KEYWORDS: Indonesia, biomass burning, wildfires, aerosols, radiative forcing, long-range transport

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1. Introduction

[2] One of the largest biomass burning events of the twentieth century occurred from September to November 1997 in Indonesia, Malaysia, and Papua New Guinea. The fires were primarily set to clear land on small farms, including by settlers as part of Indonesia's Transmigration Programme, and on large-scale oil palm and rubber plantations [Brauer and Hisham-Hashim, 1998; Ketterings et al., 1999; Vadya, 1999; Gönner, 2000]. Other fires were thought to be unintentionally set or the result of arson (e.g., for hunting and land use conflict) [Vadya, 1999; Gönner, 2000; Schindler, 2000]. The wildfires were exacerbated by (1) a land-use conversion project, the Central Kalimantan Mega Rice Project, initiated by the Indonesian government which effectively drained large expanses of tropical peat swamp forests in Central Kalimantan on the island of Borneo [Vadya, 1999; Boehm et al., 2001], (2) a nearly year-long drought induced by the most extreme El Niño-Southern Oscillation (ENSO) event of the century [Bell and Halpert, 1998; Glantz, 2001], and (3) rampant logging [Barber et al., 2002]. In this paper, we conduct simulations of September to December 1997 using the GEOS-CHEM global model of tropospheric chemistry and transport [Bey et al., 2001a] with the purpose of identifying the export pathways of trace gases and aerosols emitted from these fires and of quantifying the impacts of the pollution on the oxidizing capacity and radiative budget of the troposphere.

[3] There is evidence that anthropogenic biomass burning has occurred for thousands of years in Indonesia [Haberle et al., 2001]. The frequency and severity of large burning events, such as those in 1982-1983 and 1997-1998, are likely to increase if unsustainable exploitation of Indonesia's natural resources continues [Vadya, 1999; Walker, 1999]. Nearly 40% of Indonesian forests have been felled in the last 50 years, and the rate of deforestation is accelerating from 1 million ha/year in the 1980s, to 1.7 million ha/year in the early 1990s, and to 2 million ha/year after 1996 [Barber et al., 2002]. The total forested area in Indonesia is estimated to be 90 to 120 million ha [Makarim et al., 1998]. The potential for fire damage increases as forests are disturbed by human activities [Boehm et al., 2001; Page et al., 2002]. The recent financial collapse of (1) the Transmigration Programme in 1997, sharply cutting the number of people resettled to forested lands, and (2) the Central Kalimantan Mega Rice Project in 1999 (M. A. S. Adhiati and A. Bobsien, Indonesia's Transmigration Programme: An update, a report prepared for Down To Earth, available at http://dte.gn.apc.org/ctrans. htm#tide, July 2001) may slow the rate of deforestation. The potential for large-scale burning events also depends on the frequency and severity of droughts in the coming years. There is a high degree of uncertainty in the prediction of ENSO-related droughts in Indonesia beyond the short term, but there could be an intensification of ENSO events as global warming takes place [Intergovernmental Panel on Climate Change (IPCC), 2001]. Rainfall rates will likely decline during the dry season, even in a year not impacted by ENSO, as the forests are converted to farms, pastures and savannas [Zhang, 1994].

[4] Observations of trace gases and aerosols provide evidence for the widespread impact of the 1997 fires on tropospheric composition. Ozone (O_3) was elevated over

Indonesia and much of the tropical Indian Ocean [Fujiwara et al., 1999, 2000; Kita et al., 2000; Thompson et al., 2001; Chandra et al., 2002; Yonemura et al., 2002]. Kita et al. [2000] estimated that the Tropospheric Column O_3 (TCO) from the Total Ozone Mapping Spectrometer (TOMS) was about 40% higher than the 20-year climatology over the Indonesian region in October 1997. Matsueda and Inoue [1999] reported elevated carbon dioxide (CO₂), methane (CH₄), and carbon monoxide (CO) throughout the troposphere over Singapore and between 9 and 12 km over the South China Sea in October 1997. Matsueda et al. [1999] reported observed CO over 300 ppbv between 8 and 13 km in September and October 1997 between 10°S and 30°S in the western Pacific near Australia. Chan et al. [2001] reported an enhanced O₃ layer 10 km thick over Hong Kong with concentrations up to 130 ppbv.

[5] There have been two model studies that have contributed appreciably to the understanding of the impact of the fires on tropospheric O₃. Haughtstaine et al. [1999] performed a model sensitivity simulation of the fires on O₃ and its precursors, using the Model for OZone And Related chemical Tracers (MOZART), a global model of tropospheric chemistry and transport. They found that the emissions of trace gases and aerosols caused large perturbations to tropical photochemistry over much of the Indian and western Pacific Oceans. Their model TCO increased by 20-25 DU (1 DU = 2.69×10^{16} molec cm⁻²) over Indonesia, and their CO in the free troposphere was as high as 1 ppmv. The major transport pathways in the MOZART model of Hauglustaine et al. [1999] were to the western Pacific Ocean and Southeast Asia. The authors note that the transport in their model is driven by NCAR CCM dynamics, which represent mean conditions, and that strong export to the Indian Ocean was actually observed. Chandra et al. [2002] performed a GEOS-CHEM model simulation and estimated that the TCO was enhanced by nearly 50% over Indonesia and greater than 10% over a large portion of the tropical Indian Ocean (10°N-10°S) in October 1997 relative to October 1996, a low burning year [Chandra et al., 2002, Plates 6 and 10]. This enhancement was likely due to O₃ generation from biomass burning pollutants and from a shift in the tropical convection pattern that is typical during an ENSO year [Chandra et al., 1998; Ziemke and Chandra, 1999; Chandra et al., 2002; Sudo and Takahashi, 2001].

[6] The GEOS-CHEM model and its aerosol and trace gas emission inventories are described in section 2. In sections 3.1 and 3.2, we discuss the export mechanisms and the major long-range transport pathways of pollutants from the Indonesian region. We evaluate the model with observations of O_3 and CO where available from ozonesondes, surface measurements, and aircraft campaigns. In sections 3.3 and 3.4, we conduct model sensitivity simulations to ascertain the impact of the fires on the oxidizing capacity and radiative budget of the troposphere. We present a summary of conclusions in section 4.

2. Model Description

2.1. GEOS-CHEM Model

[7] The GEOS-CHEM model (http://www-as.harvard. edu/chemistry/trop/geos/index.html) is driven by assimilated

meteorological data from the Data Assimilation Office (DAO) of NASA and simulates tropospheric O₃-nitrogen oxides (NO_x)-hydrocarbon chemistry including about 120 chemical species and 24 tracers [Bey et al., 2001a]. For the 1997 simulation period, we use the GEOS-STRAT meteorological fields that are provided on a sigma coordinate system with 46 levels from the surface to 0.1 hPa and 2° latitude by 2.5° longitude horizontal resolution. For computational expedience, we degrade the vertical resolution by merging the vertical levels above the lower stratosphere, retaining a total of 26 levels. The photolysis rates are computed using the Fast-J radiative transfer algorithm of Wild et al. [2000], which includes Rayleigh and Mie scattering by clouds and aerosols. The emissions of NO_x from lightning are linked to deep convection following the parameterization of Price and Rind [1992] as implemented by Wang et al. [1998] with vertical profiles from Pickering et al. [1998]. Biogenic isoprene and NO_x emissions from land are computed locally using modified versions of the Guenther et al. [1995] and Yienger and Levy [1995] algorithms, as described by Wang et al. [1998] and Bey et al. [2001a]. We use version 4.31 of the model in the present study, which includes the radiative and heterogeneous chemical effects of sulfate, sea salt, black carbon, organic carbon, and dust aerosols [Martin et al., 2003], an important improvement on earlier model versions. The aerosol fields were calculated using the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model [Chin et al., 2002], which is driven by the same assimilated meteorological data from DAO as the GEOS-CHEM model. The daily average fields are coupled to the GEOS-CHEM model as described by Martin et al. [2003].

[8] Extensive evaluations of the model's output and observations are presented in a number of papers which give confidence in the ability of the model to reproduce the general features of tropospheric chemistry [*Bey et al.*, 2001a, 2001b; *Li et al.*, 2001; *Palmer et al.*, 2001; *Bell et al.*, 2002; *Chandra et al.*, 2002; *Fiore et al.*, 2002; *Jacob et al.*, 2002; *Li et al.*, 2002a, 2002b; *Liu et al.*, 2002; *Martin et al.*, 2002a, 2002b; *Liu et al.*, 2002; *Martin et al.*, 2002a, 2002b; *Liu et al.*, 2002; *Martin et al.*, 2002a, 2002b; *Palmer et al.*, 2003]. The model reproduces the seasonal and interannual variability of CO, but there is an underestimate in the tropics and Southern Hemisphere. Monthly O₃ is generally reproduced within 10 ppbv with no global bias. The model reproduces concentrations of nitrogen oxide (NO) and peroxyacetylnitrate (PAN) and columns of formaldehyde (H₂CO) and nitrogen dioxide (NO₂) generally within a factor of 2.

2.2. Aerosol and Trace Gas Emissions Inventory

[9] Figure 1 presents the fire-counts detected during the 1997 Indonesian burning event by the Along Track Scanning Radiometer (ATSR) World Fire Atlas [*Arino and Rosaz*, 1999]. The majority of fire-counts coincide with the locations of tropical peat swamp forests. *Nakajima et al.* [1999] and *Ikegami et al.* [2001] reported the optical and chemical properties of the smoke aerosols from the Indonesian fires, which indicate both wood and peat burning. The peat was made vulnerable to fire by the Indonesian government's land conversion program, which drained peat swamps. *Levine* [1999] estimated that the vast majority of aerosol and trace gas emissions in September and October



Figure 1. Total number of ATSR fire-counts detected per 1° longitude by 1° latitude grid-box in the Indonesian region from September to November 1997.

1997 on Sumatra and Borneo were from peat combustion, which typically smolders. Smoldering combustion is inefficient and produces higher emissions of CO and hydrocarbons than flaming combustion. The remainder of emissions was from forest wildfires and the burning of agricultural wastes in the fields. Most tropical peat is found in the coastal lowlands of Southeast Asia, especially Indonesia [*Rieley et al.*, 1995, 1996]. At least 20 Gt of carbon is stored in this peat, which is about 4 times the global annual carbon emission rate of fossil fuels [*Rieley et al.*, 1996].

[10] The uncertainties of the emission inventory for this burning event are large and include the total area burned, total biomass load, total biomass consumed, and emission factors. There is a wide range of estimates for the total area burned, about 4-13 million ha [Liew et al., 1998; Asian Development Bank, 1999; Fuller and Fulk, 2001; Goldhammer and Hoffmann, 2002]. Page et al. [2002] performed a detailed study of the total carbon released from peat and forest fires in a 2.5 million hectare study area of Central Kalimantan, and they extrapolated their emission estimate to the whole of Indonesia. The range of their estimate, 810-2570 Tg C, reflects the large degree of uncertainty associated with such a calculation. Yokelson et al. [1997] reported emission factors of CO₂, CH₄, CO, and various hydrocarbons for smoldering combustion from two samples of peat, one from Minnesota and the other from Alaska. Tropical peats are, however, mainly woody peats, while temperate-subarctic peats tend to be sedge and Sphagnum moss [Rieley et al., 1995; Shimada et al., 2001, and references therein]. Recently, Muraleedharan et al. [2000] conducted a laboratory experiment to quantify the emissions from tropical peat combustion, with the intent of reproducing smoldering conditions typical of peat combustion in the field in Brunei. They did not, however, account for all the carbon in the peat sample, and they reported emission factors for a few species that vary by orders of magnitude with temperature without discussing the reproducibility of the experimental results.

[11] The inventory of biomass burned used in this study was developed by J. A. Logan and R. Yevich (Harvard University) as described by *Lobert et al.* [1999] and *Duncan et al.* [2003]. This climatological, annual mean biomass burned inventory incorporates input data from many sources from 1980 to the early 1990s and is thus not representative of one individual year. We applied trace gas emission factors to the different vegetation types following the recommendations of *Andreae and Merlet* [2001] to obtain



Figure 2. Weekly total ATSR fire-counts from September to November 1997 detected in the whole Indonesian region (thick solid line) and on the islands of Borneo (dotted line), Sumatra (thin solid line), and New Guinea (dashed line).

emissions of CO, NO_x, and hydrocarbons, as described by *Staudt et al.* [2003]. A shortcoming of the inventory for our purposes is that it does not treat the combustion of peat, as the inventory was developed before the large-scale drainage of peat swamps began in 1995. We account for the trace gases emitted from the burning of peat indirectly, as discussed in the following paragraphs.

[12] We used the methodology of *Duncan et al.* [2003] to describe the timing, magnitude, and spatial distribution of biomass burned for different regions of the world. The magnitude of the biomass burned for each month from September to November 1997 was estimated using the TOMS Aerosol Index data (AI) product [Hsu et al., 1996; Herman et al., 1997; Torres et al., 1998] as a surrogate for biomass burning. We present here a brief example of the method discussed by Duncan et al. [2003] to estimate the total biomass burned for one region, Indonesia, for 1 month, September 1997. First, the climatological, annual biomass burned for Indonesia was multiplied by the ratio of the average AI for September 1997 for the Indonesian region to the mean AI of all 264 months from 1979 to 2000 for the same region. Next, fire-counts from the ATSR World Fire Atlas [Arino and Rosaz, 1999] were used to provide specific locations of the biomass burned within the Indonesian region on a 1° longitude by 1° latitude grid [Duncan et al., 2003].

[13] We performed one additional step beyond the method of *Duncan et al.* [2003] for the Indonesian region by redistributing the monthly average biomass burned into weekly averages based on the timing of the ATSR firecounts on southeastern Sumatra, southern Borneo, and southern New Guinea, the areas where the fires were concentrated (Figure 1). Figure 2 shows the fire-counts as weekly sums on Sumatra, Borneo, and New Guinea and the entire Indonesian region. The majority of the fire-counts were detected on Borneo and Sumatra, driving the variation in the timing of the entire region's fire activity. There is significant variation of the total weekly fires within each of the 3 months. The greatest number of fire-counts is seen in the second week of September, which is when the wildfires first flared. A second period of high fire-activity occurred during the last 2 weeks of October into the first week of November. The rainy season began by mid-November, suppressing fire activity.

[14] The error introduced in the emissions estimate by assuming that the biomass burned is forest, and not peat, cannot be determined, because the emission factors for tropical peat are not known, as discussed above. However, this method indirectly accounts for the burning of both peat and tropical forests as the total magnitude of the biomass burned was scaled to the AI, which includes aerosol from both types of biomass burned. We show in section 3 that the magnitude of the emissions that we use is likely reasonable, despite the exclusion of peat in the biomass burned inventory, through a comparison of the model's trace gas distributions with observations.

[15] Monthly total biomass burning emissions of CO and NO_x are shown in Table 1 for Borneo, Sumatra, New Guinea, and the world. About 45% of the Indonesian emissions occurred on Borneo, 38% on Sumatra, and 17% on New Guinea. The total CO emitted from the three islands from September to November was 124 Tg or 54% of the global biomass burning total for the same period (i.e., 228 Tg). About 6 Tg CO were emitted on other islands, giving a total of 130 Tg CO from the entire Indonesian region. Global biomass burning emissions are estimated to be about 228 Tg CO from September to November 1997 and 511 Tg CO for 1997 [Duncan et al., 2003]. Almost 2.6 Tg N was emitted as NO_x, about 44% of the global emissions from biomass burning from September to November (Table 1). The magnitude of our emissions from the burning event is higher than that of *Levine* [1999], who estimated that 38-114 Tg CO and 0.485-1.456 Tg N as NO_x were emitted in September and October 1997 on Sumatra and Borneo. The estimate of *Levine* [1999] is likely too low as it excludes emissions from New Guinea, parts of Borneo and Sumatra, and the month of November. There is the possibility that our emissions estimate is too low. Emissions may have continued even after the rains began in early November, as peat may have smoldered for months. Khandekar et al. [2000] reported that some peat deposits smoldered throughout the rainy season and rekindled in early April. Two limitations of the method for estimating emissions as presented by Duncan et al. [2003] are that smoldering fires may not be detected by ATSR and rain washes aerosol from the troposphere. In this

Table 1. Biomass Burning Emissions of CO (Tg) and NO $_x$ (Tg N) for 1997

	Borneo	Sumatra	New Guinea	Sum ^a	World
			СО		
Sept.	28.9	21.8	8.0	58.8	105.9
Oct.	17.9	17.6	7.1	42.6	77.4
Nov.	8.6	8.2	6.3	23.0	45.1
Sum	55.4	47.6	21.4	124.4	228.4
			NO _x		
Sept.	0.60	0.45	0.17	1.22	2.90
Oct.	0.37	0.37	0.15	0.88	1.94
Nov.	0.18	0.17	0.13	0.48	1.05
Sum	1.15	0.99	0.45	2.58	5.89

^aSum of emissions from the three islands.

Table 2.	Emissions	(Tg C)	of Black	Carbon	(BC)	and	Organic
Carbon (C	DC) Aeroso	ls for 1	997				

		Globe				Indonesia	
	Tg C	Biomass Burning ^a	Fossil Fuels ^a	Monoter- penes ^a	Tg C	Biomass Burning ^a	
			BC				
Sept.	3.0	82.1%	17.9%	0.0%	1.3	42.0%	
Oct.	2.3	76.0%	24.0%	0.0%	0.9	39.5%	
Nov.	1.6	65.8%	34.2%	0.0%	0.5	31.5%	
1997	18.7				2.6 ^b	14.2% ^c	
			OC				
Sept.	19.1	89.4%	4.5%	6.1%	8.8	45.8%	
Oct.	14.1	86.7%	6.3%	7.5%	6.4	45.1%	
Nov.	8.9	80.5%	9.7%	9.8%	3.4	38.5%	
1997	109				18.5 ^b	17.0% ^c	

^aPercent of global monthly source for September, October, and November.

^bSum from September to November only.

^cPercent of global annual source for 1997.

situation, only soluble trace gases would be removed by the rain, but the insoluble or slightly soluble trace gases, such as CO, would not.

[16] The 3-D aerosol concentration fields used in the GEOS-CHEM model are calculated using the GOCART model and are specific for our study period, September to December 1997. The emission rates for sea salt, sulfate, and dust aerosols in the GOCART model are monthly averages, while the emission rates for black carbon (BC) and organic carbon (OC) aerosols are weekly averages for the Indonesian region and monthly averages for the rest of the world. The 3-D aerosol concentration fields produced by the model are monthly averages, except for BC and OC aerosols, which are daily averages. The total monthly emissions of BC and OC aerosols are presented in Table 2 and contributed about 38% and 43%, respectively, to the global source of these aerosols during the study period, and 14% and 17%, respectively, for 1997. We also account for the formation of sulfate aerosols originating from sulfur dioxide (SO_2) released from the fires. In general, SO_2 emissions from biomass burning are a fraction of those from anthropogenic sources [Chin et al., 2002]. The model's sulfur emissions from biomass burning are small, and the sulfate aerosol optical depth (AOD) in the Indonesian region is generally less than 1% of the AOD of BC and OC aerosols. However, there are observations of the smoke from the Indonesian fires which suggest that the magnitude of SO₂ emissions was likely significant because of the high sulfur content in peat relative to other vegetation types [Nakajima et al., 1999; Ikegami et al., 2001; Okada et al., 2001; Langmann and Graf, 2003]. Therefore the biomass burning emissions of SO_2 from the Indonesian fires may be underestimated in this study.

[17] We estimate the total carbon (i.e., $CO + CO_2 + CH_4 + OC + BC$) emitted during the fires to be about 700 Tg. Our estimate is smaller than the estimated range of *Page et al.* [2002], 810–2570 Tg C. We believe their range is too high, as they assumed all the carbon consumed in the fires was released to the atmosphere. In reality, up to about 20% of the carbon in the biomass consumed can remain in the ash depending on several factors, such as the vegetation type [*Lobert et al.*, 1991]. We do not carry CO₂ and CH₄ as

tracers in our model simulation, as they are long-lived species and relatively well-mixed in the troposphere as compared to other trace gases. To approximate the emissions of CH₄ and CO₂, we use emission ratios (i.e., Δ CH₄/ Δ CO = 0.051 ppbv/ppbv and Δ CO/ Δ CO₂ = 0.089 ppbv/ppbv [*Matsueda and Inoue*, 1999]) inferred from observations in the free troposphere of pollution from the Indonesian fires. We estimate the emissions to be about 4 Tg CH₄ and 2300 Tg CO₂. Our estimate of the emissions represents less than 1% of the annual total emissions of CH₄, but about 10% of the annual fossil fuel emissions of CO₂ [*IPCC*, 2001]. The emissions of nonmethane hydrocarbons are small, generally less than a few percent of the total carbon content of the biomass [*Lobert et al.*, 1991; *Yokelson et al.*, 1997].

[18] The trace gas emissions used by *Chandra et al.* [2002] are the same as in the present study, except that we distribute Indonesian emissions into weekly averages instead of monthly averages as discussed above. Chandra et al. [2002] did not simulate the radiative and heterogeneous chemical effects of BC and OC aerosols from the fires. Hauglustaine et al. [1999] used the trace gas emission estimates of Levine [1999], which are likely too low, as discussed above. In addition, they artificially diluted the emissions by distributing them uniformly from August until December, instead of from the second week of September to the first week of November, when the fires occurred. Hauglustaine et al. [1999] did not account for the emission and transport of BC and OC aerosols as we do in the present study. Instead, they assumed a uniform aerosol layer from 0 to 2 km with a wavelength, independent value for the single-scattering albedo of 0.88, following measurements by Gras et al. [1999] over the burning regions of Indonesia. In section 3, we compare our model results to those of Hauglustaine et al. [1999] and discuss how the differences between the emission inventories impact the two models' predictions of trace gases and aerosols.

2.3. Model Simulations

[19] We present the results of five model simulations in section 3. The first simulation (SIM-STD) is a standard simulation including aerosol and trace gas emissions from the Indonesian fires. This simulation covers the period January through December 1997. The first 8 months are used for initialization, and we present model results for September through December. In the second simulation (SIM-NOFIRE), the emissions of aerosols and trace gases from the Indonesian fires in the GEOS-CHEM and GOCART models are "turned off" from September through December. The third simulation (SIM-NOAER) is the same as SIM-STD, except that we turned off the emissions of BC and OC aerosols from the Indonesian fires beginning in September while retaining the trace gas emissions to assess the impact of the aerosols on tropospheric chemistry. The fourth simulation (SIM-NOLIGHTNING) is the same as SIM-STD, except that we turned off NO_x emissions from lightning sources globally from September through December. The last simulation tracks several CO tracers, tagged as a function of their source regions in the GEOS-CHEM model. For example, "Indonesian" CO refers to CO emitted from the fires in the Indonesian region. The daily average concentrations of OH necessary for this simulation were archived in the standard simulation, SIM-STD. The tagged CO modeling approach is discussed by *Bey et al.* [2001b] and *Staudt et al.* [2001]. In section 3, we use the convention ΔX (e.g., X = CO, O_3 , NO_x) to signify the change in concentration, burden, or tropospheric column amount of trace gas X between two simulations. The differences that we report are due only to the effects of emissions and aerosols, as the transport (i.e., advection and convection) is the same in all simulations.

3. Results and Discussion

[20] In this study, we focus on the impact of the 1997 Indonesian fires on tropospheric chemistry, not the impacts caused by dynamical changes due to the 1997-1998 ENSO event. The changes in atmospheric circulation caused by the ENSO event were substantial [Chandra et al., 1998; Lau and Yang, 2002], however, and significantly impacted tropospheric chemistry. Chandra et al. [2002] performed sensitivity simulations using the GEOS-CHEM model and found that the ENSO-induced dynamical changes accounted for about half of the enhanced TCO in the Indonesian region in October 1997 relative to October 1996 as estimated by the CCD method using TOMS observational data [Ziemke et al., 1998]. Chandra et al. [1998] suggested that less convection in 1997 could enhance free tropospheric O₃ in the Indonesian region by (1) lofting less O_3 -poor air from the oceanic lower atmosphere and (2) lofting less water vapor, which decreases the destruction rate of O_3 by reaction with water vapor. Chandra et al. [2002] attributed the remaining portion of the TCO enhancement in 1997 to O_3 produced from the Indonesian pollution.

[21] CO in the Indonesian boundary layer during the rainy season, generally from November to April, is typically below 100 ppbv. During the burning event, however, Sawa et al. [1999] reported measured CO as high as 3-9 ppmv over Borneo, and Kunii et al. [2002] reported 20 ppmv over Sumatra. The model CO in the boundary layer over the burning regions was generally 2-4 ppmv from the second week of September to the second week of November, but the maximum daily average concentrations were as high as 7 ppmv. The maximum concentrations in the free troposphere above Indonesia were as high as 1.4 ppmv. Hauglustaine et al. [1999] reported that their model CO increased by as much as 2.9 ppmv in the boundary layer and 1 ppmv in the free troposphere. Their maximum CO is significantly lower than the maximum values in our model as their pollutant emission rates were much lower than in the present study (section 2.2). The maximum, daily average increases in model O_3 in the boundary layer and free troposphere were both about 45 ppbv because of the fires. This increase is calculated as the difference in O_3 between the model simulations with (i.e., SIM-STD) and without (i.e., SIM-NOFIRE) emissions from the Indonesian fires. Despite using different transport and emissions than in the present study, Hauglustaine et al. [1999] reported a similar increase of 45 and 50 ppbv in the boundary layer and free troposphere, respectively, over the burning regions. These high concentrations of pollution, including smoke particles, were transported to nearby cities, having deleterious effects on human health [Brauer and Hisham-

Table 3. Monthly Averaged Global Tropospheric Burdens of CO, O_3 , and NO_x , and Global Mean, Tropospheric OH for the Model Simulations With (SIM-STD) and Without (SIM-NOFIRE) the Indonesian Fires

	Sept.	Oct.	Nov.	Dec.
		CO, Tg		
SIM-STD	406	437	435	406
SIM-NOFIRE	381	380	371	367
ΔCO	25	57	64	39
$\Delta CO, \%$	6.6%	15%	17%	9.6%
		O_3 , Tg		
SIM-STD	310	319	314	307
SIM-NOFIRE	307	311	305	302
ΔO_3	3	8	9	5
$\Delta O_3, \%$	0.65%	2.6%	3.0%	1.6%
		NO _n Tg		
SIM-STD	0.163	0.173	0.184	0.204
SIM-NOFIRE	0.159	0.170	0.184	0.206
ΔNO_x	0.004	0.003	0.000	-0.002
ΔNO_x (%)	3.1%	1.8%	0.0%	-0.98%
	OH, ×	10 ⁶ molec cm ⁻¹	3	
SIM-STD	1.01	0.979	0.999	1.08
SIM-NOFIRE	1.03	1.04	1.07	1.13
ΔOH	-0.020	-0.061	-0.071	-0.05
$\Delta OH (\%)$	-2.1%	-5.7%	-6.8%	-4.4%

Hashim, 1998; Fang et al., 1999; Khandekar et al., 2000; Heil and Goldammer, 2001, and references therein; Koe et al., 2001].

[22] Table 3 shows the monthly average global tropospheric burdens of CO, O₃, and NO_x for the model simulations with (i.e., SIM-STD) and without (i.e., SIM-NOFIRE) emissions from the Indonesian fires and the corresponding ΔCO , ΔO_3 , and ΔNO_x , where a positive value means the burden was higher because of the fires. The global CO burden was 15% ($\Delta CO = 57$ Tg) higher in October and 17% ($\Delta CO = 64$ Tg) higher in November. The impact of the fires was less on the global O₃ burden (i.e., <3%).

[23] Figure 3 shows the monthly average ΔCO between the SIM-STD and SIM-NOFIRE simulations as a tropospheric column (molec cm⁻²) from September to December. The bulk of the pollution from Indonesia lay in the tropical Indian Ocean between 20°N and 20°S from September to November, where the ΔCO was generally greater than 1×10^{18} molec cm⁻², or greater than 50% relative to the column in the SIM-NOFIRE simulation in October and November. The ΔCO was greater than 5 $\times 10^{18}$ molec cm⁻² over the Indonesian region from September to November. The ΔCO was low in the equatorial Pacific, generally less than 0.2×10^{18} molec cm⁻².

[24] The pollution dispersed predominantly in the tropics and subtropics and had less impact on the extra-tropics of either the Southern or Northern Hemisphere. The column ΔCO was greater than 0.2×10^{18} molec cm⁻² over most of the tropics and subtropics from October to December (Figure 3). The ΔCO was less than 0.1×10^{18} molec cm⁻² poleward of 30°N and 30°S in September and October and poleward of 45°N and 45°S in November and December. The ΔCO was mixed zonally by December with similar distributions in both the Northern and Southern Hemisphere



Figure 3. Change in the monthly average tropospheric column of model CO, ΔCO (× 10¹⁸ molec cm⁻²), due to the fires (i.e., column in the standard simulation (SIM-STD) minus column in the simulation without trace gas and aerosol emissions from the Indonesian fires (SIM-NOFIRE)). The contour levels are 0, 0.1, 0.2, 0.5, 1, 2, 3, 4, and 5.

(Figure 3). In relative terms, however, the impact of the fires was greater in the extra-tropics of the Southern Hemisphere than in the Northern Hemisphere. For example, the ΔCO was greater than 10% between approximately

 30° N and 45° S in November and December. The ΔCO in December was 5-10% from 45° S -90° S, but less than 5% from 45° N -90° N. In general, the background CO column is higher in the Northern Hemisphere, as most fossil



Figure 4. Locations of CO and O_3 observation stations used in this study. The Japan Airlines (JAL) flight from Sydney, Australia, to Tokyo, Japan, occurred on October 14, 1997, and is represented as a solid line.

fuel burning occurs there. In addition, CO begins to accumulate at the higher latitudes of the Northern Hemisphere with the onset of winter.

[25] In the following subsections, we present comparisons of model CO and O_3 to observations taken at a number of sites. The geographical locations of these observation stations are shown in Figure 4.

3.1. Vertical Transport by Deep Convection in the Indonesian Region

[26] Deep convection is a major pathway for the transport of biomass burning pollutants from the tropical boundary layer to the free troposphere [Crutzen and Andreae, 1990; Pickering et al., 1996; Folkins et al., 1997; Jonquières and Marenco, 1998; Andreae et al., 2001]. Using a CTM driven by assimilated data provided by ECMWF, Taguchi et al. [2002] showed that deep convection significantly contributed to the vertical mixing of pollutants over Indonesia during the 1997 fires, despite the fact that there is generally less convection over Indonesia during an ENSO event. We see similar features in our model. Figure 5 shows the vertical distribution of the ΔCO (ppbv) and ΔO_3 (ppbv) between the SIM-STD and SIM-NOFIRE simulations along the equator for October. Indonesia lies on or near the equator. The ΔCO was greater than 300 ppbv throughout the tropospheric column over much of the Indonesian region. The ΔO_3 was between 10 and 20 ppbv throughout the tropospheric column, indicating the lofting of O_3 and/or its precursors from the boundary layer to the free troposphere.

[27] Figure 6 shows the monthly distribution of precipitation in the model, which we use as an indicator of the locations of deep convection. The southward progression of the monsoon rains from September to November 1997 is evident. The rains did not reach the major burning regions until mid-November. Figure 6 also shows precipitation data from the Global Precipitation Climatology Project (GPCP). GPCP is part of the Global Energy and Water Cycle Experiment (GEWEX) of the World Climate Research Program (WCRP) (http://orbit-net.nesdis.noaa.gov/arad/ gpcp/). The GPCP data is a mixture of infrared and microwave satellite estimates of precipitation with rain gauge data from more than 30,000 stations. The overall distribution of rainfall is similar between the model and GPCP data. There is a large area of convection in the model that extends over northwestern Sumatra into the South China Sea and northern

Borneo in October (Figure 6). Taguchi et al. [2002] also report strong convection near Sumatra in their model. This strong area of convection is also seen in the GPCP data, though not as pronounced as in our model. The representation of convection is a difficult problem in chemical/transport models because of the sub-grid size of these processes. Allen et al. [1997] evaluated the deep convective mass fluxes provided by DAO and found that the location of deep convective mixing is reasonable, though the frequency is generally overestimated in the tropics. Molod et al. [1996] found that the seasonal shift in the Intertropical Convergence Zone (ITCZ) is well reproduced by the model, but that the ITCZ is too broad. On the basis of the comparison with the GPCP precipitation data, it is not possible to rule out that our model overestimates deep convection during September to November 1997 in the Indonesian region.

[28] There are two sets of observations with which to evaluate our model near the fires. First, Matsueda and Inoue [1999] described CO data measured on board Japan Airlines (JAL) commercial passenger flights between Singapore and Tokyo, Japan. There was one flight during the study period on October 20, 1997. Figure 7 shows the vertical distribution of CO observed above Singapore (1.3°N, 103.8°E) during the ascent of the aircraft. CO was as high as 1.3 ppmv in the boundary layer and above 100 ppbv throughout the troposphere to 12 km indicating transport by deep convection. The model reproduces well the observations, lending some confidence to its convective scheme. Almost all model CO was attributable to Indonesian biomass burning (Figure 7). Second, ozonesonde data was collected at Watukosek (7.6°S, 112.6°E) on the island of Java, which lies south of the major burning areas on Borneo and Sumatra [Komala et al., 1996; Fujiwara et al., 2000]. The data were collected by the Earth Observation Research Center (EORC)/National Space Development Agency of Japan (NASDA) and are available from their data archive (http://www.eorc.nasda.go.jp/AtmChem/GLACE/indonesia/ index.html). Figure 8 shows the vertical profile of measured O_3 above the station compared with model O_3 . The model reproduces the general vertical structures of the observed O_3 , though it underpredicts O_3 in the boundary layer by 10-30 ppbv on most days. The model's underprediction in the boundary layer is not caused by an overestimation of aerosols, as they act to decrease the model O₃ at Watukosek by less than 5 ppbv. The underprediction is possibly associated with the horizontal resolution of the model, as



Figure 5. October-average vertical distributions of the difference in the concentrations of CO and O_3 (i.e., ΔCO (ppbv) and ΔO_3 (ppbv)) between the model simulations with (SIM-STD) and without emissions (SIM-NOFIRE) from the Indonesian fires along the equator and 15°S. Contours for the ΔCO are 10, 50, 100, 300, 500, and 700 ppbv and for the ΔO_3 are 2, 5, 10, 20, 30, and 50 ppbv. Indonesia lies between 90°E-150°E longitude and 10°S-10°N latitude.

the model box in which Watukosek lies is composed of about half ocean and half (densely populated) land.

[29] Interestingly, O_3 produced from lightning NO_x made a larger contribution to the total model O_3 than O_3 produced from its precursors emitted during the fires, despite the proximity of the station to the burning regions (Figure 8). Typically, total NO_x in the tropics is dominated by NO_x from lightning sources [*Lamarque et al.*, 1996]. The lightning NO_x is parameterized in the model as discussed in section 2.1. The model's lightning emissions over Indonesia in October 1997 are about two thirds the emissions in October 1996, a non-ENSO period, but have similar horizontal and vertical distributions. However, *Hamid et al.* [2001] reported that there was increased lightning activity over Indonesia during the ENSO event in March 1998

relative to March 1999, a non-ENSO period, as there was less, but more energetic convection.

3.2. Long-Range Transport

[30] Typically, Indonesia receives high annual precipitation as it lies under the ascending branches of two east-west circulation cells. The Walker Circulation, the large-scale circulation over the equatorial Pacific Ocean, is composed of easterly flow in the lower troposphere, rising air over Indonesia and the western Pacific Ocean, westerly flow in the upper troposphere, and descending air over the eastern Pacific Ocean [*Lau and Yang*, 2002]. A similar circulation cell, but of opposite flow, spans the equatorial Indian Ocean. Therefore one might expect pollution from Indonesia, in a typical year, to be lofted into the free troposphere



GEOS - September



Figure 6. Monthly rainfall (mm) for September, October, and November from the Global Precipitation Climatology Project (GPCP) database and the GEOS-CHEM model.

and transported to the equatorial Indian and Pacific Oceans in the upper branches of the two east-west circulation cells. During the ENSO event, the sea-surface temperatures in the eastern and central Pacific Ocean were anomalously high and, subsequently, convection shifted from the western to the eastern Pacific. The Walker Circulation was weak and not well organized, allowing only weak flow from Indonesia in the free troposphere to the equatorial Pacific Ocean.

[31] We used the NASA Langley trajectory model [*Pierce and Fairlie*, 1993; *Pierce et al.*, 1994] to compute

5-day isentropic forward trajectories to identify major export pathways of pollution from the fires in Indonesia. DAO wind fields at 4° latitude by 5° longitude were used to compute the trajectories. One trajectory was computed for each day beginning at 0000 UTC. A limitation of the trajectory model is that it does not represent sub-grid scale convective transport. The effect of latent heating on vertical (diabatic) motion is also ignored in the trajectory calculations. Since convection is found to mix pollutants throughout the column over Indonesia (see section 3.1),



Figure 7. Measured CO (asterisks) on a Japan Airlines (JAL) flight from Singapore to Tokyo on October 20, 1997. Daily average model total and "Indonesian" CO are shown as open squares and solid diamonds, respectively. The altitude of the aircraft is shown as a solid line.

we initialized parcels at three potential temperatures (i.e., 300 K, 320 K, and 340 K, representing the boundary layer, middle troposphere at about 600 mbar, and upper troposphere at about 300 mbar, respectively) on each day to investigate the transport pathways of the pollutants at various altitudes. There was little vertical evolution of the trajectories (not shown), as horizontal gradients of potential temperature are generally weak in the tropics [*Wallace and Hobbs*, 1977].

[32] Figures 9a and 9b show forward trajectories from Borneo and New Guinea, respectively, for September 10 to November 20. Forward trajectories from the burning region of southern Sumatra (not shown) indicate a very similar pattern as from Borneo. The flow in the boundary layer was generally weak in all 3 months from Borneo and New Guinea, with most of the trajectories remaining on or near the islands, especially Borneo (Figures 9a and 9b). Some pollution from Borneo and New Guinea was transported northward in the cross-equatorial surface flow, a typical feature of the Asian summer monsoon [*McGregor and* *Nieuwolt*, 1998]. The cross-equatorial flow is evident in Figure 10, which shows the October-average model winds (m s⁻¹) in the boundary layer, middle troposphere, and upper troposphere. The outflow of pollution in the middle troposphere was largely to the tropical Indian Ocean (Figures 9a and 9b) in the easterly flow, a persistent feature in all 3 months (Figure 10). The flow from Borneo in the upper troposphere was driven by the tropical easterlies in September, but underwent a transition in October to a dominant northwesterly flow in November as the Indian Ocean subtropical high, positioned at about 60°E and 30°S at the surface in Figure 10, migrated eastward toward Australia and weakened [*Yihui*, 1994]. The general flow from New Guinea in the upper troposphere became southerly by November as the high migrated.

[33] In sections 3.2.1-3.2.3, we use CO to identify the major long-range transport pathways of the Indonesian pollution, as a 5-day trajectory indicates only the initial transport pathway. The tropical lifetime of CO is relatively long, greater than 3 weeks, as compared to the timescales of transport. Three main long-range transport pathways are evident in Figure 3, to the tropical and subtropical North Pacific Ocean, South Pacific Ocean, and to the tropical Indian and Atlantic Oceans. Taguchi et al. [2002] simulated the transport of a tracer using a chemical transport model driven by assimilated meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) and identified these three major branches of transport on October 14, 1997, for the pollution from the Indonesian fires to (1) the west toward Africa over the tropical Indian Ocean; (2) the subtropical North Pacific Ocean through southern Japan; and (3) the subtropical South Pacific Ocean through Australia.

3.2.1. Transport to the Tropical Indian and Atlantic Oceans

[34] As discussed earlier, Table 3 shows the monthly average global burden of CO (Tg) resulting from the



Figure 8. Ozonesonde data (thick solid line; ppbv) and daily average model O_3 (dotted line) at Watukosek, Java (7.6°S, 112.6°E). The dashed line represents the contribution of O_3 produced from lightning emissions (i.e., the ΔO_3 (ppbv) between the SIM-STD and SIM-NOLIGHTNING simulations). The thin solid line represents the contribution of O_3 produced from Indonesian biomass burning emissions (i.e., the ΔO_3 between the SIM-STD and SIM-NOFIRE simulations).



Figure 9. (a) Forward trajectories (5 days) from Borneo initiated at potential temperatures of 300 K in the boundary layer (black lines), 320 K (\sim 600 mbar) in the middle troposphere (red lines), and 340 K (\sim 300 mbar) in the upper troposphere (green lines). The first trajectory begins on September 10 and the last one begins on November 20. (b) Same as Figure 9a, but initialized in New Guinea.

Indonesian fires (i.e., the ΔCO between the SIM-STD and SIM-NOFIRE simulations). About 89%, 73%, 63%, and 47% of the global ΔCO (Tg) in Table 3 in September, October, November, and December, respectively, lay in the tropical Indian Ocean region (0°-150°E), including the Indonesian region. The pollution was mixed throughout the troposphere over the Indian Ocean along the equator (Figure 5). The ΔCO was greater than 50 ppbv from Indonesia to central Africa, with concentrations greater than 10 ppbv over the Atlantic Ocean.

[35] Observations are sparse in the tropical Indian Ocean region for the study period. *Chandra et al.* [2002] compared the model TCO from a GEOS-CHEM simulation to the TCO estimated from the CCD method using observations from TOMS [*Ziemke et al.*, 1998; *Ziemke and Chandra*, 1999; *Ziemke et al.*, 2000, 2001]. They found that the model reproduces the main features of the estimated tropical TCO, including the Indonesian region from September to November 1997. This good agreement between the model TCO and the TCO calculated by *Chandra et al.* [2002] suggests

Figure 10. (opposite) October-average model winds (m s⁻¹) in the boundary layer, at about 600 mbar in the middle troposphere, and about 300 mbar in the upper troposphere. The sea level surface pressure is also shown in the top panel. The strength of the wind is indicated by the size of the arrow. The wind speed associated with the longest arrow is shown in the lower left-hand side of each panel. Note that the scales (i.e., the lengths of the arrows) are different between the three panels.



 $30^{\circ}S$ $120^{\circ}E$ $120^{\circ}E$ 180°



Figure 11. Daily average surface CO (ppbv) observed at Mahe Island, Seychelles (4.7°S, 55.2°E, 3 m). Total model CO is represented by the solid line, "Indonesian" CO by the dotted line, and observations by asterisks.

that the GEOS-CHEM model's assimilated meteorology captures the shift in the tropical convection pattern that is typical during an ENSO year [*Chandra et al.*, 1998; *Ziemke and Chandra*, 1999; *Chandra et al.*, 2002; *Sudo and Takahashi*, 2001].

[36] Surface CO is measured routinely as part of the cooperative National Oceanic and Atmospheric Administration (NOAA)/Climate Monitoring and Diagnostics Laboratory (CMDL) flask sampling program on Mahe Island, Seychelles (4.7°S, 55.2°E, 3 m) in the western tropical Indian Ocean [Novelli et al., 1997]. The data were provided by Paul Novelli (NOAA/CMDL, personal communication, 2002). Back trajectories from the island (not shown) indicate persistent transport of pollution from the central tropical Indian Ocean, a region with elevated levels of CO from Indonesia from September through December (Figure 3). Figure 11 compares observed, model total, and Indonesian CO. The contribution of Indonesian CO was greatest in October and November, with a maximum of about 150 ppbv in mid-November. The October to November 1997 averages of model and Indonesian CO are 215 and 85 ppby, respectively. We do not calculate an average of the sparse observations for the study period, but the average model CO from October to November 1997 is two and a half times higher than the October to November average observed CO at Seychelles from 1990 to 1996, 85 ppbv. There are only eight observations from September to December 1997, but they do suggest that the model reproduces the variation of CO at the site. The model underpredicts CO in December by about 20 ppby. The Indonesian wildfires stopped in the first week of November, about a month before the observations in December were taken. Possibly the biomass burned inventory for Indonesia underpredicts emissions from smoldering biomass in November and December as discussed in section 2.2.

3.2.2. Transport to the Tropical and Subtropical South Pacific Ocean

[37] There were two main pathways for the transport of pollutants from the wildfires to the South Pacific Ocean, a direct and an indirect pathway. The direct flow from Indonesia via Australia into the South Pacific became active in October and dominant in November in the upper troposphere (Figure 9a). *Matsueda et al.* [1999] reported trace gas concentrations measured on commercial JAL flights

between Sydney, Australia, and Tokyo, Japan, in September and October 1997. They found that the CO enhancements due to the fires maximized between 10°S and 30°S along the flight path in the western Pacific Ocean near the coast of Australia. There were eight JAL flights between Sydney and Tokyo during our study period. The measurements were taken between 8 and 13 km altitude, which is the approximate altitude range of the upper tropospheric CO maximum evident in Figure 5 at 15°S. Figure 12a shows the observed CO measured in the upper troposphere during the JAL flight on October 14, 1997. CO as high as 300 ppbv was measured above northern Australia. The model captures this high event, but the maximum total CO is only around 170 ppbv. About 100 ppbv of the model total CO is from the Indonesian wildfires. The reason for the model's underprediction is not known. Figures 12b and 12c show a horizontal cross-section of the model total and "Indonesian" CO plumes, respectively, at about 12 km on October 14, 1997. The plume was well defined and approximately 1000 km wide. It moved to the southeast over northern Australia and then headed eastward at about 20°S in the subtropical jet into the South Pacific Ocean. Similarly, observations indicate that biomass burning pollution from South America and southern Africa are commonly transported to the South Pacific Ocean in the subtropical jet during austral spring [Chatfield et al., 2002].

[38] The indirect transport to the South Pacific Ocean was persistent and occurred in the middle and upper troposphere as pollution transported over the Indian Ocean in the easterlies reversed direction around the subtropical high, located between 60°E and 90°E, and rapidly moved to the east over Australia in the southern subtropical jet. Figure 13 shows an example of this transport pathway on October 4, 1997. There is a tongue of "Indonesian" CO moving from the main plume to the southeast between Australia and southern Africa. The direct pathway over northeastern Australia is also evident in Figure 13.

[39] The October-average ΔCO (ppbv) between the SIM-STD and SIM-NOFIRE simulations along 15°S latitude, approximately the location of the axis of the plume in the South Pacific Ocean (Figure 3), was generally less than 10 ppbv below 600 mbar and between 10 and 50 ppbv above 600 mbar over the South Pacific Ocean (Figure 5). This finding indicates that little vertical mixing occurred as the pollution was transported in the subtropical jet. Our results are consistent with the findings of *Folkins et al.* [1997], who observed enhanced concentrations of trace gases in the free troposphere over Fiji in October 23–24, 1994. They attributed these enhancements to biomass burning in New Guinea.

[40] There are two sets of observations in this region with which to compare our model in addition to the JAL flight data. Figures 14a and 14b show observed and model O₃ at Samoa (14°S, 170°W) and Tahiti (18°S, 149°W), respectively, for 3 days each. The data from Samoa and Tahiti were obtained from the NASA/Global Tropospheric Experiment (GTE) data archive at NASA Langley (http://eosweb.larc. nasa.gov/project/gte/table_gte.html). The vertical distributions of ΔO_3 between the SIM-STD and SIM-NOFIRE simulations at 15°S were between 2 and 5 ppbv in the upper troposphere, generally 5% or less of the total O₃ (Figure 5).



The model simulates the general vertical structure and concentrations of the observed O_3 .

[41] Rinsland et al. [2001], using back-trajectory analyses, found that the CO column observed at Wollongong, Australia (34.5°S, 150.9°E, 30 m) during the time of the Indonesian fires was likely due to biomass burning emissions from South America and southern Africa with little impact from Indonesia. Our back-trajectory analysis (not shown) agrees that almost all of the parcels throughout the troposphere originated to the west of the station and at higher latitudes than 20°S. Figure 15 shows the CO column measured above Wollongong from September to December 1997 (Curtis Rinsland, NASA Langley Research Center, personal communication, 2001). The model reproduces the observations reasonably well. The contributions of "Indonesian," "South American," "Australian," and "African" model CO from biomass burning sources are also shown in the figure. There are four separate plumes of "Indonesian" CO affecting the site, two in early October, a week-long one in mid-November, and a prolonged one from mid-November to mid-December. While the contribution to total CO was high from "Indonesia" during the events, the contributions from "Africa," "South America," and "Indonesia" were comparable for much of October, November, and December.

3.2.3. Transport to the Tropical and Subtropical North Pacific Ocean

[42] Transport to the North Pacific Ocean was minor as compared to transport to the Indian and South Pacific Oceans. There were three major pathways for the flow of pollutants from Indonesia to the North Pacific Ocean. The first pathway occurred in the boundary layer in the crossequatorial flow associated with the East Asian Monsoon (Figures 9a, 9b, and 10). The second pathway occurred in the upper troposphere from New Guinea in November (Figure 9b). The third pathway occurred in October and November, as the pollution on the northern edge of the main plume in the Indian Ocean was rapidly transported in the northern subtropical jet in the upper troposphere. An example of this pathway is seen in Figures 12b and 12c on October 14, 1997.

[43] Figure 16 shows the total CO column observed at Mauna Loa, Hawaii (19.5°N, 155.6°W; 3.4 km). The data were provided by Curtis Rinsland (NASA Langley Research Center, personal communication, 2001). *Rinsland et al.* [1999] used back-trajectory analyses to identify possible source emission regions for high CO columns in October and November 1997. Their analyses showed that the emissions likely originated in the low northern latitudes of Southeast Asia. *Harris and Kahl* [1990] and *Jaffe et al.*

Figure 12. (opposite) (a) Concentrations (ppbv) of total model CO (squares), "Indonesian" CO (diamonds) and observed CO (asterisks) during the Japan Airlines (JAL) airliner flight on October 14, 1997, from Sydney, Australia, to Tokyo, Japan. The aircraft altitude is shown as a solid line. Model CO is daily average. Daily average model (b) total CO (ppbv) and (c) "Indonesian" CO (ppbv) at ~12 km on October 14, 1997. The solid line represents the path of the Japan Airlines (JAL) flight from Sydney, Australia, to Tokyo, Japan.



Figure 13. Daily average "Indonesian" CO column ($\times 10^{18}$ molec cm⁻²) on October 4, 1997. The contour levels are 0, 0.5, 1, 2, 3, 4, and 5 $\times 10^{18}$ molec cm⁻².

[1997] showed with back-trajectory analyses that transport from Asia is frequent at Mauna Loa. The westerly winds along the northern branch of the Pacific High transport air from Asia and veer to arrive at Mauna Loa from the north or northeast [*Merrill*, 1989]. The model simulates well the elevated CO column observed at Mauna Loa in mid-October, but it underpredicts the column observed in September by about 20% and does not capture the elevated CO observed in mid-November. The variations in model CO at Mauna Loa in September are largely due to variations in CO from "Asian" fossil fuel and "Indonesian" biomass burning sources (Figure 16).

3.3. Oxidizing Capacity

[44] In this section, we assess the impact of the fires on CO, O_3 , aerosols, and OH, the troposphere's primary oxidant. Figure 17 shows the tropospheric columns from



Figure 14. (a) Ozonesonde data (solid line; ppbv) and daily average model O_3 (dotted line) at Samoa (14°S, 170°W). (b) Same as Figure 14a, except for Tahiti (18°S, 149°W).

the standard simulation (i.e., SIM-STD) of CO, O₃, and OH, and AOD of BC and OC at 400 nm for October. The ΔCO , ΔO_3 , ΔAOD , and ΔOH are shown as percent differences relative to the SIM-NOFIRE simulation, where a positive value indicates a larger column in the SIM-STD simulation than in the SIM-NOFIRE simulation. The ΔCO was greater than 80% over Indonesia and greater than 40% over most of the tropical Indian Ocean. A ΔCO plume that was at least 20% covered about 30% of the tropics and stretched from tropical Africa eastward to the tropical South Pacific Ocean. The ΔO_3 was greater than 5% or 2 DU from tropical Africa to the South Pacific Ocean. The maximum ΔO_3 , 30% or about 15 DU, was near Sumatra. The $\triangle AOD$ was 40% over most of the tropics, except Africa, South America, and the Atlantic Ocean, where the aerosol loading was high due to biomass burning on those continents. The ΔNO_r (not shown) covered a smaller area spatially than either the ΔCO or ΔO_3 due to its relatively shorter lifetime.

[45] The wildfires had a major impact on OH in the Indonesian region where OH decreased by 80% near Sumatra and by more than 10% over the tropical Indian Ocean. OH increased 10% over a small area near New Guinea. The impact on the global, mean tropospheric OH concentration was substantial, especially since the mean OH is heavily weighted to concentrations in the tropics. Table 3



Figure 15. Tropospheric CO column ($\times 10^{18}$ molec cm⁻²) at Wollongong, Australia (34.5°S, 150.9°E, 30 m). Observations are represented as asterisks and daily average model total CO as a thick solid line. The green, fine black, red, and purple lines represent "Indonesian," "Latin American," "African," and "Australian" biomass burning CO, respectively, from the model. The sum of CO from these four source regions is shown as a dotted black line.



Figure 16. Tropospheric CO column ($\times 10^{18}$ molec cm⁻²) at Mauna Loa, Hawaii (19.5°N, 155.6°W; 3.4 km), as observed (asterisks) and modeled (upper solid line). "Indonesian" biomass burning and "Asian" fossil fuel sources are shown as a dashed and a dotted line, respectively, and their sum is shown as the lower solid line. Model CO is daily average.

shows the global mean, tropospheric OH concentrations for the model simulations with (i.e., SIM-STD) and without (i.e., SIM-NOFIRE) the Indonesian fires. The global, mean OH concentration, $\sim 1 \times 10^6$ molec cm⁻³ in the simulation with the fires, was about 6% lower in both October and November than in the simulation without the fires. The mean concentration was over 4% lower in December. In order to put the impact of the Indonesian wildfires into perspective, *Dentener and Crutzen* [1993] reported that N₂O₅ hydrolysis in aerosols decreases the annual, mean tropospheric OH by about 9% globally. *Martin et al.* [2003] reported that the radiative and heterogeneous chemical effects of aerosols beyond N₂O₅ hydrolysis in aerosols decrease the annual, mean tropospheric OH an additional 9% globally.

[46] A common indicator of the oxidizing capacity of the atmosphere is the lifetime of methylchloroform (CH₃CCl₃), a trace gas with exclusively anthropogenic sources [e.g., Lovelock, 1977; Singh, 1977; Logan et al., 1981; Prinn et al., 1995]. The dominant loss pathway of CH₃CCl₃ is by reaction with OH in the troposphere with additional minor loss pathways through photolysis and OH in the stratosphere [e.g., Volk et al., 1997] and uptake by oceans [Butler et al., 1991]. Following the method of Spivakovsky et al. [2000], we calculate the average CH₃CCl₃ lifetime during the four-month period in the SIM-STD simulation to be 6.8 years with respect to only tropospheric OH and 5.3 years with respect to all sinks. Prinn et al. [2001] reported an annual-average atmospheric lifetime with respect to all sinks of 4.9 (+0.6, -0.5) years, and Spivakovsky et al. [2000] reported 4.6 years. The lifetimes in the model with respect to only tropospheric OH for September, October, November, and December are 0.10 (1.5%), 0.31 (4.5%), 0.36 (5.1%), and 0.20 (3.0%) years longer, respectively, in the simulation with the Indonesian fires than in the simulation without.

[47] Why did the fires decrease OH so strongly? There are numerous competing effects of the trace gases and aerosols on OH. OH is predominantly formed by the reaction of water with an excited oxygen atom, $O^{1}D$, produced through the photolysis of O_{3} by certain wave-

lengths of UV light. Elevated levels of CO act to depress OH. Low concentrations of NO_x can enhance OH during the conversion of NO to NO_2 (e.g., the photolysis of NO_2 followed by NO + HO₂ \rightarrow NO₂ + OH), while high concentrations can depress OH through the formation of HNO_3 (i.e., $OH + NO_2 \rightarrow HNO_3$). Martin et al. [2003] found that BC and OC aerosols act to decrease OH and O₃ in the GEOS-CHEM model through the attenuation of UV wavelengths of light, and that OH is further decreased through the heterogeneous loss of odd oxygen species, such as HO₂, on aerosol surfaces. We performed the SIM-NOAER simulation (i.e., the same simulation as SIM-STD, except without aerosol emissions from the Indonesian fires) to quantify the radiative and heterogeneous chemical impacts of aerosols on OH. Figure 18 shows the tropospheric column ΔCO , ΔO_3 , and ΔOH as percent differences between the SIM-STD and SIM-NOAER simulations. The ΔO_3 and ΔOH are -5% (-2 DU) and -10%, respectively, over a large portion of the tropical Indian Ocean. The maximum ΔO_3 is -46% or about -21 DU. The ΔCO is more than 10%, as reaction with OH is the dominant sink for CO. The vertical distributions of the ΔO_3 (ppbv) and $\Delta OH \ (\times 10^5 \text{ molec cm}^{-3})$ along the equator were both negative throughout the troposphere. The largest negative values (i.e., >30% for O₃ and >70% for OH) occurred near the aerosol source regions in the Indonesian boundary layer. The results of the SIM-NOAER simulation indicate that aerosols from the Indonesian fires significantly depressed the O_3 production efficiency and the oxidizing capacity of the troposphere over a large portion of the tropics and subtropics.

[48] Martin et al. [2003] reported that the decreases in OH near the surface in India, $\sim 80\%$, by the radiative and heterogeneous chemical effects of aerosols in the GEOS-CHEM model were of comparable magnitudes, $\sim 40\%$. The decrease in OH due to the radiative effect is associated with the decrease in the photolysis of O₃ and NO₂ and due to the chemical effect by uptake of HO₂, mostly on OC aerosols. The chemical effects dominate at higher altitudes as the overhead aerosol column is smaller. We found similar aerosol impacts for the Indonesian fires as reported by Martin et al. [2003]. On average for October, the decrease in OH at the surface due to the presence of aerosols is about 60% near the burning regions and as high as 85% over Sumatra. At 5 km, the decrease is about 30% and as high as 50% over Sumatra. The radiative impact of the aerosols on OH is complicated as BC absorbs and OC mostly scatters. The model AOD at 400 nm of BC and OC aerosols is about 0.30 on average in October near the burning regions and as high as 0.85 in Sumatra. The smoke aerosols decrease the photolysis rate for O_3 at the surface by approximately 30-70% near the burning regions and by nearly 85% in Sumatra. A similar effect is seen for the photolysis rate for NO₂. The influence of the aerosols diminishes with height. At 5 km, the photolysis rates for O₃ and NO₂ are generally 10-20% lower and about 40% lower over Sumatra. The photolysis rate for NO₂ slightly increases in the upper troposphere.

3.4. Radiative Forcing

[49] We use the standard concept of radiative forcing [*Ramaswamy et al.*, 2001] to assess the instantaneous



Figure 17. (left) October-average tropospheric AOD of OC and BC aerosols and total tropospheric columns for CO, O₃, and OH for the standard simulation (i.e., SIM-STD). (right) The ΔCO , ΔO_3 , ΔAOD , and ΔOH relative to the simulation without Indonesian biomass burning aerosol and trace gas emissions (i.e., SIM-NOFIRE) are shown as percent differences.

change in the radiative balance of the Earth system resulting from the Indonesian wildfires. We discuss the direct radiative forcing of O₃, CO₂, and CH₄, and BC, OC, and sulfate aerosols. The forcing from O₃ is calculated using the relationship of 0.034 W m⁻² per DU change in the mean tropospheric column following *Mickley et al.* [1999]. Figure 19 shows the October-average radiative forcing (W m⁻²) from O₃ at the top of the troposphere. The forcing is positive, as tropospheric O₃ absorbs upwelling terrestrial longwave radiation. The forcing approached +0.5 W m⁻² over Indonesia and was greater than $+0.1 \text{ W m}^{-2}$ over nearly the entire tropical Indian Ocean. For comparison, *Mickley et al.* [1999] calculated a globally averaged forcing of $+0.44 \text{ W m}^{-2}$ due to anthropogenic O₃ relative to preindustrial times.

[50] Figure 19 also shows the October-average shortwave, direct radiative forcing from OC and BC aerosols at the top of the atmosphere (TOA). We do not present the forcing of sulfate aerosols, as its AOD was less than a few percent of the AOD of BC and OC aerosols as discussed in



Figure 18. October-average ΔCO , ΔO_3 , and ΔOH (molec cm⁻²) as percent differences of the tropospheric columns between the simulations with (i.e., SIM-STD) and without Indonesian biomass burning aerosol emissions (i.e., SIM-NOAER).

section 2.2. The calculation is carried out using the Goddard radiative transfer model [*Chou and Suarez*, 1999], which is coupled with the GOCART model. The shortwave radiative processes are parameterized in 11 spectral bands (0.2–10 um). The calculation assumes clear sky conditions and the aerosols are externally mixed. The forcing by OC aerosols is between -1.0 and -0.1 W m⁻² over much of the tropics, especially the southern hemisphere tropics, and -1 to -5 W m⁻² over much of the tropical Indian Ocean. The maximum forcing is -42 W m⁻² over the burning regions. A similar distribution of forcing is seen for BC aerosols though of opposite sign. The maximum forcing of

BC aerosols is +16 W m⁻² over the burning regions. The absolute magnitudes of the forcings at the TOA for OC and BC aerosols are an order of magnitude greater than the forcing of O₃. Interestingly, the net forcing of O₃ and OC and BC aerosols is nearly null over most of the tropics, except over the main burning regions of Borneo, Sumatra, and New Guinea where it is about -10 W m⁻² and south of Indonesia where it is greater than +1 W m⁻² (Figure 19). *Podgorny et al.* [2003] estimated the radiative forcing at the TOA by integrating satellite derived AOD and cloud cover with in situ observations of single scattering albedo and a Monte-Carlo Aerosol-Cloud radiation model of *Hess et al.*



Figure 19. October-average radiative forcing (W m⁻²) of O₃ at the top of the troposphere and OC and BC aerosols at the top of the atmosphere (TOA) associated with the Indonesian fires. The net forcing is shown in the bottom panel.



Figure 20. October-average radiative forcing (W m^{-2}) of OC and BC aerosols at the surface associated with the Indonesian fires. The net forcing is shown in the bottom panel.

[1998]. They performed sensitivity tests to clouds and reported that the forcings calculated by their model for clear and cloudy skies are within about 20% of one another. They reported a somewhat similar distribution and magnitude of forcing at the TOA for September to November 1997 for cloudy skies as we do even though we calculated the forcing at the top of the troposphere for October 1997 and for clear skies. Their forcing is as low as -12 W m^{-2} over the ocean in the Indonesian region and ours is about -10 W m^{-2} . Their estimated forcing is much higher over the central Indian Ocean, $-1 \text{ to } -6 \text{ W m}^{-2}$, than our estimated forcing, 0 to -1 W m^{-2} .

[51] Figure 20 shows the forcings (W m⁻²) of OC and BC aerosols at the surface. The forcings of both aerosols are negative, as all aerosols reduce solar radiation at the surface [*Ramanathan et al.*, 2001]. The absolute magnitudes of the forcings of OC and BC aerosols at the surface are higher than at the TOA and reach -65 and -113 W m⁻²,

respectively, in a few model grid boxes. The net forcing at the surface is $-10~W~m^{-2}$ over much of the tropical Indian Ocean and -150 to -178 W m⁻² over Indonesia. The forcing by OC aerosols was generally between 20 and 40% of the net forcing, despite the fact that the mass (Tg C) of OC aerosols emitted was seven times greater than BC aerosols (Table 2). Podgorny et al. [2003] also estimated the radiative forcing at the surface. Their forcing is as low as -50 W m^{-2} over the ocean in the Indonesian region and ours is between -10 and -50 W m⁻². Their estimated forcing, -10 to -40 W m⁻², is similar to ours, -5 to -50 W m⁻², over the central Indian Ocean. Satheesh and Ramanathan [2000] reported surface forcings by aerosols over the tropical Indian Ocean from -12 to -30 W m⁻² and as low as -70 to -75 W m⁻² during pollution events. Therefore our calculations show that the aerosols from Indonesian wildfires had a significant impact on the radiative balance of the tropical Indian Ocean, which may have temporarily

perturbed climate variables such as clouds or specific humidity.

[52] We now consider the potentially important direct forcing effects of CO₂ and CH₄ emitted during the burning event. As discussed in section 2.2, we do not simulate CO_2 and CH₄ as tracers in the model, but we estimate the emissions of CH₄ and CO₂ to be about 4 Tg and 2300 Tg, respectively, which represents less than 1% of the annual total emissions of CH4 and about 10% of the annual fossil fuel emissions of CO₂ [IPCC, 2001]. Though the Indonesian fires are a large source of CO₂ relative to its annual total emissions, its lifetime is long so that its tropospheric burden is large. Using the model distributions of CO in Figure 5 and the inferred emission ratios of CO, CO₂, and CH₄ of Matsueda and Inoue [1999], we estimate that the maximum ΔCH_4 and ΔCO_2 in the free troposphere above Indonesia in October are about 15 ppbv and 3.4 ppmv, respectively, which is less than 1% of the typical tropospheric concentrations of both molecules. Therefore we conclude that the instantaneous forcings of CH₄ and CO_2 from the fires are small relative to the aerosol forcing.

[53] We do not take into account the potentially important indirect effects of O_3 , CO, and aerosols (e.g., effects on clouds) on the radiative forcing [*Daniel and Solomon*, 1998; *IPCC*, 2001] or estimate the net impact of the fires on surface temperatures, as our model uses assimilated meteorology. The indirect effects must be assessed with a model that couples aerosols, chemistry, and climate [*IPCC*, 2001]. *Chan et al.* [2001] attempted to derive the surface temperature response to the Indonesian fires by using the same relationship of O_3 column change to forcing [*Mickley et al.*, 1999] and then applying a model-derived ratio of temperature change to forcing. This approach not only neglects the forcing due to aerosol, but also the complex suite of factors such as winds, clouds, and water vapor that affect local, surface temperature.

4. Conclusions

[54] The Indonesian wildfires of 1997 occurred mainly from the second week of September through the first week of November releasing, for example, ~130 Tg of CO, equivalent to $\sim 30\%$ of the annual global emissions from fossil fuel combustion. We conducted a model simulation of this extreme burning event to quantify its impacts on global tropospheric chemistry. The GEOS-CHEM model is driven by assimilated meteorology and uses an emission inventory of trace gases and aerosols specific to the study period, September through December 1997. Its ability to capture many of the daily variations of observed CO and O₃ in regions affected by the pollution (1) lends confidence for its use as a tool to study the transport pathways of the pollutants and their impacts on tropospheric chemistry and (2) indicates that the magnitude of the trace gas emission inventories is reasonable.

[55] We used isentropic forward trajectories from Indonesia and the relatively long-lived pollutant, CO, as a tracer of transport to identify the major export pathways of the pollution. Model CO ranged from 2 to 4 ppmv in the boundary layer for many weeks near the burning regions on Sumatra, Borneo, and New Guinea. Areas of deep convection lay to the north of the major burning regions, especially off the northwestern coast of Sumatra, and lofted the pollution throughout the tropospheric column above Indonesia. The dominant outflow of pollution from the Indonesian region was by tropical easterlies to the Indian Ocean in the free troposphere. Export from the islands in the boundary layer was generally weak in all 3 months. The bulk of the pollution lay in the tropical Indian Ocean between about 20°N and 20°S from September to November where the background CO column was elevated by more than 50% because of the fires. The October and November-average tropospheric burdens of CO and O₃ from the fires were about 60 and 10 Tg, respectively.

[56] Another important export pathway, to the tropics and subtropics of the South Pacific Ocean, developed as the easterly flow in the upper troposphere over Indonesia shifted to a northwesterly flow from September to November as the Indian Ocean subtropical high migrated eastward and weakened. A more persistent pathway to the South Pacific Ocean occurred as some of the pollution that was transported to the tropical Indian Ocean subtropical high and was rapidly transported in the southern, subtropical westerlies. There was episodic transport of pollution from Indonesia to the tropical and subtropical North Pacific Ocean via East Asia where the Indonesian pollution mixed with fossil fuel emissions before being exported from the continent.

[57] While the pollution from the Indonesian fires dispersed largely in the tropics and subtropics, it did have an impact on the extra-tropics of both the Northern and Southern Hemisphere. By December, CO was zonally mixed, somewhat symmetrically about the equator and its global burden was almost 40 Tg (~10%) higher than if the fires had never occurred, with half residing in the tropical Indian Ocean region. The tropospheric CO column was elevated by 10-20% from 30° N to 45° S in December, by 5-10% poleward of 45° S, and by less than 5% poleward of 45° N. The relative impact of the fires was lower in the Northern Hemisphere, as the total CO column is generally higher because most fossil fuel emissions are found there.

[58] The net effect of the trace gases and aerosols from the fires was to decrease OH and, subsequently, the oxidizing capacity of the troposphere. OH decreased by more than 50% near Indonesia and by more than 20% over much of the tropical Indian Ocean through consumption by CO, heterogeneous loss of HO_x on aerosols, and reduction of UV light by aerosols. The global mean, tropospheric OH decreased by about 6% in October and November. The global CH₃CCl₃ lifetime, an indicator of the oxidizing capacity of the atmosphere, increased by about 5% in October and November. Through a comparison of model simulations with and without BC and OC aerosols from the Indonesian fires, we found that the presence of the aerosols decreased O₃ and OH throughout the tropospheric column by more than 20% and 40%, respectively, in the Indonesian region and by more than 5% and 10%, respectively, over most of the tropical Indian Ocean.

[59] The fires significantly impacted the Earth's radiative budget, especially over Indonesia and the tropical Indian Ocean. The net radiative forcing of OC and BC aerosols at the surface was -10 W m^{-2} over much of the tropical

Indian Ocean and as low as -150 W m^{-2} over the burning regions in Indonesia. The forcing at the top of the atmosphere was an order of magnitude less than at the surface because the individual forcings for the two aerosol types were of similar distributions, but opposite signs. We estimate that the forcings associated with O₃, CO₂, and CH₄ from the fires were minor relative to the forcings of the OC and BC aerosols.

[60] We estimate that about 700 Tg C were emitted during the burning event, which is about 3-4% of the total carbon stored in peat in Indonesia [*Rieley et al.*, 1996]. Although the Indonesian fires of 1997 appear to be an extreme and isolated event, the frequency of such intense fires in the region may increase over the coming decades if the unsustainable exploitation of the region's natural resources continues and/or the severity of ENSO-induced droughts increase in response to possible climate change [*IPCC*, 2001]. Thus our work can be seen as an attempt to quantify the impact of future climate change on tropospheric chemistry.

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