Comparison between global chemistry transport model results and Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) data

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Abstract. Ozone distributions from state-of-the-art global three-dimensional chemistry transport models are compared to O_3 data collected on Airbus A340 passenger aircraft as part of the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) project. The model results are compared to monthly averaged data at cruise altitudes in the upper troposphere and lower stratosphere and monthly averaged vertical profiles collected over particular cities during takeoff and landing. The models generally show good agreement with the data in regions which have previously been well documented and where the meteorology is well understood/captured by meteorological models (e.g., over Europe). However, in the upper troposphere and lower stratosphere, models often fail to capture sharp gradients across the tropopause and from the subtropics to the tropics. In some models, this is related to deficiencies in model transport schemes and upper boundary conditions. Also, regions of the globe where our understanding of meteorology is poorer and emissions are less well known (e.g., tropics, continental Africa, Asia, and South America) are not simulated as well by all models. At particular measurement locations, it is apparent that emission inventories used by some global models underestimate emissions in certain regions (e.g., over southern Asia) or have incorrect seasonal variations (e.g., biomass burning over South America). Deficiencies in chemical schemes may also explain differences between models and the data.

1. Introduction

Well validated global chemistry transport models are required to study the impact of changing source gas emissions on the chemical composition of the troposphere and lower stratosphere, and in particular, the O_3 budget of this region. O_3 is an important urban pollutant and greenhouse gas. Recent work has shown that radiative forcing resulting from O_3 change is sensitive to changes occurring in the middle troposphere [*Hansen et al.*, 1997; *Forster and Shine*, 1997] as well as around the tropopause region [*Lacis et al.*, 1990]. The latter is a region where subsonic aircraft emit NO_x which may be perturbing the O_3 distribution [see *Brasseur et al.*, 1998a, and references therein]. The tropospheric budget of O_3 is complex. In the troposphere, the lifetime of O_3 (weeks to a few months) is such that its distribution is governed by transport processes as well as photochemistry. The major components are downward flux from the stratosphere, transport of O_3 (or its precursors) from the surface by convective or frontal processes, loss at the surface by dry deposition, and photochemical production or destruction. The latter depends largely on levels of NO_x and hydrocarbons.

Ozonesonde data have shown that there is a large day-to-day variability in the vertical distribution of O_3 [e.g., *Oltmans et al.*, 1996] and often distinct layers are evident. On seasonal timescales, the vertical distribution shows O_3 increasing with altitude from the surface up to the tropopause where strong gradients exist into the lower stratosphere. In general, O_3 fields from global tropospheric chemistry models [e.g., *Roelofs and Lelieveld*, 1996; *Muller and Brasseur*, 1995; *Hauglustaine et al.*, 1998] have been compared to seasonally av-

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eraged ozonesonde data which are available at a limited number of locations worldwide [*Oltmans et al.*, 1989; *Komhyr et al.*, 1992; *Oltmans and Levy*, 1994]. This climatological data set, often calculated from biweekly or weekly ascents collected over many years, has provided a valuable benchmark against which to compare models on seasonal time scales. Recently, the models used in this paper have been compared to ozonesonde data as part of an International Global Atmospheric Chemistry/Global Integration and Modelling (IGAC/GIM) project model comparison exercise [*Kanakidou et al.*, 1999].

In this paper, we compare results from several state-of-the-art chemistry transport models (CTMs) with O_3 data from the European Union (EU) Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) project. Recent papers by *Marenco et al.* [1998] and *Thouret et al.* [1998a, b] describe the MOZAIC project and O_3 data set in more detail. MOZAIC data have already been used to evaluate the TOMCAT model which was run for summer and winter periods and compared on a flight by flight basis [*Law et al.*, 1998]. Here we compare output from five CTMs against monthly averaged O_3 observations calculated using 2 years of MOZAIC data. The data were collected at aircraft cruise altitudes (8 to 12 km) spanning the tropopause region and during ascent and descent of the aircraft over major cities. This is the first time that data have been available on a reasonably regular basis at many of the locations discussed here.

In this paper, the MOZAIC data were used to evaluate the models and to draw conclusions about model performance following the validation exercise. Also, the observed seasonal cycle in O_3 at 31 cruise locations and 23 vertical profile locations have been examined for new features. Models were compared to the observed annual cycle, discrepancies were noted, and reasons were put forward to explain the differences. Model and measured standard deviations were also compared to see if models can capture the observed variability. In order to draw general conclusions about model performance, the observations were divided into specific regions, and systematic discrepancies were identified. It was also important to compare models with locations where data coverage was sufficient. Problems with sampling in both the data and the models are discussed in this context. A subset of the available observations and model results have been selected to illustrate the main findings.

The MOZAIC data are discussed in section 2, the general characteristics of each model are discussed in section 3 and the methodology for the comparison exercise is discussed in section 4. The results are described region by region in section 5, and general conclusions about model performance are summarized in section 6.

2. MOZAIC Data

The MOZAIC project consists of automatic instrumentation, to measure O_3 and water vapor, installed on five long-range Airbus A340 aircraft in normal airline operation. Regular flights started in September 1994 and are still being collected. The main routes are from Europe to North America, Europe to South America (especially Rio–Sao Paulo), Europe to China and Japan (Bejing, Seoul, and Tokyo-Osaka), Europe to Southeast Asia (Bangkok, Hanoi, and Saigon) and Europe to South Africa (namely, Johannesburg). Further details are given by *Marenco et al.* [1998] and *Thouret et al.* [1998a, b]. At cruise altitudes, the aircraft typically fly at five constant pressure heights between 300 hPa and 200 hPa. Approximately 90% of the MOZAIC measurements are collected during the cruise phase (i.e., 8 to 12 km altitude range), and the remainder are collected during takeoff and landing at airports. The cruise data and the vertical profile data are used separately in this study.

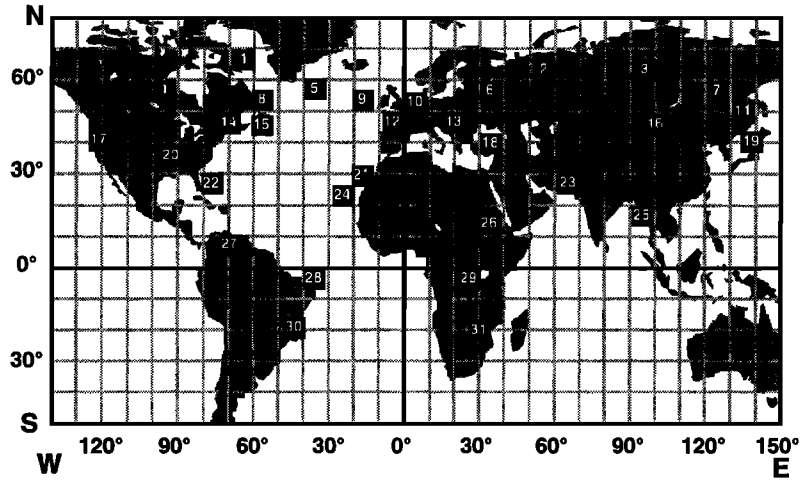

Figure 1. Geographical locations of the 31 altitude stations at the five cruise levels used for the comparison between monthly mean MOZAIC O_3 and global CTM results, with an indication of their code number.

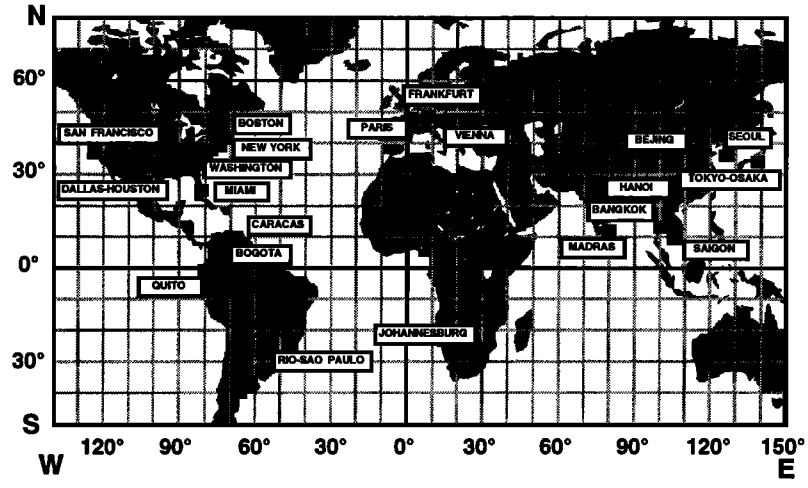

Figure 2. Geographical location of the 21 cities used for the comparison between MOZAIC O_3 vertical profiles and global CTM results.

The spatial coverage of the MOZAIC data at cruise altitudes has been shown by *Marenco et al.* [1998, Figure 5]. The North Atlantic flight corridor is the most well documented region with over 40% of flights in this region. A 2 year period has been considered in this study using data from September 1994 to August 1996. Monthly mean O_3 values were calculated for 31 cruise level locations, shown in Figure 1. They were chosen on the basis of good data availability and global coverage. First, the 4 s O_3 data were reduced to 1 min average data along each flight and then averaged over 5° by 5° latitude–longitude boxes at each location and over the five A340 flight levels which have midlevels (and range in brackets); 287 hPa (290-285 hPa) or 9.4 km; 262 hPa (263-258 hPa) or 10.0 km; 238 hPa (242-237 hPa) or 10.6 km; 216 hPa (223-215 hPa) or 11.2 km; and 196 hPa (206-195 hPa) or 11.8 km. The pressure altitudes are relative to a surface pressure of 1013.25 hPa, and the heights in kilometers are calculated from pressure levels using a standard atmosphere.

MOZAIC data were also collected over 21 cities during the period considered here. Their locations are shown in Figure 2. Out of these, nine were selected based on a high frequency of data collection and to have as good global coverage as possible. Figure 3 shows the number of flights available per month for this subset of sites together with their latitude and longitude. For one case, Sao Paulo and Rio, data from these two cities, which are within a few hundred kilometers of each other, were averaged together in order to increase the measurement density and to improve the statistics for this location. In Figure 3, the coordinates correspond to the midpoint between Sao Paulo and Rio. Monthly mean vertical profiles of O_3 were calculated by averaging 1 min mean data within 50 hPa layers (~150 m) from 1000 hPa to 200 hPa. For example, the mean at 550 hPa corresponds to the average of all measurements between 575 and 525 hPa. Again, the data were averaged flight by flight before calculating monthly average O_3 concentrations and standard deviations for each layer.

3. Model Characteristics

As stated in the introduction, the distribution of O_3 in the troposphere and lower stratosphere is governed by a range of physical and chemical processes. The ability of current CTMs to reproduce observed distributions of O_3 will depend on various inputs, such as emission inventories, as well as the schemes used for transport and chemistry of trace species. A wide variety of schemes are in use although there is movement toward a consensus about which schemes may be preferable. The main characteristics of the models used in this study are given in Table 1.

Model comparison exercises organized by the World Climate Research Programme (WCRP) and the Intergovernmental Panel on Climate Change (IPCC) have helped to identify model formulations which are more desirable. For example, WCRP comparisons have shown that convection schemes developed for weather forecasting models perform better, when compared to observations of short-lived species such as ^{222}Rn, as opposed to more stochastic approaches based on climatological data [*Jacob et al.*, 1997]. In fact, three of the models used here use a mass flux type scheme [e.g., *Tiedtke et al.*, 1989] which produces more realistic results [*Stockwell and Chipperfield*, 1999; *Lawrence et al.*, 1999a; *Hauglustaine et al.*, 1998] in the Cambridge off-line chemistry transport model (TOMCAT) the Model of Atmospheric Transport and Chemistry - Max-Planck-Institute for Chemistry version (MATCH-MPIC) and the Model for Ozone and Related Chemical Tracers (MOZART) than in the Intermediate Model for the Annual and Global Evolution of Species (IMAGES) and the Model of Global Universal Tracer Transport

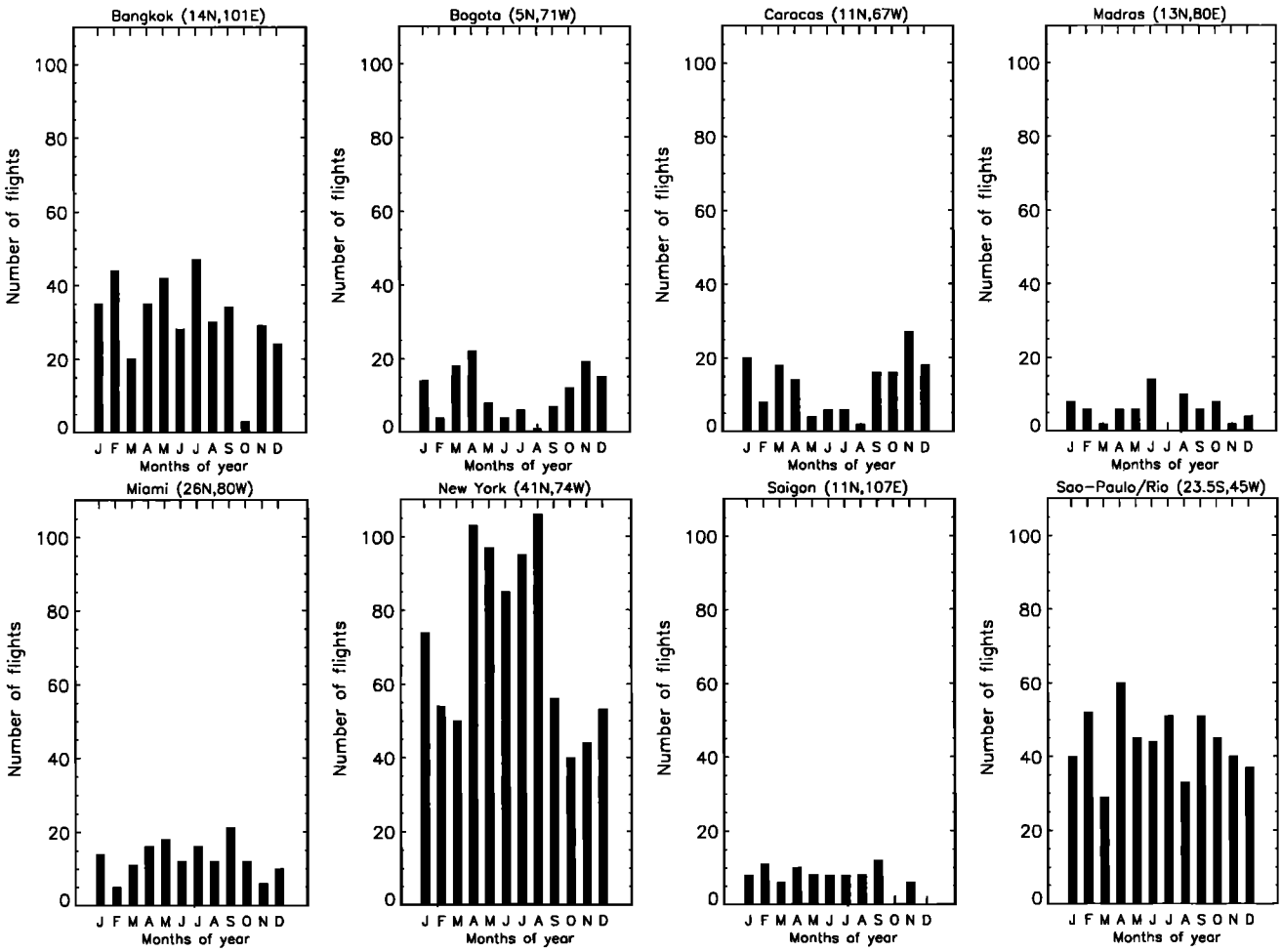

Figure 3. The number of MOZAIC flights available for each month in the 2 years from August 1994 to September 1996 together with the latitude and longitude for the vertical profile sites discussed in the paper.

in the Atmosphere (MOGUNTIA) (see Table 1). Similarly, models which use deposition schemes which are calculated as an interactive process within the model (e.g., dry deposition linked to the boundary layer height and exchange of heat and momentum or wet deposition linked to convective and dynamical rainfall) will produce more realistic results. This was demonstrated in the TOMCAT model when comparing to observations of ^{210}Pb, the product of ^{222}Rn decay [*Giannakopoulos et al.*, 1999]. This was also the conclusion of another WCRP model comparison looking at the performance of wet and dry deposition schemes in many CTMs [*Rasch et al.*, 1998]. Here, two models, IMAGES and MOGUNTIA, use simpler schemes for convection and deposition which are based on a more climatological approach and will not necessarily capture short-term variations.

The models used in this study have been run with chemical schemes ranging from CH_4 and CO chemistry in MATCH-MPIC to more detailed descriptions of the chemistry including the oxidation of alkanes in TOMCAT and other shorter-lived nonmethane hydrocarbons (NMHCs) in IMAGES, MOZART, and MOGUNTIA. Emission data sets are also from different sources and some models, such as TOMCAT and MOZART, have lightning NO_x emissions which are linked to the convection scheme [*Stockwell et al.*, 1999; *Hauglustaine et al.*, 1998]. Intuitively, it would be expected that models run with more complex chemistry would produce results which agree better with observations, but this is not found to be the case. In the recent IGAC/GIM model intercomparison exercise [*Kanakidou et al.*, 1999], various CTMs were used, including the ones in this study, and no consistent pattern emerged when comparing to O_3 and CO data. This is because the distributions of these trace gases depend also on transport processes, which, together with the model chemistry, will also be affected by the resolution at which a model is run. Models run at higher horizontal resolution [*Stockwell and Chipperfield*, 1999; *Van Velthoven and Kelder*, 1996] perform better than lower resolution simulations. Therefore MOZART and MATCH-MPIC could be expected to perform better than other models in this comparison with MOZAIC data.

Another important factor, which varies between the models, is the type of advection and diffusion schemes applied to tracers and the treatment and altitude of the

Table 1. Model Characteristics

Models	TOMCAT(A)	MOZART(z)	IMAGES(l)	MATCH(m)	MOGUNTIA(g)
Horizontal resolution	T21 (5.6° x 5.6°)	T42 (2.8° x 2.8°)	5° x 5°	T63 (1.9° x 1.9°)	10° x 10°
Vertical resolution	31 levels	25 levels	25 levels	28 levels	10 levels
Upper boundary	10 hPa	3 hPa	50 hPa	2 hPa	100 hPa
Meteorological data	ECMWF (reanalyses) (1994-1995)	NCAR (CCM2)	Monthly average wind variance from ECMWF (85-89)	NCEP reanalyzed (1993)	Monthly average winds and temperature from *Oort* (1983)
Frequency	6 hours	3 hours		6 hours	
Chemical time step	15 min	20 min	30 min	30 min	2 hours
Dynamical time step	30 min	20 min	6 hours	30 min	2 hours
Advection	*Prather* [1986]	semi-Lagrangian	semi-Lagrangian	semi-Lagrangian	Deep cumulus convection
Convection	*Tiedtke* [1989]	*Hack* [1994]	*Costen et al.* [1988]	*Pan and Wu* [1995]	and eddy diffusion
Boundary layer mixing	*Holtslag and Boville* [1993]	*Holtslag and Boville* [1993]	vertical diffusion	*Holtslag and Boville* [1993]	*Feichter and Crutzen* [1990]
Chemistry scheme	CH_4, CO, and alkanes, 50 species (27 advected)	CH_4, CO, and NMHC, 55 species (50 advected)	CH_4 and NMHC, 150 species (55 advected)	CH_4 and CO	CH_4, CO, and NMHC, reactions on aerosols
Top boundary conditions	O_3, CH_4, NO_y at 20 hPa	H_2O, O_3, NO_x, HNO_3 above 60 hPa	H_2O, O_3, NO_x, HNO_3 above 50 hPa	surface CH_4 O_3 and NO_y at 57 hPa	O_3 and NO_y at 100 hPa
Emissions	CO, NO_x, CH_4, ethane, propane, formaldehyde, and acetone	CO, NO, CH_4, N_2O, ethane, ethene, propane isoprene, terpene, propene, and acetone	CO, NO, CH_4 ethane, ethene, propane isoprene, propene, and acetone	CO and NO_x	CO, NO_x, CH_4, and ethane, ethene, propane isoprene, propene, n-butane, and acetone
References	*Law et al.* [1998] *Stockwell and Chipperfield* [1999]	*Brasseur et al.* [1998] *Haughustine et al.* [1998]	*Muller and Brasseur* [1995]	*Lawrence* [1996] *Lawrence et al.* [1999 a,b]	*Zimmerman* [1988] *Kanakidou and Crutzen* [1999]

top boundary condition for O_3. Table 1 shows that the top boundary varies from 100 hPa in MOGUNTIA to 2 hPa in MATCH-MPIC; this will affect the ability of models to simulate the downward flux of ozone in the stratosphere and the flux across the tropopause. Previous model calculations show cross-tropopause fluxes for O_3 ranging from 391 Tg yr^{-1} in MOZART [*Hauglustaine et al.*, 1998] to 600 Tg per yr^{-1} in MATCH-MPIC [*Lawrence et al.*, 1999a]. Problems with excessive downward transport of stratospheric O_3 have already been identified in the IMAGES model [*Muller and Brasseur*, 1995] when comparing with ozonesonde data. Smearing out of gradients across the tropopause were also found in TOMCAT when compared previously to MOZAIC O_3 profiles [*Law et al.*, 1998]. Model resolution around the tropopause could be a contributing factor to these findings, but further studies are required to confirm this.

All the models in this comparison are off-line CTMs; that is they use meteorological fields (winds, temperatures, etc.) from weather forecasting or climate models to force the model dynamics. Two types of model have been run: type A models used monthly mean climatological data with additional variability built in (i.e., IMAGES and MOGUNTIA), and type B models used meteorological fields with a higher time resolution varying between 3 to 6 hours (i.e., TOMCAT, MATCH-MPIC, MOZART). MOZART used climatological data based on the output from the National Center for Atmospheric Research (NCAR) Community Climate Model (CCM2), whereas TOMCAT and MATCH-MPIC used analyzed data based on observations from the European Center for Medium Range Weather Forecasts (ECMWF) and the National Centers for Environmental Prediction (NCEP) respectively (see Table 1). Therefore type B models are more likely to capture day-to-day variations and perform better when compared to the data than type A models which have little variation throughout each month. It is also possible to calculate more meaningful standard deviations and other statistics (e.g., interquartile range) as well as monthly averaged concentrations for type B models. However, type A models do have some inherent variability produced by their convection schemes.

In addition, the climatological winds and temperatures used in type A models are based on data averaged over many years, that is much longer than the 2 year period chosen for this comparison. However, the meteorological fields used in type B models did not correspond exactly to the 2 year period chosen. The TOMCAT model used meteorology for 1995; MATCH-MPIC used meteorology for 1993, and MOZART used output from a climate model, CCM2. Thus output from MOZART does not represent any particular year. Meteorological conditions vary from year to year, and this alone will explain some of the differences between individual model results and the MOZAIC measurements.

In summary, the model characteristics which should lead to a model comparing well with the data are high

resolution, use of meteorological analyses or assimilated data to drive the model, convection and deposition schemes linked to model physics, and a complex chemical scheme including a wide range of NMHCs together with seasonally varying emission inventories.

4. Comparison Methodology

In the previous section, differences in the types of meteorological fields used to drive the CTMs were outlined. The models may also not be directly comparable with the MOZAIC data due to differences in the sampling and averaging methods used. These points should be borne in mind in section 5 when model results are compared to the measurements. Note also that the MOZAIC data are relative to 1013 hPa.

Models represent averages over a specific spatial regime, which might contain sharp gradients (e.g., near the tropopause), whereas observations are collected at particular locations (latitude, longitude, and altitude). At cruise levels, this has been accounted for by interpolating between model levels to the pressure level at the midpoint of each of the five cruise altitudes. This can still produce errors since the models have different vertical resolutions around the tropopause, and this will affect their ability to capture sharp gradients in O_3 across the tropopause. For example, the MOGUNTIA model has very low vertical resolution around the tropopause, and the five MOZAIC flight levels correspond to only two model levels. Therefore the interpolated model results are unlikely to show any irregular changes because O_3 gradients are smoothed out considerably in this model. In fact, no model has a vertical resolution better than 1 km around the tropopause.

There is another reason why the modeled concentrations can be different from those measured. The measurements are representative of distinct flights on particular days, during which the meteorological conditions do not necessarily represent the average conditions during the month. Models have different output periods (e.g., every 6 hours or once a day), and in particular, type A models have little day-to-day variability. These differences may also lead to differences in the modeled average concentrations. Also, MOZAIC flights often occur during certain time windows within a day which may lead to a systematic bias in the data. Therefore it is best to run CTMs using meteorological data for the same period when the measurements were taken and to sample the model along individual flights [e.g., see *Law et al.*, 1998]. It was not possible to obtain model output in this way as not all models were able to obtain output in this manner.

For the type A models, it is only sensible to compare the monthly mean O_3 concentrations with the measured mean concentrations. For the type B models, it is possible to calculate and compare standard deviations as well as monthly mean concentrations. However, it should be noted that the observed distribution may be quite different from a Gaussian distribution, and for that reason a standard deviation may not always be the most appropriate statistical measure. This is especially true if the actual distribution is bimodal, such as around the tropopause where sharp gradients in O_3 exist and either tropospheric or stratospheric air can be sampled. This may lead to larger measured standard deviations than in the model results which tend to smooth out gradients. In addition, the short sampling time of the measurements (4 s), which, even though they have been averaged over 1 min and 5° by 5°, may still favor the occurrence of extreme values due to small-scale features which models are unable to capture at the present time.

Lastly, there are points related to the way in which the data have been averaged which need to be taken into account. During the cruise phase of each flight the statistics are calculated from 1 min averaged O_3 concentrations. Usually, more than one averaged value or "measurement" was collected within each 5° box. Note that the size of each box varies from 500 km x 550 km in the tropics to about 250 km x 550 km at 65° latitude. The speed of the aircraft is ~800 km h^{-1}, so a diagonal path (~750 km) through a box at midlatitudes would result in about 60 averaged measurements. However, the path through a 5° box can be much shorter if the aircraft passes across the corner of a box resulting in fewer average measurements being collected. In reality, the O_3 concentration in a grid box will vary a lot during each month, and the average of the "snap shots" collected by the MOZAIC aircraft will not necessarily provide the true mean of the O_3 distribution and its associated variability given by the standard deviation in the measurements.

It is clear that to calculate a monthly mean O_3 concentration which is as close as possible to the real monthly mean, a certain minimum number of measurements is required. The larger the number of measurements used to calculate the monthly mean, the more representative it is likely to be, but in the MOZAIC data set, this reduces the number of boxes which can be included in the study. Model results and measurements were examined as a function of the minimum number of measurements used to calculate the average O_3 concentration. It was decided that at least 50 measurements in the 2 year period should be available in each 5° grid box to calculate the monthly O_3 statistics at cruise altitudes. The choice of 50 measurements is arbitrary. As additional measurements become available over more years, more stringent criteria can be used in the future. The same reasoning was applied to the vertical profiles. All calculated and measured profiles were compared, and at least eight ascents or descents per month had to be available to calculate the monthly averages and standard deviations at a particular location.

5. Results

In the following section, the comparison between model results and MOZAIC O_3 data is described. The results from the comparison have been divided into different geographical regions which are discussed separately. In this study, the general aims were to examine the observed seasonal cycle of O_3 at different locations for interesting features and to see whether the CTMs were capable of reproducing the observed monthly mean concentrations and their associated variability. The latter is given by the standard deviation in the data. As stated previously, some models (IMAGES, MOGUNTIA) were not able to calculate standard deviations in a sufficiently meaningful way. The other models calculated means and standard deviations from 6 hourly (TOMCAT, MATCH-MPIC) or 24-hour average output (MOZART). Therefore some differences between the models and data may occur due to the way in which the statistics have been calculated. The main aim was to identify systematic discrepancies between the different models and the data in order to reach some general conclusions about model performance in different regions of the troposphere and lower stratosphere. The main findings of this study are discussed below together with possible reasons for discrepancies between models and MOZAIC data.

5.1. Northern Hemisphere Midlatitudes

5.1.1. Cruise altitudes. The observed O_3 concentrations at cruise altitudes show a relatively well-defined seasonal variation at all sites between 40° and 60°N (sites 5–19) with a maximum in the spring and a minimum in the fall/early winter. Four examples are shown in Figure 4 for locations over the mid-Atlantic (site 5), western Europe (site 10), northeast Russia (site 3), and the east coast of North America (site 14) at 238 hPa. The peak-to-peak ratio is about 2 to 2.5 at most sites. The magnitude of the maximum concentration, generally found in late winter and spring, at the different sites depends, to a large extent, on the position of this flight level relative to the tropopause

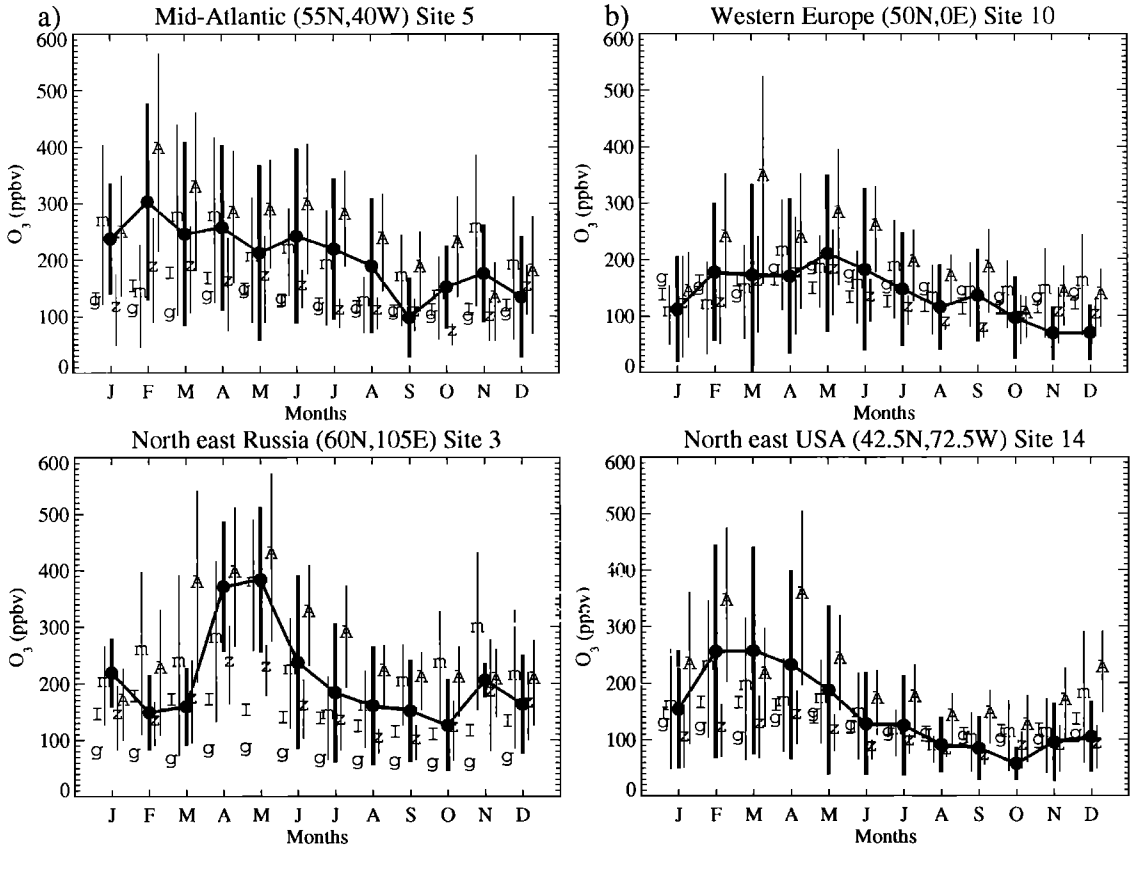

Figure 4. Monthly mean concentrations of O_3 in ppbv at aircraft cruise level 238 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) at (a) site 5, (b) site 10, (c) site 3, and (d) site 14. Site locations are shown in Figure 1. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

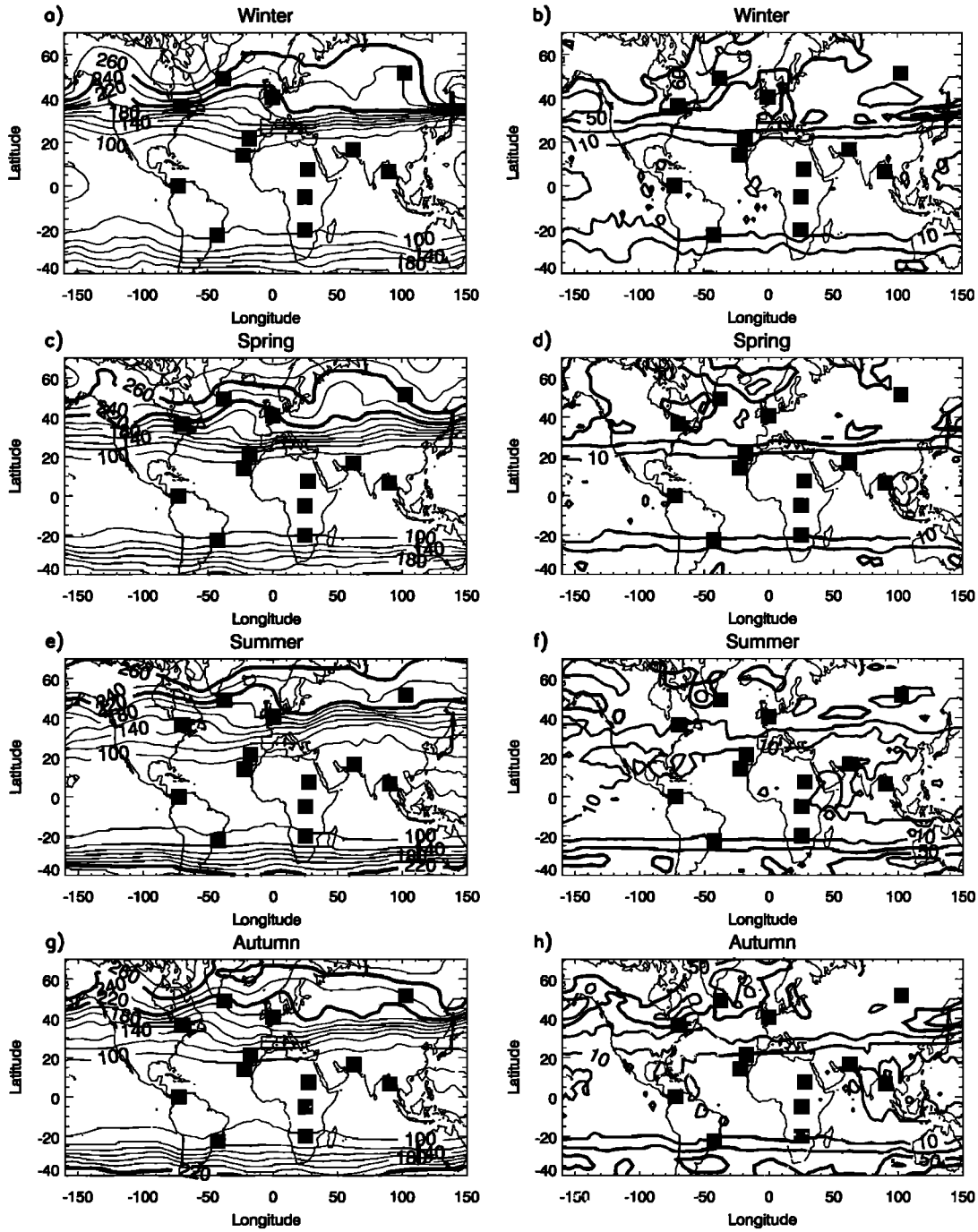

Figure 5. Seasonally averaged tropopause heights and standard deviations in hPa calculated from 1995 ECMWF data (see text for details of the calculation); (a) mean tropopause height for winter (DJF), (b) standard deviation in the tropopause height for winter (DJF), (c) mean tropopause height for spring (MAM), (d) standard deviation in the tropopause height for spring (MAM), (e) mean tropopause height for summer (JJA), (f) standard deviation in the tropopause height for summer (JJA), (g) mean tropopause height for autumn (SON), and (h) standard deviation in the tropopause height for autumn (SON).

which is governed by the position of the large planetary scale ridge and trough pattern in the upper troposphere. Figure 5 shows the variation in the seasonal mean tropopause height and the standard deviation around the mean (in hPa), calculated from 6-hourly ECMWF analyses for 1995. The criteria of potential vorticity equal to 3.5 PVU was used to determine the tropopause height in middle and high latitudes [*Hoerling et al.*, 1991, 1993], while the 380 K potential temperature surface was used in the tropics (30°N to 30°S) [*Holton et al.*, 1995]. The locations of the sites discussed in this paper are also shown. This figure shows that data collected at 238 hPa at sites 10 and 14, which are farther south than sites 3

and 5, are, on average, below the tropopause in all seasons. However, as shown by the standard deviations, the tropopause height varies greatly at midlatitudes and therefore sites 10 and 14 sampled stratospheric as well as tropospheric air, particularly in the winter and the spring. This is reflected in the large standard deviations in the observed O_3 concentrations. Figures 4 and 5 also show that, in midlatitudes, regions with lower tropopauses, associated with trough regions (50°–100°W; 100°–150°E), have higher O_3 concentrations than regions with higher tropopauses, associated with ridge regions (40°W–40°E), at a particular pressure height. This was also pointed out by *Thouret et al.* [1998a]. For example, contrast the peak O_3 concentrations over site 10 (western Europe), a ridge region, which are 100 ppbv lower than peak concentrations over site 14 (northeast United States), a trough region. The site over northeast Russia (site 3) has the highest maxima. This site is also within a trough region but farther north where tropopause heights are even lower. Therefore data at site 3 are largely stratospheric, although again due to variations in the tropopause height, tropospheric air was also sampled.

All models involved in this comparison are able to produce at least qualitative agreement with the observed seasonal cycles at these locations, with a maximum in spring and a minimum in the fall/early winter with the latter usually best reproduced by the models. The variability in the data and in the type B models is largest in the spring months when stratosphere-troposphere exchange processes are at a peak [*Holton et al.*, 1995]. However, variability in these model results is less than in the data which ranges from, for example, 70 ppbv to 440 ppbv at site 14 over North America. The best overall agreement with the data is found at sites over western Europe (see Figure 4b). At the other locations, where little or no data were available before, agreement is noticeably worse in certain models (MOGUNTIA, IMAGES, MOZART). Here these models underestimate the spring maximum in O_3. Discrepancies with the data may be due to several reasons, some of which have been quantified previously. The meteorological fields used to drive the models may not reproduce the trough/ridge pattern in the tropopause height, particularly if a model was run at low resolution. Also, the number of levels and location of the top boundary will affect the ability of models to capture the stratospheric circulation and seasonal variations in O_3 in the lower stratosphere and in the tropopause region. This is true for MOGUNTIA which has a top boundary at 100 hPa and low vertical resolution and has been noted in comparisons with ozonesonde data [*Kanakidou et al.*, 1999]. In the MOZART model, the cross-tropopause flux is quite low compared to other estimates [*Hauglustaine et al.*, 1998] indicating that the downward transport of O_3 from the stratosphere is too weak in this model thus leading to the underestimation in O_3 seen here around the tropopause. The IMAGES model also underestimates O_3 concentrations in the spring months. This is due to excessive horizontal transport of stratospheric O_3 into the subtropics, rather than downward at midlatitudes [*Rasch et al.*, 1997; *Muller and Brasseur*, 1999]. In general, the O_3 seasonal cycle calculated by MATCH-MPIC agrees well with the observations, although the peak-to-peak amplitude is too low in some cases. This may be due to using winds for 1993, but this has yet to be confirmed. It is somewhat surprising that MATCH-MPIC agrees so well with the data. In a study by *Lawrence et al.* [1999a], the flux across the tropopause for a newer model version was found to be rather high compared to most recent estimates (1100 Tg yr^{-1}), and agreement with sonde data was found to be better with a lower flux (600 Tg yr^{-1}). The model version with the lower flux has been used here.

A strong vertical gradient, characteristic of the tropopause which is the lowest in late winter and spring (February to May), was found in the vertical profile data over sites 10 and 14 (not shown here). Most of the time, the shape is reproduced quite well by the models except in spring when they simulate a weaker gradient than observed and a lower O_3 tropopause than observed. Problems with capturing the sharp gradients across the tropopause were also found in TOMCAT when comparing to individual MOZAIC profiles; the model smears out the gradients leading to an overestimation of O_3 concentrations in the tropopause region [*Law et al.*, 1998]. Subsequent results (unpublished) have shown that this was due to excessive O_3 at the top boundary and too rapid downward transport in the stratosphere.

5.1.2. Vertical profiles under continental influence. All profiles were examined for those sites falling into the latitude band, 40°–60°N. In this section, results from the comparison over New York are used to illustrate the salient points. Figure 6 shows the modeled monthly average O_3 results compared to the observations at four pressure altitudes over New York. In the lower and middle troposphere, there is a clear summer maximum in the data. In the upper troposphere, the data show a broad spring maximum with higher extreme values (over 100 ppbv) when stratospheric air has been encountered. The same pattern was found at other continental midlatitude locations although the peak summer concentrations vary. For example, over Frankfurt (not shown) mean O_3 concentrations only reach ~40 ppbv at 950 hPa which is lower than over New York. However, at 750 hPa in the summer months, Frankfurt mean O_3 concentrations are higher (up to ~70 ppbv) than over New York. Near the ground, lower observed O_3 may be a result of NO titration in the possibly more polluted Frankfurt area or enhanced vertical exchange of pollutants out of the boundary layer over central Europe.

In general, the CTMs reproduce the observed seasonal cycle at different altitudes in the troposphere at midlatitudes. However, there is a tendency for models to underestimate the summer maximum in the lower troposphere over New York and at other northern mid-

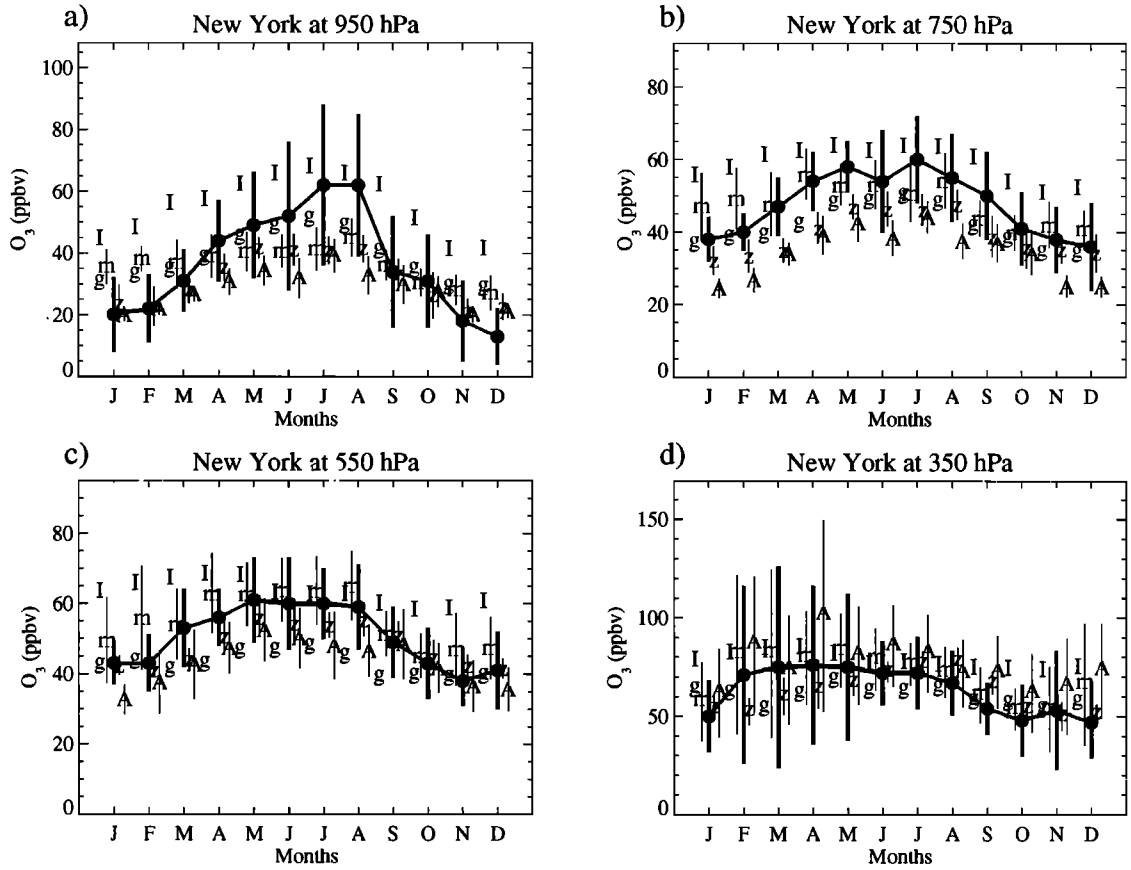

Figure 6. Monthly mean concentrations of O_3 in ppbv over New York, United States, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) over 50 hPa intervals centered on (a) 950 hPa, (b) 750 hPa, (c) 550 hPa, and (d) 350 hPa. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

latitude sites, such as Frankfurt (not shown). One exception to this is IMAGES which overestimates O_3 in the summer and the winter. This was also found when comparing to other surface O_3 data [*Muller and Brasseur*, 1999]. The ability of models to reproduce the summer maximum is related to whether they are able to simulate pollution episodes. Examination of the modeled O_3 standard deviations (type B only) shows that models have lower variability than the data. For example, in Figure 6a, models which capture daily variations (i.e., MOZART, MATCH-MPIC, and TOMCAT) have standard deviations lower than observed by about 20 ppbv. This discrepancy may be due to differences in sampling (see discussion in section 4). In TOMCAT, this underestimation may be due to the lack of short-lived hydrocarbons and the coarse horizontal resolution used [*Law et al.*, 1998]. However, MOZART, which was run at T42 (~2.8°) resolution with a large range of NMHCs, also underpredicts summertime O_3 concentrations. MATCH-MPIC, which was run at higher horizontal resolution (T63 or ~1.9°), simulates monthly mean O_3 concentrations which are closer to the observed mean values even though it was run with only CH_4 and CO chemistry. In contrast, MOGUNTIA, which was run at very coarse resolution (10°) and with many NMHCs, reproduces the mean O_3 concentrations reasonably well. Therefore major factors affecting O_3 concentrations predicted by models are the complexity of the NMHC chemistry included and the resolution of the integrations. The latter will affect models' ability to reproduce synoptic scale events and sub-grid scale processes (convection, PBL mixing) which can produce polluted plumes. Interestingly, certain models (MATCH-MPIC, MOGUNTIA, and IMAGES) overpredict O_3 in the winter. Again, chemistry, boundary layer mixing, and model resolution may be affecting model results leading to an underestimation of O_3 titration by NO_x in polluted regions. This range of results was also found in the recent IGAC/GIM comparison against surface data [*Kanakidou et al.*, 1999].

In the mid and upper troposphere, there are some noticeable discrepancies with the observations. At 550 hPa, IMAGES predicts a spring maximum possibly related to too much downward transport from the stratosphere. At 350 hPa, MOZART incorrectly predicts a summer maximum. This discrepancy is not seen higher

up at cruise altitudes (see Figure 4). Overall, at northern midlatitude sites, models are able to capture the seasonal cycle of O_3 reasonably well, but the summer maximum in the mid and lower troposphere is underestimated.

5.1.3. Vertical profiles under summertime marine influence. At several coastal locations (e.g., Tokyo/Osaka and Miami), MOZAIC O_3 observations are clearly influenced by clean maritime air during the summer months resulting in a summer minimum and a spring maximum in the lower and middle troposphere. This is in contrast to New York, discussed in the previous section, which although located on the coast, is influenced much more by transient weather systems picking up polluted air [*Fehsenfeld et al.*, 1996] leading to higher O_3 in the summer months. Over the North Atlantic and the Pacific Oceans, large subtropical high-pressure systems exist during the summer resulting in the transport of tropical maritime air with low levels of O_3 in a northwestward direction to North America and Asia [*Thouret et al.*, 1998b]. To illustrate this, Figure 7 shows the modeled and observed seasonal cycle of O_3 at Miami, again at four pressure levels. Results for Tokyo are similar. Very low surface mixing ratios are observed during the summer (less than 10–20 ppbv). This has also been observed at other surface coastal/oceanic sites influenced by clean maritime air such as Mace Head, Ireland [*Simmonds et al.*, 1997] and Bermuda [*Oltmans and Levy*, 1994].

The models generally overestimate O_3 in the lower troposphere in the summer months over Miami. TOMCAT and MATCH-MPIC reproduce the observations relatively well at most levels except near the surface where they overestimate O_3. Both IMAGES and MOZART predict too much O_3 up to an altitude of 350 hPa. MOGUNTIA tends to underestimate the O_3 seasonal cycle, mainly due to an underestimation of the O_3 mixing ratios in the summer months. The variability in the data is quite large showing that this location is affected by polluted continental air masses as well as clean marine air masses. Coastal sites are difficult to model and will be sensitive to model resolution which determines whether a location is primarily under land or ocean influence in each model [*Thouret et al.*, 1998b].

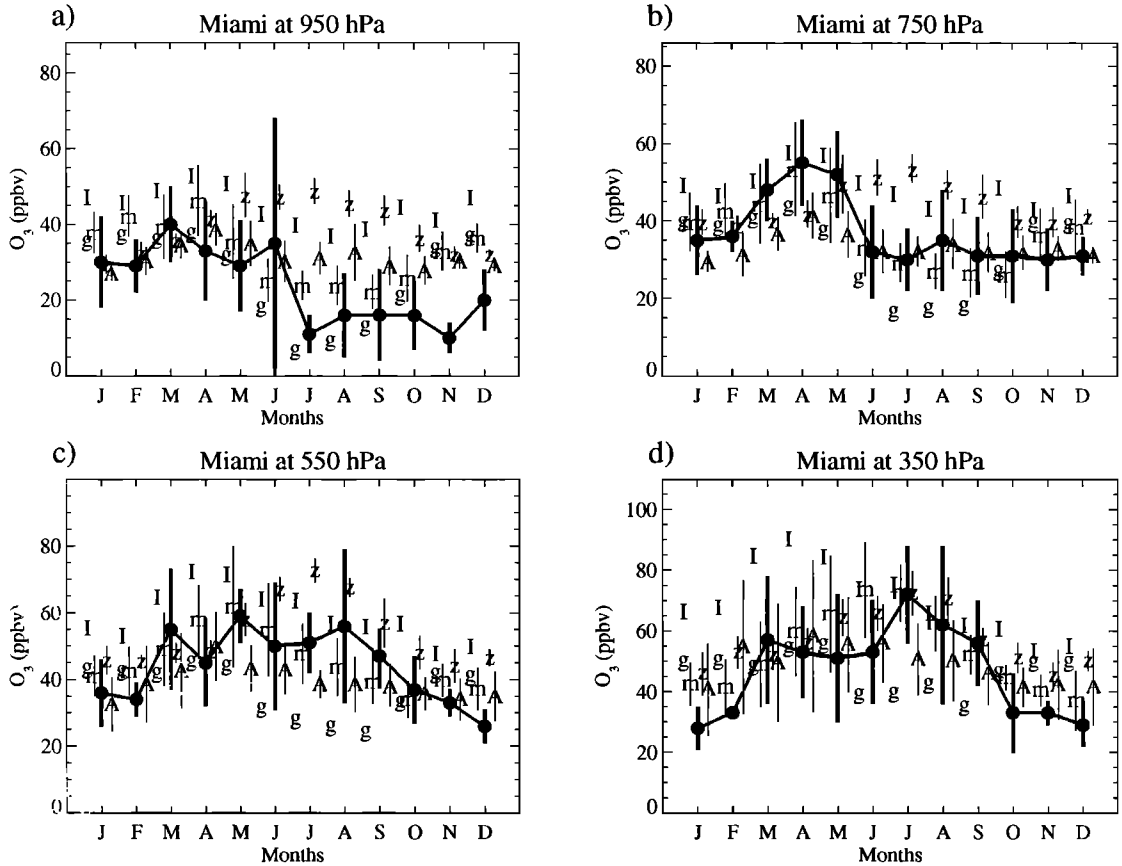

Figure 7. Monthly mean concentrations of O_3 in ppbv over Miami, United States, at vertical profile altitudes centered on (a) 950 hPa, (b) 750 hPa, (c) 550 hPa, and (d) 350 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART). Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

The standard deviations produced by MATCH-MPIC, TOMCAT, and MOZART agree well with the data in the midtroposphere but are too low in the lower troposphere. In this study, model results have been interpolated to the center of the 5° box used to calculate the observed averages. Therefore the model results should reflect how well models capture land/sea gradients.

At this site, the CTMs driven by analyzed winds (i.e., MATCH-MPIC and TOMCAT) seem to provide a better representation of transport processes associated with the formation of summertime high-pressure gyres over the oceans. However, much of the data was collected over Miami in 1995. While TOMCAT was run using meteorological fields for this year, MATCH-MPIC was run with data for 1993. This suggests that running models at high horizontal (probably greater than T42 or 2.8°) and vertical resolution (probably 1–2 km) and using meteorological data for the same period when the measurements were collected will produce better agreement between model results and the data. While this conclusion is not that surprising, it still remains to be verified.

5.2. Africa

5.2.1. Cruise altitudes over the mid-Atlantic. The mid-Atlantic cruise locations (labeled 21 and 24 in Figure 1) are both close to the coast of Africa. Figure 5 shows that the MOZAIC aircraft which collected data at these sites were flying in the troposphere. The average tropopause height is always higher than the cruise altitudes of the aircraft even when the standard deviation in the tropopause height is taken into account. However, the measured O_3 concentrations at site 21 (see Figure 8) do show evidence of air which is stratospheric or which has been transported from the stratosphere into the troposphere. Incidences of transport of high O_3 into the troposphere have already been seen in the MOZAIC data by *Suhre et al.* [1997]. Further study showed that these events are due to transport across the subtropical tropopause [*Cammas et al.*, 1998]. Large variations in observed O_3 at site 21 are only found in the winter and spring when this type of transport is more active [*Holton et al.*, 1995]. At site 24, farther south, O_3 concentrations are lower, on average, than at site 21. Therefore this site exhibits essentially tropospheric characteristics because the measurements were collected well below the tropopause. Even so, concentrations exceeding 100 ppbv were occasionally observed in the late winter, but there is no evidence for the large variations seen in winter and spring farther north. No clear seasonal cycle is apparent for site 24.

Most models have problems capturing the differences between these two sites. In general, it appears that model resolution (vertical and horizontal) and the position of the tropopause and the subtropical jet are influencing the CTMs ability to model O_3 correctly in this region. Of the models which are able to capture day-to-day variations in O_3, MATCH-MPIC and TOMCAT show the most variability, which is more in line with the observations at both sites. They also reproduce the observed gradient between the two locations. However, MOZART simulates little difference between the sites leading to an underestimation of O_3 in the first 6 months of the year at site 21. It is not clear whether this is due to the positioning of the subtropical jet and its associated lower tropopause too far to the north or whether there is too little transport of O_3 across the tropopause in this model. As stated previously, it is known that this model has rather a low global cross-tropopause O_3 flux [*Hauglustaine et al.*, 1998]. The same is true for the MOGUNTIA model, but the lack of gradient between the sites is most likely to be due

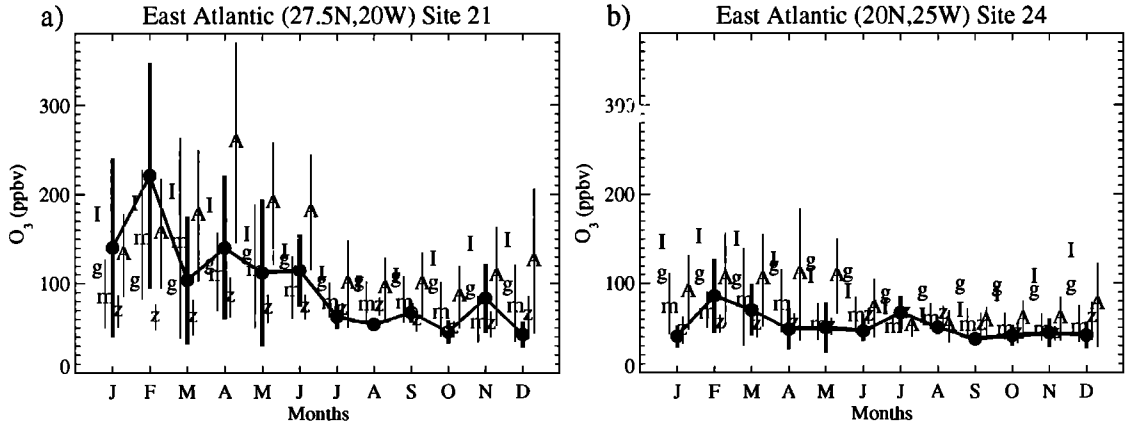

Figure 8. Monthly mean concentrations of O_3 in ppbv at aircraft cruise level 216 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) at (a) site 21 and (b) site 24. Site locations are shown in Figure 1. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

to sites 21 and 24 lying in the same model grid cell. IMAGES also has problems simulating these two sites, and in particular, it overestimates the O_3 levels at site 24 during the winter and spring months. This is due to this model overestimating the flux of ozone across the tropopause in the subtropics [*Rasch et al.*, 1997; *Muller and Brasseur*, 1999].

5.2.2. Cruise altitudes over the African continent. Relatively few MOZAIC data were collected over Africa at cruise altitudes making it difficult to determine seasonal cycles in the data. Out of the three locations shown in Figure 1, site 31 over South Africa is the most well documented. Examination of Figure 9, which shows data at 262 hPa at site 26 north of the equator over Sudan, site 29 on the equator over Zaire, and site 31 south of the equator over South Africa, reveals that O_3 concentrations decrease toward the equator; that is, site 29 has lower O_3 levels than the other sites. At all these sites, O_3 mixing ratios remain below 100 ppbv, indicating an absence of stratospheric influence. This is confirmed by Figure 5 which shows that data collected at these sites are well below the tropopause. Sites 26 and 31 seem to have opposing seasonal cycles with a spring peak and fall minimum at each location although this is less obvious at site 26. At site 29, there is no clear seasonal cycle although the data suggest a possible double peak in the spring and the fall. Additional data in the future will help to confirm this.

The distribution of O_3 around the equator over Africa will be dominated by the seasonal movement of the Intertropical Convergence Zone (ITCZ), seasonal variations in emissions, notably biomass burning and regional circulation patterns. Over southern Africa, results from the SAFARI experiments also show the presence of a spring O_3 maximum in ozonesonde data [*Diab et al.*, 1996] between 0° and 30°S largely due to the uplift by deep convection of O_3 precursors from biomass burning emissions into stable haze layers in the upper troposphere [*Thompson et al.*, 1996]. At this time of year, an anticyclonic ridge generally exists, and pollutants can become trapped in gyres for many days allowing levels of O_3 to build up [*Garstang et al.*, 1996].

Nearer to the equator, ozonesonde data collected at Brazzaville, Zaire, have a maximum lasting from June to September which is somewhat different from the pos-

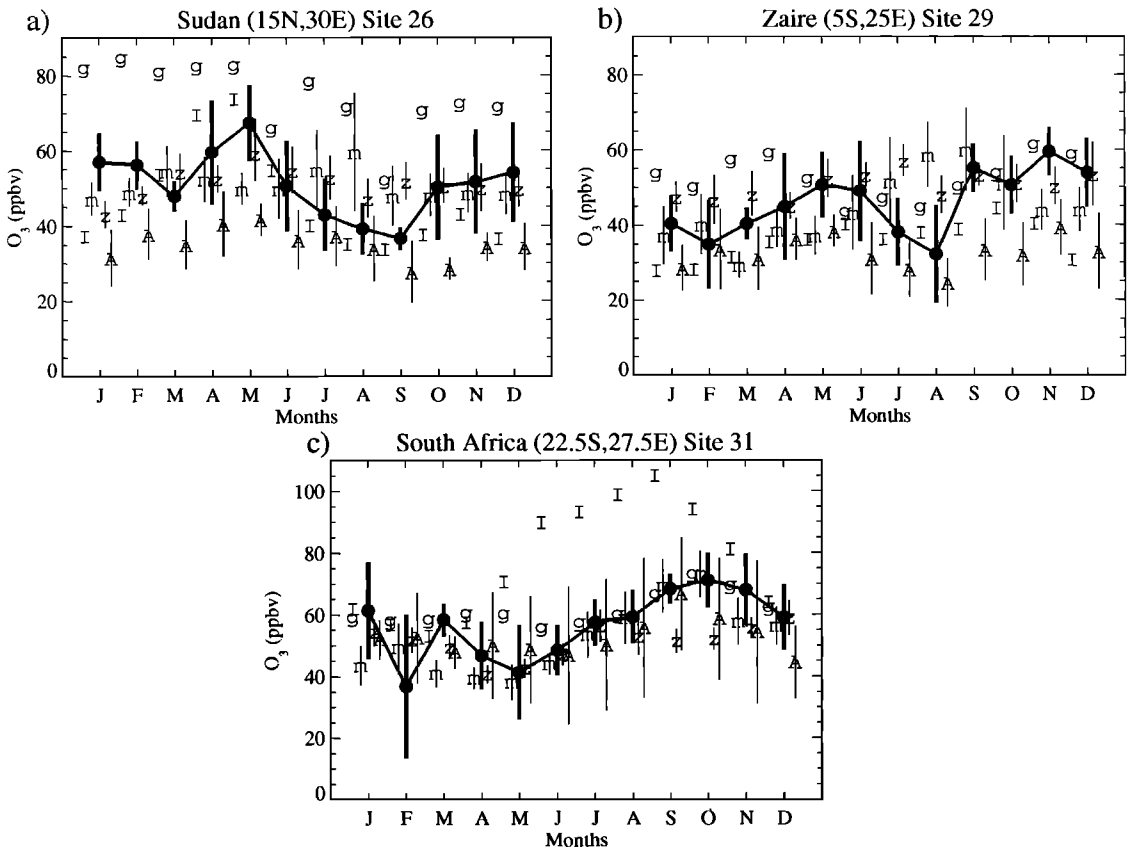

Figure 9. Monthly mean concentrations of O_3 in ppbv at aircraft cruise level 262 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) at (a) site 26, (b) site 29, and (c) site 31. Site locations are shown in Figure 1. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

sible double peak seen in the MOZAIC data over Zaire. North of the equator, it is possible that O_3 concentrations are enhanced by biomass burning emissions just north of the ITCZ in the first few months of the year [*Jonquieres et al.*, 1998]. One possibility is the southward transport of O_3 and precursors toward the ITCZ followed by upward (by deep convection) and northward transport into the westerly upper level flow associated with the subtropical jet region. However, a detailed study of O_3 over eastern Africa still remains to be carried out.

MOGUNTIA overestimates the O_3 mixing ratios, particularly at site 26, north of the equator although it captures the seasonal cycle to a certain extent. The other models do not capture the observed seasonal cycles very well either, particularly at sites 26 and 29. It is possible that the seasonal variation in biomass burning emissions north of the equator is incorrectly specified, especially in TOMCAT which underestimates O_3 levels. However, deep convection in this model may also be too weak. Other studies with TOMCAT have shown that these results are probably a combination of these two effects [*Stockwell and Chipperfield*, 1999; P.-H. Plantevin et al., manuscript in preparation, 1999].

Overall, the models perform reasonably well for site 31. This is probably due to the wealth of information from the SAFARI/TRACE experiments which have provided much needed details about emissions and circulation patterns in southern Africa. All of the models correctly predict O_3 mixing ratios below 100 ppbv except for the IMAGES model. Here this model has a large spring maximum indicative of too much stratospheric air being mixed into the upper troposphere [*Rasch et al.*, 1997]. However, there is still some spread in the other model results. As well as enhanced O_3 in the upper troposphere, MOZAIC vertical profiles over Johannesburg, South Africa, in August (not shown) also show a maximum in the midtroposphere [*Thouret et al.*, 1998b]. This is again due to entrapment of pollution in stable haze layers following uplift by convection [*Diab et al.*, 1996]. Models appear able to capture this feature.

5.3. South America

5.3.1. Vertical profiles influenced by biomass burning. MOZAIC vertical profile data collected over South America show clear evidence for enhanced O_3 levels south of the equator during July to October and north of the equator from December to March. Again, comparison with model results may highlight deficiencies in biomass burning emissions of NO_x, CO, CH_4, and NMHCs (both magnitude and seasonality), photochemical O_3 production efficiency, and transport of polluted plumes by deep convection and the large-scale flow. Tropical forested regions, such as the Amazon basin, also have the potential to emit large amounts of biogenic NMHCs, primarily as isoprene, which can also produce significant quantities of O_3, if sufficient NO_x is present. Vertical transport of local pollution from large urban areas can also not be ruled out.

Figure 10 shows the seasonal variation of observed and modeled O_3 over Sao Paulo/Rio, south of the equator, at four altitudes. There is a clear austral spring maximum in September/October over the entire depth of the troposphere. This is also observed at cruise altitude site 30 (see Figure 13). At this time of year, there is general northwesterly flow of air out of the South American continent and large-scale burning in the Amazon forest and savannah regions northwest of Sao Paulo/Rio. These factors, combined with deep convection can loft pollutants emitted over the Amazon into the mid and upper level flow producing plumes which move out into the southern Atlantic Ocean as was observed during TRACE-A [*Thompson et al.*, 1996]. Also, the O_3 concentrations in the wet season (April/May) are noticeably lower. This has also been seen in ozonesonde observations [*Kirchoff et al.*, 1996].

In the lower troposphere, the MOZART model captures the seasonal cycle in O_3 better than the other models. The other models overestimate O_3 by varying amounts, particularly from April to August suggesting that their biomass burning emissions are too high and the seasonality is incorrect. However, biomass burning emissions, which are known to vary significantly from year to year, were not specific to the time period when the MOZAIC data were collected. In the middle and upper troposphere, most models perform reasonably well except for IMAGES which overestimates O_3 from May to September. Again, this is due to an excessive stratospheric O_3 flux at this location in this model. Also, the variability is generally higher in the data than in the type B models at all altitudes, except at 350 hPa. There is also a secondary maximum in January/February, which becomes more apparent above 550 hPa and is probably due to long-range transport of O_3 produced from biomass burning north of the equator in Brazil or Africa or from industrial emissions [*Jonquieres et al.*, 1998].

Around and north of the equator, MOZAIC data were collected rather sporadically at Quito (Ecuador), Bogota (Columbia), and Caracas (Venezuela). When sufficient data were available (i.e., eight or more profiles in 1 month), monthly means and standard deviations were calculated. To illustrate the influence of biomass burning on this region, observed and modeled O_3 vertical profiles in March and October are shown in Figures 11 and 12 for Caracas and Bogota, respectively. In March, which is toward the end of the burning season north of the equator, both sites show higher O_3 in the middle and upper troposphere than at the surface. In October, slightly higher concentrations (greater than 25 ppbv) are only evident in the upper troposphere. These data are derived from rather few vertical profiles but suggest either long-range transport of O_3 and/or its precursors from south of the equator or Africa or convective

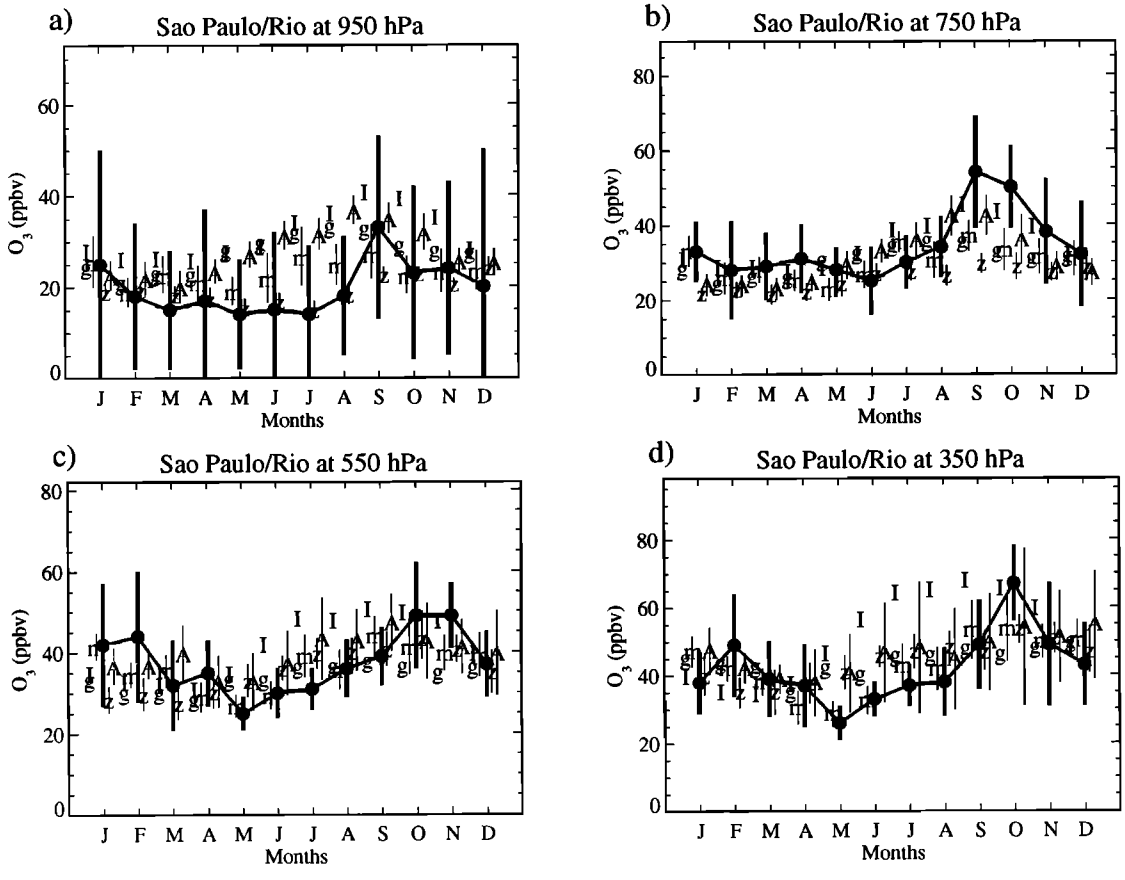

Figure 10. Monthly mean concentrations of O_3 in ppbv over Sao Paulo/Rio, Brazil, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) over 50 hPa intervals centered on (a) 950 hPa, (b) 750 hPa, (c) 550 hPa, and (d) 350 hPa. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

uplift of local pollution [*Jonquieres et al.*, 1998]. The models generally capture the observed behavior of both these months better over Caracas than over Bogota, that is higher concentrations in the midtroposphere in March. However, none of the type B models reproduce the higher variability in the data in March compared to October although this may be an artifact resulting from relatively few observations in these 2 months. Also, over Caracas, apart from TOMCAT, models overestimate the low O_3 mixing ratios (sometimes less than 10 ppbv) in the lower troposphere. Humidity fields, used to calculate photochemical destruction of O_3 via the reaction $H_2O + O(^1D)$, may be the reason for this discrepancy. TOMCAT used analyzed specific humidity fields from ECMWF for the year 1995 when the majority of profiles were collected over Caracas. Alternatively, it could be that O_3 is being titrated by high NO_x from local pollution in the vicinity of the airport [*Law et al.*, 1998].

In October, over Bogota, MATCH-MPIC and MOGUNTIA overestimate, and TOMCAT underestimates, the O_3 concentrations in the mid and upper troposphere. This may indicate deficiencies in the modeled strength of the Hadley circulation and deep convection which can lead to transport of O_3 from south of the equator. Variations in the amount of NO_x produced from lightning in regions of deep convection may also lead to differences between models. The MOGUNTIA model clearly has problems in the upper troposphere which may be due to excessive downward flux of O_3 from the stratosphere at both locations.

5.3.2. Cruise altitudes.

Data collected at cruise altitude 216 hPa, over South America at sites 27 and 30 (see Figure 1), are shown in Figure 13, together with the model results. The data at site 27 over Venezuela clearly show that the MOZAIC aircraft were flying in the upper troposphere, due to the low levels of O_3 recorded (mean values less than 60 ppbv). At this location, most of the models are within the range of the observations, with the exception of MOGUNTIA; this model's parameterization of the O_3 upper boundary at 100 hPa and deep convection lead to an overestimation of O_3 mixing ratios at these altitudes. MOZART also predicts higher O_3 than

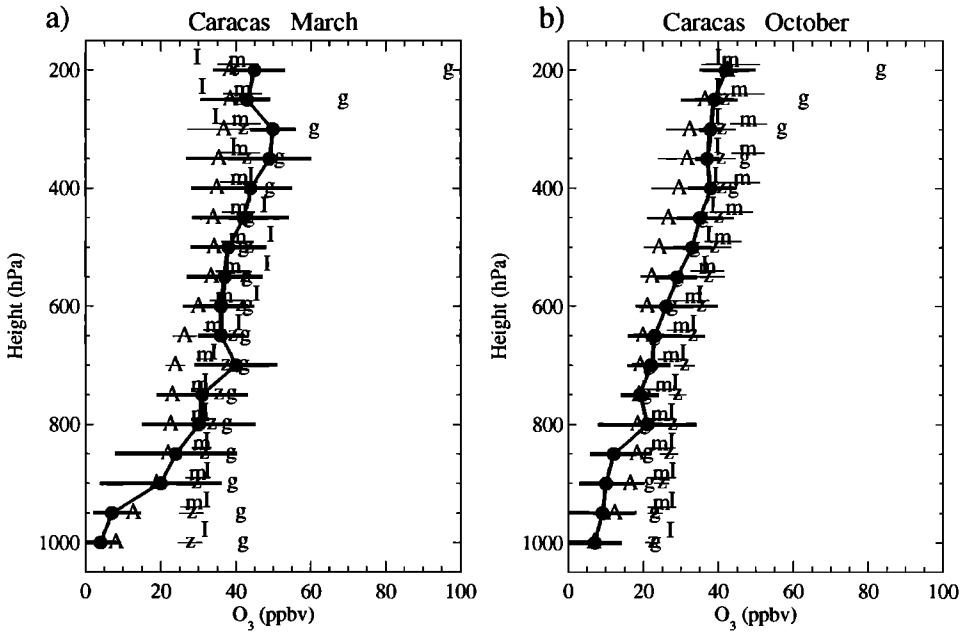

Figure 11. Monthly mean concentrations of O_3 in ppbv over Caracas, Venezuela, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) centered on 50 hPa intervals from 1000 hPa to 200 hPa for (a) March and (b) October. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results.

observed during a couple of summer months. Again, differences between model results and the data are likely to be due to variations in model treatment of deep convection and advection which affects long-range transport of pollutants.

At site 30, in the Southern Hemisphere over Sao Paulo/ Rio, high values of O_3 were sometimes encountered, particularly during the austral spring. As noted for the vertical profile data over Sao Paulo /Rio, this is due to long-range transport of O_3 produced from

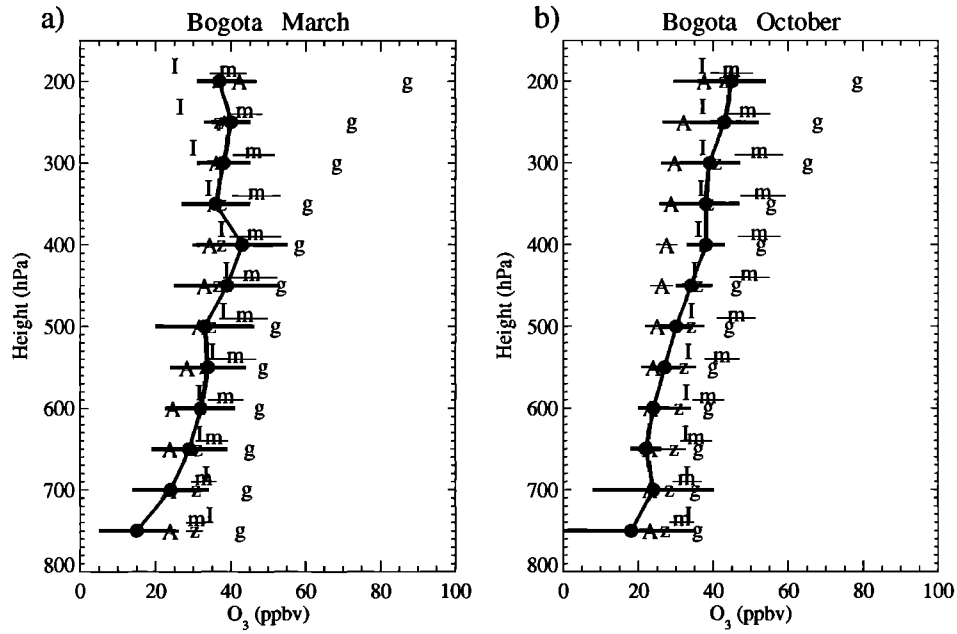

Figure 12. Monthly mean concentrations of O_3 in ppbv over Bogotá, Columbia, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) centered on 50 hPa intervals from 1000 hPa to 200 hPa for (a) March and (b) October. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results.

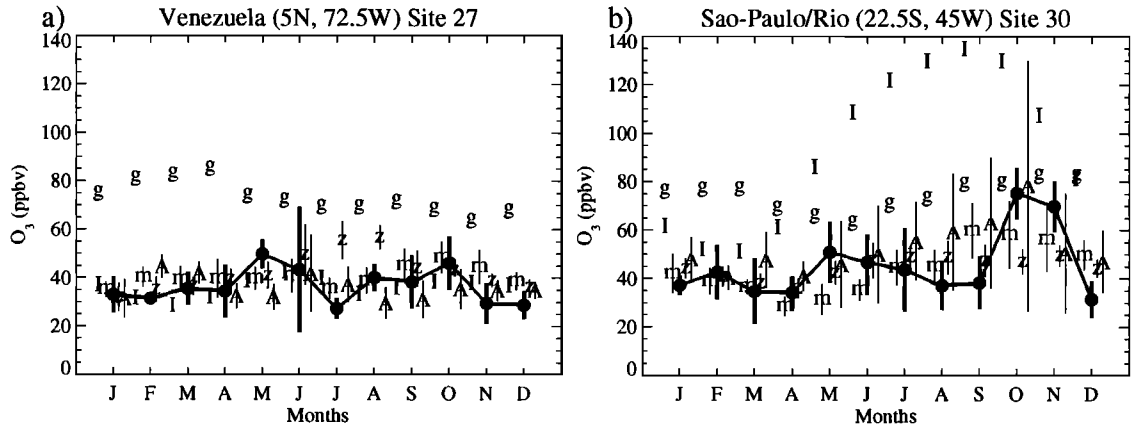

Figure 13. Monthly mean concentrations of O_3 in ppbv at aircraft cruise level 216 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) at (a) site 27 and (b) site 30. Site locations are shown in Figure 1. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

biomass burning over the interior of Brazil. This was also observed in the TRACE-A experiment [*Collins et al.*, 1996]. As was seen for site 27, MOGUNTIA also overestimates O_3 mixing ratios at this location. IMAGES also shows the same problem found over South Africa and in the Northern Hemisphere subtropics, that is too much stratospheric O_3 in the upper troposphere at these locations. Of the other models, MATCH-MPIC and TOMCAT peak too early in the austral spring, and MOZART misses the austral spring peak altogether.

5.4. Indo-Asian Subcontinent

5.4.1. Cruise altitudes. Figure 14 shows model results and data for site 23 over Pakistan and site 25 over Burma, respectively, at 216 hPa when 12 months of data were available to calculate the statistics. Figure 5 shows that data collected at these cruise altitudes were always in the troposphere even when the standard deviation in the tropopause height is taken into account. MOZAIC observations over Pakistan have a spring maximum probably produced by incursions of stratospheric air across the subtropical tropopause of the nature described by *Suhre et al.* [1997] and *Cammas et al.* [1998]. Site 23 also exhibits a summer minimum with monthly mean concentrations as low as 40 ppbv during the summer monsoon season. At site 25, over Burma, there is no influence from the stratosphere, and mean observed O_3 concentrations range from 25 ppbv to 50 ppbv. There is only a tenuous seasonal cycle with a slight spring maximum and a weak summer/fall minimum when this region is located close to a region of deep convection. Occasionally, O_3 concentrations as low as a few ppbv

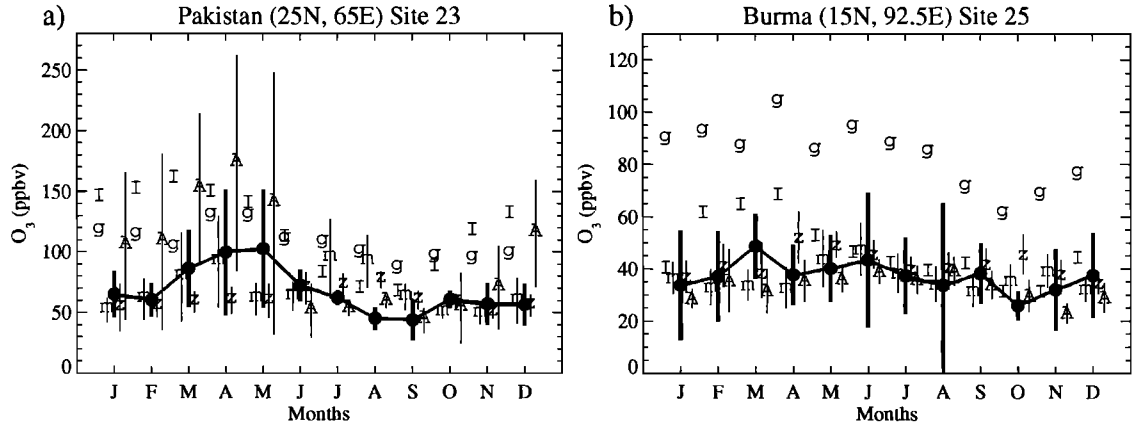

Figure 14. Monthly mean concentrations of O_3 in ppbv at aircraft cruise level 216 hPa calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) at (a) site 23 and (b) site 25. Site locations are shown in Figure 1. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results. Note that all averages are centered on the middle of the month.

were observed in the upper troposphere at this location.

The models capture the observed seasonality with varying degrees of success. At both locations, deficiencies in the modeled cross-tropopause flux of O_3 or the position of these sites relative to the modeled subtropical tropopause are again apparent in MOGUNTIA and IMAGES. Also, over Pakistan (site 23), TOMCAT overestimates O_3 probably due to a smearing out of the O_3 gradients around the tropopause [*Law et al.*, 1998]. This has already been noted for other Northern and Southern Hemisphere locations. However, over Pakistan, the other two models, MOZART and MATCH-MPIC, are not able to capture the spring peak in O_3 very well either; MATCH-MPIC peaks rather early, and MOZART peaks too late in the fall. Only TOMCAT calculates lower O_3 concentrations during the summer months in good agreement with the data at site 23. Again, this could be related to using meteorology for the correct year, 1995.

Over Burma (site 25), the models driven by analyzed meteorology reproduce the observed monthly mean concentrations quite well. MOZART, which was forced using climate model winds, tends to slightly overpredict O_3. However, none of the models simulate the very low concentrations seen during August in the observed O_3 standard deviation and only TOMCAT predicts O_3 levels below 30 ppbv. The low observed O_3 may be caused by chemical reactions which are not included in models at the present time or by inadequacies in the humidity fields used in the model integrations.

5.4.2. Vertical profiles over tropical Asia. MOZAIC data were collected at several locations over Asia. Here data and model results for Bangkok (note only three profiles available in October) at four altitudes (Figure 15), vertical profiles for Saigon in February and September (Figure 16) and Madras in January and October (Figure 17) are discussed. There are several features which are common to the observed seasonal cycles in the profiles collected over tropical Asian locations. In the lower and mid troposphere, Bangkok, Saigon, Hanoi (not shown), and Madras all have higher concentrations in the winter/ early spring. This maximum which broadens with increasing altitude could be due to long-range transport of pollutants by northwesterly

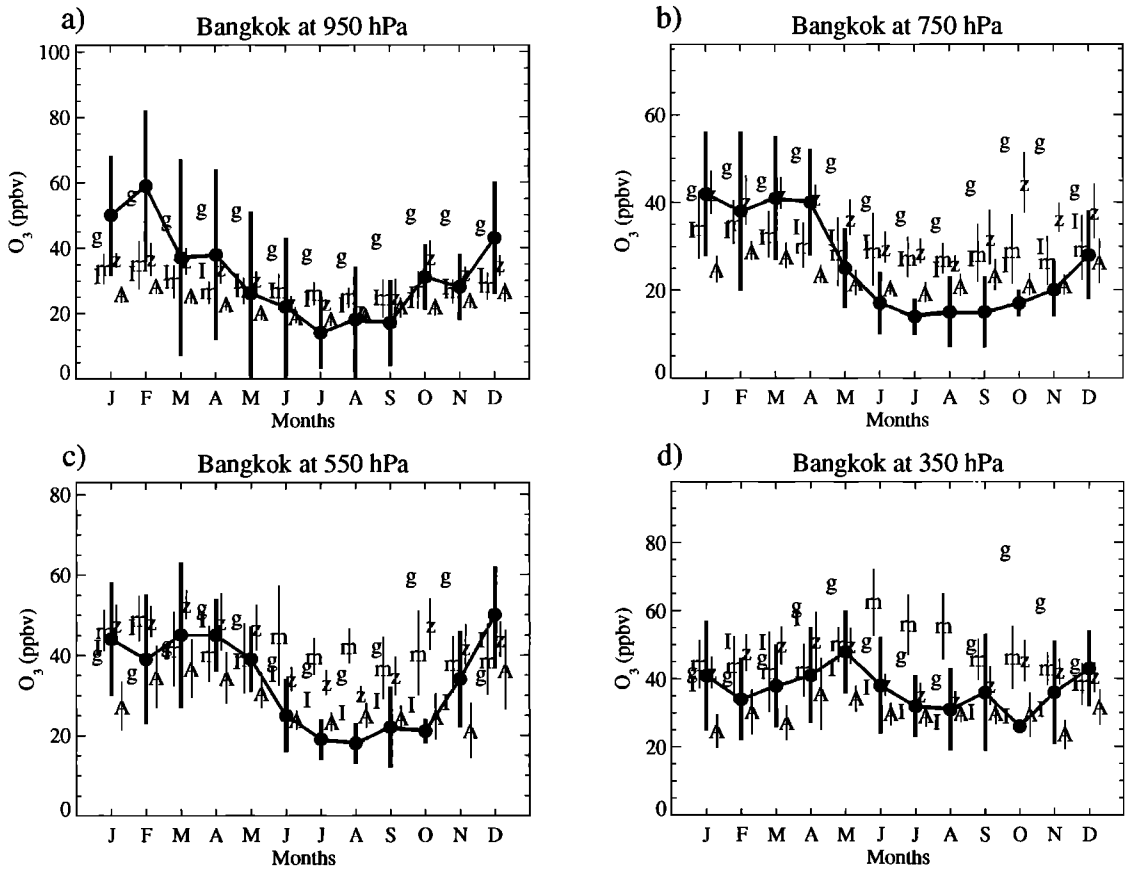

Figure 15. Monthly mean concentrations of O_3 in ppbv over Bangkok, Thailand, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) over 50 hPa intervals centered on (a) 950 hPa, (b) 750 hPa, (c) 550 hPa, and (d) 350 hPa. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART and TOMCAT model results. Note that in October only three MOZAIC O_3 profiles were used to calculate the statistics and that all averages are centered on the middle of the month.

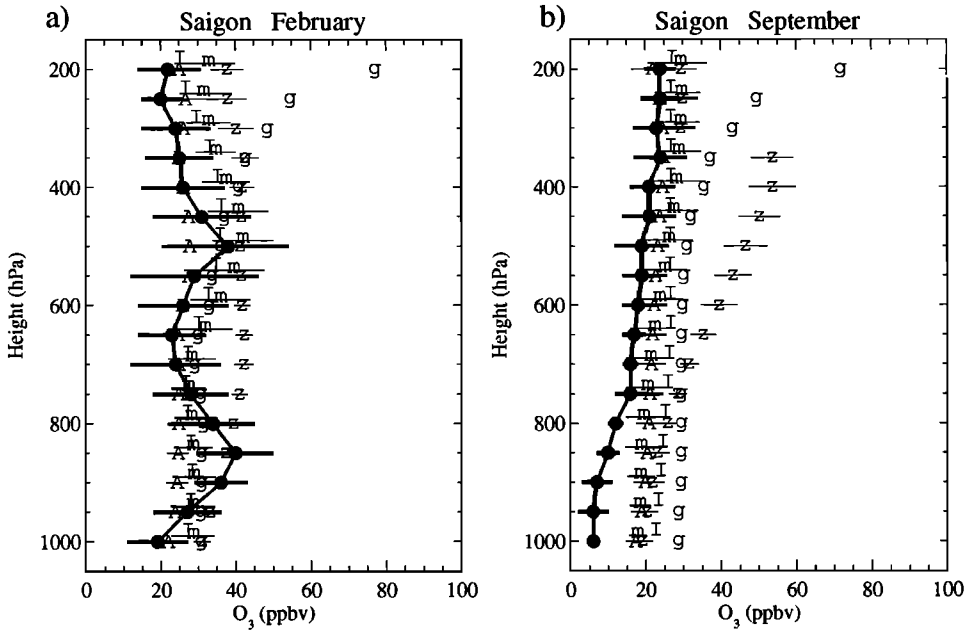

Figure 16. Monthly mean concentrations of O_3 in ppbv over Saigon, Vietnam, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) centered on 50 hPa intervals from 1000 hPa to 200 hPa for (a) March and (b) October. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results.

winds associated with the winter monsoon circulation bringing air from the the Asian continent [*Hastemrath*, 1985]. The presence of higher O_3 concentrations in the midtroposphere at this time of year may be due to uplift farther north by convection or possibly frontal systems. Conversely, these locations show a summer minimum with mean concentrations as low as 15 ppbv in the period from July to September at the surface. This is

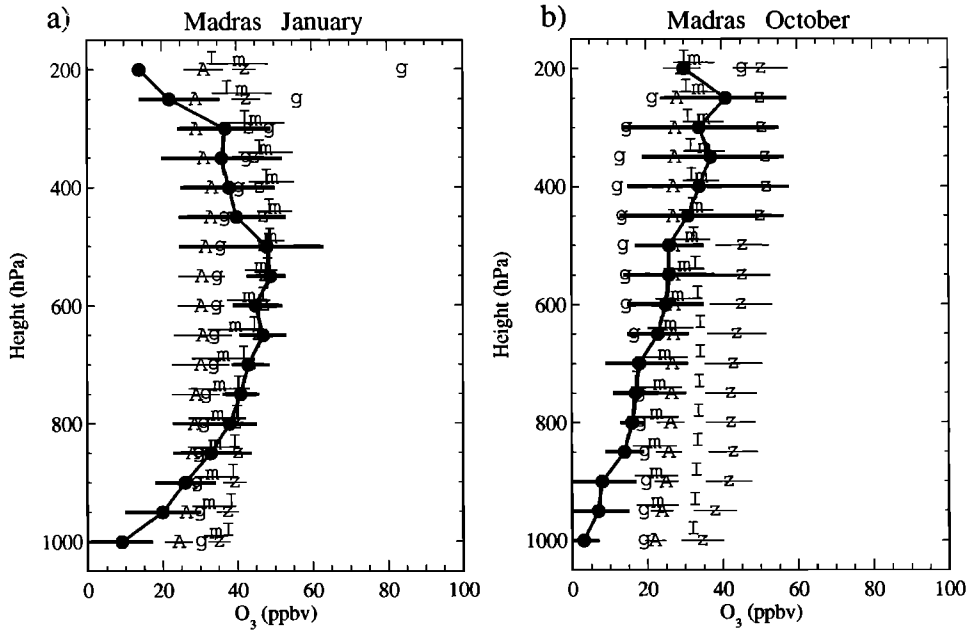

Figure 17. Monthly mean concentrations of O_3 in ppbv over Madras, India, calculated from MOZAIC data (solid circle denotes mean concentration) and model results (I, IMAGES; g, MOGUNTIA; m, MATCH-MPIC; A, TOMCAT; z, MOZART) centered on 50 hPa intervals from 1000 hPa to 200 hPa for (a) March and (b) October. Standard deviations (denoted by the bars) are also shown for the measurements and MATCH-MPIC, MOZART, and TOMCAT model results.

related to the monsoon circulation advecting clean air from the southwest over the Indian Ocean to the ITCZ region which is well north of the equator in the summer months.

At Bangkok, most models miss the spring peak in the lower troposphere possibly due to a lack of emissions farther north over China resulting in an underestimation of photochemically produced O_3 in this region. Interestingly, 1995 was an El-Niño year and biomass burning emissions were higher in the region of Southeast Asia. None of the models take into account year-to-year variability in the emissions. MOGUNTIA overestimates O_3 in the lower troposphere, particularly in October and November. The models also slightly overestimate the summer/fall minimum as a result of having rather flat seasonal cycles. Again, TOMCAT is the only model to capture the low observed mean O_3 in the summer over Bangkok but extreme events, with very low O_3, are again not simulated by any model. The models also overestimate O_3 near the surface at Saigon (see Figure 16b) and Madras (see Figure 17b) where observations are as low as 5 ppbv in the late summer. In the mid to upper troposphere, the models are able to capture the winter/spring maximum over Bangkok reasonably well but the summer minimum is overestimated.

Over Saigon, in February (see Figure 16a), the models have rather invariant vertical profiles, whereas the data exhibit two maxima; one in the lower troposphere and one in the upper troposphere. However, it should be noted that rather few measured profiles were collected (see Figure 3). Again, lack of emissions in the tropics or deficiencies in long-range transport of pollutants may be responsible for this. The same points can be made for the model results over Madras in January (Figure 17a) although the observed maximum is broad and spans the midtroposphere region. MATCH-MPIC and MOZART reproduce the mean vertical profile over Madras reasonably well in January, and MATCH-MPIC also predicts lower concentrations over this location in October in the midtroposphere, which is in agreement with the observations. However, MOZART overestimates O_3 in the fall over Madras and over Saigon. MOGUNTIA, on the other hand, underestimates O_3 in the upper troposphere in October and in the midtroposphere in January over Madras (as does TOMCAT). These differences may suggest discrepancies in the emissions used by the models over Asia.

6. Summary of Model Performance

This comparison has highlighted several discrepancies between MOZAIC O_3 observations and the model results as well as some regions where agreement is very encouraging. In this section, each model is discussed in turn before making some general conclusions about model performance.

1. IMAGES overpredicts O_3 around the tropopause in the subtropics and northern midlatitudes indicating that the horizontal flux of stratospheric O_3 is too strong; a fact that has been noted previously [*Rasch et al.*, 1997; *Muller and Brasseur*, 1999]. It is important to note that IMAGES (like MOGUNTIA) calculates monthly average concentrations. The effect of wind variability is accounted for as a diffusion process. The diffusion coefficients (calculated from the variances of the observed winds) are largest at midlatitudes just below the tropopause in IMAGES. Excessive meridional diffusion in the winter and spring has resulted in excessive O_3 at these altitudes in the model runs shown here. IMAGES also tends to produce too much O_3 photochemically near the surface in the tropical biomass burning season and in the Northern Hemisphere summer. The chemical scheme used in this model may be the reason for this discrepancy [*Muller and Brasseur*, 1999].

2. MOGUNTIA underestimates O_3 concentrations at cruise altitudes outside the tropics but overestimates O_3 in the tropics suggesting problems related to this model's top boundary condition at 100 hPa, the flux of stratospheric O_3, and the model's poor vertical resolution, which will affect its ability to model the location of the tropopause correctly. Note that the global exchange of mass through the tropopause estimated by *Holton* [1990] and the O_3 concentrations at 100 hPa measured by *Komhyr et al.* [1989] have been used to define the flux of ozone at 100 hPa level in this model. The points made about the IMAGES model, relating to the use of climatological input data, are also relevant to the MOGUNTIA model. Considering this model has relatively coarse horizontal resolution, it reproduces O_3 near to the surface in the Northern Hemisphere reasonably well although it does overpredict O_3 in the tropics. It would be interesting to see whether this model, which includes a comprehensive NMHC scheme, would overpredict O_3 if run at higher horizontal and vertical resolution.

3. MOZART has a tendency to underestimate O_3 above ~300 hPa during spring. This was also seen when comparing to ozonesonde data [see *Hauglustaine et al.*, 1998], and it is known that this model has a rather low cross-tropopause flux of O_3. However, this model does show day-to-day variability with seasonal cycles which are generally in phase with the observations and spring maxima at cruise altitudes in midlatitudes. However, in common with most other models, MOZART shows less variability than seen in the observations. MOZART also tends to produce too much O_3 in the mid and upper troposphere in the tropics over Asia.

4. TOMCAT overestimates O_3 concentrations at cruise altitudes in midlatitudes and in the subtropics. This is partially due to the gradients being smeared out but tracer experiments have also shown that this appears to be due to an overly strong stratospheric circulation in this model. It may also be related to the lack of full stratospheric chemistry in the lower strato-

sphere and the treatment of O_3 at the top boundary [H. Teyssèdre et al., manuscript in preparation, 1999]. TOMCAT also underpredicts summertime photochemical O_3 production at northern midlatitudes and also in the tropics during, for example, the biomass burning season over Asia. Lack of NMHC chemistry or low emissions can account for this discrepancy. Interestingly, TOMCAT is able to simulate the low O_3 concentrations over tropical coastal locations, though not as low as observed.

5. MATCH-MPIC often compares well with the MOZAIC data, probably due to the simulations being run at higher horizontal resolution compared to the other models. This is an interesting result, given that MATCH-MPIC only includes CH_4 and CO oxidation chemistry. Around the tropopause, it appears that modeled O_3 distributions are being controlled more by meteorology than by (NMHC) chemistry. Agreement between this model and the data is also reasonably good in the troposphere. However, it would be interesting to see if inclusion of NMHC chemistry would result in an overestimation of O_3 concentrations. The MATCH-MPIC model does have problems reproducing the seasonal cycle correctly in certain regions. For example, maximum O_3 concentrations occur too early in the Northern Hemisphere spring at cruise altitudes and in areas affected by biomass burning emissions in the tropics. This may be due to the use of analyzed meteorological data for 1993 or the seasonality in the stratospheric O_3 flux or biomass burning emissions which are not representative of the years when the measurements were made.

7. Conclusions

Results from five global CTMs have been compared with 2 years of MOZAIC O_3 data. At cruise altitudes, areas generally showing reasonably good agreement are found over Europe and North America although some models have difficulty reproducing the spring maximum around the tropopause. In regions where data have been limited or were nonexistent previously (e.g., over Asia), the agreement is much poorer. Reasons for these discrepancies may be due to the input meteorological data, the model formulation of O_3 transport downward from the stratosphere, including the way O_3 is prescribed at the top boundary, and poor vertical and/or horizontal resolution around the tropopause. Some models also have problems capturing latitudinal and longitudinal gradients seen in the observations, particularly in the region of the subtropical jet stream. Again, this is probably related to model resolution which affects the ability of models to correctly position the tropopause, rather than a lack of chemistry.

In the troposphere, models tend to underestimate O_3 concentrations at sites in the Northern Hemisphere which exhibit a summer maximum, probably due to a lack of photochemical O_3 production. This may result from a lack of chemistry, deficiencies in transport schemes, as well as inadequate resolution. Conversely, at certain coastal sites, which exhibit a summer minimum, most models tend to overestimate O_3 concentrations either due to deficiencies in the specified wind and/or humidity fields or missing chemistry. Discrepancies in the seasonality or magnitude of emissions may account for differences between modeled and measured O_3 over Asia and South America.

In summary, this study has highlighted many interesting features in the MOZAIC O_3 data set and has allowed regions where the models perform well, and not so well, to be identified. This shows the value of the MOZAIC data set for model evaluation. Overall, the models are able to reproduce the seasonality of O_3 at many locations within the limits of observed variability which is very encouraging. Reasons for differences have been identified, and further investigations are now required to pinpoint exactly why each model agrees or differs from the data at various locations. It is apparent that, perhaps not surprisingly, model simulations run at high vertical and horizontal resolution and with analyzed/assimilated meteorological fields at high temporal resolution are better able to capture the observations. As more MOZAIC data is collected, it will be possible to build a more complete climatology against which global CTMs can be compared.

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