Trace gases and air mass origin at Kaashidhoo, Indian Ocean

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[1] Carbon monoxide (CO) was measured at the Kaashidhoo Climate Observatory (KCO, Republic of Maldives) between February 1998 and March 2000 to assess the regional pollution of the remote atmosphere in the northern Indian Ocean. CO showed a distinct annual cycle with maximum daily mixing ratios of around 240 parts per billion (ppb), a seasonal difference of about 200 ppb, and high variability during the dry seasons. Detailed air mass trajectory analysis for 1998, 1999, and 2000 was used to identify source regions and to associate them with various levels of pollution encountered at KCO. We conclude that most significant changes in local pollution throughout the year are caused by changes in air masses. Air at KCO generally originated from three main regions with decreasing pollution: India and southeast Asia, the Arabian Sea, and the Southern Hemisphere. We show that isentropic air mass trajectories can be used to predict CO pollution levels at KCO to a certain extent and vice versa. Nitrous oxide, CFC-11, CFC-12, CCI_4, and SF_6 were measured during the Indian Ocean Experiment (February to March 1999) to support pollution analysis and to confirm that India is the main source for heavy pollution measured at KCO. Correlations between CO and other gases and aerosol properties measured at the surface illustrate that CO may also be used as a proxy for aerosol loading and general pollution at the surface. *Index Terms:* 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 9340 Information Related to Geographic Region: Indian Ocean; 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; *Keywords:* INDOEX, Maldives, carbon monoxide, CFC, Indian Ocean, atmospheric chemistry

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1. Introduction

[2] The Indian Ocean Experiment (INDOEX) was a multiagency, multiplatform effort to study the impact of pollution on clouds, climate, and regional air quality and the regional and long-range transport of aerosols and trace gases during the dry seasons of 1998 (First Field Phase (FFP)) and 1999 (Intensive Field Phase (IFP)) in the remote, northern Indian Ocean [*Coakley et al*., 2002]. The main reasons for choosing the Republic of Maldives as a base for the measurements were the high population density and limited air quality restrictions in the surrounding continents as well as a circulation pattern over India that transports large amounts of pollution into the remote Indian Ocean during the dry season, which alternates with quasi-pristine conditions during the wet season [*Lelieveld et al*., 2001; *Ramanathan et al*., 2001]. Very few long-term data exist for this region, which increasingly impacts regional and global environmental change.

[3] Carbon monoxide (CO) is an important intermediate in the oxidation cycles of hydrocarbons and methane and affects the oxidizing capacity of the atmosphere through its role as a main sink for the hydroxyl radical (OH) in the troposphere [*Crutzen and Zimmermann*, 1991; *Thompson*, 1992] and as an intermediate in tropospheric ozone production [*Crutzen*, 1973]. For a recent, comprehensive compilation on CO science, see *Khalil et al*. [1999]. With a mixing ratio of about 40–50 parts per billion (ppb, 10^{-9} mole per mole) in the unpolluted, Southern Hemispheric air [*Novelli et al*., 1998], a reasonably long lifetime of about 2 months, and a lack of large sources in the remote Indian Ocean, CO is an excellent tracer for pollution from incomplete combustion such as fossil fuel use [e.g., *Bradley et al*., 1999] and biomass burning [e.g., *Crutzen et al*., 1979].

[4] Chlorofluorocarbons (CFC), sulfur hexafluoride (SF_6), tetrachloromethane (carbon tetrachloride, CCl_4), and nitrous oxide (N_2O) have lifetimes of decades to more than a century and significant implications on global warming and stratospheric ozone depletion [*Schimel et al*., 1996; *Albritton et al*., 1999]. Owing to their long lifetimes, the halogenated gases do

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not undergo appreciable decay throughout their transport to the Indian Ocean and can, thus, be used to confirm and locate anthropogenic activities as they also do not have significant sources and sinks over the ocean.

[5] Carbon monoxide (CO) was measured at the Kaashidhoo Climate Observatory (KCO) in the Maldives between February 1998 and March 2000, and some halogenated compounds and N_2O have been measured during the FFP and IFP to characterize the intensity and spatial and temporal outflow of pollution from the Indian subcontinent.

[6] Here we present the first long-term record of any trace gas for this region in the Indian Ocean along with the seasonality and features in the mixing ratio of CO at KCO, a detailed air mass trajectory analysis for the dry seasons, and some important correlations between different trace gases and between CO and aerosol properties.

2. Location and Methods

[7] KCO is located in the remote Indian Ocean at 4.966°N and 73.466°E (Figure 2, center point). This location is subjected to the air of two hemispheres because the interhemispheric tropical convergence zone (ITCZ) moves across the site twice per year. In addition, air mass trajectories also show that air from many distinctly different regions within one hemisphere are encountered throughout the year, which makes the Maldives unique to study air mass origin, pollution levels of several regions and their possible impact on climate; see also section 5 for further resources.

2.1. Carbon Monoxide Measurements

[8] For the determination of CO in this study, we used a continuous nondispersive infrared (NDIR) absorption instrument, model 300 (Advanced Pollution Instrumentation), which is described in detail by *Cogan and Lobert* [1998]; the basic functionality and main measurement interferences have also been discussed in detail by *Dickerson and Delaney* [1988]. Briefly, the commercial instrument includes a zero and span calibration module and was complemented by an external, membrane gas dryer (NAFION type, model PD625-24-AFS, Permapure), which was operated at a reduced pressure (pump and needle valve arrangement to keep drying air at around 800 hPa) to reduce the sample dewpoint to −40°C. Sampling was carried out through ~15 m of 6.3 mm OD FEP fluoroethylene-propylene (FEP) and Decoron™ tubing (Alltech, Goodrich Sales) with an intake of 47 mm, 5-μm PTFE (Teflon) membrane filters (Cole-Parmer). Residence time of air in the sampling system at a flow rate of 0.7 L min^{-1} was about 30 s prior to entering the instrument; the lag time for the 95% level signal was approximately 4 min.

[9] Calibration of the instrument was carried out with an ambient, real air standard in an Aculife™ treated aluminum cylinder with brass valve (Scott Specialty Gases). This secondary standard was filled at Niwot Ridge, Colorado, and calibrated for CO by the National Oceanographic and Atmospheric Administration's Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) at a mixing ratio of 125.8 ± 1.2 parts per billion (ppb, 10^{-9} mole per mole). Within the precision of this calibration, the mixing ratio did not change during the first 2 years of this study (2-year recalibration yielded 124.9 ± 1.2 ppb). These calibrations reflect the latest change in the CMDL calibration scale for CO (P. C. Novelli, personal communication, June 2000). Daily checks of the instrument's sensitivity were carried out with a commercial gas standard (Scott Specialty Gases) at 497.3 ± 4.8 ppb and did not reveal any drift in instrument sensitivity during the first 2 years of operation.

[10] Data were collected continuously by the instrument and averaged for output over 30 min for this study. Thirty-minute means (which alternated with a 20-min zero calibration every 50 min) were then averaged to derive daily, weekly, and monthly means. The precision of ambient air measurements, defined as 1 standard deviation of the measured, 30-min, mean CO mixing ratio over 3 months during the wet season (detrended), was 9.6 ppb and varied only slightly throughout the year with elevated (worse) precision during the dry seasons due to higher variability of ambient mixing ratios. Similarly, the standard deviation of observations within any daily mean was, on average, 10.1 ± 3.9 ppb (3 to 38 ppb) over the entire data record (not detrended). The precision of repeated zero-level measurements was only 5.0 ppb, reflecting the instrumental noise level [*Cogan and Lobert*, 1998]. The lower detection limit of this instrument, defined as 3 times the precision, is then between 15 and 30 ppb. The lowest, observed, mean, daily mixing ratios at KCO were 45–50 ppb. Data were screened for instrumental problems and local pollution and then subjected to a recursive filter of 3 standard deviations, rejecting an additional 1.5% of the 30-min means.

[11] For quality assurance, continuous CO data were compared with flask data collected by NOAA/CMDL (Figure 1, *P. C. Novelli et al*., NOAA/CMDL, unpublished data, 1999). The mean difference between CMDL flask pairs and KCO 3-hour means was 8.1% for the intercomparison period March 1998 to July 1999 (CMDL being higher, $N = 45$). The agreement is somewhat better during the wet season, where mixing ratios do not fluctuate as much as during the dry season, when gradients in excess of 80 ppb per day have been observed. A brief intercomparison with similar, continuous data collected on the R/V *Ronald H. Brown* on March 5, 1999, yielded an agreement within 10%, with KCO data being lower (*R. Dickerson*, University of Maryland, personal communication, 1999).

2.2. Measurements of Other Gases

[12] Halogenated compounds and N_2O were measured with a gas chromatograph equipped with dual-electron capture detectors (GC/ECD, Agilent Technologies). The commercial GC system was modified with stream selection and injection valves (Valco), a membrane dryer (NAFION type, model MD-110-96-SF, Permapure), and a pumping station as described by *Lobert et al*. [1995] except for using a metal bellows pump at 15 L min^{-1} (model MB-158, Senior Flexonics), 0.95-mm OD Decoron™ tubing (Goodrich Sales) and a glass-fiber-filled inlet filter in the current setup. Chromatographic methods have been published by *Lobert et al*. [1995] for CFCs, CCl_4, and N_2O and by *Geller et al*. [1997] for SF_6. Any two ambient air measurements were bracketed with a calibration standard injection for a mean frequency of 45 min per ambient air measurement.

[13] Calibration of these gases was carried out with ambient air standards linked to the above mentioned secondary standard and bracketing the ambient concentrations by about 10% to compensate for the nonlinear character of the ECD [*Bullister and Weiss*, 1988; *Butler and Elkins*, 1991]. Calibration of CFCs, N_2O, and SF_6 in the standard gas tanks was carried out by NOAA/CMDL and Scripps Institution of Oceanography (SIO) in 1997 and 1999. Only small drifts (less than 1%) in the mixing ratios of compounds were encountered between

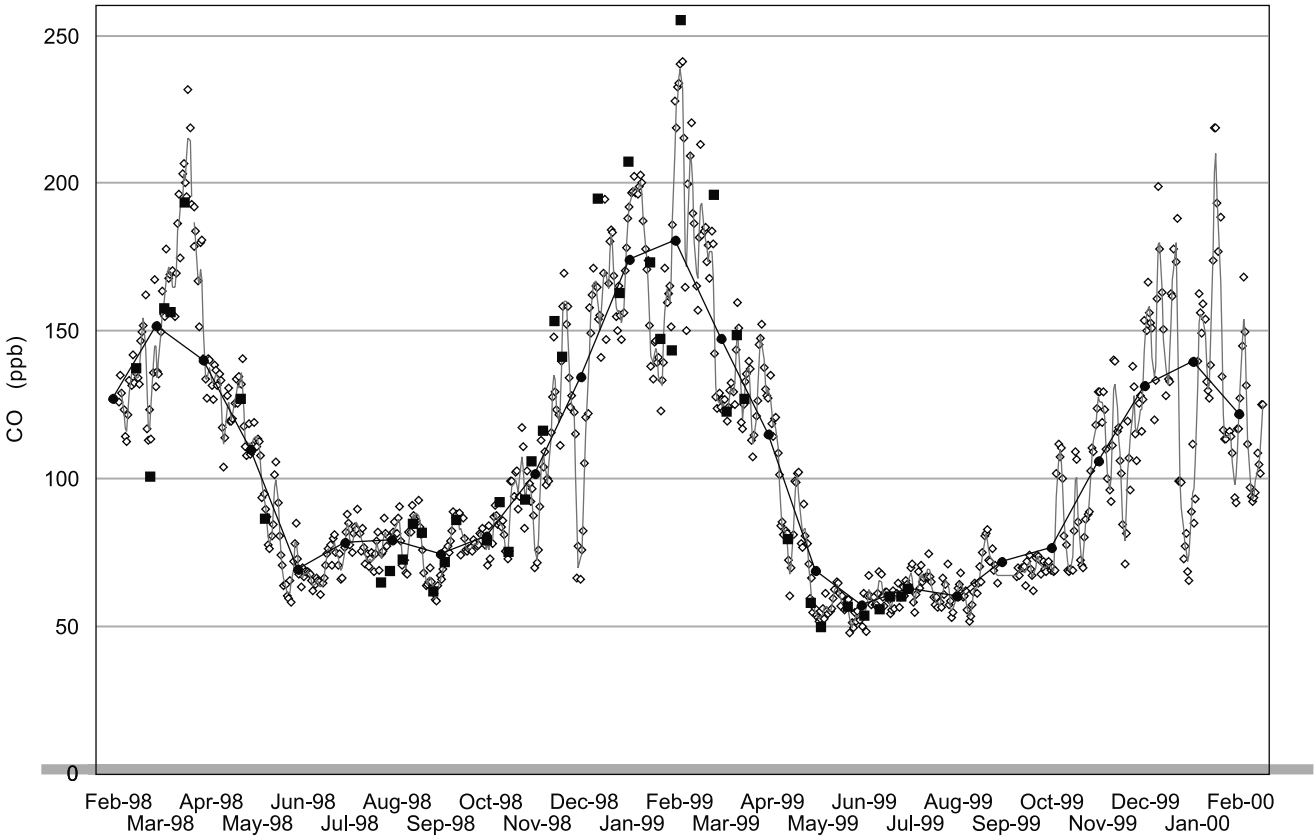

Figure 1. Time series of daily (open diamonds) and monthly (solid circles) mean mixing ratios of surface CO at KCO with flask data (solid squares) from the NOAA/CMDL flask network. The light dotted line is a running mean ($N = 3$) through the daily means; the heavy solid line connects monthly means to guide the eye. Unpublished CMDL data courtesy of *P.C. Novelli et al*. (1999).

standard calibrations. Air intakes for all trace gas studies were mounted on top of the observation tower at KCO, about 13 m above ground, pointing northeast, which is the prevailing wind direction during the dry season. Measurement precision of the compounds was between 0.3% (CFCs, N_2O) and 5% (SF_6); accuracy of the CMDL and SIO calibration scales is within 3% except for SF_6, which is 5% (CMDL scale, *Hall et al*. [2000]; SIO scale, *Weiss et al*. [1981] and *Prinn et al*. [2000] SF_6 scale, *Geller et al*. [1997]).

2.3. Trajectory Analysis

[14] For trajectory analysis, we employed an isentropic trajectory model of NOAA/CMDL [*Harris and Kahl*, 1994], which uses gridded data from the European Centre for Medium-Range Weather Forecasts (ECMWF). This model estimates the 10-day pathway of air parcels arriving at KCO twice daily at 0000 and 1200 universal time (UT) with positions given every 3 hours. The model uses a dual-mode, vertical transport assumption: if the trajectory falls below 100 m above ground level (AGL), horizontal advection is modeled using winds vertically averaged through a 500-m layer. When the parcel is not close to the ground, the isentropic transport assumption is used. Uncertainties in trajectories are introduced by interpolation, diabatic effects, subgrid-scale processes, and observational errors. Trajectories are generally good indicators of large-scale flow, but because of uncertainties, any one trajectory should not be viewed as the guaranteed path of an air parcel. Trajectory uncertainties are discussed by *Merrill et al*. [1985] and *Harris* [1992] and references therein.

[15] Trajectory cluster analysis was employed to determine the transport pathways to KCO as shown in Figure 2. This multivariate statistical method separates the trajectories into groups or clusters by maximizing the similarity of cluster members (both length and shape) while keeping the groups as distinct as possible [*Harris and Kahl*, 1990]. The mathematical algorithm does not result in a unique solution but rather gives an objective approximation. For this reason we use the technique as a tool to describe or summarize the data, not as immutable fact. We chose to calculate six clusters for each figure because we have found by experience that this number gives some detailed information concerning transport to the site, while at the same time it usually does not result in redundant clusters (except where air mass transport is very uniform, as can be seen in monthly clusters for KCO). Cluster mean trajectories are assigned a percent frequency of occurrence; the cluster mean represents the average pathway of trajectories in that cluster. Because of the natural variability of atmospheric processes, each cluster contains a range of trajectory shapes and origins upwind.

3. Results and Discussion

3.1. Carbon Monoxide and Air Mass Trajectory Analysis

[16] Carbon monoxide at KCO describes a distinct annual cycle with daily mean mixing ratios varying between 50 ppb in

Table 1. Seasonal and Annual Statistics for CO (in ppb) at KCO Calculated From Daily Means[a]

	Dry 1998[b] Feb.–May 15	Wet 1998 May 16–Oct.	Dry 1999 Nov.–April	Wet 1999 May–Oct.	Dry 2000[b] Nov.–March	Dry Mean	Wet Mean	1999 Mean	1998–2000 Mean[b]
Mean	146	78	142	66	123	137	72	105	107
Std. dev.	28	11	39	12	33	36	13	48	43
Median	136	77	139	64	117	132	70	85	94
Minimum	. . .[c]	58	. . .[c]	48	. . .[c]	. . .[c]	48	48	48
Maximum	232	. . .[c]	241	. . .[c]	219	241	. . .[c]	241	241
N, days	86	169	181	172	122	389	341	352	730

[a]Dry and wet refer to the dry (NE monsoon) and wet (SW monsoon) seasons at KCO. Std. dev., standard deviation.
[b]Partial dry seasons only in 1998 and 2000.
[c]The assignment of seasons is somewhat arbitrary owing to a lack of a sharp transition point; hence we did not calculate dry minimum and wet maximum values. For the same reason, the mean seasonal values are somewhat uncertain.

the wet (SW monsoon) season and around 240 ppb in the dry (NE monsoon) season (Figure 1). The Maldives (a republic with a population of about 300,000 on a land area of 300 km^2 spread over 1200 islands and 8° of latitude) do not represent a significant mesoscale source of pollution, and the observed annual CO cycle is rather based on the influx of polluted and clean air masses caused by the movement of the ITCZ, which separates the troposphere of the Southern Hemisphere (SH) and Northern Hemisphere (NH) and moves across the site twice per year around April-June and October-December. The ITCZ can extend as far north as 30°N during the wet season, when the station is exposed to very clean Southern Hemispheric air, and as far south as 20°S during the dry season, when KCO is subjected to the pollution outflow from the Indian subcontinent, SE Asia, and Arabia [*Pant and Kumar*, 1997; *Krishnamurti et al*., 1997]. Basic statistics of annual and seasonal CO data are given in Table 1.

[17] The annual, mean CO mixing ratio at KCO for the entire 1998-2000 record was 107 ppb with a spread of about 200 ppb between minimum and maximum observations (Figure 1, Table 1). During the wet season, the day-to-day variability of mixing ratios is much lower and reflects a more consistent air mass source than that of the dry season. Most of the high CO variability during the dry season is a result of varying source regions. Changes of CO as high as 80 ppb per day were observed in the dry season of 2000, whereas wet season changes typically did not exceed 15 ppb between any 2 days. Some of the large changes are due to vertical transport and nearby cyclonic systems such as the large drops in CO mixing ratios in December 1998, February 1999 (discussed by *Verver et al*. [2001]), and January 2000, but most of the remaining variability can be explained by different air masses transporting varying levels of pollution to KCO. Prevalent wind direction at KCO, and hence air mass origin, can change within hours and impacts the local pollution accordingly.

[18] A detailed cluster analysis of trajectories for the site reveals several main air mass source regions. For Figure 2, we averaged 7828 individual, twice-daily trajectories over a period of 11 years (1989 to 1999) to derive a mean, annual air mass transport scheme represented by the six cluster means. Near-surface level air (500 m, 950 hPa) arriving at KCO basically originates from either the Northern Hemisphere or the Southern Hemisphere. Monthly cluster means reveal that transport of Northern Hemispheric air to KCO is limited to the dry season, and that of Southern Hemispheric air is limited to the wet season with considerable overlap during the transition seasons (not shown here).

[19] Dry season air masses are transported to KCO along two major paths, the Bay of Bengal and the Arabian Sea. Air masses generally follow the main pathways indicated by the clusters, but variations along these paths with measurable implications on KCO pollution occur through the proximity of the air to the continents. A third source region is Southeast (SE) Asia, which does not show for the 950-hPa level in the 11-year mean owing to its infrequent occurrence. Nonetheless, at higher altitudes (1500 and 2500 m, Figure 2), the influence of SE Asian air increases and contributes 18% and 25% to the long-term clusters of NH air and can substantially pollute the troposphere over the Indian Ocean around KCO. SH air generally originates in the midlatitude Indian Ocean within 10 days prior to arrival and typically travels faster than NH air. However, its pathways vary to a large degree in their proximity to the African continent and the Somali Basin, which is more pronounced at higher altitudes.

[20] Associations of individual, semidaily trajectories with CO measured at KCO reveal a general pattern of pollution levels at KCO, which we define as high (CO > 140 ppb),

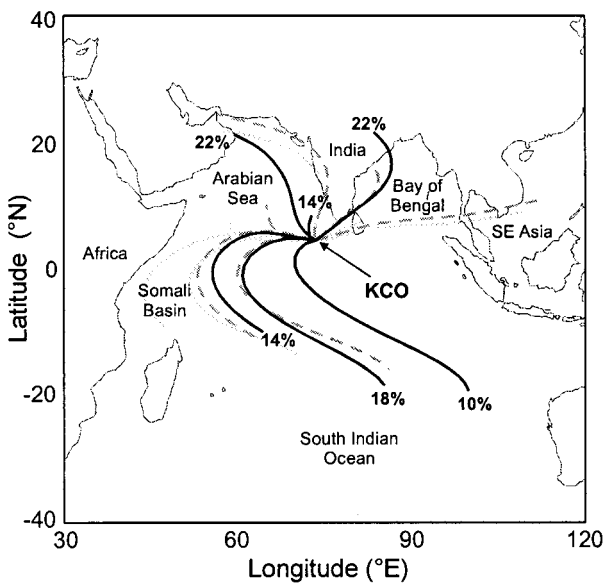

Figure 2. Map of the Indian Ocean with the location of KCO indicated at the center point. Lines merging at KCO are 11-year mean, annual, 10-day back-trajectory clusters for air masses arriving at KCO. Heavy solid lines are for 950-hPa (500 m) arrival level, dashed lines are for 850 hPa (1000 m); and light dotted lines are for 750 hPa (2500 m). Percentages are the frequency of individual trajectories at the 950-hPa level.

moderate (140 > CO > 80 ppb), and low (CO < 80 ppb). High pollution at KCO was typically produced through air originating from within India and traveling either through the Bay of Bengal or directly from within the continent, with typical transport times of 6 to 10 days until arrival at the site. The curved nature of the cluster trajectory through the Bay of Bengal is due to a persistent anticyclonic circulation over the Indian subcontinent during the dry season. Along with a strong subsidence of tropospheric air over this region and a suppressed, horizontal dispersion [*Verver et al*., 2001], continental outflow from this region is heavily polluted and, accordingly, generates the highest CO mixing ratios at KCO. A less frequent channel for high pollution is from SE Asia (~10°S to ~20°N).

[21] Moderate CO pollution at KCO was observed whenever the air originated from the northwest and was transported through the Arabian Sea. Air from this channel can vary considerably in CO depending on its original source region and proximity to the Indian subcontinent, where it can mix with the continental outflow from the "Bombay plume." The end of April 1999 demonstrated a clear transition from higher to lower CO pollution when air mass pathways slowly departed eastward from the Indian continent and remained increasingly in the west Arabian Sea. During the 1999 IFP, air from as far away as Europe was encountered from this channel. For transport across such large distances within 10 days, air typically traveled at higher altitudes (faster) and subsided into the Indian Ocean upon arrival over the Arabian Sea.

[22] A significant change in source regions from India/Bay of Bengal to the Arabian Sea was encountered during the 1999 IFP, when CO mixing ratios sharply dropped from 140–240 ppb in February and early March to about 100–150 ppb in April. This was caused by a subtropical high over India that shifted eastward to the Bay of Bengal and cut off the outflow of Indian air to KCO, where winds changed accordingly from northeasterly to northerly flow. This episode and the changes in CO are discussed in more detail in other papers [*de Laat et al*., 2001; *Verver et al*., 2001]; J. M. Lobert and J. M. Harris (unpublished manuscript, 2000, see section 5 for more data on INDOEX) discuss the vertical structure of the troposphere for that period. Monthly trajectory clusters for this dry season are included in Figures 3e-3h. Notable is a distinct shift from 91% Indian air in February (Figure 3f) to 24% in March (Figure 3g) of 1999.

[23] Air masses arriving at KCO during the wet season all originated from the middle to high latitudes of the Southern Hemisphere (Figure 2). Pristine SH air contains about 35–45 ppb CO [e.g., *Novelli et al*., 1998], but daily means at KCO never dropped below 47 ppb and that only for a few days in the 1999 wet season (Figure 1). During this period with lowest CO (end of May to beginning of June 1999), KCO encountered air masses that had traveled quickly from the SH around 30°–40°S to KCO on a trajectory through the ocean without contacting any continent and without deviation through the Somali Basin. On average, however, wet season CO at KCO was 78 ppb in 1998 and 66 ppb in 1999 (Table 1), indicating prevalent low pollution in arriving air masses. Daily trajectories reveal that during the 1998 wet season, air masses traveled, on average, closer to the African continent compared with the same period in 1999. Southern Africa during its dry season of June to September (the wet season at KCO) is subjected to a similar continental gyre as is found over India during the months of January to March [*Swap and Annegarn*, 1999]. Consequently, the Somali Basin and South Indian Ocean are outflow regions for southern Africa and can exhibit significant levels of pollution from biomass burning. This explains the nonpristine character of air associated with western, SH cluster means in Figure 2 and associated low pollution even during the wet season in the Maldives.

[24] The six cluster means for each altitude depicted in Figures 2 and 3 are usually enough to describe the mean air mass transport at the surface for any given location. However, when we associated daily trajectories with CO mixing ratios, we identified some minor flow paths that are of distinct interest to the KCO location. These minor flow paths are, of course, included in (but visually not represented by) the clusters of Figure 2. Hence we have defined three more discrete air flow bins in addition to the cluster means of Figure 2 for a total of nine source bins (Figure 4a). Bins 2, 3, 4, 7, 8, and 9 are basically identical to those of the cluster means in Figure 2; the three additional bins (1, 5 and 6) were defined as follows. Bin 1 represents air from SE Asia, bin 5 is associated with air circulating in the open ocean around KCO without contact to land (varying directions, low wind speeds), and bin 6 is air from the west Arabian Sea without contact to any continent but possibly under influence from the ITCZ. Bins 1 through 5 are associated with the dry season, i.e., air from north of the ITCZ; bins 7–9, with the wet season or air from south of the ITCZ. We associated individual, daily trajectories with these bins such that air had to spend most of its time (≥5 days) in any given bin.

[25] Surface mixing ratios were correlated with air masses traveling through the nine bins and arriving at KCO at an altitude of 500 m (Figure 4b and Table 2). Bins 1–3 exhibit the highest pollution levels and can be combined into a general north to northeasterly flow pattern. Bins 4 and 5 describe the northwesterly and westerly flow regime and represent moderate pollution at the site. The air from the west Arabian Sea (bin 6) is, on average, lower than any of the other dry season air masses but very similar to the air from bin 7, which is air from south of the ITCZ. This pollution level appears to be characteristic for the Somali Basin. As expected, bins 8 and 9 are very clean but, on average, still higher than the expected background level of pristine air. This may be, in part, due to pollution outflow from Australia, caused by biomass burning. Biomass burning emissions from Australia can be expected yearround but peak between August and October, the tropical burning season [*Cheney et al*., 1980].

[26] When comparing measurements taken at other sites with those of KCO, an increasing trend in the absolute levels of CO in the Indian Ocean can be discerned from the south to the north. Daily, mean CO at Amsterdam Island (AMS, 37°S, 77°E [*Gros et al*., 1999]) and at Crozet Island (CRZ, 56.5°S, 51.9°E (P. C. Novelli et al., unpublished data, 1996); see section 5) shows a seasonal cycle of about 30 to 60 ppb, which is mostly driven by the seasonal cycle of OH but also indicates some pollution during the austral winter through outflow from the African continent and Madagascar. CO measured on Mahe in the Seychelles (SEY, 4.7°S, 55.2°E [*Novelli et al*., 1998]) varies from 50 to 150 ppb between seasons compared with CO at KCO between 50 and 240 ppb. The Seychelles encounter similarly enhanced CO in the wet season and high pollution during the dry season as KCO but are further away from the main source of pollution, the Indian subcontinent. Thus even in air from south of the ITCZ, CO maximum weekly means increase from 60 ppb (CRZ, AMS), to over 70 ppb (SEY), to 82 ppb (KCO), suggesting a positive gradient of CO to the north.

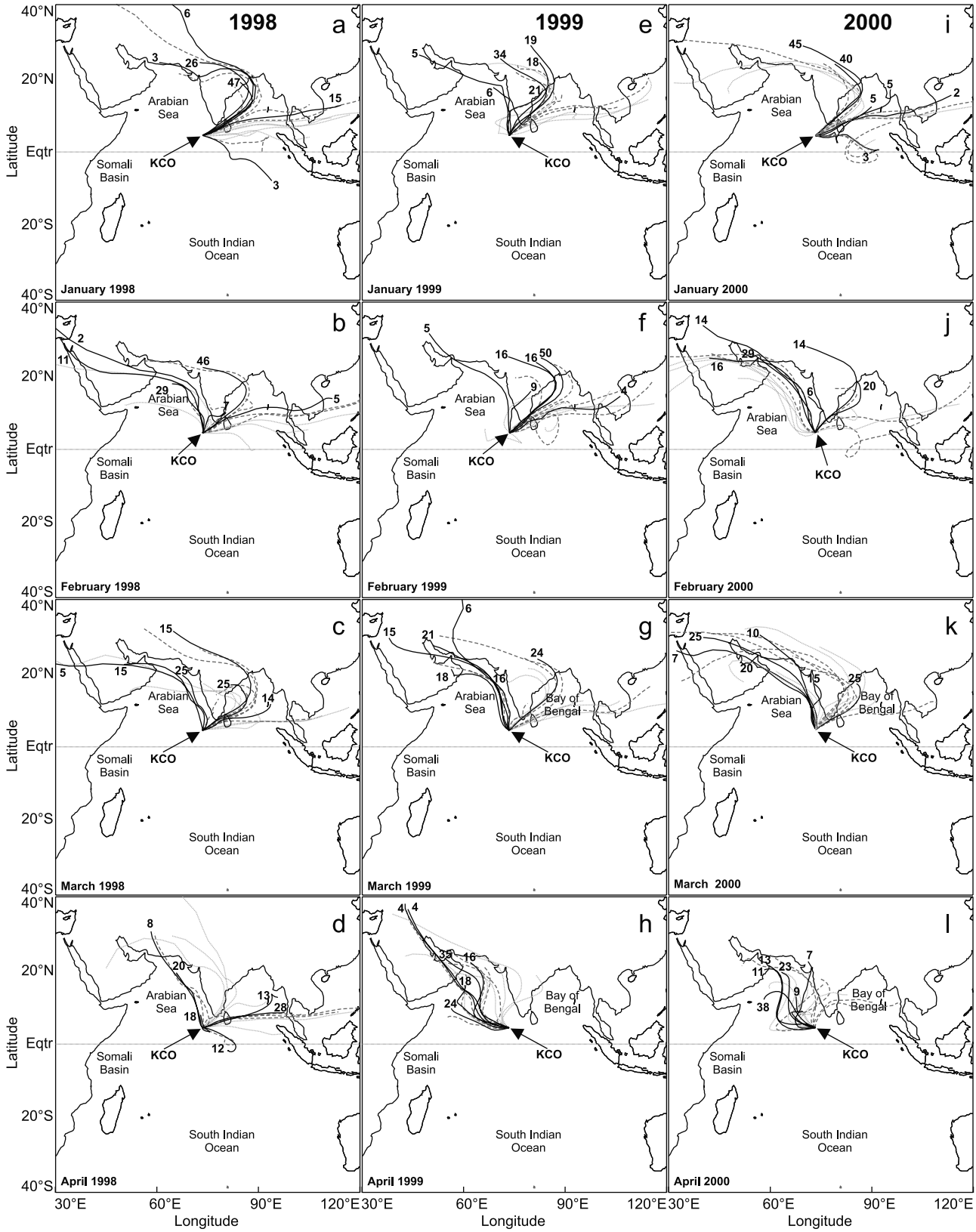

Figure 3. Monthly mean trajectory clusters for air masses arriving at KCO at 950 hPa/500 m (heavy solid lines), 850 hPa/1500 m (dashed lines), and 750 hPa/2500 m (light dotted lines) for the dry season months January through April of (a-d) 1998, (e-h) 1999, and (i-l) 2000. Numbers on the cluster starting points are percentages for the 500-m level during that month.

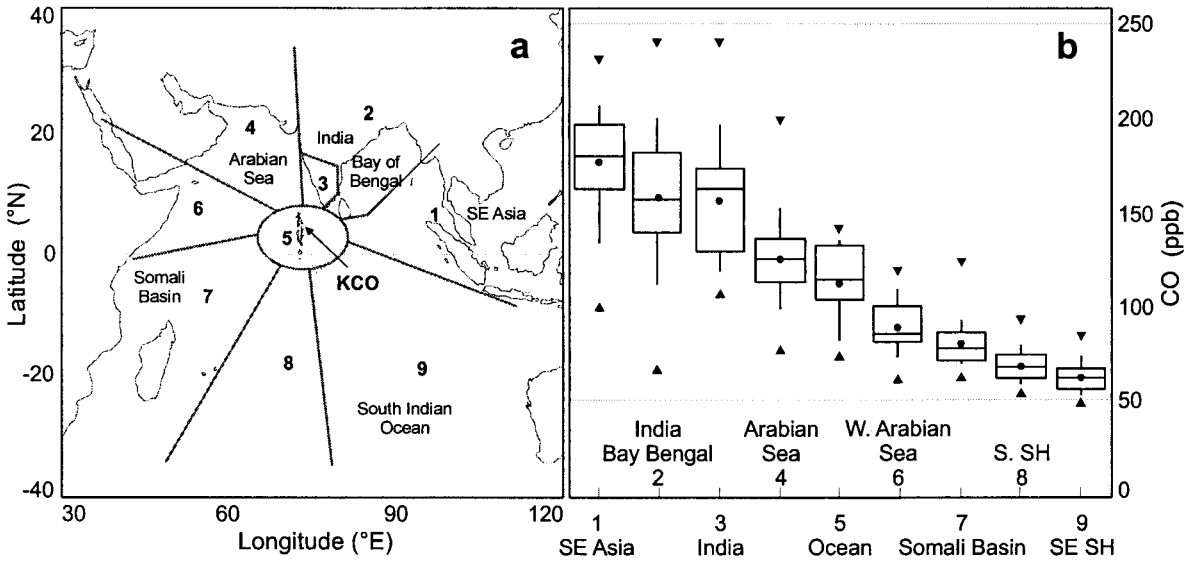

Figure 4. (a) Nine most important air mass flow bins for KCO and (b) daily mean CO mixing ratios distributed over those nine air mass flow bins. The boxes describe the upper and lower quartile, the solid dot is the mean, and the bar is the median. The whiskers are the 10% and 90% quantiles, and the black triangles are minima and maxima. Data are listed in Table 2.

Likewise, maxima in CO from north of the ITCZ increase from 150 ppb at SEY to 240 ppb at KCO.

3.2. Differences Between the Dry Seasons of 1998, 1999, and 2000

[27] It was shown that the ITCZ movement across the site defines the main seasons at KCO. The onset of each season typically extends over 1 to 3 months of time and can be identified by the highest variability in the changes of wind direction and respective air mass origin. The months with highest variability are, on average, November and December for the onset of the dry season, which peaks in January to March and includes most of April (not shown here). The core wet season from June to September is about 1 month longer than the dry season and does not start until the middle of May. However, considerable variability in the transition between seasons up to 1 whole month from year to year can be expected.

[28] Some variability in air mass origin and transport patterns and, accordingly, differences in the observed CO were seen between the years of 1998, 1999, and 2000. In particular, we were interested to see if the 1999 dry season was much different from the 11-year mean. Figure 5 contains CO data from the three dry seasons, plotted such that the start of the record is set to 1 October of each respective dry season; the end is the middle of June of the following year. We refer to the dry season of a year as the months of January to April of that year, including the last months of the previous year. The reference in Figure 5 is the dry season of 1999 (heavy solid line), which is also the only period that we fully covered with CO measurements. The dry season typically extends over 5–6 months and usually starts in October, when the ITCZ begins to move across the site. This transition, however, is not sharp, can vary in time and space, and can also revert several times; i.e., KCO is exposed to winds and air masses changing back and forth until the ITCZ clearly moves away from the site. The final transition from dry to wet season (when the ITCZ does not move back) consistently happened around the middle of May and was sharper during the observation period as the transition from wet to dry seasons, which usually extended over several months. The reversible ITCZ movement across the site explains the sharp drops in CO mixing ratios observed during the transition periods in November and December, whereas sharp gradients in CO during the core dry season can be caused

Table 2. Carbon Monoxide for Nine Air Mass Flow Bins According to Figure 4[a]

Bin	Definition	Mean	Median	Std. Dev.	Std. Err.	Min.	Max.	75%	25%	*N*
1	SE Asia	177	180	31	7	99	232	196	163	21
2	India/Bay of Bengal	158	156	35	3	65	240	182	139	125
3	India	156	159	33	6	106	241	173	129	28
4	Arabian Sea	126	125	22	2	76	200	135	112	162
5	Open ocean	112	114	21	5	73	141	130	102	19
6	W. Arabian Sea	89	85	14	2	60	119	99	81	51
7	Somali Basin	80	77	12	1	61	124	86	71	120
8	S. SH clean	68	67	8	1	53	93	74	61	105
9	E. SH very clean	62	61	8	1	48	84	67	56	98
All	Mean for 1998–2000	106	94	42.9	1.6	48	241	134	70	729

[a]Abbreviations are as follows: std. dev., std. err., min., max., 75%, and 25% are the standard deviation, standard error, minimum, maximum, 75% quantile, and 25% quantile values, respectively, all in ppb. *N* is the number of days used to derive these values.

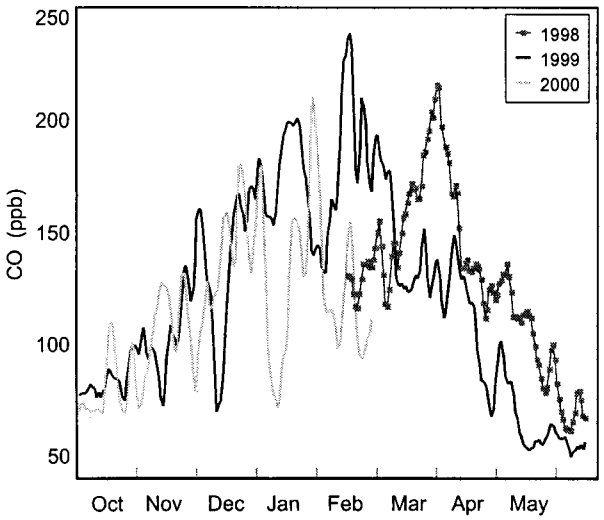

Figure 5. CO during the dry seasons of 1998, 1999, and 2000. Lines are running means through daily mean mixing ratios ($N = 3$).

by different air masses within the Northern Hemisphere or vertical mixing during cyclonic events [*Verver et al.*, 2001].

[29] High CO pollution (>140 ppb) can be seen in the record from November to April. The year 1999 had a rather extended dry season with high pollution levels on 87 days. The dry season of 2000 was initially very similar to what was observed in 1999, but CO mixing ratios dropped significantly starting in February and remained low until the end of the record and likely remained at moderate levels in March and April of 2000 (Figures 3k and 3l) as air masses originated from the Arabian Sea channel (representing moderate pollution) to 75% in March and 100% in April. Assuming that CO levels remained moderate, the 2000 dry season exhibited high CO (>140 ppb) on only 31 days.

[30] In contrast to both 1999 and 2000, the year 1998 showed a peak in CO pollution during April, when CO in both other years was only moderate. The transition to the wet season in 1998 was considerably steeper, and we observed CO pollution throughout May, whereas it tapered off earlier in 1999 (and presumably even earlier in 2000). CO measurements did not start until the middle of February 1998, but trajectory clusters of Figure 3 indicate that the pollution was probably high during January and early February of 1998 (Figures 3a and 3b) as air at KCO predominantly originated from within India, with some contribution from SE Asia. The minimum number of days with high CO levels was then 37 during the observation period in 1998, but likely much higher for the entire dry season.

[31] Daily, maximum CO mixing ratios in 1999 were similar in magnitude compared with 1998 and 2000 and exceeded both other years only by about 10–20 ppb (Figure 5). However, the extent of the dry season pollution was, on average, significantly longer than that in 2000 and likely also somewhat longer than that in 1998. In terms of prevalent air mass transport, 1999 was similar to the 11-year, monthly means in January and April but deviated somewhat in February and March with significant shifts between the Indian and Arabian Sea transport channels: more air from India in February (91% of all days in 1999 versus 70% long term) and twice the contribution from the Arabian Sea than the long-term mean in March (76% versus 32%). Those anomalies did probably not change the mean dry season impact

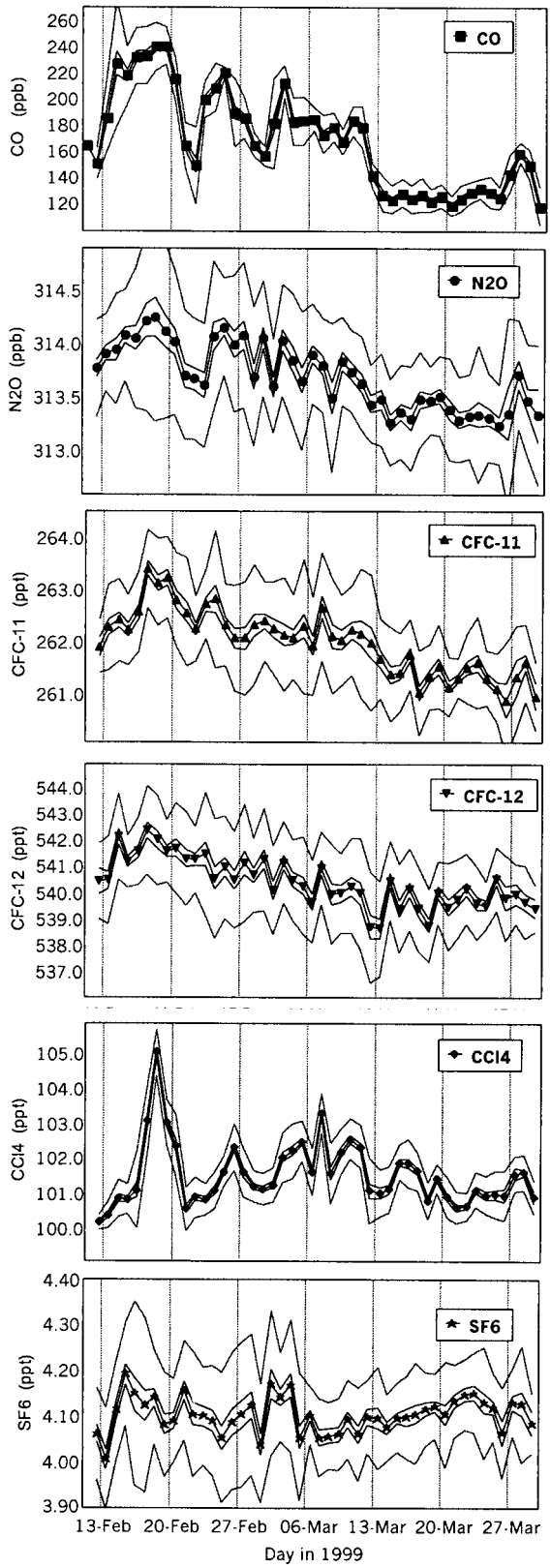

Figure 6. Time series of daily mean mixing ratios of surface CO, N_2O, CFC-11, CFC-12, CCl_4, and SF_6 at KCO during the INDOEX IFP. The center line connects the means to guide the eye, the outer thin lines describe the 90% confidence levels ($\pm 1\sigma$), and the thin lines closer to the means are ± 1 standard error.

Table 3. Measurement Statistics for all Gases for the Dry Season of 1999 at KCO

Gas and Unit	Range[a]	Mean[b]	Standard Deviation[b]	Standard Error[b]	Minimum[b]	Maximum[b]	N[b]
CO, (ppb)	110–245	169	40.8	1.2	84.4	290.3	1194
N_2O, (ppb)	313.3–314.3	313.7	0.73	0.02	310.4	317.1	1261
CFC-11, (ppt)	260.9–263.4	262.1	1.19	0.03	257.9	266.2	1231
CFC-12, (ppt)	538.7–542.4	540.5	2.48	0.07	532.2	553.6	1250
CCl_4, (ppt)	100.2–105.1	101.6	1.14	0.03	98.8	106.5	1247
SF_6, (ppt)	3.91–4.10	4.11	0.13	0.004	3.66	4.93	1263

[a]Range of daily means during the Indian Ocean Experiment Intensive Field Phase (12 February to 29 March 1999).
[b]Statistical data calculated from individual, hourly measurements for the entire period.

on the region but significantly increased the variability and ultimately the value of observations during the INDOEX IFP.

3.3. CFCs, CCl_4, N_2O, and SF_6

[32] Measurements of halogenated compounds and nitrous oxide (N_2O) were carried out between 12 February and 29 March, 1999 (Figure 6); measurement statistics are given in Table 3. Variations in the mixing ratios of these compounds were very subtle in comparison with CO and usually well within the measurement precision, but some of the major features observed in CO can also be seen in the records of long-lived gases. Particularly N_2O and CFC-11 follow the pronounced maximum, sharp drop, and subsequent rise in pollution during the middle of February as well as the overall trend from higher to lower pollution between the 2 months that was caused by the change in air masses described earlier.

[33] CCl_4 shows a somewhat different behavior; its mixing ratio does not change much during the IFP except for the large pollution event encountered at KCO between 16 and 21 February and maybe a small decrease after 6 March caused by less polluted air from the Arabian Sea. The air during that particular pollution event came from India through the Bay of Bengal channel and was crossing the Calcutta metropolitan region. However, that was also the case for most days in February. We have to assume that air from this period contained unusually large amounts of CCl_4, which otherwise may not be produced or emitted in large quantities in India at all times. However, the occurrence of CCl_4 amounts this high indicates that this compound is still in use in India, whereas it was phased out for any use in developed countries. Local contamination of measurements can be ruled out, as CCl_4 was not used at KCO at any time and the extent of this event over 4 days makes emission from inside the Maldives or ships around the islands unlikely. SF_6, finally, does not show any significant features during the measurement period; its mixing ratio remained constant at about 4.1 parts per trillion (ppt).

[34] Correlations of daily mean mixing ratios with those of CO are reasonably good for N_2O, CFC-11, and CFC-12 (Table 4) and indicate that these gases all have common source regions and regional distributions. The Indian subcontinent seems to be a moderate source for these gases as mixing ratios all drop off with the change of air masses away from the continent after 11 March. CFC-12 does not correlate as well with CO as CFC-11, which could be due to a smaller source in India. However, this difference may also be, in part, due to different emission patterns or seasonally variable emissions. Both CFC-11 and CFC-12 (as well as CCl_4) are still being used in developing countries in accordance with the Montreal Protocol and its amendments [*United Nations Environment Programme*, 1999]. However, CFC-12 is mainly used in refrigerators and air conditioners [*Alternative Fluorocarbon Environmental Acceptability Study* (*AFEAS*), 1993], which are not as common in India as in more industrialized countries; hence consistent leakage from this source is probably low. CFC-11, on the other hand, is mainly used as a foam blowing agent and for other industrial uses [*AFEAS*, 1993] and consistent outgasing in India is more likely for CFC-11 than for CFC-12. Emitted amounts for CFC-11 and CFC-12 were reported by *McCulloch et al*. [1994] to be rather low at 5000 and 7000 tons, respectively. However, it seems that these reported emissions may be associated with large error bars and also underestimated, as they are based solely on publicly available data (A. McCulloch, Marbury Technical Consulting, personal communication, 2000).

[35] The good correlation of N_2O with CO is somewhat surprising, as it indicates similar sources and, hence, a dominant continental and anthropogenic signal for both gases. Atmospheric N_2O originates mainly from natural sources such as denitrification in soils and outgasing from the ocean, most prominently in upwelling regions of the oceans. Anthropogenic production is thought to be mainly for fertilizer use [*Albritton et al*., 1999], and globally, fossil fuel and biomass burning contribute only a minor portion. The Indian Ocean does have large areas of strong upwelling and a significant signal of N_2O emissions has been reported [*Bange et al*., 1996]. However, the very good correlation with CO, a good tracer for biomass burning, indicates that significant amounts of atmospheric N_2O in the remote Indian Ocean may originate from within India. This is supported by the change from higher to lower N_2O mixing ratios from February to March, when KCO encountered a change in air masses away from the Indian continent. Even though biomass burning globally does not constitute a large source for N_2O, *Mitra et al*. [1997] reported significant, albeit very uncertain, emission estimates of pyrogenic N_2O. In addition to direct emissions, N_2O can also be produced in sulfur-rich burning plumes during their transport to the open ocean [*Muzio and Kramlich*, 1988]. However, *Parashar et al*. [1998] reported that most N_2O emissions in India are from fertilizer applications. Either way, the measurements suggest that most of the enhanced N_2O signal at KCO seems to be from anthropogenic activity in India and not of natural origin.

Table 4. Correlations of Trace Gases With CO[a]

Gas	Slope	Intercept	r^2	R	N
N_2O	7.10×10^{-3}	312.5	0.79	0.89	46
CFC-11	1.38×10^{-2}	259.7	0.72	0.85	46
CFC-12	1.83×10^{-2}	537.3	0.59	0.77	46
CCl_4	1.17×10^{-2}	99.6	0.24	0.49	46
SF_6	4.24×10^{-5}	4.1	0.002	0.04	46

[a]Variables r^2, R, and N are the variance, correlation factor, and number of daily means used for the least squares fits, respectively.

[36] SF_6, finally, is mainly produced for use in transformers, from which it slowly leaks to the atmosphere, and for some other minor industrial applications [*Geller et al*., 1997]. The complete lack of a pollution signal and correlation with any of the other gases measured at KCO suggests that India does not emit substantial amounts of this gas. SF_6 was also measured in flasks taken at KCO by NOAA/CMDL (unpublished data courtesy of *E. Dlugokencky*, NOAA/CMDL (1999)). These data were in excellent agreement with our measurements; the mean deviation was 1.2%, or 0.05 ppt.

[37] All other data were compared with NOAA/CMDL measurements taken at the Mauna Loa (22°N) and Samoa (15°S) observatories (unpublished data courtesy of J. W. Elkins et al. (1999); see section 5 for data sources). We calculated the mean between those two stations for the months of February and March 1999 and compared it with an average of the lower mixing ratios at KCO toward the end of March (less polluted air). However, these values may not be comparable because Samoa is in the Southern Hemisphere and variations in the location of the ITCZ may introduce some incompatibility. Also, distribution and mean mixing ratios are likely to be somewhat different in the Pacific and Indian Oceans, at least during the dry season. Using these reference points, data collected at KCO were in reasonable agreement with CMDL station data: deviations of N_2O, CFC-11, CFC-12, and CCl_4 data from the CMDL means were −0.5% (1.4 ppb), 1.1% (5 ppt), −1.9% (5.8 ppt), and −0.1% (0.1 ppt), respectively, all within or close to the uncertainty of KCO values.

3.4. Correlations of CO With Aerosol Properties

[38] Considering the high pollution levels of both trace gases and aerosols in the Indian Ocean [*Lelieveld et al*., 2001; *Ramanathan et al*., 2001] the question arises if one measurement can be used as a proxy for the other. Aerosol optical depth (AOD) was measured through AERONET (section 5) with a CIMEL photometer [*Eck et al*., 2001]. To a first approximation, 500-nm AOD and surface CO generally seemed to track each other throughout the year (Figure 7a, $r^2 = 0.31$), but there is considerable scatter diminishing the usability of this correlation. A calculation of least squares fits at all seven wavelengths of the CIMEL instrument using daily means during 1999 produces an anticorrelation between the regression coefficient of AOD and CO versus the wavelength with the best correlation at lowest wavelengths, corresponding to smaller aerosol particles (Figure 7b). This trend with aerosol size is not too surprising, as typical transport times of 6–10 days from the pollution sources to KCO can give rise to possibly significant alterations in aerosol properties, e.g., through dry and wet deposition, more pronounced for larger than for smaller particles. Small particles may also change in time during transport, owing to aging processes such as coagulation, condensation, and gas-to-particle conversion. Some fine mode particles may also be hygroscopic and increase in size at high relative humidity. In addition, the lower correlation between AOD and CO at longer wavelengths may be due in part to the much larger contribution of coarse mode aerosol, such as dust, to the AOD at longer wavelengths (especially 675, 870, and 1020 nm). The AOD at shorter wavelengths (<500 nm) is dominated by fine mode particles which are more strongly associated with the same anthropogenic processes which produce CO [*Eck et al*., 2001]. CO, on the other hand, has a lifetime of months and its mixing ratio does not change appreciably within 6–10 days. However, even the best fit at 340 nm is poor ($r^2 = 0.40$) and cannot be considered for use as a simple proxy for CO pollution or vice versa. We found similar correlations for both 1998 and 1999.

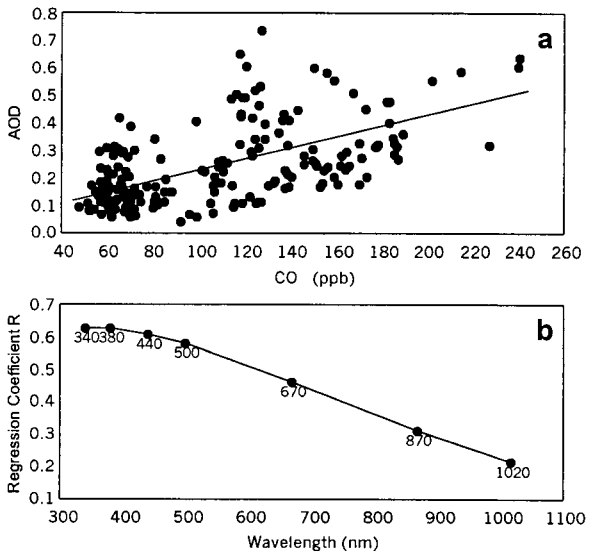

Figure 7. (a) Daily means of aerosol optical depth at 500 nm correlated to surface CO for 1999. (b) Regression coefficients of AOD versus CO for seven wavelengths between 340 and 1020 nm. AOD data courtesy of NASA Goddard Space Flight Center (GSFC) [*Eck et al*., 2001].

[39] In addition, AOD is a column-integrated measurement, which considers all aerosols that typically extend up to 5- to 7-km altitude, whereas CO was measured at the surface and does not usually represent more than the first 500 m of altitude. The generally low correlation demonstrates that both measurements are not necessarily comparable, at least over extended periods of time. A much better correlation with CO was achieved by using the surface aerosol scattering and absorption coefficients, instead of AOD, both of which are measured from the same location and air inlet height (Figures 8b and 8c). This confirms (1) that pollution-level trace gases and aerosols at the surface have the same origin and can probably be used as a proxy for each other in this location and season and (2) that surface pollution measured in the Maldives is not always indicative of column-integrated pollution due to the stratification of the troposphere.

[40] At higher altitudes, the air mass composition was, indeed, significantly different from that at the ground. In February 1999, air masses arriving at KCO at both surface and higher altitudes originated mainly from India (Figure 3f). In contrast, most air masses arriving at 500 m during March originated from the Arabian Sea, whereas those arriving at 1500 and 2500 m still came from India/Bay of Bengal (Figure 3g). This explains the drop in CO mixing ratios at the surface from February to March and the persistent pollution layers found in CO data from aircraft measurements at higher altitudes [*Williams et al*., 2002]. J. M. Lobert and J. M. Harris (unpublished manuscript, 2000, see section 5 for more data on INDOEX) also show persistent layers of heavy O_3 pollution around 1–4 km altitude during March 1999. Such layers would significantly increase column pollution (and hence AOD) but would not be detected at the surface. The correlation of AOD with CO in Figure 3a is, indeed, much better if calculated only for the month of February, where significant layers of heavy pollution aloft were not observed and the surface aerosol dominated the AOD measurements.

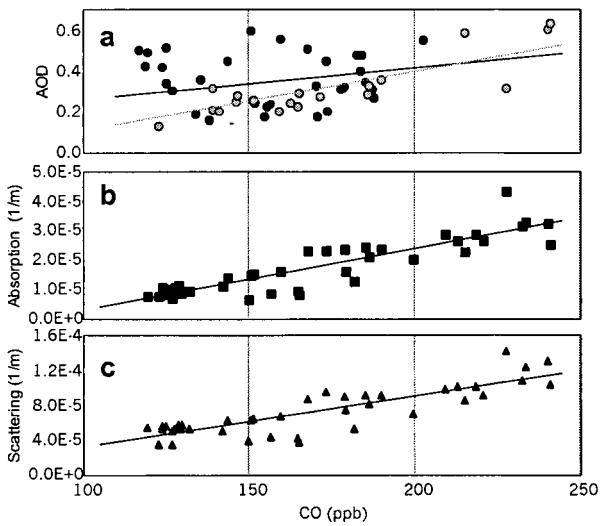

Figure 8. Daily mean aerosol properties versus CO during the 1999 IFP: (a) AOD February, (gray circles) and March (black circles), (b) absorption coefficient (black squares), and (c) scattering coefficient (black triangles). AOD is for 500 nm; scattering and absorption, for 565 nm. The solid lines are least squares fits to the full data pairs; the light dotted line in Figure 8a is for February AOD only. Aerosol optical depth courtesy of B. Holben, NASA/GSFC [*Eck et al*., 2001]; scattering and absorption data courtesy of NOAA/CMDL (J. Ogren et al., unpublished manuscript, 2000; A. Jefferson et al., unpublished manuscript, 2000). See Table 4 for correlation data.

[41] *Lelieveld et al*. [2000] and *Eck et al*. [2001] reported that, on average, AOD increased between January and March 1999, whereas CO showed an increase only from January to February but then decreased significantly in March. Judging from O_3 mixing ratios from soundings, elevated layers in March appeared to carry highly polluted air from within India (J. M. Lobert and J. M. Harris, unpublished manuscript, 2000, see section 5 for more data on INDOEX). Furthermore, dust particles, which made up about 15% of the AOD signal in March [*Satheesh and Ramanathan*, 2000] were much lower during February (D. Savoie et al., unpublished manuscript, 1999). Taken together, this can, in part, explain the observed anticorrelation between AOD and CO during the period of February to March 1999.

4. Summary and Conclusions

[42] This study gives an overview of the regional CO pollution in the Maldives between February 1998 and March 2000 with emphasis on the dry seasons of these years. Carbon monoxide mixing ratios showed a distinct annual cycle of 50–240 ppb with a mean of 107 ppb for the study period, indicating urban-level pollution in the remote Indian Ocean during the dry seasons. Trajectory analysis reveals that most of the significant changes in CO mixing ratios can be attributed to changes in air mass origin and that CO can be used to broadly identify the sources of air masses. We assigned nine different bins of possible air mass origin and associated CO pollution levels at KCO to each bin. Simplifying this scheme, air measured at KCO appears to be highly polluted (CO $>$ 140 ppb) when air originates from within India or SE Asia, moderately polluted (140 ppb $>$ CO $>$ 80 ppb) from the Arabian Sea and Somali Basin, and rather clean (CO $<$ 80 ppb) from the Southern Hemisphere with roughly equal contributions from each of these regions throughout each year. We showed that significant changes in CO can be caused through the change in air mass proximity to the continents and through meteorological events such as cyclones and vertical mixing. We also found that air from the Southern Hemisphere seems to be significantly impacted most of the time and exhibits CO mixing ratios that are much higher than expected for pristine environments.

[43] Considering (1) the pollution levels found in the northern Indian Ocean, (2) the significantly enhanced CO levels from the Somali Basin, (3) pollution events reported by other investigators in the midlatitude Southern Hemisphere, and (4) the transport of pollution from Africa across the southern Indian Ocean to Australia, we conclude that the remote Indian Ocean cannot be considered pristine anymore at all times.

[44] CFCs and N_2O correlated well with CO during the dry season of 1999 and confirm the outflow of heavy pollution from India, which seems to be a significant source for these compounds. In contrast, SF_6 does not appear to be emitted in significant quantities from India or any other region surrounding KCO. Correlations of CO with aerosol properties illustrate that CO can be used as a proxy for surface aerosol loading in this region but not for column aerosol abundance due to a heavy stratification of the troposphere at times.

[45] Long-term measurements from within the Maldives cover a unique mixture and wide range of air mass source regions and will prove to be very useful as baseline measurements for models and future investigations in a region that is increasingly affected by heavy pollution. The comparison of heavily polluted and quasi-pristine air masses enables a unique assessment of the effect of aerosols and trace gases on regional and global climate.

5. Further Resources

[46] Data from this publication are available at http://www.joss.ucar.edu/indoex/. Information on KCO can be found at http://www-indoex.ucsd.edu/observatory/, and information for INDOEX, at http://www-indoex.ucsd.edu/. More information on data, projects, and publications is at http://Jurgen Lobert.org/projects/indoex/. The AERONET website for CIMEL data can be accessed at http://aeronet.gsfc.nasa.gov:8080/, and the NOAA/CMDL data archive is at http://www.cmdl.noaa.gov/info/ftpdata.html. Information about the NOAA/CCCG cooperative air sampling network is published at http://www.cmdl.noaa.gov/ccgg/flask/, information about trajectory analysis is at http://www.cmdl.noaa.gov/ozwv/traj/.

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