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Shipboard and Satellite Views of Elevated Tropospheric Ozone over the Tropical Atlantic in January-February 1999

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Abstract.

During the Aerosols99 trans-Atlantic cruise from Norfolk, VA, to Cape Town, South Africa, daily ozonesondes were launched from the NOAA *R/V Ronald H Brown* between 17 January and 6 February1999. A composite of tropospheric ozone profiles along the latitudinal transect shows 4 zones, which are interpreted using correlative shipboard ozone, CO, water vapor, and overhead aerosol optical thickness measurements. Elevated ozone associated with biomass burning north of the ITCZ (Intertropical Convergence Zone) is prominent at 3-5 km from 10-0N, but even higher ozone (100 ppbv, 7-10 km) occurred south of the ITCZ, where it was not burning. Column-integrated tropospheric ozone was 44 Dobson Units (DU) in one sounding, 10 DU lower than the maximum in a January-February 1993 Atlantic cruise with ozonesondes [Weller et al., 1996]. TOMS tropospheric ozone shows elevated ozone extending throughout the tropical Atlantic in January 1999. Several explanations are considered. Back trajectories, satellite aerosol observations and shipboard tracers suggest a combination of convection and interhemispheric transport of ozone and/or ozone precursors, probably amplified by a lightning NO source over Africa.

1. Introduction: Tropical tropospheric ozone from satellites & sondes

Tropospheric ozone over the tropical Atlantic is a seasonally varying feature, which can be tracked from satellite with moderately high spatial (1x2 deg) and temporal (5-15 day means) resolution. The 14-year Nimbus 7/TOMS satellite record [Thompson and Hudson, 1999] reveals periods of pollution-level ozone (> 30 DU, column integral) occurring over the south tropical Atlantic outside the well-known September-October maximum [Fishman et al., 1991].

Confirmation of high-ozone episodes in January and February is limited to the 1990-1992 and 1998-1999 Ascension Island (8S, 15W) ozone soundings [Thompson and Witte, 1999]. Ships offer a moving platform for ozonesonde launches [Smit et al., 1989; Weller et al.,1996] that extend coverage for satellite comparison. The Aerosols99 cruise on the *R/V Ronald H Brown*, from Norfolk, Virginia (37N, 76W) to Cape Town, South Africa (34S, 22E) in January-February 1999 provided the first shipboard ozone soundings from North America to southern Africa, as well as a suite of chemical tracer data [Parsons and Dickerson, 1999]. The route included a sector downwind of northern equatorial biomass fires and a southeast Atlantic segment previously sampled only during the September-October 1992 ozone maximum [Fishman et al.,1996; Thompson et al., 1996].

The most remarkable finding was that although northern equatorial biomass burning led to elevated surface ozone and CO north of the ITCZ, the highest tropospheric ozone was south of the ITCZ, at 7-10 km altitude. Ozone, water vapor, CO and overhead aerosol measurements from the *R/V Ronald H Brown* are used with back-trajectories and satellite data to elucidate chemical and dynamical influences on tropospheric ozone during the cruise. The mechanism causing the southern hemisphere (SH) ozone maximum appears to be a combination of interhemispheric advection with convection [Jonquières et al.,1998], possibly augmented by NO from lightning, but stratospheric and biogenic influences cannot be ruled out.

2. Measurements and Methods. Remote Sensing Data.

Ozone and temperature profiles on the *R/V Ronald H. Brown* were determined with an electrochemical concentration cell ozonesonde (ENSCI 2Z) in combination with a RS-80/15 Väisäla radiosonde and a HumiCap humidity sensor; 1-sec data were recorded. The procedures for sonde preparation and data acquisition were developed by NOAA/Climate Diagnostics and Monitoring Laboratories [Komhyr et al, 1989]. All but four of 22 launches reached 30 km. Launches were from 30N to 30S (17 January to 6 February 1999).

Marine boundary layer (MBL) ozone and shipboard CO were measured with a commercial UV photometer (TEI model 49) and a modified commercial nondispersive infrared gas filter correlation instrument (TEI model 48C), respectively [Parsons and Dickerson, 1999]. Hourly averages of 1 minute data are reported here. AOT (aerosol optical thickness) was measured using a multichannel sun photometer (Solar Light Co.) on days for which midday sun was uninterrupted by clouds.

TOMS tropospheric ozone data between 20N and 20S are provided by near-real-time processing at the University of Maryland and NASA/Goddard [http://metosrv2.umd.edu/~tropo]. Absorbing aerosol gridded data are from the TOMS archive. Lightning flash counts were obtained from the Lightning Imaging Sensor (LIS) aboard the Tropical Rainfall Measuring Mission (TRMM) satellite.

Back trajectories were run with the GSFC isentropic trajectory model initialized from the ozonesonde launch locations. NCEP analyses from standard 2.5x2.5deg 0 and 12Z fields were used in the model. These compared favorably to winds from shipboard radiosondes. To look at shipboard tracer origins, air parcels were followed along potential temperature surfaces at 1 and 4 km altitudes for 8 days. Mid- and upper tropospheric (UT) air parcel origins were computed at 8 and 11 km.

3. Ozonesonde Latitudinal Cross-section with Four Zones.

Figure 1 shows a latitudinal cross-section of tropospheric ozone derived from the Aerosols99 sondes. Four distinct ozone profile types appeared along the transect, roughly coincident with variations in other tracers [T. S. Bates, Personal Communication, 1999].

➳ Zone 1: Northern hemisphere (NH) mid-latitude air of mixed troposphere-stratospheric origins. UT ozone >/~ 100 ppbv on the first launch (30.6N, 66W, 17 January 1999) coincided with low water vapor traceable back to high latitudes.

➳ Zones 2 and 3 (14N to 23S). A broad band of high column ozone straddling either side of the ITCZ, which was located at 0-5S (Figure 1). In Zone 2 (14N - 0), surface ozone and CO are highly correlated, with ozone at 42 ppbv when CO jumps to > 200 ppbv (Figure 2). Ozone peaks > 60 ppbv at 3-5 km (Figure 3a) appeared at the same location as elevated CO and AOT (Figure 2). Zone 3 (0-23S) denotes a shift to a clean MBL with maximum ozone in the mid-troposphere (Figure 2), up to 100 ppbv (Figure 3B).

➳ Zone 4: Subtropical SH (23-30S). The 5-10 km ozone peaks diminished to typical background tropospheric values. Surface data and sondes show MBL ozone to be 10-20 ppbv. Surface CO of 45-50 ppbv is consistent with remote SH air quality.

4. Interpretation of Tropospheric Ozone in Zones 2 and 3

The high ozone mixing ratio layers of Zones 2 and 3 translate into high column ozone

which TOMS (Figure 4A) shows to be a large regional feature, extending from South America to Africa and 5N to 15S. Integrated tropospheric ozone in Zone 2 ranges from 32-39 DU and from 37-44 DU in Zone 3. Tropospheric ozone from the sondes and from TOMS tropospheric ozone are within experimental error of the modified-residual method ($\pm$ 4 DU), with the sondes serving as validation for the satellite measurement.

Figures 2 and 4B suggest an explanation for the 2-5 km high ozone mixing ratios in Zone 2. CO and AOT (values in Figure 2 caption) co-vary according to the influence of biomass burning. Pronounced increases in CO and AOT occur at 14N (23 January) and at 8N (25 January). AVHRR satellite observations identify the first jump with a transition from background aerosol to mixed dust and smoke and the second increase to an all-smoke regime (P. Durkee, personal communication, 1999). Other tracers not shown (e.g. elevated aerosol K, black carbon, nitrate) also signify biomass burning from in Zone 2. As a result, ozone at 2-5 km increases from 60 ppbv (24 January) to 90 ppbv (26 January).

The link between biomass burning and high ozone layers in Zone 2 is evident from satellite. Back trajectories initialized from the sonde launch positions at ~4 km pass over regions of biomass burning from 23-27 January (3rd-7th white dots in Figure 4B) as given by a proxy, TOMS absorbing aerosol. Previous investigations [Thompson et al.,1996; Jonquières et al.,1998] show ozone photochemical formation over African burning regions allowing ozone to accumulate to 35-45 DU. This is 10-20 DU greater than background Atlantic ozone [Hudson and Thompson, 1998].

What happened during the Zone 2-Zone 3 transition? On 26-27 January, CO and surface ozone (0-5N, Figure 2) reached their maxima when convection mixed polluted air from aloft toward the surface. During the encounter with the most intense ITCZ activity (28 January, ~1S), strong winds destroyed the ozonesonde at ~600 hPa. The incomplete trace was nearly uniform, indicating that lower tropospheric ozone (and presumably ozone precursors) were transported upward. From the ITCZ, air parcels maintain relatively constant altitude (layers in Figure 3B) as they are advected from NE to SW (Figures 4B, C), as suggested by the shape of the highest ozone pool in Figure 4A.

Thus, the ITCZ acts as a mechanism for interhemispheric transport by pumping low-level ozone from the NH to higher altitudes in the SH. This is apparent in Zone 3. The highest ozone mixing ratios are above 5 km (Figure 3B), where individual layers of 90-105 ppbv ozone persist from 29 January through 1 February. Figure 4B shows 4-km air parcels for these dates (white dots 7-10) intersecting high-smoke aerosol (index > 0.5) regions. Ozone enhancements also appear to result from lightning, which injects NO directly into the middle and upper troposphere [Thompson et al., 1996]. Superposition of ~8 km back trajectories initiated from sonde locations at which ozone > 60 ppbv (dots 9-12 in Figure 4C) shows parcels intersecting African areas over which lightning is detected. The highest-ozone layer in Figure 1 (0-16 S) is typically associated with convective cloud outflow.

Other processes may contribute to elevated free tropospheric ozone in Zone 3. (1) Soil release of NO over southern African savannas following precipitation has been observed [Harris et al., 1996; Swap et al., 1996]; convective transport of such NO could supply mid-tropospheric ozone. (2) Folkins et al. [1999] pointed out signs of stratospheric influence at14-17 km in Samoan ozonesondes. These may explain Zone 3 ozone peaks above 12 km.

5. Discussion & Conclusions

The only previous Atlantic cruise ozonesonde data for January and February, is by

Weller et al [1996] during a 1993 *R/V Polarstern* transect from South America to Europe. They show a pattern similar to that on the *Ronald H Brown*. Tropospheric ozone was markedly greater in the SH (>50 DU for two soundings) than in the NH. A March-April 1987 South America to Europe *Polarstern* transect [Smit et al.,1989] also showed ozonesonde maxima south of the ITCZ at 8-10 km, whereas north of the ITCZ, downwind of northern African biomass burning, peak ozone occurred at 3-6 km. Maximum SH free tropospheric ozone (and other tracers) were observed from aircraft over the tropical Atlantic in January 1991 [Jonquières and Marenco, 1998]. As for the Aerosols99 cruise, Jonquières et al. [1998] showed northern African burning interacting with convection over South America and Africa. Layers of high ozone mixing ratio at 5-10 km appear in Ascension Island ozonesondes in January 1998 and 1999 [Thompson and Witte, 1999] and in some January and February 1990-1992 Natal, Brazil (6S, 35W) and Ascension profiles [Thompson et al.,1996; Hudson and Thompson, 1998].

In summary, the first northwest-to-southeast Atlantic cruise with ozone profiles, tracer and radiative measurements (January-February 1999), displayed four zones of ozone profiles. The most striking feature was elevated SH tropospheric ozone, which satellite observations show to be a large regional feature. Tropospheric column ozone at 1-20S was 38-44 DU, in agreement with TOMS tropospheric ozone. Shipboard tracers, trajectories and satellite observations suggest that mid-tropospheric ozone maxima are modulated by biomass burning, convection, cross-hemispheric transport and lightning.

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Figure Captions.

Fig. 1. Cross-section of tropospheric/ lower stratospheric ozone mixing ratio based on 22 soundings taken aboard R/V R H Brown between 17 January and 6 February 1999, usually at 0900-1300Z. 1 km averages used; all but 3 soundings reached 10 hPa. Four zones: 1: 30-14N (17-22 Jan.); 2, 14N-0 (23-27 Jan); 3, 0-23S (ITCZ, and SH subsidence, 28 Jan - 3 Feb). 4: 23-30S (subtropical, "background" ozone profiles).

Fig. 2 Time-series of tracers along *R/V Ronald H Brown* track. Surface ozone and CO (x1/2) mixing ratios in ppbv, from ship measurements; 0-5 km, 5-10 km mean ozone from sondes. AOT (at 380 nm) from midday sun photometer observations on cloud-free days shows dust and smoke influence in Zone 2 and ITCZ segment. Zone 1 mean, 19-22 Jan.: 0.102.

Date	23 Jan	24 Jan	25 Jan	26 Jan	27 Jan	28 Jan	29 Jan	30 Jan	31 Jan	1 Feb.	2 Feb.	3 Feb
Lat.	14N	11.3N	8.5N	5.5N	2.25N	1.6S	5.3S	7.6S	11S	14S	17S	20S
AOT	0.34	0.25	0.36	n/a	0.45	n/a	0.26	0.15	(stratus cloud)		0.12	S0.10

Fig. 3 (A, B) Ozone (ppbv, mixing ratio) for Zones 2 (23-27 January 1999) and 3 (28 January-3 February). Origins of Zone 2 2-5 km ozone peaks traced to biomass burning (Fig 4B). Individual profiles (black) plotted along with 0.25 km interval means of all profiles (red). Near zero mixing ratio in one Zone 3 profile at 0-0.4 km occurred when sonde passed through *Ronald H Brown* stack plume. Zone 3: high ozone at 8-10 km could also originate from lightning NO (Fig 4C). Ozone profiles at Ascension, January-February 1999, and Natal, Brazil (not shown) have mid-tropospheric ozone maxima similar to those in Zone 3.

Fig 4. (A) TOMS tropospheric ozone, averaged over January 1999, with integrated tropospheric ozone from the sondes shown in white. Sondes on 29 January not used due to incomplete profiles (cf Fig. 1, 1-5S). Comparisons only valid where modified-residual method is reliable, in equatorial air, from ~5N to 20S. Gaps in modified-residual tropospheric ozone correspond to regions of persistent clouds, where TOMS data are not used; compare lightning locations in (C). (B) Eight-day back trajectories from position of ozonesonde launches (21 Jan.-6 Feb.) along *R/V R H Brown* cruise track, initialized at ~4 km; TOMS aerosol index averaged from 23-30 Jan. 1999, in false color. Aerosol over Africa from 15-8N due to dust. From 8N-5S aerosol corresponds to smoke [P. Durkee, personal communication, 1999]. Back trajectories from the 3-5 km high ozone peaks in Fig. 3A. (C) same as (B) except for trajectories at ~ 8 km, superimposed over total LIS lightning flashes for 28 Jan.-4 Feb. 1999. Clusters of back trajectories were run to capture uncertainties associated with analyzed winds (NCEP 2.5 x 2.5 deg) in this case. Line shown corresponds to weighted central point in each cluster. Some of the more southerly points included some So. American parcel origins at 8 km. Time of trajectory initiation is 12Z. Model was also run with parcels at 11 km. In Zone 3, 11-km origins clustered at the ITCZ over high aerosols.

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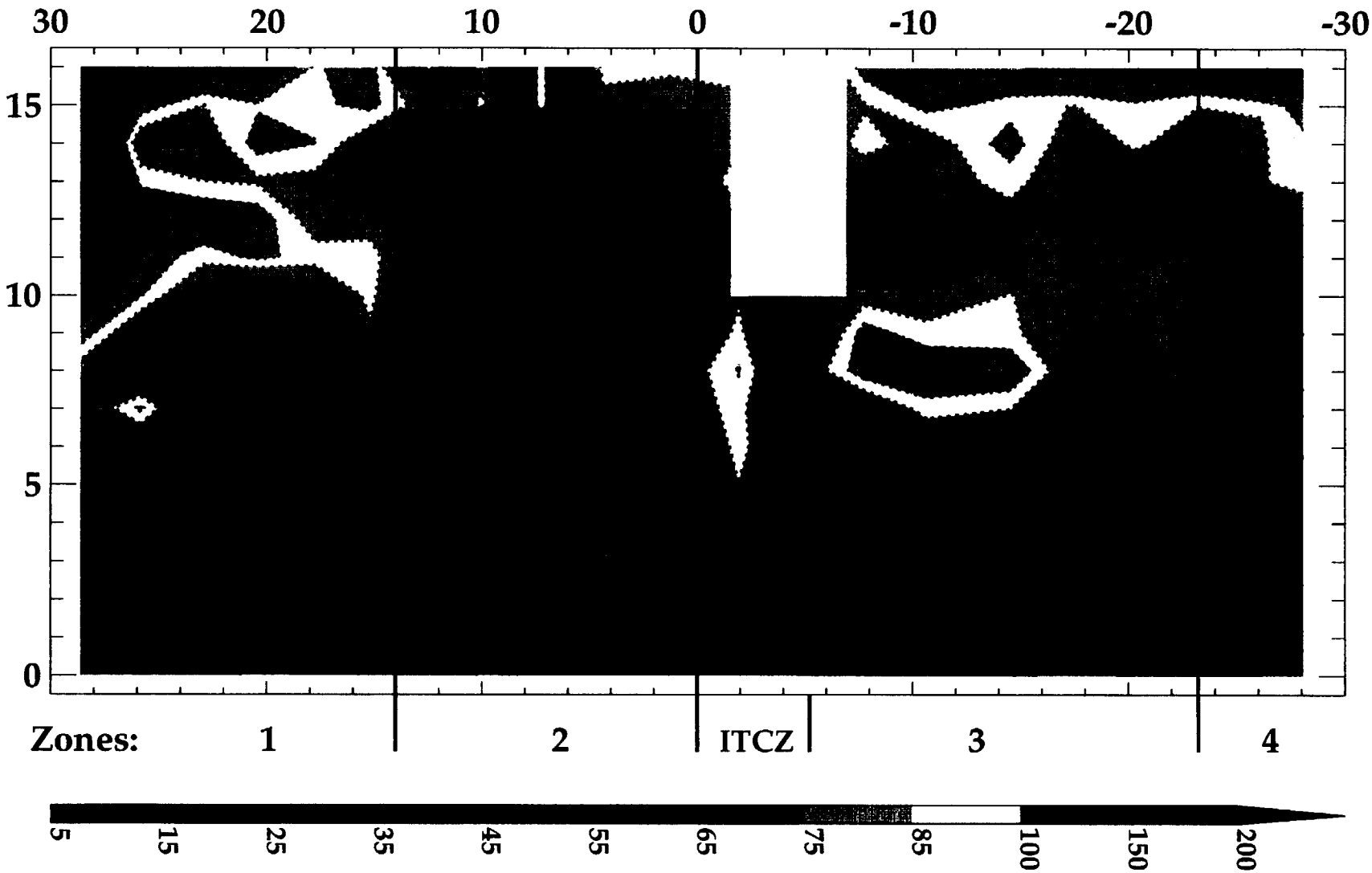
Cruise Ozone Mixing Ratio (ppbv)
Altitude vs. Latitude
30
20
10
0
-10
-20
-30
15
10
5
0
Zones:
1
2
ITCZ
3
4
5
15
25
35
45
55
65
75
85
100
150
200

Fig. 1

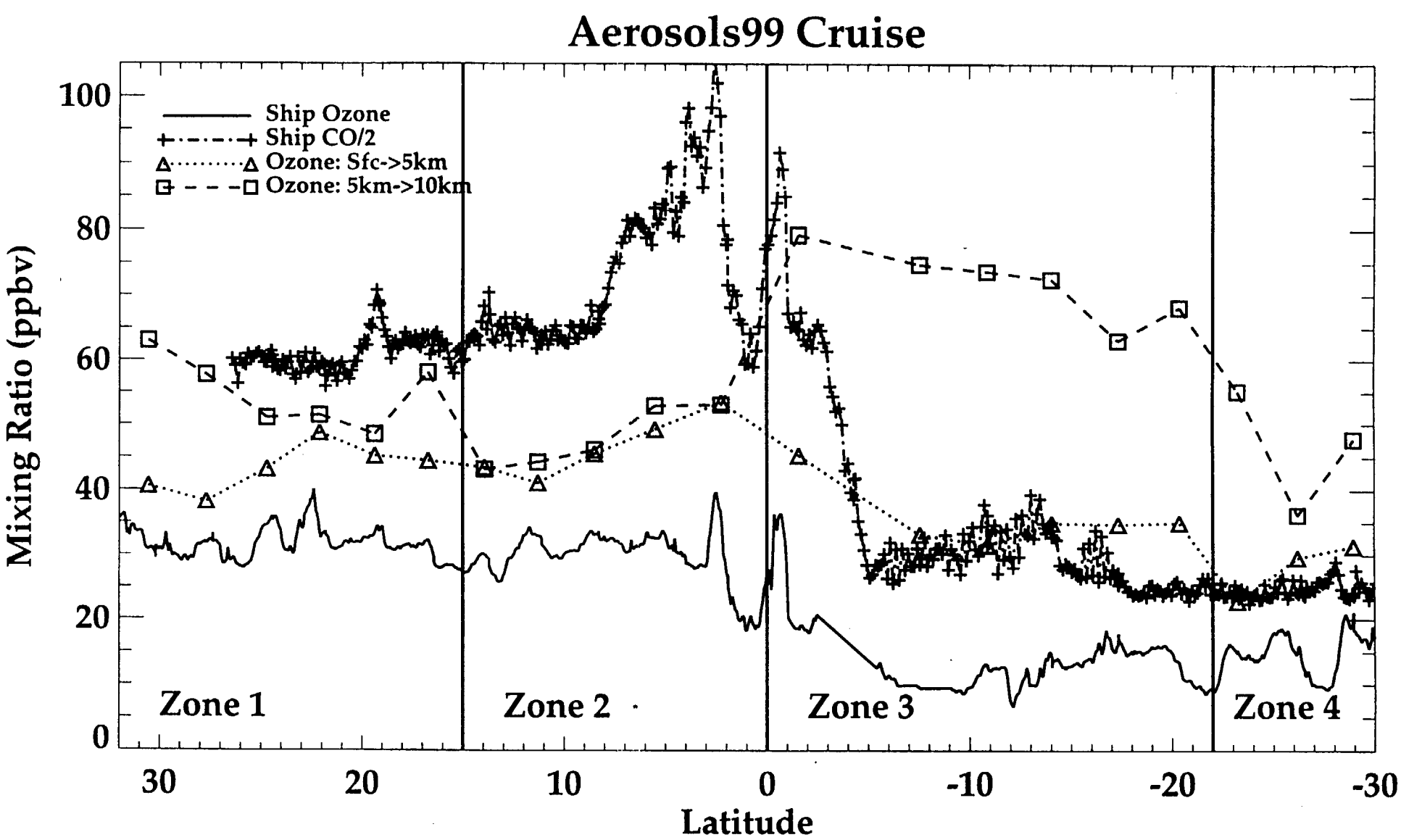

Fig. 2

/GSFC/UMD/1999

AEROSOLS '99 Cruise - Mean Profile .25 km average.

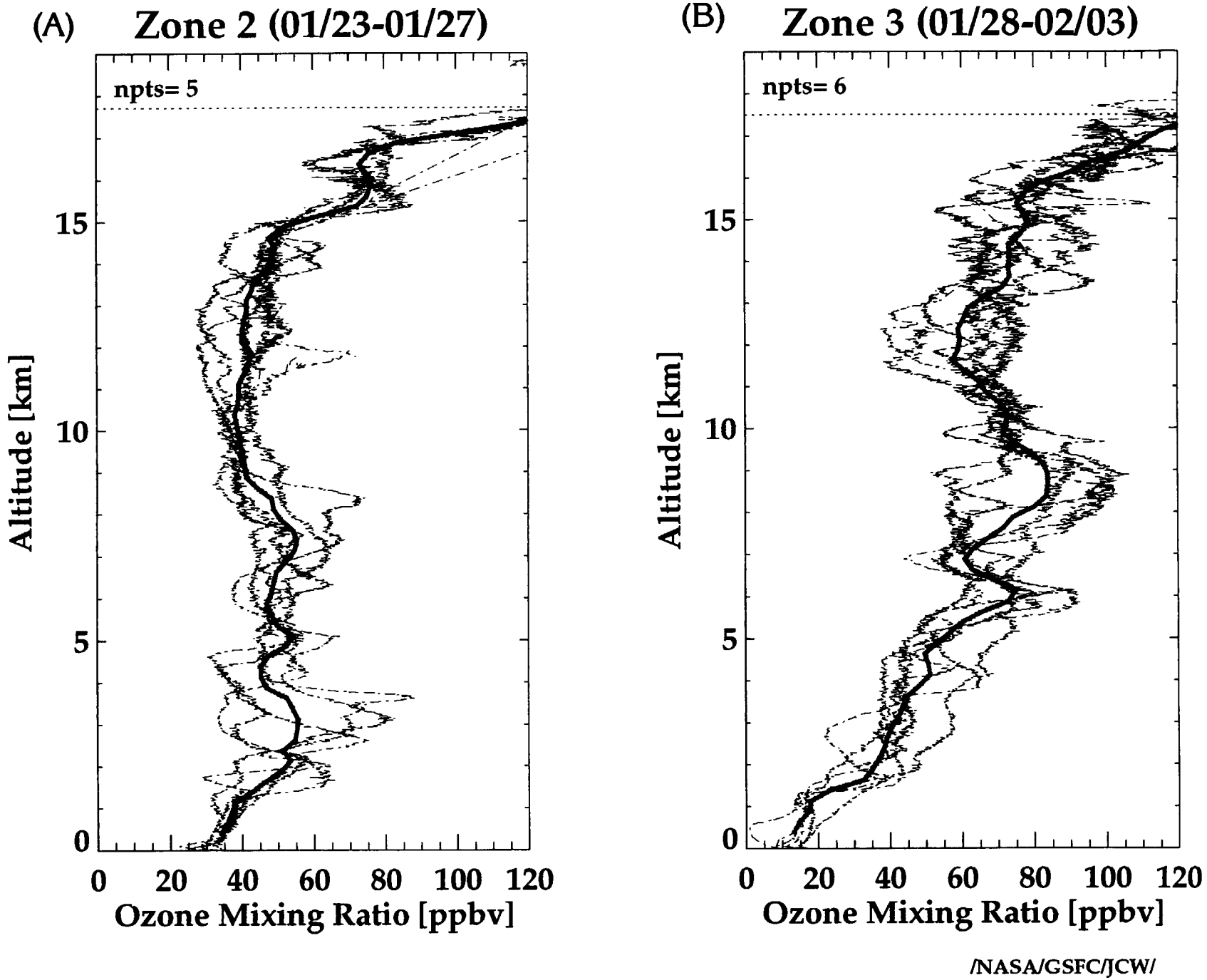

Fig. 3

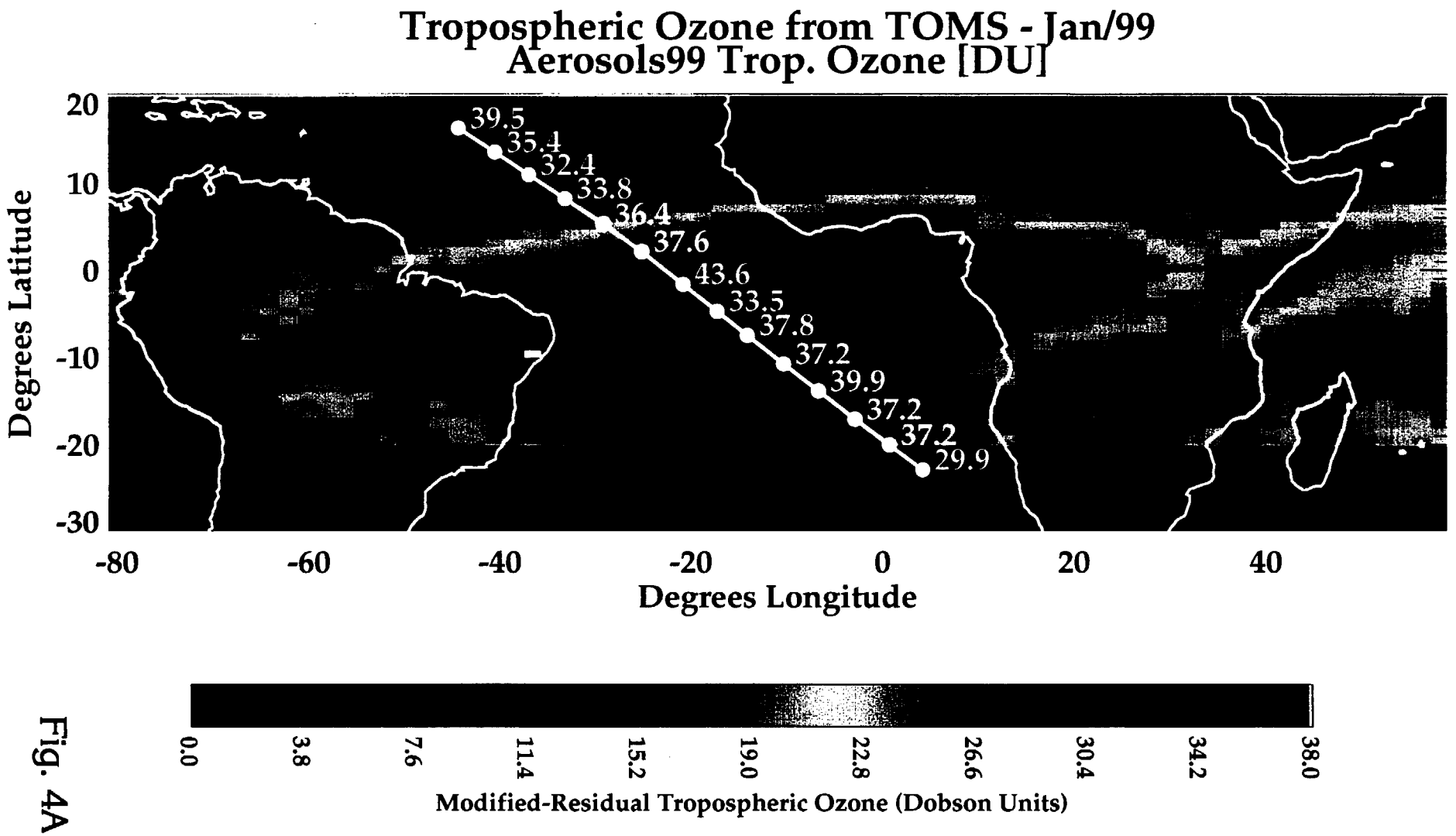
Tropospheric Ozone from TOMS - Jan/99
Aerosols99 Trop. Ozone [DU]
39.5
35.4
32.4
33.8
36.4
37.6
43.6
33.5
37.8
37.2
39.9
37.2
37.2
29.9
20
10
0
-10
-20
-30
Degrees Latitude
-80
-60
-40
-20
0
20
40
Degrees Longitude
0.0
3.8
7.6
11.4
15.2
19.0
22.8
26.6
30.4
34.2
38.0
Modified-Residual Tropospheric Ozone (Dobson Units)

Fig. 4A

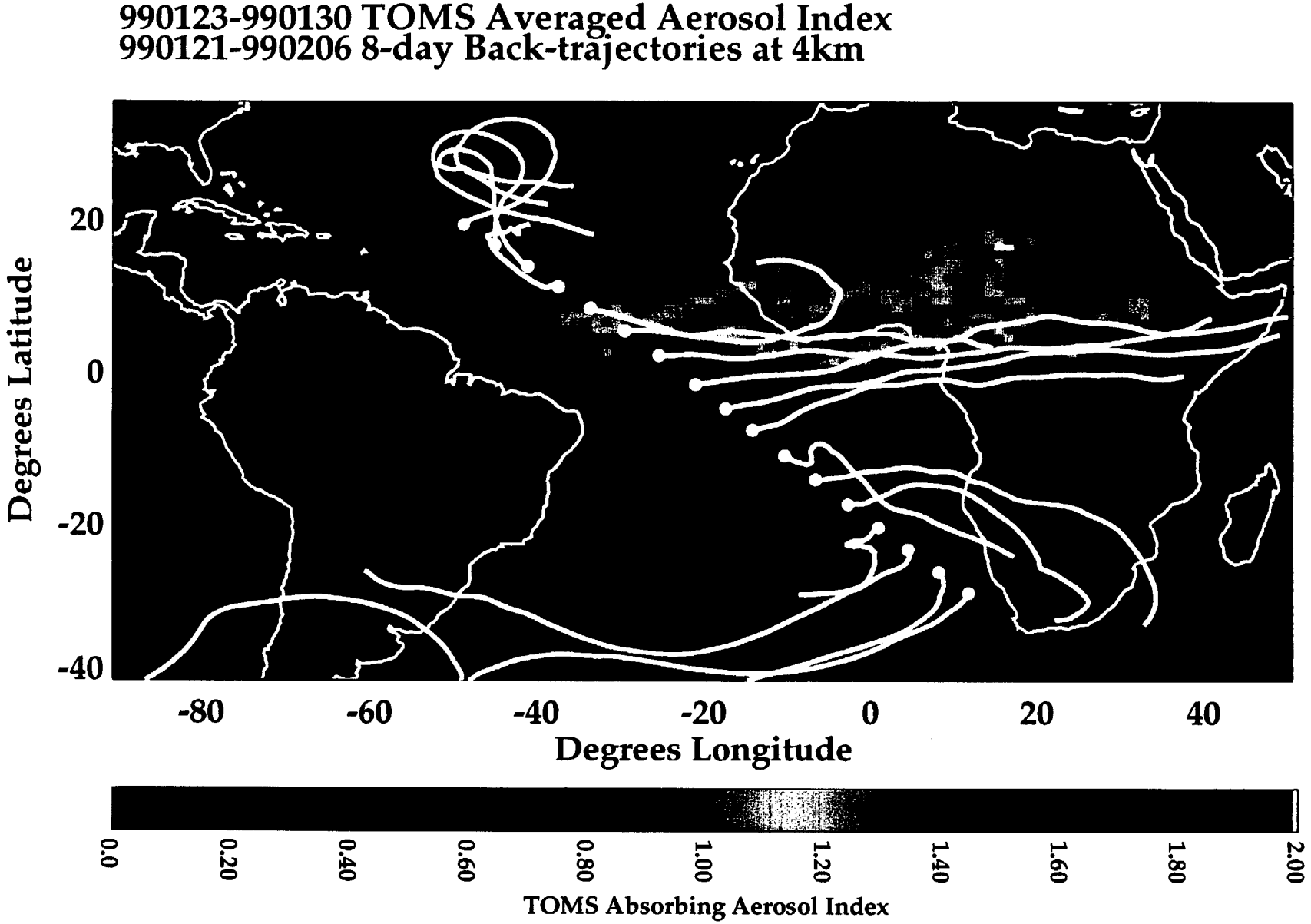
990123-990130 TOMS Averaged Aerosol Index
990121-990206 8-day Back-trajectories at 4km
Degrees Latitude
20
0
-20
-40
-80
-60
-40
-20
0
20
40
Degrees Longitude
0.0
0.20
0.40
0.60
0.80
1.00
1.20
1.40
1.60
1.80
2.00
TOMS Absorbing Aerosol Index

Fig. 4B

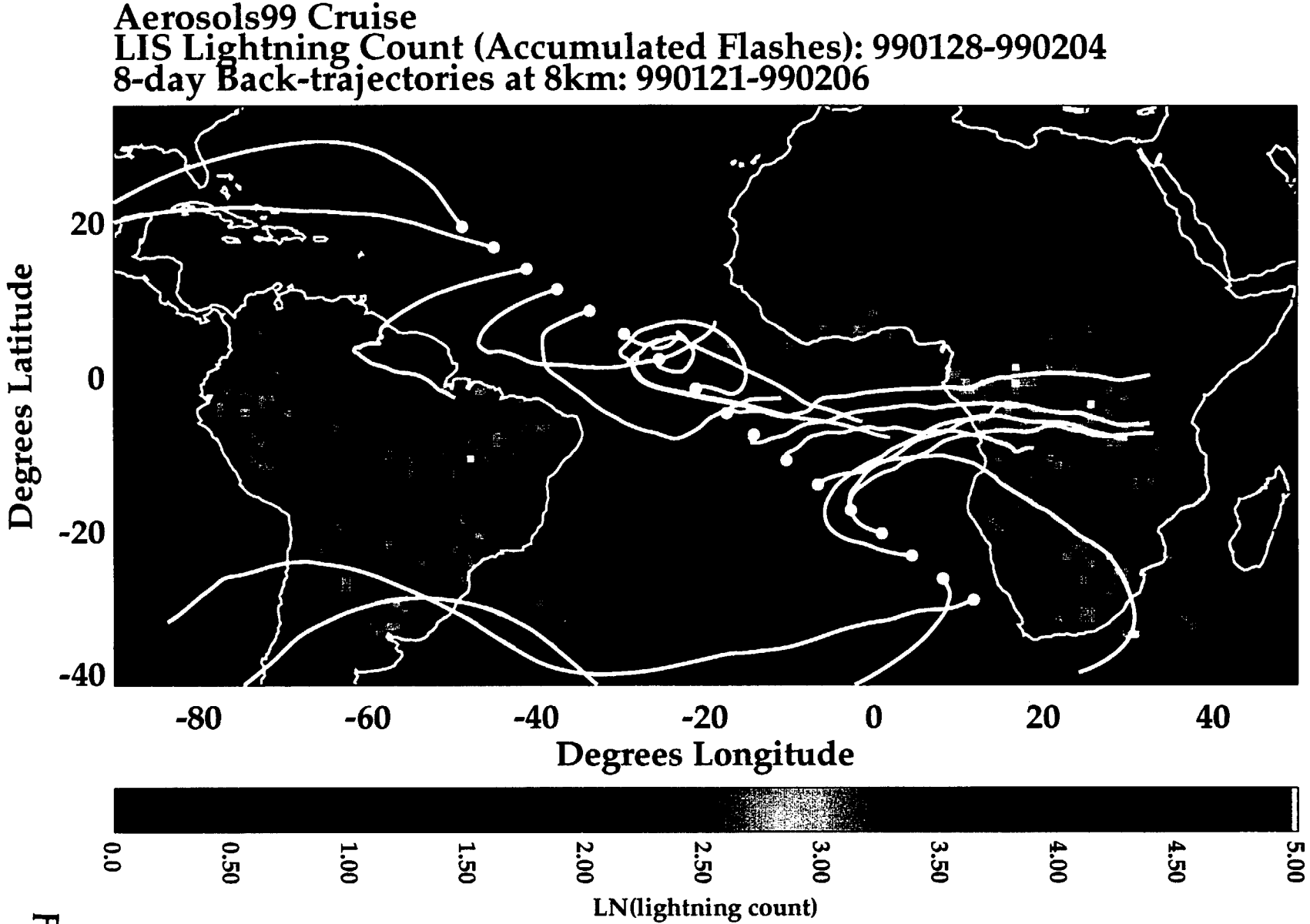
Aerosols99 Cruise
LIS Lightning Count (Accumulated Flashes): 990128-990204
8-day Back-trajectories at 8km: 990121-990206
Degrees Latitude
20
0
-20
-40
-80
-60
-40
-20
0
20
40
Degrees Longitude
0.0
0.50
1.00
1.50
2.00
2.50
3.00
3.50
4.00
4.50
5.00
LN(lightning count)
/NASA/GSFC/AMT+JCW

Fig. 4C