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Tibetan middle tropospheric ozone minimum in June discovered from GOME observations

Yi Liu,[1] Yong Wang,[1] Xiong Liu,[3,4,5] Zhaonan Cai,[1,2] and Kelly Chance[4]

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[1] Global Ozone Monitoring Experiment (GOME) observations from 1997–2000 have revealed the Tibetan Middle Tropospheric Ozone Minimum (TMTOM), a low-ozone layer that occurs in the middle troposphere over the Tibetan Plateau (TP) in June. Ozone profiles were derived from GOME observations and validated by ozonesonde measurements at two stations (Lhasa and Xining) over the TP. The mean bias is 5–10% within the troposphere. The ozone profiles reveal the TMTOM phenomenon occurs in the middle troposphere (8–13 km) over the middle and eastern TP in June. Dynamical field analyses showed that the TMTOM accompanies the onset of the Asian summer monsoon. The low-ozone air from the Bay of Bengal is transported into the middle troposphere over the TP by southwest currents while the lower troposphere over the TP is still occupied by ozone-rich air blocked by the transport barrier of the Himalayas. The TMTOM was most prominent in June 1998 likely linked to the occurrence of the intense El Niño of 1997–1998 because the Tibetan anticyclone is of large area and has strong intensity during El Niño years. The occurrence of the TMTOM can serve as an indicator of the phase of evolution of the Asian summer monsoon.

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1. Introduction

[2] Ozone is a critical atmospheric gas in the troposphere, playing important roles in atmospheric chemistry, air quality and climate change. Tropospheric ozone has two sources: photochemical production within the troposphere and downward transport from the stratosphere [*Danielsen*, 1968]. The combined effects of these two sources and the transport processes within the troposphere control the temporal and spatial distribution of ozone. Over the tropical-subtropical Asia-Pacific region, the middle and lower tropospheric ozone has a distinct meridional gradient during the summer, characterized by low concentrations over the tropics and high concentrations over the subtropics. As a result, meridional transport greatly influences the ozone distribution [*Logan*, 1985]. *Chan et al.* [1998] found a relative minimum of ozone mixing ratio in the upper troposphere at Hong Kong (22°N, 114°E) in late autumn and winter associated with enhanced intrusion of tropical air with low ozone concentrations. At the stations of Hong Kong and Naha (26°N, 128°E), ozone minima in the upper troposphere during wintertime are also reported by *H. Y. Liu et al.* [2002]. The Tibetan Plateau of China (TP), the highest plateau in the world, extends over 27°N–45°N, 70°E–105°E at an average elevation of approximately 4 km. Owing to its dynamic barrier and thermodynamic effects, the TP profoundly influences the general circulation of the atmosphere. The meteorological causes of ozone variation are thus an important research topic over the Asia-Pacific region.

[3] During the Asian summer monsoon period (from May/June to August/September), monsoon circulation and heating anomalies created by the TP form the South Asian anticyclone over the TP. The resulting change of monsoon circulation significantly affects the ozone distribution over this area. The behavior of total column ozone has been studied in the TP regions [e.g., *Reiter and Gao*, 1982; *Zhou et al.*, 1995; *Zou*, 1996; *Tobo et al.*, 2008]. *Reiter and Gao* [1982] reported that low total column ozone occurs over the TP as the South Asian anticyclone moves to the TP in mid-April. *Zhou et al.* [1995] and *Zou* [1996] found a summer minimum of total column ozone over the TP. Based on ozonesonde measurements over Lhasa station of the TP, *Tobo et al.* [2008] reported that the minimum of total column ozone is accompanied by the upper tropospheric monsoon anticyclone.

[4] Because of the lack of observations, tropospheric ozone over the TP has not been satisfactorily assessed. Satellite observations have the advantage of providing continuous coverage of ozone over a large spatial range, thereby allowing the study of its spatiotemporal variation. Recently, *Liu et al.* [2005, 2007] derived ozone profiles from the surface to the stratosphere from GOME data using an optimal estimation technique. These retrieved ozone profiles were validated with various measurements over the globe [*Liu et al.*, 2005, 2006a, 2006b] and over China [*Cai et al.*, 2009]. In this paper, we use the retrieved tropospheric ozone from GOME and analyze the meteorological causes of an ozone minimum in the mid-troposphere over Tibet during the summer monsoon season.

[5] In Section 2, we describe the GOME ozone profiles used in this study and their validation using ozonesonde measurements at two stations on the TP. In Section 3, we exploit GOME ozone profiles and ECMWF reanalysis data to explain the Tibetan Middle Tropospheric Ozone Mini-

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[1] Key Laboratory of Middle Atmosphere and Global Environment Observation, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.

[2] Graduate University of the Chinese Academy of Sciences, Beijing, China.

[3] Goddard Earth Sciences and Technology Center, University of Maryland, Baltimore County, Baltimore, Maryland, USA.

[4] Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, USA.

[5] Also at NASA Goddard Space Flight Center, Greenbelt, Maryland, USA.

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Table 1. Validation of GOME Ozone Profiles With Ozonesonde Observations at Lhasa and Xining[a]

Altitude Range (km)	Lhasa (%)	Xining (%)
30–32	−2.2 ± 0.5 (13)	−3.8 ± 0.4 (12)
28–30	−2.2 ± 0.6 (15)	−5.0 ± 0.4 (12)
25–28	−2.0 ± 0.7 (15)	−7.2 ± 0.6 (14)
23–25	−1.0 ± 0.9 (17)	−8.0 ± 1.0 (14)
21–23	1.2 ± 1.0 (17)	−5.8 ± 1.7 (14)
19–21	5.9 ± 1.7 (17)	−2.7 ± 2.1 (15)
16–19	10.0 ± 2.7 (17)	4.5 ± 3.3 (15)
14–16	7.2 ± 3.2 (17)	5.2 ± 4.8 (15)
12–14	1.0 ± 3.5 (17)	2.8 ± 6.7 (15)
10–12	−3.4 ± 3.1 (17)	0.4 ± 5.5 (15)
8–10	−4.9 ± 2.5 (17)	0.9 ± 3.6 (15)
6–8	−5.0 ± 2.2 (17)	1.8 ± 2.7 (15)
4–6	−3.8 ± 1.9 (17)	2.3 ± 2.1 (15)

[a] Second and third columns list (GOME-sonde)/sonde × 100% ± standard error (number of samples).

mum (TMTOM) phenomenon and its formation mechanism. Section 4 provides a summary and a discussion.

2. Description and Validation of GOME Ozone Data

[6] GOME was launched in 1995 on the European Space Agency (ESA) Remote Sensing-2 (ERS-2) satellite to measure backscattered radiance spectra from the Earth's atmosphere over the wavelength range 240–790 nm [*European Space Agency*, 1995]. Observations with moderate spectral resolution of 0.2–0.4 nm and high signal-to-noise ratios in the Hartley (200–320 nm), Huggins (320–350 nm), and Chappuis (400–700 nm) bands make it possible to retrieve the vertical distribution of ozone in the stratosphere as well as in the troposphere [*Chance et al.*, 1997]. The ozone profiles used in this study were retrieved by *Liu et al.* [2005, 2007] from GOME measurements. The retrievals involve detailed wavelength and radiometric calibrations of GOME level-1 data and degradation correction of GOME reflectance spectra by comparing the average reflectance to that at the beginning of observations [*Liu et al.*, 2007]. The retrieved ozone was found to agree well with ozonesonde, TOMS, and Stratospheric Aerosol and Gas Experiment II (SAGE-II) measurements [*Liu et al.*, 2005, 2006b]. The total errors of the retrievals were estimated to be 20–30% in the troposphere and lower stratosphere and 5–10% in the stratosphere [*Liu et al.*, 2005]. There are 4.5–5.5 degrees of freedom for signal in the atmosphere and up to ∼1.2 in the troposphere. The vertical resolution is 7–10 km in the stratosphere and 8–15 km in the troposphere. Ozone profiles used in this study are retrieved at 24 approximately 2.5 km layers from surface to about 60 km with 4–7 layers in the troposphere, depending on the terrain and tropopause height. Please refer to *Liu et al.* [2005] for more details about the retrieval algorithm and its retrieval characterization.

[7] For this research, whether GOME ozone profiles are suitable for tropospheric ozone studies over the TP is a key issue. Thus we compared ozone profiles from GOME to ozonesonde measurements at Lhasa (29.7°N, 91.1°E, 3650 m above sea level, located in the southern part of the TP) from June to October in 1998 and 1999 [*Tobo et al.*, 2008] and at Xining (36.63°N, 101.75°E, 2296 m above sea level, located in the northeastern TP) from April to August 1996. There are 17 and 15 coincidences at these two stations, respectively. Table 1 summarizes the comparison from *Cai et al.* [2009]. In the middle and lower troposphere, the bias was within ±5% at both Lhasa and Xining; in the upper troposphere and lower stratosphere (UT/LS), GOME had positive biases of 10% at Lhasa and 5% at Xining; in the middle stratosphere, GOME retrievals compared well to ozonesonde values at Lhasa but had a negative bias of up to 8% at Xining.

[8] The retrieved ozone profiles were processed into a monthly mean dataset with a horizontal resolution of 2° latitude × 2.5° longitude. The ozone concentration was converted from Dobson units to the mean volume mixing ratio (VMR, in ppbv), using the method described by *Ziemke et al.* [2001].

3. Formation Mechanism of the TMTOM in June Over the Tibetan Plateau

[9] In remote areas, the ozone mixing ratio usually increases from the surface to the tropopause due to the effect of downward transport of stratospheric ozone. Therefore, the lowest ozone mixing ratio in the vertical distribution of monthly mean ozone is seldom found in the middle troposphere. Figure 1 (left) shows the GOME monthly mean ozone profiles along 31°N longitude-altitude cross-sections. Figure 1 clearly illustrates minimum ozone values centered in the middle troposphere (8–13 km) over the middle and eastern parts of the TP. This minimum occurs in June for all the years investigated (1997–2000). The ozone mixing ratio is noticeably lower at this level than at higher and lower levels. We call this regional phenomenon the TMTOM.

[10] Based on the ECMWF reanalysis dynamical fields data shown in Figure 1 (left), there is strong ascending circulation (strong convection) from the surface to ∼12–13 km with remarkable westerly winds over the TP. Southerly winds prevail in the lower troposphere over the TP; in the middle and upper troposphere, southerly winds are found over the western TP and northerly winds over the eastern TP. The winds are organized by the South Asia anticyclone that is a main part of the Asian summer monsoon. The monsoon circulation consists of cyclonic flow and convergence in the lower troposphere together with strong anticyclonic circulation and divergence in the upper troposphere. Usually the Asian summer monsoon circulation consists of the South Asian anticyclone over the TP and the strong convections over the Bay of Bengal (6°–20°E, 80°–100°N), the South China Sea (7°–20°N, 110°–120°E), and South Asia [*Y. Liu et al.*, 2002]. Ascending flow of the convection over the Bay of Bengal carries ozone-poor air from the lower troposphere up to the middle troposphere, where it is transported northward to the middle troposphere over the TP. The blocks formed by the westerly jet over the northern TP (from the north) and northerly winds over the eastern TP (from the east), and the transport barrier created by the Himalayas to lower troposphere flow over the southern TP (from the south), cause ozone-poor air to accumulate in the middle troposphere over the middle and eastern TP in June, forming the TMTOM phenomenon.

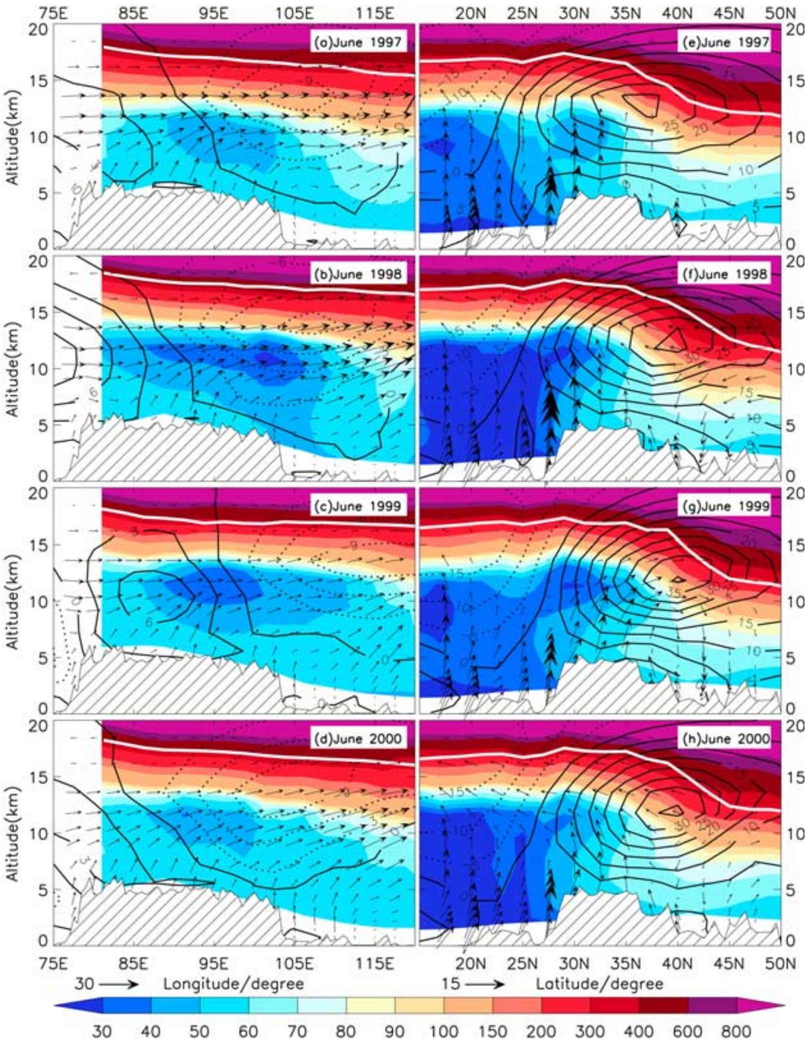

Figure 1. GOME monthly mean ozone mixing ratio (color shaded, ppbv), European Center for Medium-Range Weather Forecasts (ECMWF) vertical circulation (vector, horizontal wind: m/s; vertical wind: −Pa/s × 100) and wind (contour), and the National Centers for Environmental Protection (NCEP) thermal tropopause (white line) in June from 1997 to 2000. The solid diagonal lines indicate topography. (a–d) Longitude-altitude cross-sections along 31°N and meridional wind (positive: south wind; negative: north wind, interval: 3 m/s). (e–h) Latitude-altitude cross-sections along 93.75°E and zonal wind (positive: west wind, negative: east wind, interval: 5 m/s).

[11] Figure 1 (right) shows the GOME monthly mean ozone profiles along 93.75°E latitude-altitude cross-sections. There are lower ozone plumes with ozone of 30–40 ppbv from the surface to heights of 12–13 km; these plumes spread northward from the Bay of Bengal to the middle troposphere (8 ~ 13 km) over the TP. The contour of 50 ppbv extends to the middle troposphere of the northern TP (35°N), where the westerly jet at approximately 40°N blocks the ozone-poor air transported to the northern TP and causes ozone-poor air to accumulate over the middle and eastern TP.

[12] During May, the convection over the Bay of Bengal is not strong enough to take the ozone-poor air up to the middle troposphere, and the Himalayas create a transport barrier at the southern part of the TP so that the middle troposphere over the TP is still occupied by ozone-rich air (Figures 2a and 2c). However, in July, the South Asian anticyclone is fully developed and centered more westward over the TP and Iranian Plateau, which increases the northerly winds in the middle troposphere of the TP. These northerly winds transport ozone-rich air originating from higher latitudes and make the TMTOM insignificant (Figures 2b and 2d). This explains why the TMTOM occurs in June but not in May or July during the 4 years studied.

[13] The intensity of the TMTOM is associated with the strength and the position of the southerly wind, which is organized by monsoon anticyclone. As examples, the stronger southerly winds in 1998, with maximal wind speed greater than 6 m/s (Figure 1b); and those in 1999 with maximal wind speed of 6 m/s (Figure 1c) are associated with the stronger TMTOMs in these years, as compared to 1997 and 2000 (Figures 1a and 1d). A weaker TMTOM in 2000, compared to that in 1999, is clearly attributable to the northerly winds in the middle troposphere of the TP, as

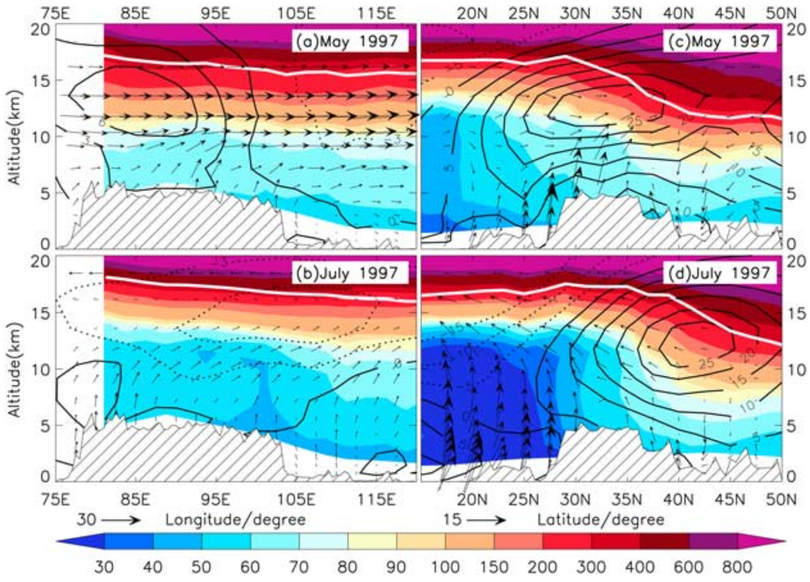

Figure 2. As in Figure 1, but for May and July of 1997.

shown in Figures 1g and 1h. The relationship can be simply explained by the meridional transport of ozone-poor air from the lower latitude to the TP. The intensity of the TMTOM is also associated with the strength and the position of the ascending flows over the Bay of Bengal and TP, especially over the steep slopes of the southern TP (Figures 1e–1h), which are attributed to the evolution of the Asian summer monsoon. The ascending flows have a positive correlation with the low ozone transported over the TP. For example, the strongest ascending flow, in 1998, led to the lowest ozone mixing ratio of the TMTOM during the 4 years.

[14] Since the TMTOM appears most noticeable around 200 hPa (∼12 km) in altitude, we show (Figure 3) the horizontal distribution of the monthly mean ozone mixing ratio at 200 hPa in June. There are two low-ozone centers. One is located over the Bay of Bengal, in which strong convection transports the ozone-poor air from the boundary layer to the middle troposphere. The other is located over the south and east of the TP, which is attributed to the thermal convection over the TP. As discussed in the previous section, the strength of the TMTOM intensifies with increasing ascending flow and southerly wind. In 1998, these two favorable factors created a larger area of

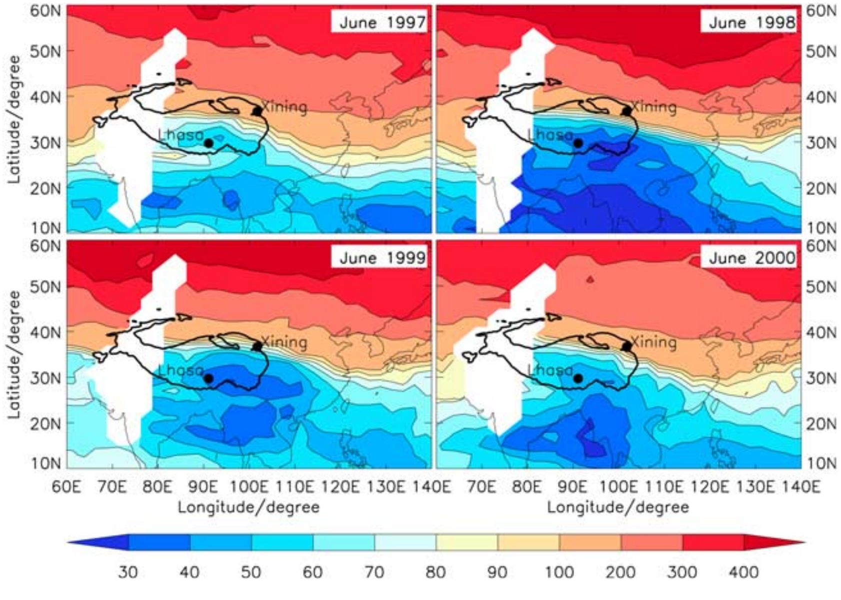

Figure 3. Horizontal distribution of the GOME monthly mean ozone mixing ratio (ppbv) at 200 hPa in June from 1997 to 2000. The thick line shows the part of the plateau that is above 3 km in elevation. No GOME data is available over the white area due to the downlinking of GOME data to ground stations.

lower ozone mixing ratio than those found in the other 3 years at the 200 hPa pressure surface. The ozone mixing ratio was 50–60 ppbv in 1997, 30 ppbv in 1998, 30–40 ppbv in 1999, and 40–50 ppbv in 2000 at Lhasa.

4. Summary and Discussion

[15] Using GOME ozone profiles, we reveal the TMTOM phenomenon, an ozone minimum in the middle troposphere (~200 hPa/12 km) over the TP that occurs annually in June. Its relationship with dynamic fields shows that its formation is mainly due to wind transport of ozone-poor air, which is controlled by the evolution of the Asian summer monsoon. Ascending flow of the convection over the Bay of Bengal brings ozone-poor air from the lower troposphere up to the middle troposphere, where it is transported northward with the southerly wind. The blocks created by the westerly jet over the northern TP and northerly winds over the eastern TP, and the transport barrier of the Himalayas to the lower troposphere in the southern TP, combine to cause the accumulation of ozone-poor air in the middle troposphere over the TP and the formation of the TMTOM in June. The intensity of the TMTOM is associated with the strength and the position of the southerly wind over the TP and the ascending flows over the Bay of Bengal and TP, which are attributed to the evolution of the Asian summer monsoon. The relationship can be simply explained by the upward and meridional transport of ozone-poor air from the lower latitudes to the TP. For example, the strongest ascending flow and southerly wind, in 1998, led to the lowest ozone mixing ratio of the TMTOM during the 4 years studied.

[16] The Asian summer monsoon plays a primary role in forming the TMTOM. The Asian summer monsoon circulation consists of the South Asian anticyclone and the strong convections over the Bay of Bengal and South China Sea. These two convective regions are characterized by lower ozone and higher water vapor and are the origin of the lower ozone air of the TMTOM. Thus, the TMTOM can serve as an indicator of the evolution of the Asian summer monsoon. The Asian summer monsoon experiences strong intraseasonal and interannual variations. The detail relationship between these variations and occurrences of TMTOM can be investigated using high temporal resolution ozone data (e.g., using GOME-2 or OMI data) and chemical transport model.

[17] The analyses in this study show that the TMTOM phenomenon has significant interannual variation. The lowest ozone mixing ratio for the four years studied occurred in 1998. This likely has a close relationship with the intense El Niño event that occurred during 1997–1998, as the Asian monsoon is significantly linked to the El Niño-Southern Oscillation (ENSO). *Zhang et al.* [2000] found that South Asian anticyclone of large area and strong intensity always corresponded with El Niño years. For example, in 1998, the strong El Niño corresponded to the strong, east phase of the South Asian anticyclone [*Zhang et al.*, 2000]. This strong, east phase of the South Asian anticyclone would enhance the TMTOM, consistent with the findings of this study. *Shapiro et al.* [2001] showed that ENSO has an important impact on dynamical features in the extra-tropical Pacific upper troposphere, including the strength of the sub-tropical jet. Some studies have examined the effects of ENSO on tropospheric ozone in the tropics [*Doherty et al.*, 2006; *Ziemke and Chandra*, 2003]. However, further study is needed to clarify precisely how ENSO influences the tropospheric ozone over the TP and how much it contributes to the TMTOM.

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Z. Cai, Y. Liu, and Y. Wang, Key Laboratory of Middle Atmosphere and Global Environment Observation, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China. (liuyi@mail.iap.ac.cn)

K. Chance, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138-1516, USA.

X. Liu, Goddard Earth Sciences and Technology Center, University of Maryland, Baltimore County, Baltimore, MD 21228, USA.