Signature of stratospheric air at the Tibetan Plateau

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[1] Current estimates of gross carbon flux tend to ignore the downwelling flux of CO_2 from the stratosphere. Observations showed that there is a phase shift between the time series for the concentration of the standard isotopologue $C^{16}O^{16}O$ and $C^{16}O^{18}O$ at Waliguan, China (36°17′N, 100°54′E, 3816 m) and several other places. Previous attempts to explain the shift have not been satisfactory. Here we show that the phase shift could be explained by the downwelling air from the stratosphere, and demonstrate that this source of CO_2 provides a useful tool for constraining the carbon cycle. Using O_3 as a proxy of stratosphere-troposphere exchange, we find excellent correlation between O_3 and $C^{16}O^{18}O$ observed at the Waliguan Observatory. The observed variability of $C^{16}O^{18}O$ is consistent with model predictions, thus supporting that the surface air has significant contributions from the stratosphere. Quantitative modeling may provide a powerful tool for constraining the sources and sinks of CO_2 using the isotopically enriched CO_2 from the stratosphere as a tracer. **Citation:** Liang, M.-C., J. Tang, C.-Y. Chan, X. D. Zheng, and Y. L. Yung (2008), Signature of stratospheric air at the Tibetan Plateau, *Geophys. Res. Lett.*, *35*, L20816, doi:10.1029/2008GL035246.

[2] The isotopic composition of CO_2 in the atmosphere is an integrated signal of atmospheric and biogeochemical processes, and thus provides a tool for studying biogeochemical cycles involving CO_2 [*Ciais et al.*, 1997; *Intergovernmental Panel on Climate Change* (*IPCC*), 2001]. To separate the different processes, several tracers must be used. For example, atmospheric $\delta^{13}C(CO_2)$ can differentiate the uptake fluxes of CO_2 by the land and ocean, while $\delta^{18}O(CO_2)$ provides a constraint on terrestrial gross primary production [*Ciais et al.*, 1997; *Cuntz et al.*, 2003; *IPCC*, 2001]. In the atmosphere, the primary mechanism that modifies the isotopic composition of CO_2 is the exchange reaction with $O(^1D)$ in the stratosphere [*Thiemens et al.*, 1991; *Yung et al.*, 1991, 1997; *Liang et al.*, 2007, 2008]. This exchange process conserves the concentration of CO_2 but leaves an isotopic signature that is distinct from tropospheric/biospheric CO_2. A two-box model [*Hoag et al.*, 2005] has been used to investigate the contribution of stratospheric CO_2 to tropospheric/biospheric CO_2 based on the concept of mass-dependent and mass-independent isotopic fractionation processes. The authors found that the predicted $\Delta^{17}O(CO_2)$, on the order of tenths ‰, could be measurable.

[3] The Waliguan Observatory (36°17′N, 100°54′E, 3816 m; site code is WLG) is one of the highest Global Atmospheric Watch stations in the world located on the northeastern edge of the Tibetan Plateau, where the averaged height is greater than 3 km. The observatory is remote, making it an ideal station for longterm monitoring of the background atmosphere. The great elevation difference between the plateau and the surrounding regions causes a large differential heating, induces a large scale circulation [e.g., *Flohn*, 1968; *Yanai et al.*, 1992], and exerts a profound influence on the regional climate [e.g., *Webster*, 1983; *Chase et al.*, 2003]. The discovery of elevated O_3 concentration at Waliguan in the summertime leads to two competing hypotheses: intrusion from the stratosphere [*Ding and Wang*, 2006; *Wang et al.*, 2006], long-range transport from polluted areas nearby [*Zhu et al.*, 2004], or a combination of the two [*Lee et al.*, 2007]. Here we will pursue the first hypothesis and connect it to intrusion events that carry heavy CO_2 from the stratosphere [*Liang et al.*, 2007, 2008].

[4] The observed phase lag between normal CO_2 (and $^{13}CO_2$) and $C^{16}O^{18}O$ at Waliguan [*Cuntz et al.*, 2003] could be explained by an additional isotopic flux from the stratosphere (see below); this additional source has not been included in current models [*Cuntz et al.*, 2003]. Thus, this site provides an ideal test-bed for monitoring the terrestrial carbon cycle. Available measurements used to support the proposal are summarized below.

[5] The O_3 concentrations were measured independently by two UV absorption analyzers (US Thermal Environment, Model 49C) running in parallel [*Tang et al.*, 1996] with a detection limit of 1 ppbv and a precision of ±2.0 ppbv. A detailed description of the site environment of the Waliguan Observatory and an audit program on the measurements of this station are described elsewhere [e.g., *Tang et al.*, 1996; *Klausen et al.*, 2003]. In this study, the monthly average concentrations were derived from the hourly averaged concentrations; we defer comprehensive data analysis to a later paper (J. Tang et al., manuscript in preparation, 2008). The near surface concentrations of CO, $C^{16}O^{16}O$, and $^{13}CO_2$ were obtained from a complete and accurate proxy record of the Cooperative Atmospheric Data Integration Project [*Earth System Research Laboratory*, 2007a, 2007b, 2007c]. The $C^{16}O^{18}O$ flask measurements and analysis were carried out by the Carbon Cycle Cooperative Global Air Sampling Network of Earth System Research Laboratory (ESRL) Global Monitoring Division (GMD) at the National Oceanic and Atmospheric Administration (NOAA) [*Zhou et al.*, 2005].

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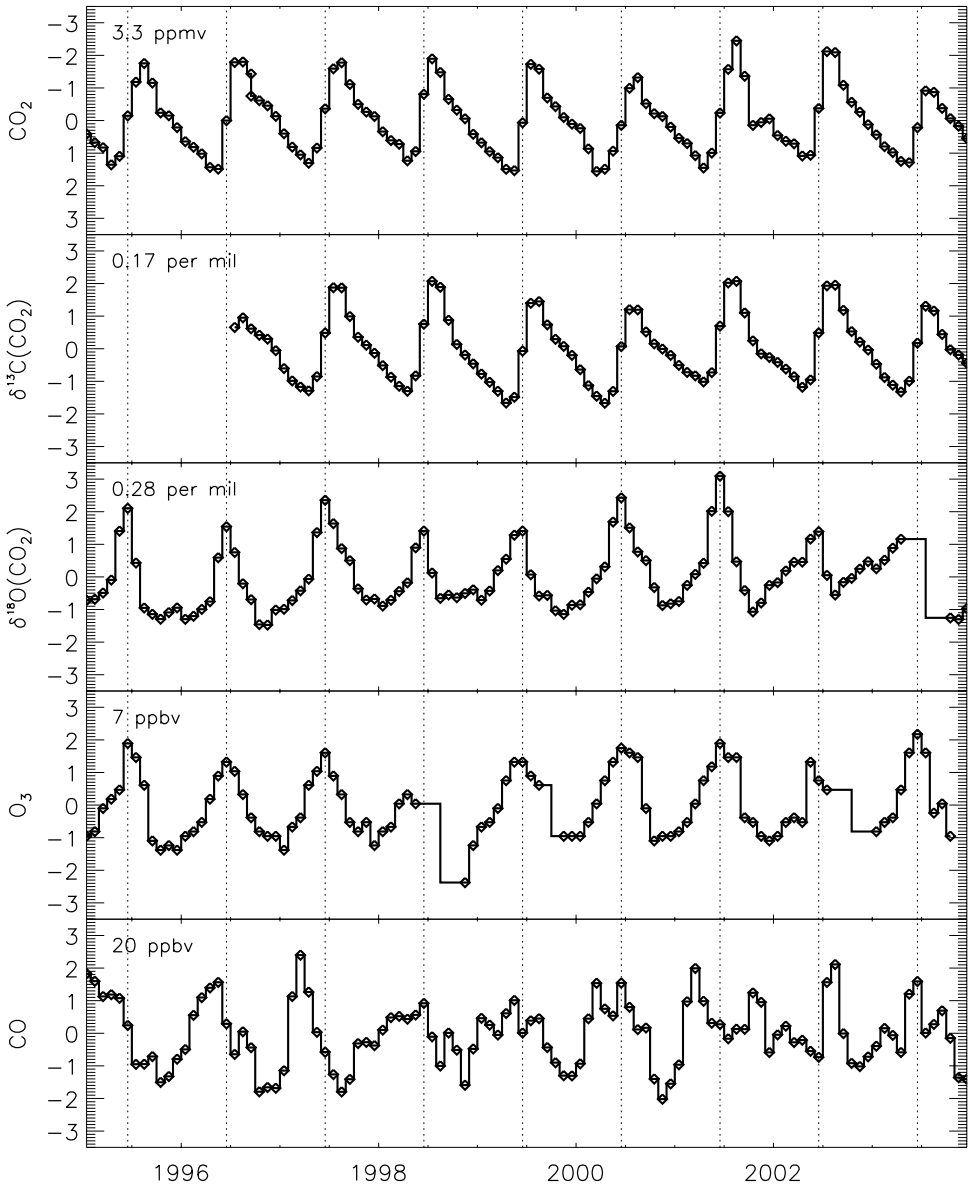

Figure 1. Time series of the volume mixing ratio of CO_2, $\delta^{18}O(CO_2)$, $\delta^{13}C(CO_2)$, the volume mixing ratio of O_3, and the volume mixing ratio of CO, scaled by their standard deviations (3.3 ppmv, 0.28‰, 0.17‰, 7.0 ppbv, and 20.0 ppbv, respectively). The trends have been removed. The dotted lines indicate June of each year. The scale of CO_2 has been inverted.

[6] Figure 1 shows the time series of O_3, CO, CO_2, $\delta^{13}C(CO_2)$, and $\delta^{18}O(CO_2)$ observed at the Waliguan Observatory. A strong seasonal cycle is clearly seen in CO_2, $\delta^{13}C(CO_2)$, and $\delta^{18}O(CO_2)$ (upper panel), suggesting that CO_2 and its isotopologues are controlled primarily by biogeochemical processes near the surface. The concentration of CO_2 is basically anticorrelated with $\delta^{13}C(CO_2)$; the phase coherence of seasonal cycles of both species is good. However, there is a phase lag of more than one month between normal CO_2 (and $\delta^{13}C(CO_2)$) and $\delta^{18}O(CO_2)$, suggesting that in addition to biologically controlled processes, hitherto unknown processes are present. We argue below that the stratosphere-troposphere exchange plays a role. The surface concentration of O_3 has a well-defined seasonal cycle, possessing a sharp peak in June (labelled by vertical dotted lines). In contrast, CO varies without a significant seasonal cycle. This demonstrates that O_3 originates from well-defined source(s) and is transported to the site by large scale circulation, while CO has many local sources. The former is attributable to the stratosphere-troposphere exchange processes (the downwelling branch of the Brewer-Dobson circulation) and the latter is by long-range transport from nearby cities. See later paragraphs for more detailed discussion.

[7] Figure 2 (top) shows the correlation plots of O_3 versus $\delta^{18}O(CO_2)$ and Figure 2 (bottom) shows O_3 versus CO. In general, good phase correlation ($R^2 = 0.67$) is observed between $\delta^{18}O(CO_2)$ (upper) and O_3, and both these species have maximal values in the middle of the year (see Figure 1). There are two sources of O_3: the stratosphere and distant polluted areas such as China and Europe. However, the relative contribution from each to the

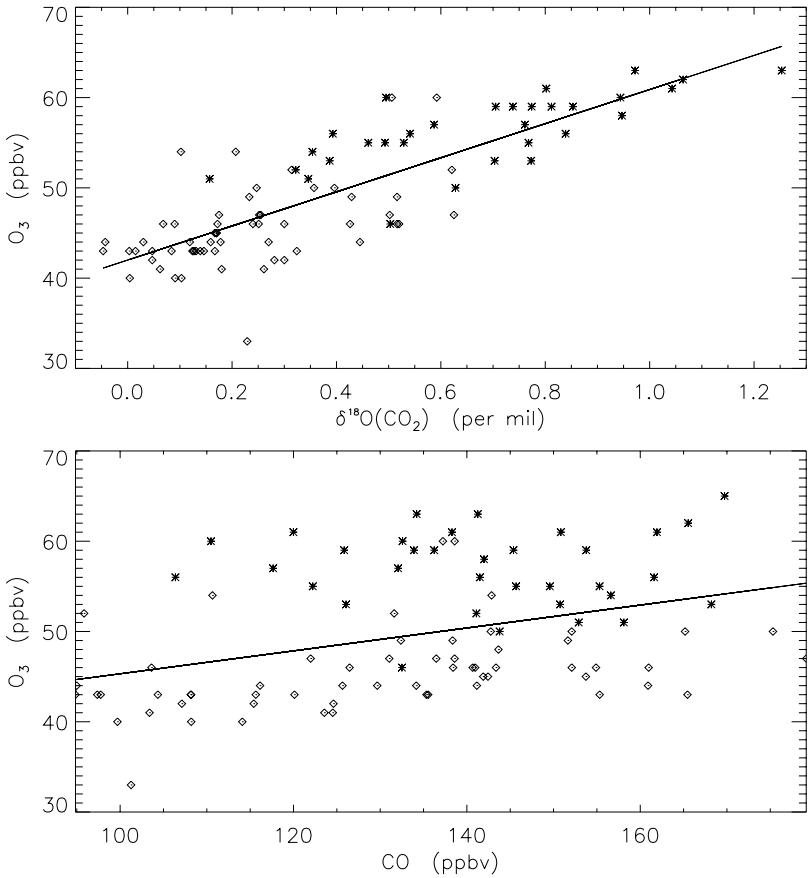

Figure 2. Scatter plot of (top) O_3 versus $\delta^{18}O(CO_2)$ and (bottom) O_3 versus CO. The data between the months April and August are shown by the asterisks.

Waliguan O_3 is debated [*Zhu et al.*, 2004; *Ding and Wang*, 2006]. We believe that most of O_3 during spring-summer time is from the stratosphere, while that in other seasons is affected primarily by long-range transport of polluted air. This argument is supported by the correlation between CO and O_3 (see below).

[8] The downwelling of air from the stratosphere maximizes in spring-summer time [see, e.g., *Appenzeller et al.*, 1996; *Rosenlof and Holton*, 1993], carrying stratospheric signatures such as high O_3 and low CO_2 to the troposphere. This stratospheric source of CO_2 is enhanced in $\delta^{18}O(CO_2)$ and $\Delta^{17}O(CO_2)$ [*Thiemens et al.*, 1995; *Boering et al.*, 2004], and has a seasonal cycle that is different from that originating from the surface. (See *Thiemens* [2006] for an intensive review.) For example, at the Waliguan site we study here, biogeochemical models [*Cuntz et al.*, 2003] predicts extensive effects due to respiration in March–April (maximum in $\delta^{18}O(CO_2)$) and August (minimum in $\delta^{18}O(CO_2)$) and due to assimilation in ~March (minimum in $\delta^{18}O(CO_2)$) and July–August (maximum in $\delta^{18}O(CO_2)$), while the Brewer-Dobson circulation has maximum strength in ~March–July (see Figure 3). The 2-D model predicts $\delta^{18}O(CO_2) \approx 0.5‰$ in the downwelling air [*Liang et al.*, 2008] (see also Figure 3), close to the observed peak-to-peak $\delta^{18}O(CO_2)$ of 1‰ at Waliguan (Figure 1). We stress that the effect from the stratosphere could be underestimated by the 2-D model, because the model has no longitudinal variation and the latitudinal grid is too coarse (10°) to capture individual intrusion events that occur frequently in spring-summer time at the location of the Waliguan Observatory.

[9] Figure 3 shows the 2-D modeled isoflux (diamonds) of CO_2 across the tropopause, taken from by *Liang et al.* [2008]. The zonally averaged flux peaks in May and has a magnitude of ~8 PgC/year‰ per 10 degree latitude bin (or $\sim 10^{-6}$ mole m^{-2} s^{-1} ‰), a value that is only a factor of a few less than that the biological fluxes (such as assimilation and respiration) [*Cuntz et al.*, 2003].

[10] In the mid latitudes of East Asia, ozonesonde and surface observations revealed a distinct spring maximum [e.g., *Oltmans et al.*, 2004, and references therein]. The finding is consistent with the fact that stratospheric intrusions of O_3 into the troposphere reach their seasonal maximum due to unusually strong winds associated with the polar and subtropical jet streams over the east Asian coast and the persistence of cyclogenesis over Japan that result in frequent tropopause folding and thus significant intrusion of O_3 into the troposphere [*Austin and Midgley*, 1994]. In subtropical south China and north western Pacific, there is an important part of the spring O_3 maximum in the lower free troposphere that is due to photochemical production associated with biomass burning emission from the Southeast Asia subcontinent [*Chan et al.*, 2003a]. The measurements on the edge of subtropics of east China in spring 2001 revealed that while stratospheric O_3 is still the predominant source of O_3 in the middle and upper tropo-

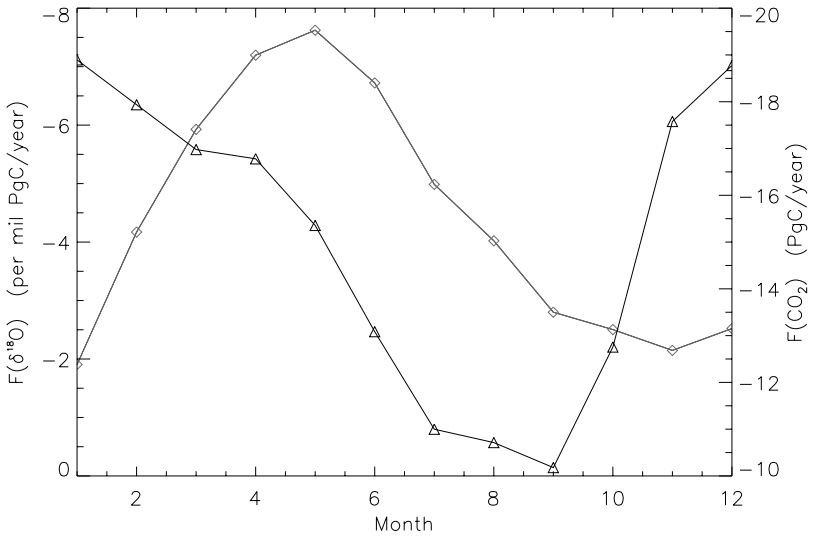

Figure 3. Seasonal cycle of the cross tropopause fluxes of $\delta^{18}O(CO_2)$ (diamonds, $F(\delta^{18}O)$) and CO_2 (triangles, $F(CO_2)$). Negative values denote downward transport.

sphere, anthropogenic sources caused high O_3 pollution episodes in the boundary layer [*Chan et al.*, 2003b].

[11] In Central Asia at the Waliguan Observatory, tropospheric O_3 shows a unique seasonal maximum extending from spring to summer [*Tang et al.*, 1996]. Ozonesonde measurements in the summer of 1996 revealed that high O_3 accompanying low relative humidity propagates continuously from the upper to the lower troposphere [*Zheng et al.*, 2004]. In a recent modelling study, *Zhu et al.* [2004] proposed that the seasonal O_3 transitions were associated with the Asian monsoon system and transport from Eastern/Central China. Central/South Asia and Europe are responsible for the distinct seasonal O_3 cycle at the Waliguan Observatory. However, an alternative explanation argued that the downward transport of air from the upper troposphere/lower stratosphere is an important factor causing the summer O_3 maximum [*Ma et al.*, 2005]. The stratospheric origin of summer O_3 is also supported by independent evidence of good correlation between O_3 and radioactive ^{7}Be and ^{210}Pb in aerosols [*Zheng et al.*, 2005] and meteorological simulation that the summertime O_3 enhancements at Waliguan are due to stratospheric intrusion, rather than transport of anthropogenic pollution [*Ding and Wang*, 2006]. A combination of the two sources was later proposed [*Lee et al.*, 2007].

[12] The good correlation between O_3 and $\delta^{18}O(CO_2)$ clearly suggests that O_3 and $\delta^{18}O(CO_2)$ come from the same region, i.e., the stratosphere we propose here (see Figures 1 and 2). The stratospheric origin is supported by the fact that the spring-summer O_3 is likely transported from the stratosphere to the troposphere by the stratosphere-troposphere exchange (mainly intrusions at the Waliguan Observatory), which can be seen from Figure 2 that the correlation between O_3 and CO in spring-summer is poor ($R^2 = 2 \times 10^{-4}$; asterisks). The correlation is better ($R^2 = 0.13$; diamonds) in other seasons, suggesting that the O_3 is long-range transported from nearby polluted areas [e.g., *Parrish et al.*, 1998]. (At the remote site such as Waliguan Observatory, there are two known significant sources of O_3: stratospheric origin and photochemical origin from pollution.)

[13] In short, the stratosphere-troposphere exchange carries stratospheric air to the troposphere. The air has oxygen isotope signature in CO_2 distinct from that originating from the surface. The nonzero $\Delta^{17}O(CO_2)$ may provide an additional constraint on the CO_2 biogeochemical cycles through the evolution/destruction of $\Delta^{17}O(CO_2)$ in biogeochemical processes. The availability (the amount of the isotopically enhanced CO_2) could be greatly enhanced in the regions where intrusion events occur frequently. This happens in the Central/East Asia (see above), especially the Tibetan plateau we examine here. Measurements of $\Delta^{17}O(CO_2)$ are in progress. The new and direct observationally-based budget of stratospheric and biogeological CO_2 will soon be available to improve our understanding of the sources and sinks of CO_2 at/in the surface.

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