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## MULTIYEAR AVERAGE CHARACTERISTICS OF CO<sub>2</sub> VARIATIONS IN THE FREE ATMOSPHERE OVER COLORADO (40° N, 104° W).

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### Abstract

A statistical analysis is made of vertical changes in CO<sub>2</sub> mole fraction and its seasonal variations in the free troposphere from the data of flask aircraft measurements over Briggsdale and Carr, Colorado, USA (~ 40° N, ~ 104° W) during years 1992 - 2011. Quadratic functions give good approximations for the general CO<sub>2</sub> 19-year growth rates at different altitudes in the troposphere. The averaged over altitudes 4 – 8 km 19-year mean CO<sub>2</sub> mole fraction related to year 2002 is  $372.1 \pm 0.1$  ppm, its mean growth rate is  $1.97 \pm 0.02$  ppm/yr and acceleration of the growth is  $0.019 \pm 0.01$  ppm/yr<sup>2</sup>. Observed CO<sub>2</sub> seasonal cycles, also amplitudes and phases of their spectral components are less variable in the troposphere above altitude 4 – 5 km than below. This may reflect better mixing and larger influence of atmospheric circulation there, than at lower altitudes. Annual and semiannual components could prevail in average CO<sub>2</sub> seasonal cycle in the troposphere above altitude 4 – 5 km, while shorter period components are more important at lower altitudes. The amplitude of the semiannual component grows in time faster than the amplitude of annual component. In the lower part of the troposphere, transitions from low-altitude CO<sub>2</sub> characteristics (partly influenced by local sources) to more homogeneous ones in the upper troposphere are observed.

### 1. Introduction

For attaining a better understanding of the carbon global cycle and its impact on climate change, it is necessary to analyze characteristics of atmospheric constituent variations over time intervals tens of years. Until recently, the majority of CO<sub>2</sub> monitoring stations have employed the flask sampling method of CO<sub>2</sub> mole fraction determination at the Earth's surface (Conway et al., 2003) or in the troposphere using aircraft. Extensive results of multiyear CO<sub>2</sub> measurements in the tropo-stratosphere with scientific and commercial aircraft were published previously (Pearman and Beardsmore, 1984; Matsueda et al., 2002; Levin et al., 2002; Gerbig et al., 2003; Font et al., 2008; Sawa et al., 2012, etc.). Most of these studies were concentrated on studies of horizontal CO<sub>2</sub> structures at several selected levels in the tropo-stratosphere. At the same time knowledge of average vertical profiles of CO<sub>2</sub> mole fraction in different locations is essential for different applications and assessments. Gavrilov et al. (2005) made a preliminary analysis of vertical changes in the CO<sub>2</sub> mole fraction and its seasonal variations in the troposphere from the data of flask aircraft measurements over Colorado, USA. In this study we extend this analysis to a longer time interval from 1992 to 2011. The aircraft measurements are made near the Rocky

Mountains. The aircraft data are compared with surface data from relatively nearby flask sampling sites.

The statistics of multiyear aircraft observations in the free atmosphere may be useful for the study of climate changes in the atmosphere and for other purposes. For example, the data may be essential for interpreting the data of ground-based and satellite optical observations, which have been extensively developed recently (Yang et al., 2002; Yokota et al., 2009; Deutscher et al., 2010; Wunch et al., 2011, Oshchepkov et al., 2012).

## 2. Aircraft CO<sub>2</sub> Measurements and Data.

Aircraft measurements of the CO<sub>2</sub> mole fraction over Colorado, USA, were started in 1992 after launching the NOAA/(CMDL-ESRL) Carbon Cycle Greenhouse Gases (CCGG) aircraft program that has been dedicated to collecting air samples in vertical profiles over (mostly) North America. The program's goals are to capture seasonal and inter-annual changes in trace gas mole fractions throughout the boundary layer and free troposphere (up to 8500m). Samples are collected over the flat, Pawnee National Grassland, and agricultural land near Briggsdale (40.37° N, 104.3° W, 1490 masl) in northeastern Colorado, and have been collected over nearby Carr, Colorado. The Carbon Cycle group's longest vertical-profile record comes from this area, with sample collection beginning in November 1992 during the initial development and testing phase of the automated flask sampling system. Sampling flights were originally conducted over Carr (surface elevation 1740 m), but in December 2004 the site was moved  $\approx$  40 miles southeast to Briggsdale (surface elevation 1490 m). Measurements are made at altitudes 2 – 8 km using laboratory analysis of the flasks for CO<sub>2</sub> with a precision of  $\pm 0.07$  ppm (Conway et al., 1994). We used more than 500 vertical profiles obtained during years 1992 – 2011.

Vertical profiles of the CO<sub>2</sub> mole fraction measured with aircrafts are variable from day to day. Sometimes substantial vertical CO<sub>2</sub> changes are observed, while near constant CO<sub>2</sub> mole fraction in altitude may occur.

## 3. Multiyear average CO<sub>2</sub> profiles.

Problems for calculating multiyear average CO<sub>2</sub> mole fraction,  $X$ , and its temporal trends may be caused by substantial variability and seasonal variations, which should be filtered out. For estimating the mean value and linear trend of the CO<sub>2</sub> mole fraction from multi-year non-equidistant airplane measurements we first calculate least-square fits to polynomials in different layers having thickness  $\Delta z$  and average height  $z_i$ . To obtain smoother results these fitting layers are shifted with altitude steps less than  $\Delta z$ . Multiyear climatological CO<sub>2</sub> trend within each  $i$ -th height layer may be expressed as follows:

$$X(z_i) = X_i + \gamma_i(t - t_{ref}) + \beta_i(t - t_{ref})^2, \quad (1)$$

where  $t$  is time in years,  $t_{ref}$  is the reference time close to the middle of analyzed time interval,  $X_i$  is the average CO<sub>2</sub> mole fraction,  $\gamma_i$  and  $\beta_i$  are constants. Analysis shows that reliable climatological  $\beta_i$  values can be obtained for time intervals longer than 15 – 20 years. Therefore, we approximated long term CO<sub>2</sub> trends with quadratic function (1) for the entire 19-year interval only. For shorter time intervals we estimated linear trends omitting the last term in (1). The obtained quadratic or linear fits are subtracted and residuals are approximated with the sum of annual, semiannual, 4- and 3-month periodical components using least-square approximation for the analyzed time intervals. These approximated seasonal variations are subtracted from the data for each altitude layer. Then previously obtained CO<sub>2</sub> average values and linear trends are corrected using filtered data after removal of excessively large “outliers”, determined as residuals from the long-term trend and harmonic fit  $\pm 2$ -sigma. We used  $\Delta z = 1.5$  km.

Obtained average characteristics of CO<sub>2</sub> climatological trend (1) and amplitudes and phases of mentioned periodical components for the entire 19-year analyzed interval are shown in Figure 1. The entire set of Colorado aircraft measurements consists of two parts: measurements over Carr in years 1992 – 2004 and over Briggsdale in 2005 – 2011 (see section 2). To study possible differences in CO<sub>2</sub> statistical characteristics in different places and different time intervals, we analyzed those time intervals separately and presented the results in Figure 2.

One can see that differences between the mean CO<sub>2</sub> mole fractions  $X_i$  at different altitudes in Figures 1 and 2 do not exceed 1 ppm at low altitudes and are less than 0.3 ppm above altitude 4 km. Differences in CO<sub>2</sub> trends in Figures 1 and 2 are not larger than 0.02 ppm/year at altitudes 4 – 7 km. Parameters of CO<sub>2</sub> climatological trend (1) averaged over altitudes 4 – 8 km for all analyzed time intervals are presented in Table 1.

Figures 1 and 2 also show amplitudes,  $A_k$ , and phases,  $t_{mk}$ , expressed as days of maximums of spectral components  $A_k \cos[2\pi(t_{day} - t_{mk})/\tau_k]$  of CO<sub>2</sub> average seasonal cycles at different altitudes, where  $t_{day}$  is time in days of year;  $A_k$  and  $\tau_k$  are amplitude and period;  $k = 24, 12, 6, 4, 3$  correspond to biannual, annual, semiannual, 4- and 3-month components, respectively. Figures 1 and 2 also show the total numbers of measurements ( $N_{tot}$ ) and the total used ( $N$ ) in different 1.5 km thick sliding altitude layers. Figures 1 and 2 show that the annual component of CO<sub>2</sub> seasonal variations has maximum amplitudes (about 2.9 - 3.5 ppm). Amplitudes of other spectral components in Figures 1 and 2 do not exceed 1 ppm. Height variations of annual component amplitude in Figures 1 and 2 do not exceed 1 ppm above altitudes 4 – 5 km. Amplitudes of 6-month component grows with height, while amplitudes of 4- and 3-month components decrease in Figures 1 and 2. This reveals possible differences in spectral composition of average seasonal cycles at different altitudes. According to Figures 1 and 2, annual and semiannual components could prevail in average CO<sub>2</sub> seasonal cycles in the troposphere above altitude 4 – 5 km, while shorter period components are more important at lower altitudes, suggesting a regional surface source for the shorter components. This is illustrated in Figure 3 below, where the CO<sub>2</sub> seasonal cycle at altitude 2 km for aircraft measurements exhibits more shorter-period structures than those at higher altitudes.

Phases of CO<sub>2</sub> spectral components in Figures 1 and 2 are expressed in terms of the days of year,  $t_{mi}$ , when the maxima of respective components occur. For 12- and 6-month components phases grow with height in bottom plots of Figures 2, which means that their maxima occur later at larger altitudes. Shift in time between maxima of 4- and 3-month components at different altitudes are smaller than that of annual and semiannual components (see Figures 2). Vertical gradients of the spectral component phases may reflect speeds of their vertical propagation. The phase gradients of annual and semiannual components in Figures 1 and 2 are smaller above altitudes 4 – 5 km than below. This means that the upper part of the troposphere may be better mixed. Analysis of Figures 1 and 2 shows larger variability of the mean CO<sub>2</sub> mole fractions, amplitudes and phases of annual and semiannual components in the lower part of the troposphere below altitudes 4 – 5 km. At larger altitudes Figures 1 and 2 reveals more uniform values of  $X_i$  and characteristics of annual and semiannual components. Table 1 represents characteristics of CO<sub>2</sub> trends and components of seasonal cycles averaged over altitudes 4 – 8 km.

#### 4. Interannual CO<sub>2</sub> Variations.

To analyze time variations of CO<sub>2</sub> trends we use the GLOBALVIEW-CO2 (2011) data obtained using the standard method developed in NOAA/(CMDL-ESRL). The trend is calculated as the low-frequency part of the spectrum of CO<sub>2</sub> variations. The method uses least squares approximation of the curve consisting of the third-order polynomial and annual, semi-annual, 4- and 3-month harmonics that provide the best fit to the experimental data. Then the interannual variations and seasonal anomalies are estimated by smoothing the residuals from the curve fit (see Thoning et al., 1989). Figure 3 shows measured CO<sub>2</sub> mole fractions and their trend

components obtained from the Colorado airplane data and from nearby surface CO<sub>2</sub> flask observations. One can see the similarity of variations at different altitudes.

Dashed lines in the upper plots of Figure 3 show the average quadratic approximations (1) calculated using parameters from the right column of Table 1. One can see that (1) gives good enough approximations for climatological trends of CO<sub>2</sub> mole fractions at altitudes above 4 km over Colorado. At lower altitudes dashed lines in Figure 3 underestimate observed CO<sub>2</sub> mole fractions. This is consistent with increases in average CO<sub>2</sub> mole fractions  $X_i$  at low altitudes in Figures 1 and 2. The GLOBALVIEW-CO<sub>2</sub> (2011) trends shown with solid lines in Figure 3 in different years may be larger or smaller than the mean  $\gamma$  values. Kashin et al. (2008) showed an existence of long-term spectral components with periods 21 – 183 months in CO<sub>2</sub> interannual variations. The GLOBALVIEW-CO<sub>2</sub> (2011) uses low-pass filters with approximately 1-year cutoff, therefore its trends contain contributions from these long-term interannual changes (Francey et al., 2010).

Upper plots of Figure 3 show very close seasonal variations above altitudes 4 – 5 km in most years. Figure 4 presents average for years 1992 - 2011 annual cycles of CO<sub>2</sub> mole fractions at different altitudes from GLOBALVIEW-CO<sub>2</sub> (2011) data. One can see very close variations at altitudes 4 – 7 km from aircraft measurements over Colorado (thin lines), which correspond to the seasonal variation calculated using parameters of spectral components presented in Table 1 and shown in Figure 4. Amplitudes of seasonal variations in the upper troposphere are smaller than those observed at nearby surface flask stations Niwot Ridge (3520 m altitude) and Utah (1320 m). Figure 4 shows time shifts between the seasonal maxima and minima of CO<sub>2</sub> mole fractions at Niwot Ridge and Utah. Differences between surface and upper tropospheric CO<sub>2</sub> variations may be connected with larger dependences of surface data on local CO<sub>2</sub> sources and sinks. One of the reasons might be a differences in vegetation cycles and differences in air mass transported to different altitudes.

## 5 Discussion.

Coefficient  $\beta$  in (1) is related to the acceleration of the CO<sub>2</sub> growth  $dy/dt = d^2X/dt^2 = 2\beta \approx 0.019 \pm 0.01$  ppm/yr<sup>2</sup> for the  $\beta_{aver}$  value presented in Table 1 for years 1992 – 2011. This value is smaller than average  $dy/dt = 0.023$  ppm/yr<sup>2</sup> obtained from the Mauna Loa 50 year CO<sub>2</sub> measurements (Francey et al., 2010). Also, the  $\beta_{aver}$  value in Table 1 is smaller than  $\beta \sim 0.02$  ppm/yr<sup>2</sup>, which can be obtained for year 2002 from the expression for exponential growth of anthropogenic CO<sub>2</sub>, obtained by Hofmann et al. (2009). Francey et al. (2010) showed that  $dy/dt$  is highly variable and may contain components with periods tens of years. Such long-term variations and possible relocation of the observation site from Carr to Briggsdale may cause substantial statistical errors of  $\beta_{aver}$  value given in Table 1. Therefore, further accumulating the data of multiyear CO<sub>2</sub> measurements is essential for more accurate estimations of  $\beta$ .

As mentioned above, Figures 1 – 4 show that observed CO<sub>2</sub> seasonal cycles and amplitudes and phases of their spectral components are more stable in the troposphere above altitude 4 – 5 km than below. The evolution of the CO<sub>2</sub> seasonal cycle at different altitudes may result from the interaction between the local/regional sources and mixing, and large-scale circulation. Above mountains heights (4-5 km in Colorado) a mixture of air parcels coming from far away CO<sub>2</sub> sources may prevail and CO<sub>2</sub> characteristics are more homogeneous in Figures 1 - 4. Larger stability of average CO<sub>2</sub> mole fractions and seasonal cycles in the upper part of the troposphere may show better mixing and larger influence of large-scale circulation there than at lower heights. This may be especially true for the latitude band, where Colorado (USA) is located. Air parcels circulating along the 40° N latitude circle may go above a number of the main mountain systems of the northern hemisphere (Rocky Mountains, Pyrenees, Tien-Shan, etc.). These mountains may produce orographic waves, turbulence and other mesoscale and small-scale perturbations, which propagate to the tropo-stratosphere. An analysis of mesoscale temperature variances at different heights along the latitude circle of 40° N by Gavrillov (2007) from

observations with the low-orbit GPS satellite CHAMP showed substantial perturbations in the tropo-stratosphere over the above mentioned mountains, which may produce increased mixing there.

Influence of local sources on CO<sub>2</sub> mole fractions and seasonal cycles could be larger near the ground and in the planetary boundary layer (see, for example, Haspra et al, 2012; Jiang et al., 2012). At Carr and Briggsdale below altitudes 4 - 5 km a quasi-horizontal CO<sub>2</sub> transport from sources located at elevated surfaces of the Rocky Mountains and from the plains to the east of the mountains may become essential due to local circulation cells. The proportion between air parcels coming from local and far away sources may increase at lower altitudes. Therefore, a broad transition zone may occur from low-altitude CO<sub>2</sub> characteristics (influenced by local sources) to more homogeneous ones in the middle troposphere above 4 – 5 km. Sometimes such transition may be smooth (see, for example, phases of annual component in Figures 1 and 2) or sharp (phases of semiannual component at low altitudes). Therefore, CO<sub>2</sub> mole fractions and their seasonal cycles may be more variable at low altitudes than in the upper troposphere, as it is seen in Figures 1 – 4.

Also CO<sub>2</sub> characteristics in the lower troposphere may have larger dependences on locations of observation sites. For example, Haspra et al. (2012) reported substantially larger amplitudes of CO<sub>2</sub> seasonal cycle (up to 14 ppm) at altitudes 10 -100 m than those at 2.5 – 3 km over Europe in years 2001- 2008. Comparisons of curves in Figure 2 for observations in years 1992 - 2004 over Carr and in 2005 - 2012 over Briggsdale show substantial differences in amplitudes of annual, semiannual and 3-month components of CO<sub>2</sub> seasonal variations at low altitudes. Part of these differences may be caused by differences in local CO<sub>2</sub> sources and sinks influencing the two observation sites. Differences in CO<sub>2</sub> seasonal variations at low altitudes for the two mentioned time intervals were also obtained using the traditional method (Thoning et al., 1989). Further studies of CO<sub>2</sub> seasonal cycles at different altitudes and locations are required.

Multiyear average amplitudes of the annual and semiannual components in Figures 1, 2 and in Table 1 grow in time. One of the reasons for these components could be seasonal cycles of fossil-fuel CO<sub>2</sub> emissions (Jiang et al., 2012). Observed summer maxima of these emissions may form semiannual CO<sub>2</sub> variations (Blasing et al., 2005). The summer maxima of CO<sub>2</sub> anthropogenic emissions may be caused, for example, by air-conditioning activity, which requires increasing in coal and natural gas burning at power station. Transporting upwards and mixing in the free atmosphere CO<sub>2</sub> from these sources may produce substantial semiannual oscillations (Jiang et al., 2012) having maxima in summer and winter (see Figures 1, 2 above 3 – 4 km and Table 1). Air-conditioning is less required in cool mountain regions, and local fossil-fuel emissions in clean regions near Carr and Briggsdale may have no substantial increase in summer. This may explain small amplitudes of semiannual CO<sub>2</sub> variation at low altitudes in Figures 1 and 2.

Analysis of Figure 2 and Table 1 reveals substantial height and temporal increases in the amplitudes of CO<sub>2</sub> semiannual component in the troposphere above altitudes 4 – 5 km. This is consistent with observed increases in amplitudes of summer maxima of fossil-fuel CO<sub>2</sub> emission with time (Blasing et al., 2005). Table 1 shows 30% increase in average semiannual CO<sub>2</sub> amplitudes at altitudes 4 – 8 km from years 1992 – 2004 to 2005 – 2011. This is larger than the respective increase of 7% in CO<sub>2</sub> annual amplitudes in Table 1. So big increase in CO<sub>2</sub> semiannual amplitude in Table 1 may be partly caused by relocation of aircraft observations from Carr to Briggsdale and requires further clarification. Jiang et al. (2012) analyzed data of different surface GLOBALVIEW-CO<sub>2</sub> stations and they found larger amplitudes of semiannual CO<sub>2</sub> component in Central and Eastern Asia at latitudes 35 - 45° N, where air-conditioning is probably more extensive. Mixed in the atmosphere and transported by circulation to other places, CO<sub>2</sub> from these sources may produce amplitudes of semiannual component in the free troposphere exceeding respective local surface amplitudes, as we may observe in Figures 1 and 2. These Figures show also that phases of semiannual CO<sub>2</sub> component at altitudes above 4 – 5 km are quite stable in different time intervals and locations. Therefore, observations of

semiannual component in the free troposphere could be useful for further understanding peculiarities of global CO<sub>2</sub> sources and transport.

Approximations of CO<sub>2</sub> seasonal cycles include traditionally the 4- and 3-month components (see Thoning et al., 1989). Such components may arise due to asymmetry of seasonal cycles of fossil-fuel CO<sub>2</sub> emission, which have the main maximum in December-January and the secondary maximum in August (Blasing et al., 2005). Time differences between these maxima are 7 - 8 months in spring and 4 - 5 months in autumn. Similar asymmetry is observed for photosynthesis-respiration cycles, when the CO<sub>2</sub> drawdown always takes less time (may through august) than the increase during fall-winter-early spring. Decomposition of such asymmetric seasonal cycles into Fourier series may give substantial amplitudes not only for annual and semiannual, but also for higher order components, which may produce respective CO<sub>2</sub> variations in the atmosphere. Other sources of 4- and 3-month components could be weather conditions, droughts, forest fires, etc., which may produce local variations of vegetation and CO<sub>2</sub> emission with time scales of several months.

Figures 1 and 2 show small amplitudes of 4- and 3-month components in the troposphere above 4 - 5 km, where annual and semiannual components may prevail in CO<sub>2</sub> seasonal variations. At lower altitudes, amplitudes of higher order components are larger in Figures 1 and 2, and they may depend on time and measurement locations. Analysis of Figure 2 reveals larger amplitudes of 4-month component and smaller amplitudes of 3-month component at low altitudes in years 2005 - 2011 over Briggsdale, than those in 1992 - 2004 over Carr in Figure 2. This may show differences in local CO<sub>2</sub> sources at the two locations. In Figures 1 and 2 phases (times of maxima and minima of 4-month component) are decreasing at altitudes 3 - 6 km. This may be caused by transition from later maxima of 4-month component produced by local sources at low altitudes to earlier its maxima observed in the mixed air parcels coming from far-away sources to the upper troposphere. For all other components times of their maxima are larger in the upper troposphere than at low altitudes. Therefore, their phases generally grow with height in Figures 1 and 2.

One of the main assumptions of spectroscopic methods of CO<sub>2</sub> determination is uniformity of CO<sub>2</sub> mole fraction at all altitudes in the atmosphere. Figures 2 and 3 show larger changes in the mean CO<sub>2</sub> mole fraction and amplitudes and phases of its annual and semiannual components in the lower part of the troposphere compared to the upper troposphere. The transition height between these parts of the troposphere over Colorado is 4 - 5 km according to Figures 2 and 3, which corresponds to the pressure level  $p_1 \approx 600$  mb. Highest altitude of used airplane measurements in Colorado is 8.5 km (or  $p_2 \approx 330$  mb). Taking standard surface pressure to be  $p_0 \approx 1000$  mb, we can estimate the weights of contributions of mentioned lower and upper troposphere layers to the total column CO<sub>2</sub> content at a sea level station making spectroscopic observations to be 40% and 27%, respectively. The rest 33% of column CO<sub>2</sub> is located in the middle atmosphere above the altitude region covered with aircraft flask measurements. Therefore, seasonal variations in total column CO<sub>2</sub> abundance observed with spectroscopic ground-based measurements may combine local peculiarities of CO<sub>2</sub> sources and sinks in the lower atmosphere, processes of transport and mixing of atmospheric gases, and global-scale CO<sub>2</sub> changes at different altitudes up to the middle atmosphere. Contributions of different layers to the column abundance depend on mentioned above weights of different layers and the activities of CO<sub>2</sub> variations in these layers. Comparison of the spectroscopic and aircraft flask measurements may help in determination of relative contribution of local and global-scale cycles into total CO<sub>2</sub> contents in atmospheric columns.

Our analysis includes the 24-month spectral component of CO<sub>2</sub> variations, parameters of which are calculated using least-square fit to detrended aircraft measurements and are shown in Figures 1, 2 and in Table 1. Phases of 24-month component in Table 1 correspond to its maxima in winters of

odd years and minima in winters of even years at altitudes 4 – 8 km. This component may be connected with quasi-biannual oscillations frequently observed in variations of other atmospheric parameters (Wang et al., 2011). Kashin et al. (2008) made spectral analysis of the data of about 30-year long ground-based spectroscopic CO<sub>2</sub> observations at Issyk-Kul Monitoring Station (see ISK 242 N00 in WMO WDCGG, 2006) in Kyrgyzstan, 42° N, 77° E, 1650 masl. This station is located at latitude close to that of Colorado aircraft observations. Kashin et al. (2008) found a broad spectral maximum at periods 20 – 25 months, which can be attributed to the quasi-biannual oscillations. Also, they found several other spectral maxima having longer periods up to 183 months. Similar maxima are observed in interannual variations of atmospheric temperatures, winds and gas species (Jiang et al., 2010; Visheratin, 2012). Therefore, long-term aircraft CO<sub>2</sub> measurements may help in studies of interannual changes in atmospheric dynamics and their influence on the transport of gas species and atmospheric composition at different altitudes.

## 6. Conclusion

A statistical analysis of airplane and surface CO<sub>2</sub> measurements over Colorado, USA, during years 1992 – 2011 shows that general trends and seasonal CO<sub>2</sub> variations are almost the same at altitudes 4 – 8 km in well mixed region over the Rocky Mountains. Least-square fitted polynomial functions give good enough approximations for general CO<sub>2</sub> 19-year grows at different altitudes in the troposphere. The 19-year CO<sub>2</sub> mole fraction averaged over altitudes 4 – 8 km and related to year 2002 is  $372.1 \pm 0.1$  ppm, its mean growth rate is  $1.97 \pm 0.02$  ppm/yr and acceleration of the grows is  $0.019 \pm 0.01$  ppm/yr<sup>2</sup>. Observed CO<sub>2</sub> seasonal cycles and amplitudes and phases of their spectral components in Figure 2 are less variable in the troposphere above altitude 4 – 5 km than below. This may reflect better mixing and larger influence of large-scale circulation there, than at lower altitudes. Annual and semiannual components could prevail in average CO<sub>2</sub> seasonal cycle in the troposphere above altitude 4 – 5 km, while shorter period components are more important at lower altitudes. Amplitudes of semiannual component grow in time faster than amplitudes of annual component. Phases of semiannual component of CO<sub>2</sub> seasonal cycle are quite stable in time in the upper troposphere. In the lower part of the troposphere, transitions from low-altitude CO<sub>2</sub> characteristics (partly influenced by local sources) to more homogeneous ones in the upper troposphere are observed. Long-term aircraft CO<sub>2</sub> measurements may be useful in studies of interannual atmospheric variations and their influence on the transport of gas species and composition at different altitudes.

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**Figure captions.**

**Figure 1.** Total,  $N_{tot}$ , and used in the analysis,  $N$ , numbers of aircraft CO<sub>2</sub> measurements at different altitudes over Colorado in years 1992 – 2011; average CO<sub>2</sub> mole fractions  $X$ , in ppm and its grows rate  $\gamma$  in ppm/year; amplitudes,  $A_k$ , in ppm and phases,  $t_{mk}$ , as the day of maximum of 24-, 12-, 6-, 4-, 3-month components of CO<sub>2</sub> variations.

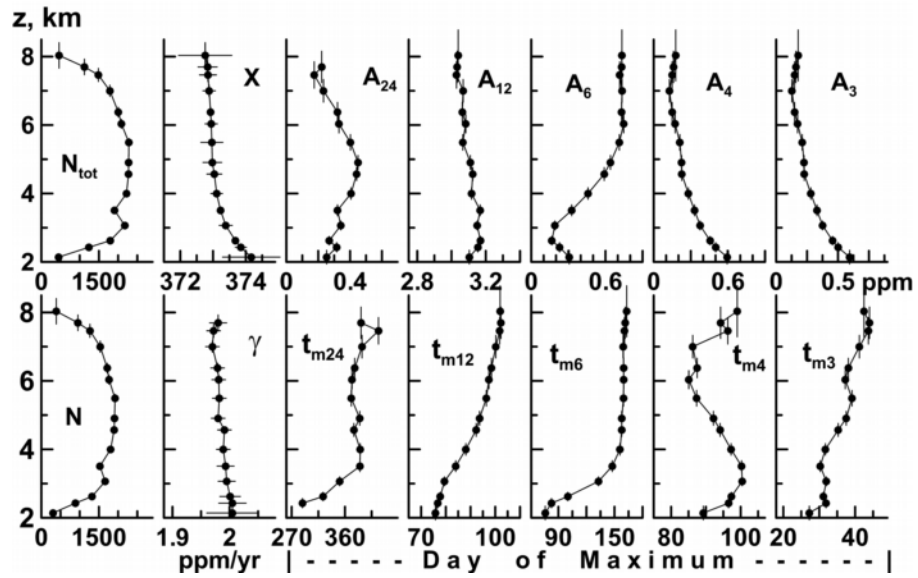
**Figure 2.** Same as Figure 1, but for CO<sub>2</sub> measurements at different altitudes over Carr in years 1992 – 20004 and Briggsdale in 2005 - 2011.

**Figure 3.** Variations and trends of CO<sub>2</sub> mole fraction from Colorado aircraft measurements at different altitudes (upper plots) and from ground-based flask measurements in stations Utah (UTA) and Niwot Ridge (NWR) according to GLOBALVIEW (2011) data. Dashed lines show average quadratic trends (1) calculated using coefficients given in Table 1 for years 1992-2011.

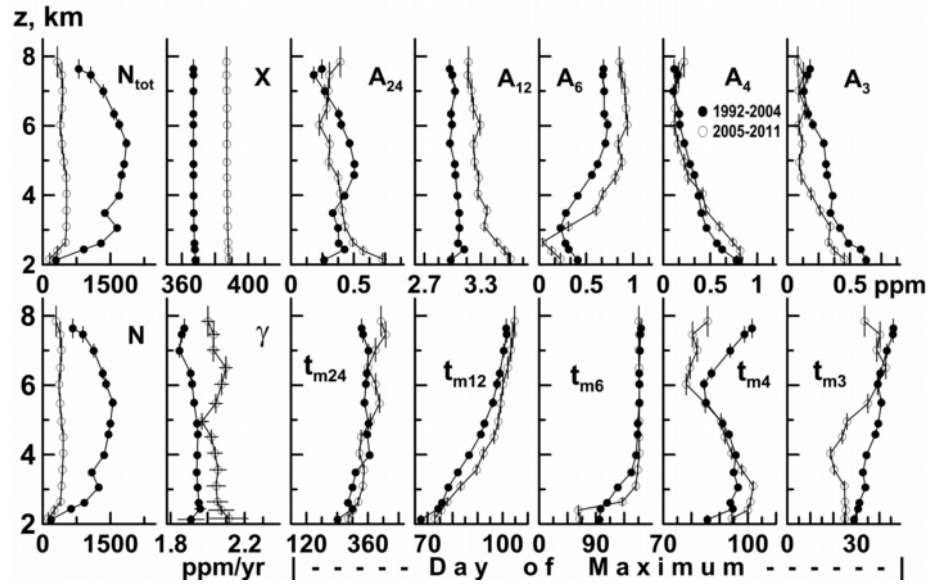
**Figure 4.** Comparison of average seasonal CO<sub>2</sub> variations observed with aircrafts in Colorado at altitudes 4, 5, 6, 7 km (1), at ground flask stations Niwot Ridge (2), Utah (3), and calculated from parameters of Table 1 (4).

Table 1. Average CO<sub>2</sub> statistical characteristics at altitudes 4 – 8 km in different time intervals.

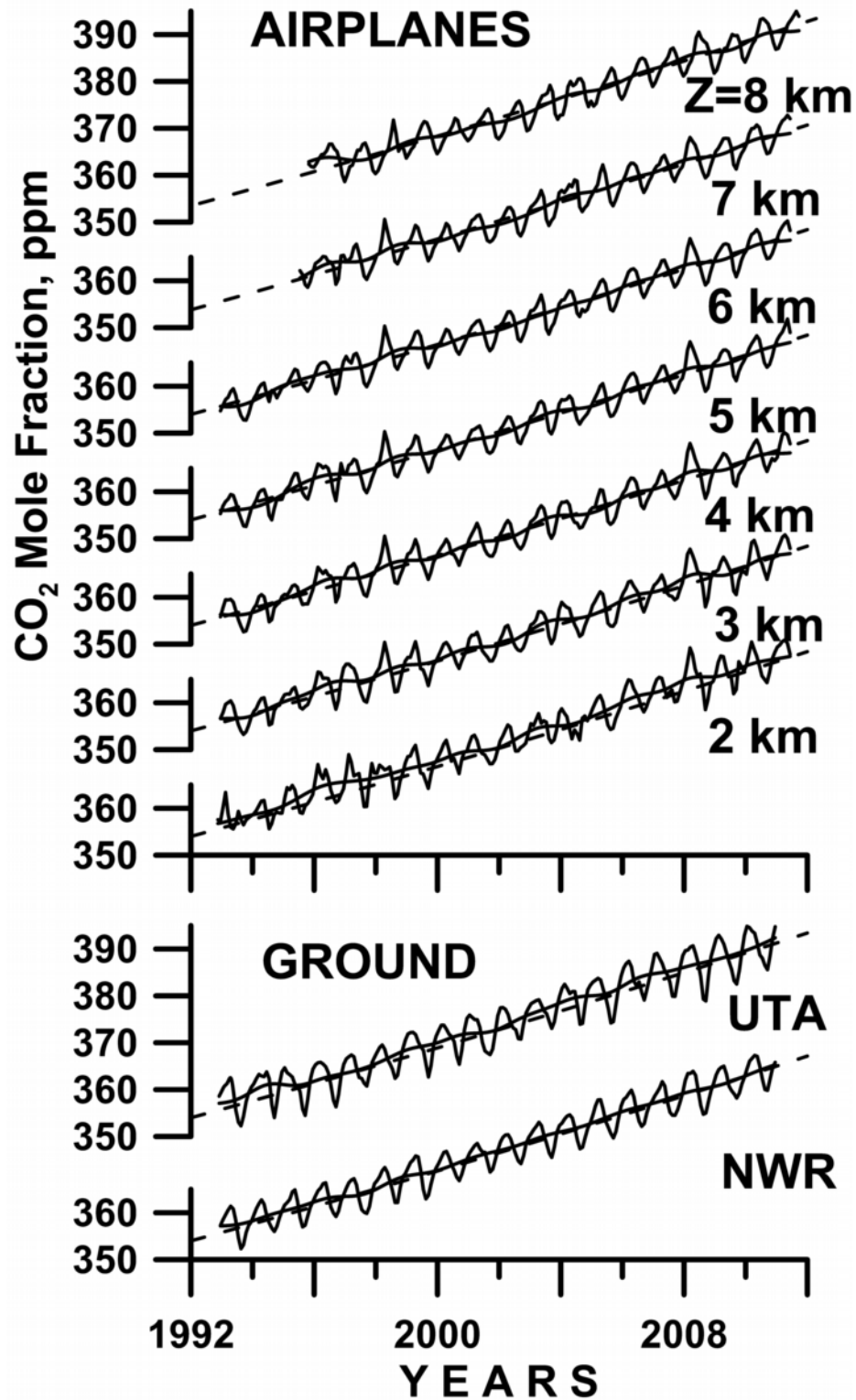
Parameter/Years	1992-2004	2005-2011	1992-2011
N	3175	1043	4322
t <sub>ref</sub> , year	1999	2009	2002
X <sub>aver</sub> , ppm	367.0±0.1	387.1±0.1	372.7±0.1
γ <sub>aver</sub> , ppm/yr	1.91±0.03	2.06±0.03	1.97±0.02
β <sub>aver</sub> , ppm/yr <sup>2</sup>			(9.6±5)10 <sup>-2</sup>
A <sub>24</sub> , ppm	0.4±0.1	0.3±0.1	0.3±0.1
t <sub>m24</sub> , day	351±10	386±30	384±15
A <sub>12</sub> , ppm	3.00±0.02	3.22±0.05	3.07±0.02
t <sub>m12</sub> , day	97±4	101±3	98±4
A <sub>6</sub> , ppm	0.67±0.05	0.88±0.04	0.70±0.06
t <sub>m6</sub> , day	159±2	159±1	159±1
A <sub>4</sub> , ppm	0.20±0.08	0.18±0.06	0.17±0.08
t <sub>m4</sub> , day	92±6	84±5	90±4
A <sub>3</sub> , ppm	0.22±0.07	0.11±0.02	0.18±0.04
t <sub>m3</sub> , day	41±4	35±7	39±3



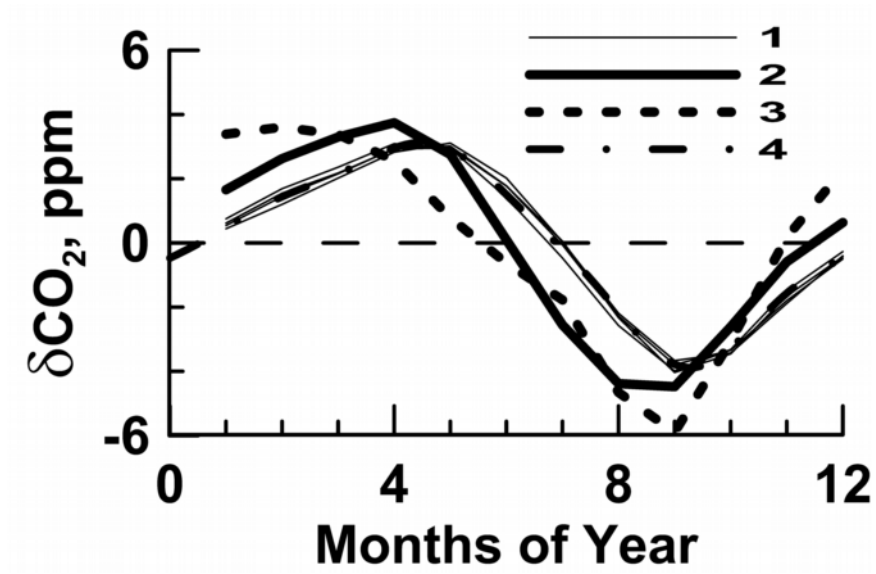
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