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2 **CO₂ Semi-annual Oscillation in the Middle Troposphere and at the**
3 **Surface**
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1 **Abstract:**

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3 Using in-situ measurements, we find a semi-annual oscillation (SAO) in the mid-
4 tropospheric and surface CO₂. Chemistry-transport models (2-D Caltech/JPL model, 3-D
5 GEOS-Chem, and 3-D MOZART-2) are used to investigate possible sources for the SAO
6 signal in the mid-tropospheric and surface CO₂. From model sensitivity studies, it is
7 revealed that the SAO signal in the mid-tropospheric CO₂ mainly originates from surface
8 CO₂ with a small contribution from transport fields. It is also found that the source for the
9 SAO signal in surface CO₂ is mostly related to the CO₂ exchange between biosphere and
10 atmosphere. By comparing model CO₂ with in-situ CO₂ measurements at surface, we find
11 that models are able to capture both annual and semi-annual cycles well at surface.
12 Models can also simulate annual and semi-annual cycles of mid-tropospheric CO₂
13 reasonably well in the tropical region by comparison with aircraft measurements.
14

1. Introduction

Atmospheric CO₂ has a global trend of ~2 ppm/year [Keeling *et al.*, 1995]. The increasing of atmospheric CO₂ has a significant impact on the global climate change [Dickinson and Cicerone, 1986]. Superimposed upon this trend is an annual cycle resulting from the uptake and release of CO₂ by vegetation whose amplitude is greatest in the northern hemisphere (NH). Using CO₂ measurements at Mauna Loa, Buermann *et al.* [2007] found that variations of CO₂ seasonal cycle amplitudes are closely related to the carbon sequestration in the biosphere, and are influenced by precipitation and circulation. In addition to the trend and annual cycle, atmospheric CO₂ also shows intra-seasonal and inter-annual variabilities.

El Niño is the most important tropical interannual variability that can influence the CO₂ concentrations. During El Niño (La Niña) events, the atmospheric CO₂ growth rate increases (decreases) at tropical surface stations [Keeling *et al.*, 1995; Jones *et al.*, 2001; Nevison *et al.*, 2008]. Using mid-tropospheric CO₂ data from AIRS, Jiang *et al.* [2010] found that El Niño can influence the mid-tropospheric CO₂. Mid-tropospheric CO₂ is enhanced in central Pacific Ocean and diminished in the western Pacific Ocean during El Niño [Jiang *et al.*, 2010]. In the high latitudes, mid-tropospheric CO₂ concentration can be influenced by the strength of the polar vortex. Polar mid-tropospheric CO₂ is reduced (enhanced) when the polar vortex is strong (weak) [Jiang *et al.*, 2010]. Recently, Li *et al.* [2010] demonstrate that mid-tropospheric CO₂ concentrations can be influenced by the Madden-Julian Oscillation.

In this paper, we will focus on investigating the intra-seasonal variability of mid-tropospheric CO₂, especially on the semi-annual oscillation (SAO) of CO₂ and its possible sources. This work will yield a quantitative study of how SAO influences the mid-tropospheric CO₂. It also offers an opportunity to investigate the possible source for the SAO signal in the mid-tropospheric CO₂.

2. Data and Models

1
2 In this paper, we use aircraft CO₂ from *Matsueda et al.* [2002], which are incorporated
3 into Comprehensive Observation Network for Trace gases by AirLiner (CONTRAIL).
4 Aircraft CO₂ from *Matsueda et al.* [2002] are measured at 8-13 km biweekly since April
5 1993 to present. CO₂ over the western Pacific from Australia to Japan are measured. The
6 latitudinal coverage is approximately from 25°S to 35°N. The longitudinal coverage is
7 from 135°E to 150°E. We also use surface CO₂ flask measurements from NOAA ESRL
8 network [*Tans et al.* 1998; *GLOBALVIEW-CO₂*, 2007]. Site information for NOAA
9 surface CO₂ is available at http://www.esrl.noaa.gov/gmd/dv/site/site_table.html.

10
11 To investigate possible sources of the semi-annual oscillation of mid-troposphere and
12 surface CO₂, we use three different chemistry-transport models. These models are the
13 Caltech/JPL 2-D chemistry-transport model (CTM) [*Shia et al.*, 2006], 3-D GEOS-Chem
14 [*Suntharalingam et al.*, 2004], and 3-D MOZART-2 [*Horowitz et al.*, 2003]. The 2-D
15 CTM has 18 latitudes, equally spaced from pole to pole. It has 40 vertical layers, equally
16 spaced in log scale of pressure from the surface to the upper boundary at 0.01 hPa.
17 Transport in the model is by the stream function and the horizontal and vertical
18 diffusivities taken from *Jiang et al.* [2004]. The stream function is derived from the
19 National Center for Climate Prediction (NCEP) Reanalysis 2 data [*Jiang et al.*, 2004]. An
20 important feature of the 2-D CTM is its ability to reproduce the age of air in the
21 stratosphere [*Morgan et al.*, 2004].

22
23 GEOS-Chem (v7.3.3) is driven by the Goddard Earth Observing System (GEOS-4)
24 assimilated meteorological data from the NASA Global Modeling Assimilation Office
25 (GMAO). Spatial resolution for GEOS-Chem is 2° (latitude) × 2.5° (longitude). There are
26 30 levels in the vertical from the surface to about 0.01 hPa (~70 km). Advection is
27 computed every 15 minutes with a flux-form semi-Lagrangian method [*Lin and Rood*,
28 1996]. Moist convection is computed using the GEOS convective, entrainment, and
29 detrainment mass fluxes described by *Allen et al.* [1996a, 1996b]. The physics in the
30 GEOS-4 analysis system is adopted from the National Center for Atmospheric Research
31 (NCAR) Community Climate Model, Version 3 (CCM3) and Whole Atmosphere

1 Community Climate Model (WACCM) with important modifications to make it suitable
2 for data assimilation [Bloom *et al.*, 2005].

3
4 MOZART-2 is driven by the meteorological inputs every 6 hours from the NCEP
5 Reanalysis 1 [Kalnay *et al.*, 1996]. Advection is computed every 20 minutes with a flux-
6 form semi-Lagrangian method [Lin and Rood, 1996]. The horizontal resolution is 2.8°
7 (latitude) \times 2.8° (longitude) with 28 vertical levels extending up to approximately 40 km
8 altitude [Horowitz *et al.*, 2003]. MOZART-2 is built on the framework of the Model of
9 Atmospheric Transport and Chemistry (MATCH). MATCH includes representations of
10 advection, convective transport, boundary layer mixing, and wet and dry deposition.

11
12 Surface emissions and vertical transport in CTMs are both crucial for CO₂ simulation in
13 the free troposphere. We will use two different boundary conditions to investigate how
14 boundary conditions affect the mid-tropospheric CO₂. The GLOBALVIEW-CO₂ mixing
15 ratio data [Tans *et al.* 1998; GLOBALVIEW-CO₂, 2007] is used as the lower boundary
16 condition for the Caltech/JPL CTM, GEOS-Chem, and MOZART-2. For convenience,
17 we refer this hereforth as the GLOBALVIEW-CO₂ boundary condition [Jiang *et al.*,
18 2008]. Since the GLOBALVIEW-CO₂ data are limited in space, especially over ocean,
19 we used the GLOBALVIEW-CO₂ to rescale the CO₂ mixing ratio at the surface for the
20 GLOBALVIEW-CO₂ boundary condition. First, monthly mean model CO₂ mixing ratios
21 at surface are regressed against GLOBALVIEW-CO₂ surface flask measurements. Then
22 model surface CO₂ mixing ratios are rescaled by the scale derived from regression. The
23 monthly mean GLOBALVIEW-CO₂ flask data are close to the GLOBALVIEW-CO₂
24 boundary condition when they are co-located.

25
26 We will also use the prescribed CO₂ sources and sinks as the boundary condition for
27 GEOS-Chem and MOZART-2. The exchange of CO₂ between the terrestrial biosphere
28 and atmosphere is based on net primary productivity and respiration fluxes from the
29 Carnegie-Ames-Stanford (CASA) ecosystem model [Randerson *et al.*, 1997]. Monthly
30 mean biospheric CO₂ fluxes from 2000 to 2004 are used in the models. Air-to-sea
31 exchange of CO₂ is from Takahashi *et al.* [1997]. Estimates of fossil fuel emissions are

1 from *Marland et al.* [2007]. Monthly mean biomass burning emissions of CO₂ are
2 derived based on *Duncan et al.* [2003]. Since there is an unbalanced CO₂ budget
3 associated with the prescribed source and sink boundary condition [*Suntharalingam et al.*,
4 2003; *Suntharalingam et al.*, 2004], we regress surface CO₂ mixing ratio in the GEOS-
5 Chem restart file against the GLOBALVIEW-CO₂ surface flask measurements. As a
6 result, the unbalanced CO₂ budget is resolved to some degree [*Jiang et al.*, 2008].
7 Discrepancies between the model CO₂ simulations (driven by the same meteorological
8 fields) with the above-mentioned two boundary conditions would help identify potential
9 issues with the surface sources and/or sinks on simulating CO₂ annual and semi-annual
10 cycles.

12 **3. Results**

14 Figure 1 presents a comparison between Matsueda's aircraft CO₂ (red dots) and model
15 CO₂ mixing ratios averaged between 9 km and 13 km (solid lines) from 2000 to 2004.
16 The panels are for 25°S, 15°S, 5°S, 5°N, 15°N, and 25°N, respectively. Different color
17 lines are for different model simulations. There are two GEOS-Chem model outputs. One
18 is forced by the GLOBALVIEW-CO₂ boundary condition (green line). The other is
19 forced by the prescribed CO₂ sources and sinks boundary condition (orange line). GEOS-
20 Chem CO₂ forced by the prescribed CO₂ source/sink boundary condition (orange line)
21 have higher CO₂ concentrations in the summer seasons than that forced by
22 GLOBALVIEW-CO₂ boundary condition (green line). It suggests that there might be a
23 missing sink in the prescribed CO₂ source/sink boundary condition in the summer season.
24 Purple line is CO₂ from Caltech 2D model. Blue line is CO₂ from MOZART2 forced by
25 NCEP1 meteorology. The model results match the high precision aircraft measurements
26 of CO₂ in the middle troposphere remarkably well. Seasonal cycle and trend for CO₂ are
27 simulated well by different models. The amplitude of CO₂ seasonal cycle increases with
28 latitudes, with larger seasonal cycle in the northern hemisphere compared with that in the
29 southern hemisphere. In addition to the annual cycle, there is a six-month signal
30 appearing in the CO₂ from both aircraft and model simulations. To investigate the six-
31 month signal in more details, power spectral analysis is applied to the detrended CO₂

1 from aircraft and model simulations. Linear trends have been removed from CO₂ time
2 series. Power spectra for the detrended CO₂ are shown in Figure 2. In addition to the
3 spectral peak at 12 months, 6-month signal appears in both Matsueda's CO₂ and model
4 CO₂.

5
6 To investigate sources of the semi-annual oscillations in the mid-tropospheric CO₂, we
7 first apply sensitivity studies to the 2-D Caltech/JPL chemistry and transport model.
8 Averaged CO₂ at 9-13 km forced by the GLOBALVIEW-CO₂ boundary condition and
9 NCEP2 Meteorology field is shown by solid line in Figure 3. To investigate the annual
10 cycle and semi-annual cycle amplitudes in the mid-tropospheric CO₂, we fit the data by a
11 series of Legendre polynomials and harmonic functions [Jiang *et al.*, 2008]. We use the
12 sum of the first, second, and third Legendre polynomials to remove the trend from the
13 data. The harmonic functions represent annual and semi-annual cycles. Annual cycle,
14 calculated by $e \cos(2\pi t) + f \sin(2\pi t)$, is shown in Fig. 3b, where e and f are the
15 amplitudes of the annual cycle. The amplitude for the annual cycle of 2-D model CO₂ at
16 25°N is about 2.3 ppm. Semi-annual cycle, calculated by $g \cos(4\pi t) + h \sin(4\pi t)$, is
17 shown in Fig. 3c, where g and h are the amplitudes of the semi-annual cycle. The
18 amplitude for the semi-annual cycle of 2-D model CO₂ at 25°N is about 0.8 ppm. In a
19 sensitivity test, we removed the annual and semi-annual oscillations from surface CO₂
20 and used linear trends as the boundary condition at the model surface. As such, there is
21 no semi-annual cycle and annual cycle source originating from surface in this model run.
22 This results in the reduction of the amplitude of semi-annual cycle for mid-tropospheric
23 CO₂ to 0.09 ppm, which is about 11% of the total amplitude of the semi-annual
24 oscillation in the mid-tropospheric CO₂. It clearly suggests that the dominant source for
25 the semi-annual cycle in the middle troposphere is propagated from the surface sources.
26 Weak semi-annual and annual cycles in the mid-tropospheric CO₂ shown by dashed lines
27 in Figs. 3b and 3c originate from transport fields. The phase of the CO₂ seasonal cycle
28 due to the transport (dashed line in Figure 3b) is shifted relative to that forced by the
29 GLOBALVIEW-CO₂ boundary condition and NCEP2 reanalysis meteorology field (solid
30 line in Figure 3b). The CO₂ seasonal signal due to the transport (dashed line in Figure 3b)
31 is related to the strength of the vertical velocity in the 2D CTM. The vertical velocity is

stronger in the summer season and weaker in the winter season. More CO₂ can be lifted to the middle-troposphere during the summer than in the winter season. Thus mid-tropospheric CO₂ (dashed line in Figure 3b) reaches maximum value in the summer season at 25°N. Phase for the CO₂ seasonal cycle (forced by the GLOBALVIEW-CO₂ boundary condition and NCEP2 reanalysis meteorology field; solid line in Figure 3b) represents the contribution from surface CO₂ sources, which reach maximum in April. In another sensitivity test, we force the model with linear CO₂ trend boundary condition and climatological transport fields. Climatological transport fields are the average of the transport fields from 2000 to 2004. There is no semi-annual oscillation originating from either the surface or the transport fields. As a result, semi-annual oscillation disappears in mid-tropospheric CO₂ as shown by the dotted line in Fig. 3c. In independent sensitivity studies, we also find that the semi-annual oscillation in the mid-tropospheric CO₂ originates from the surface semi-annual cycles and not the surface annual cycles.

Annual and semi-annual oscillations in surface CO₂ are also examined. Similar spectral analysis is applied to the GLOBALVIEW-CO₂ and GEOS-Chem model CO₂ at the surface. In addition to the annual cycle, semi-annual oscillation signals are also present in the surface CO₂ from GLOBALVIEW-CO₂ and model CO₂. To compare the annual cycle and semi-annual cycle amplitudes in surface CO₂ from observations and model results, we calculate the annual cycle amplitude ($\sqrt{e^2 + f^2}$) and semi-annual cycle amplitude ($\sqrt{g^2 + h^2}$) for surface CO₂ from GLOBALVIEW network and GEOS-Chem model output. Results are shown in Figure 4. The amplitudes for annual and semi-annual cycles from GEOS-Chem CO₂ are very close to those from GLOBALVIEW-CO₂ at the surface. The amplitudes of annual and semi-annual cycles are larger in the northern hemisphere compared with those in the southern hemisphere. The maximum amplitude for the annual cycle of surface CO₂ is about ~10 ppm. The maximum amplitude is about ~3.5 ppm for the semi-annual cycle of surface CO₂. Scatter plots of the observed and model simulated amplitudes for semi-annual cycle and annual cycle of surface CO₂ are shown in Figure 5. As revealed in Figure 5, the GEOS-Chem model seems to over-estimate the semi-annual oscillation amplitudes compared with those from observations. The latitudinal

distributions of the amplitudes for the semi-annual and annual cycles are shown in Figure 6. The amplitude increases with latitude, which appears from both surface GLOBALVIEW-CO₂ data and models. Semi-annual and annual cycle amplitudes are larger in the northern hemisphere than those in the southern hemisphere. This is because semi-annual and annual cycles in surface CO₂ sources (e.g. the net exchange between biosphere and atmosphere) are larger in the northern hemisphere than those in the southern hemisphere.

To investigate possible sources for the semi-annual oscillation of CO₂ at the surface, we examine the signals from different CO₂ surface sources, which include biomass burning, fossil fuel emission, ocean, and biosphere. Fossil fuel emission contributes to the positive trend in CO₂. CO₂ semi-annual cycle and annual cycle are mainly from exchange between the atmosphere and the biosphere. Biomass burning also contributes to the semi-annual cycle of CO₂. Gross primary production, respiration, and net ecosystem production at 30°N and 110°E, shown in Figure 7, are an example to illustrate the semi-annual oscillation in CO₂ source from the biosphere. Gross primary production (Fig. 7a), is related to carbon uptake by plants during photosynthesis. The values are negative since CO₂ is uptaken by vegetation from the atmosphere. Ecosystem respiration (Fig. 7b), is related to the autotrophic and heterotrophic respirations from biosphere. In the winter season, photosynthesis is largely reduced. The peak for gross primary production (Fig. 7a), is relatively flat in winter. However, there are still CO₂ emitted to the atmosphere by respirations from the biosphere in winter, which has a relatively sharp peak compared with the photosynthesis term. The sum of the two terms, gross primary production and ecosystem respiration, leads to the double peaks in each year in the net ecosystem production, as shown in Fig. 7c. Thus, phase differences in the gross primary production (photosynthesis) and ecosystem respiration lead to the semi-annual oscillation in CO₂ at surface. Surface semi-annual oscillation can propagate to the middle-troposphere, which is the dominant source for the semi-annual oscillation in the mid-tropospheric CO₂.

4. Conclusions

1 In addition to the annual cycle, the semi-annual oscillation of mid-tropospheric and
2 surface CO₂ is discussed in this paper by combining the in-situ measurements with
3 chemistry-transport models. Chemistry and transport models, driven by different
4 transport schemes, are used to simulate the middle tropospheric CO₂. We also apply
5 different boundary conditions to force the 3-D CTMs. The seasonal cycle and semi-
6 annual oscillation of surface CO₂ are well simulated by chemistry-transport model with
7 the prescribed CO₂ sources and sinks boundary condition. Semi-annual oscillation is also
8 found in the mid-tropospheric CO₂. From the sensitivity study, we found that the semi-
9 annual oscillation in the mid-tropospheric CO₂ is mainly originated from sources at
10 surface. Possible reason for the semi-annual oscillation of surface CO₂ is the CO₂ surface
11 source due to net ecosystem production.

12
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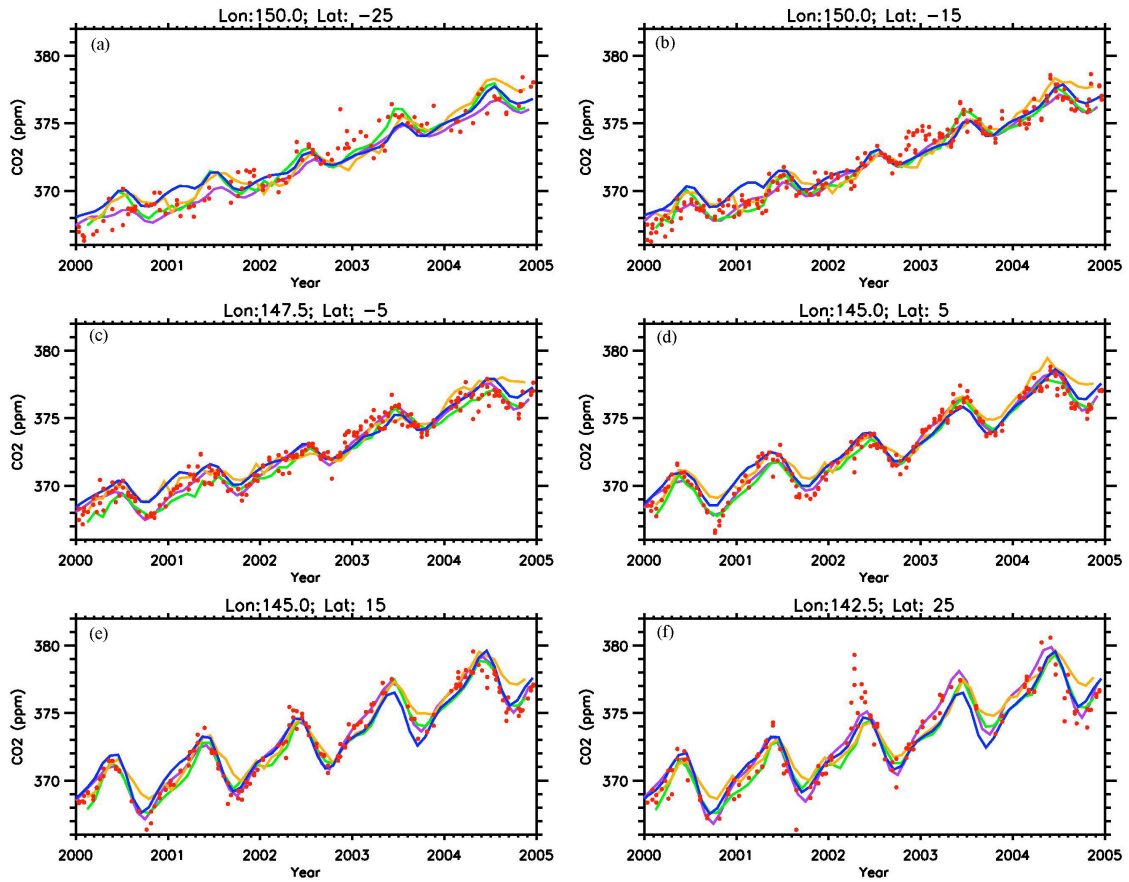
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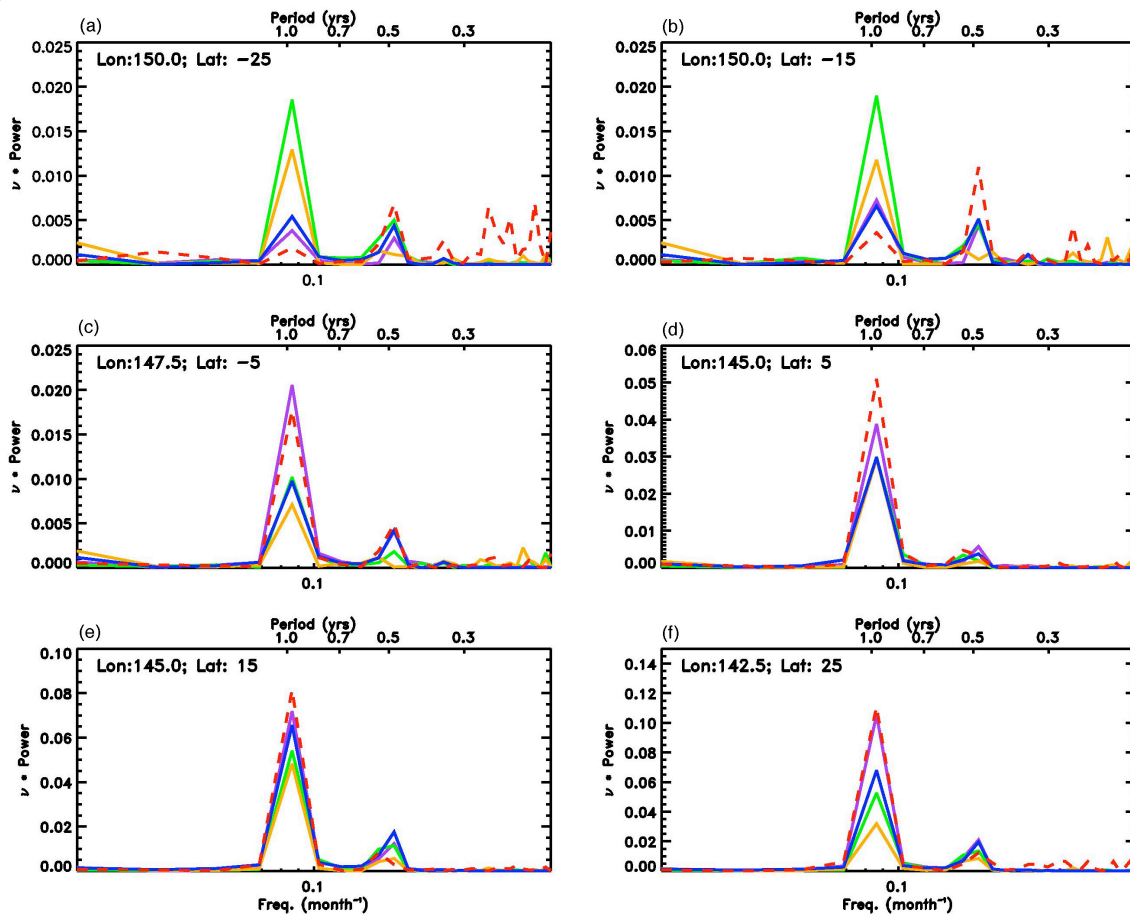
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16 from <http://www.cmdl.noaa.gov/>)

1 Figure 1: Aircraft observations between 9 km and 13 km (red dots) [Matsueda *et al.*,
2 2002] and model CO₂ mixing ratios (color lines). The CO₂ mixing ratios from the GEOS-
3 chem model forced by GLOBALVIEW-CO₂ boundary condition (BC) and prescribed
4 CO₂ source/sink BC are shown by the green and orange lines, respectively. The CO₂
5 mixing ratio from the Caltech/JPL 2-D model forced by NCEP2 and GLOBALVIEW-
6 CO₂ BC are shown by purple line. The CO₂ mixing ratios from MOZART-2 forced by
7 NCEP1 and GLOBALVIEW-CO₂ BC are shown by the blue line.
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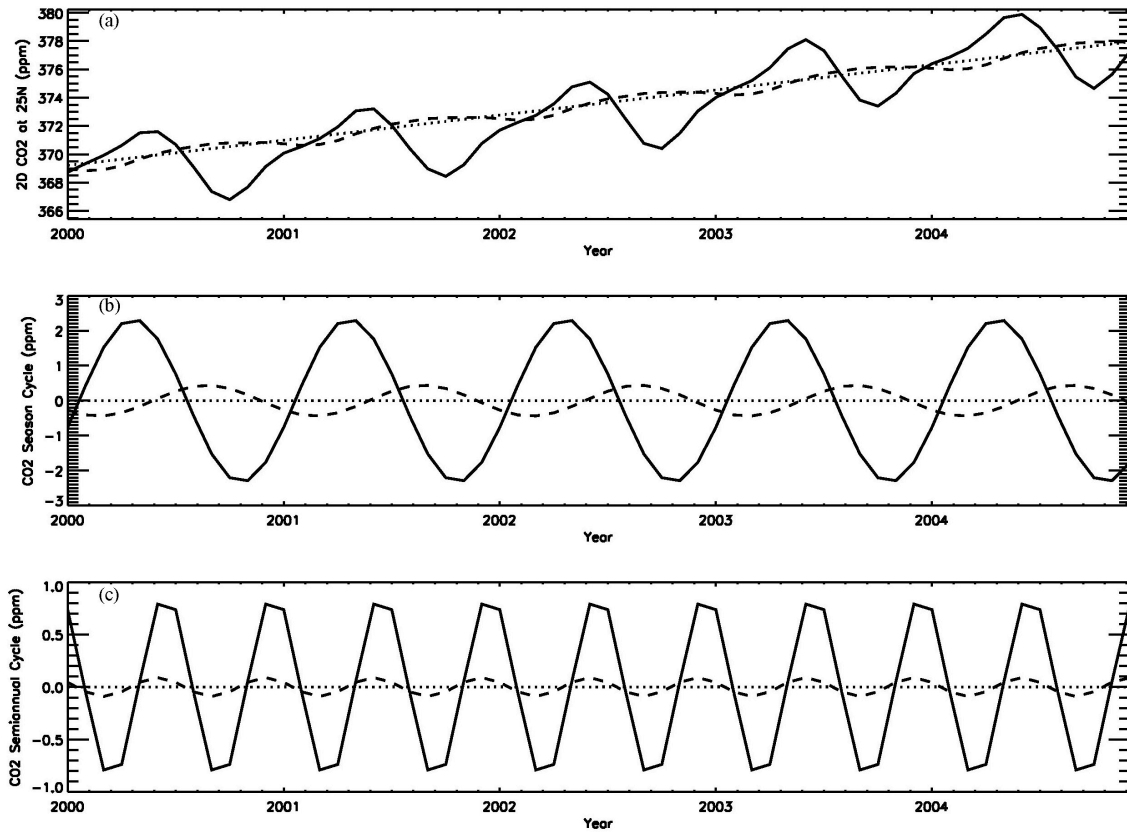
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- 1 Figure 2: Power spectra for aircraft and model CO₂ time series. Red dash line is the
- 2 power spectra for the Matsueda's data. Colors for solid lines are the same as in Figure 1.



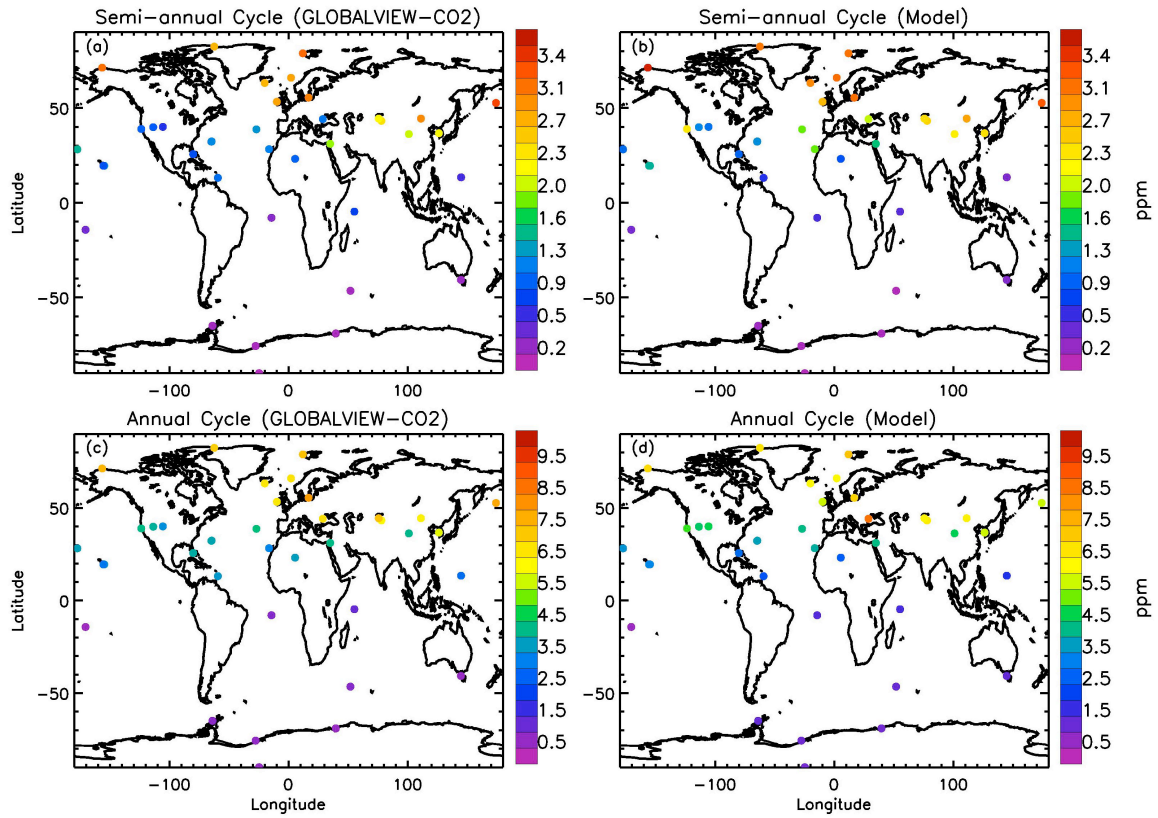
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1 Figure 3: (a) Caltech/JPL 2-D model CO₂ at 25°N. (b) Seasonal cycle of model CO₂ at
2 25°N. (c) Semiannual cycle of model CO₂ at 25°N. Model CO₂ forced by the
3 GLOBALVIEW-CO₂ boundary condition and NCEP2 reanalysis meteorology field are
4 shown by solid line. Model CO₂ forced by the linear CO₂ trend boundary condition and
5 NCEP2 reanalysis meteorology are shown by dashed line. Model CO₂ forced by the
6 linear CO₂ trend boundary condition and climatology transport field are shown by the
7 dotted line. Units are ppm.
8



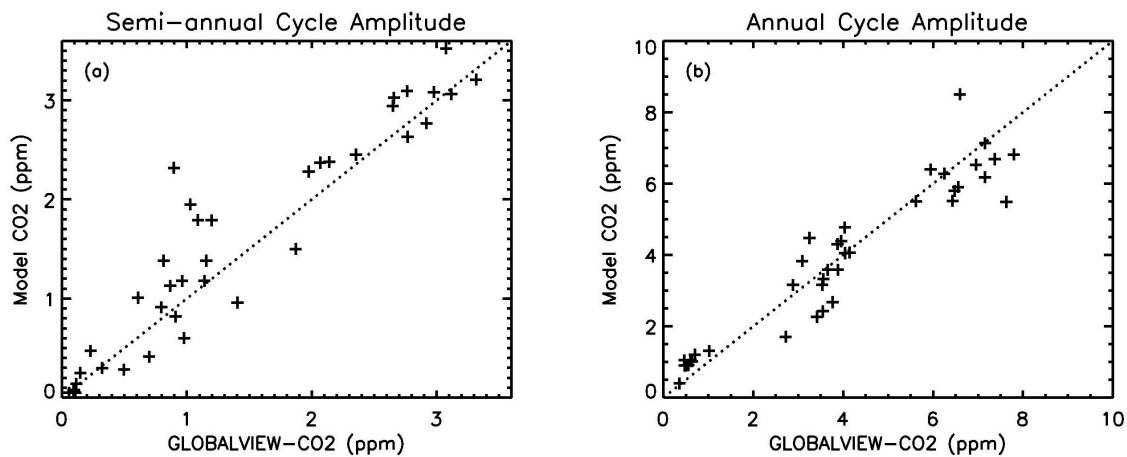
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1 Figure 4: (a) Semi-annual oscillation amplitude from GLOBALVIEW-CO₂ measurement.
 2 (b) Semi-annual oscillation amplitude from GEOS-chem model CO₂. (c) Annual cycle
 3 amplitude from GLOBALVIEW-CO₂ measurement. (d) Annual cycle amplitude from
 4 GEOS-chem model CO₂. Units are ppm.
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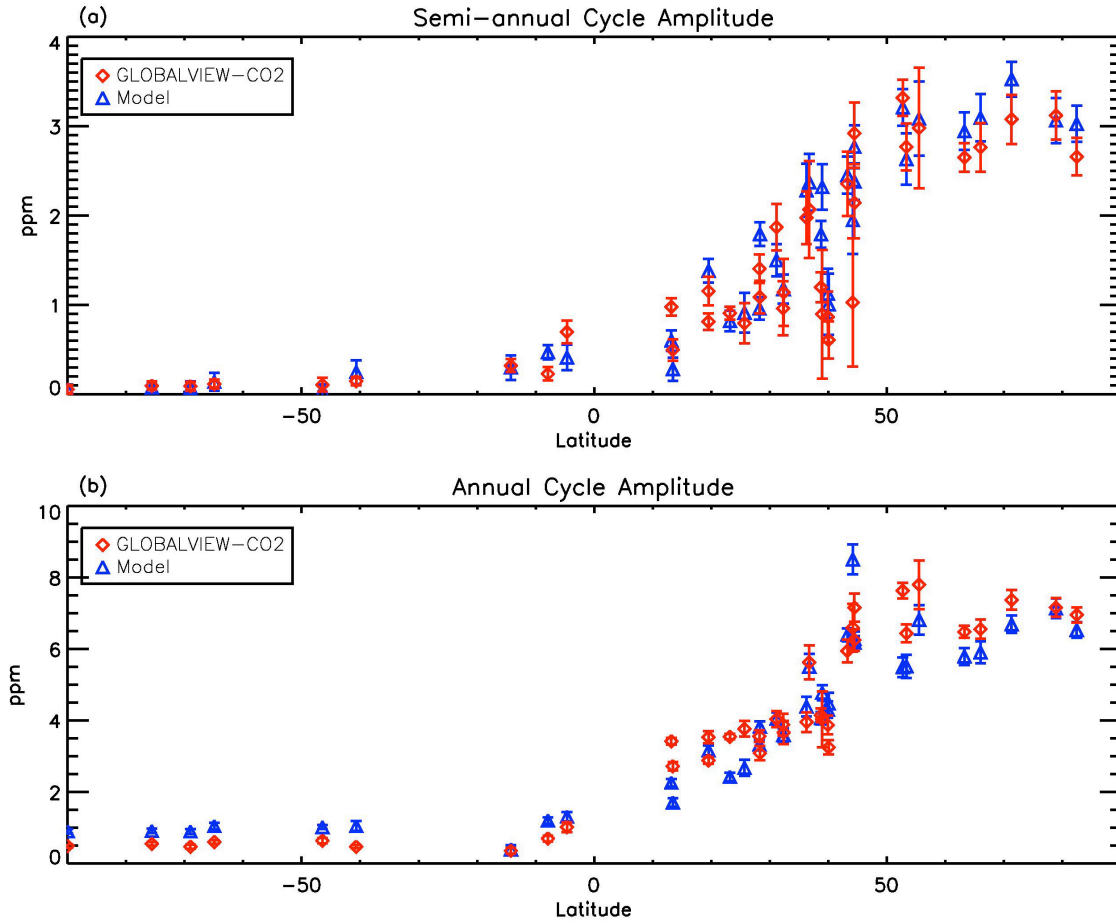
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1 Figure 5: (a) Scatter plot of the semi-annual cycle amplitude for GLOBALVIEW-CO₂
 2 and GEOS-chem model CO₂. (b) Scatter plot of the annual cycle amplitude for
 3 GLOBALVIEW-CO₂ and GEOS-chem model CO₂.
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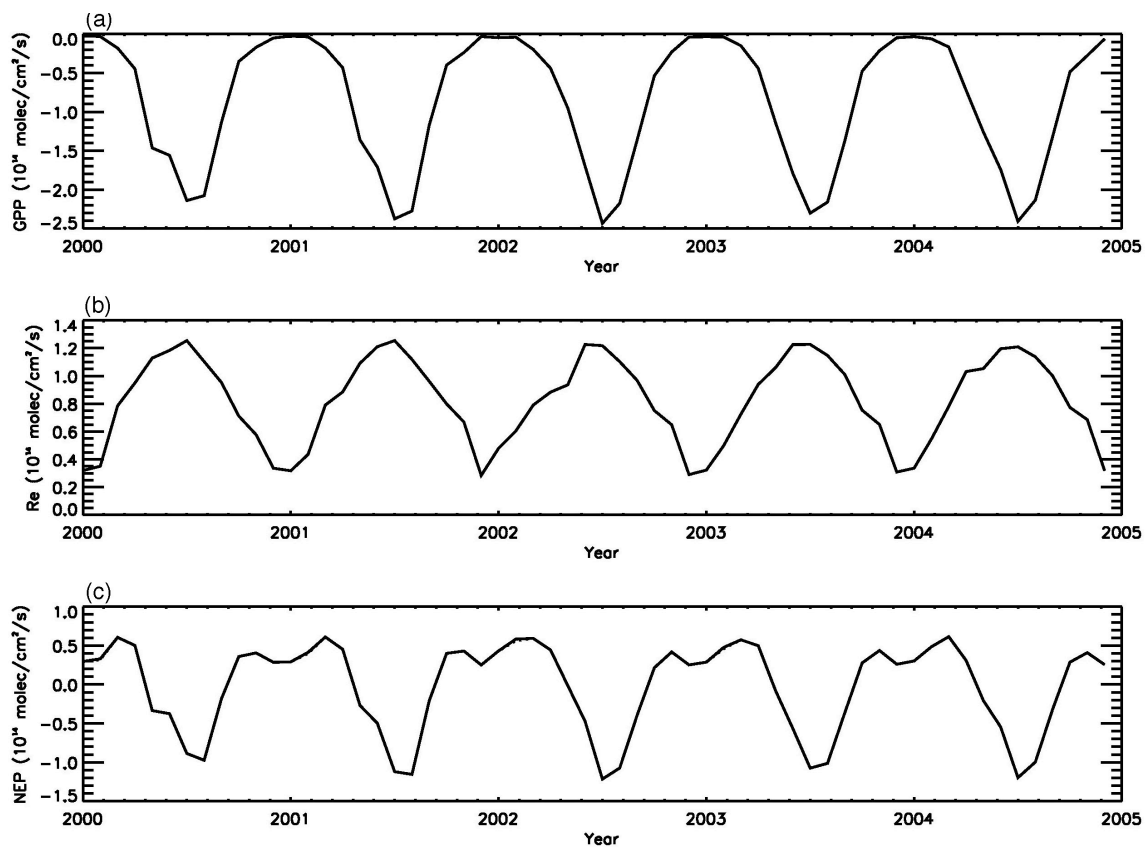
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1 Figure 6: (a) Latitudinal distribution of semi-annual cycle amplitude. (b) Latitudinal
2 distribution of annual cycle amplitude. Diamonds are the results from GLOBALVIEW-
3 CO₂. Triangles are the results from GEOS-chem model. Error bars are the standard
4 deviations of the results at each latitude band.
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1 Figure 7: (a) Gross Primary Production, (b) Ecosystem Respiration, and (c) Net
 2 Ecosystem Production from Carnegie-Ames-Stanford (CASA) ecosystem model at 30°N
 3 and 110°E.
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