Large Scale Temporal and Spatial Gradients of Carbon Dioxide as Derived from the NOAA/ESRL Aircraft Profiles

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1. Abstract

To assess the impacts of fossil fuel combustion on atmospheric composition and climate, Dave Keeling began measurements of carbon dioxide (CO_2) at the Mauna Loa Observatory in 1957. While such measurements are still useful in quantifying the atmospheric burden of CO_2, other measurement strategies are necessary to determine the fate and source of fossil-derived CO_2. To address this need the NOAA/ESRL GMD Carbon Cycle Greenhouse Gases Group created the aircraft sampling network in 1992. Currently this network consists of 20 aircraft sites primarily located in North America. The air samples collected are analyzed in Boulder, Colorado for CO_2, carbon monoxide (CO), hydrogen (H_2), nitrous oxide (N_2O), sulfur hexafluoride (SF_6), and methane (CH_4) as well as halocarbons and the isotopic ratio of CO_2. In this study we examine the spatial and temporal variability of CO_2 throughout a climatological year.

2. Sampling Equipment

Figure 1

Air samples are collected using two different instruments, the programmable flask package (PFP) and the programmable compressor package (PCP) (Figure 1). Each PFP holds twelve 0.75L glass flasks connected by a stainless steel welded manifold and a pressure sensor. The PCP contains the systems power, pumps, flow meter, and electronics. These two units are deployed on small aircrafts for programmable time-interval or event based sampling of atmospheric CO_2 and other trace gases.

3. Aircraft Profiles

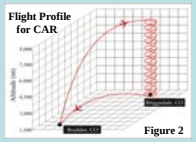

Figure 2

This program collects discrete air samples from 500-8,000 m above ground. Figure 2 shows the aircraft profile taken at Briggsdale, CO (CAR). At present, 20 aircraft sites are being sampled every two to three weeks as a part of the North American Carbon Program. These samples are collected using single and twin engine Cessnas. Trace gases from the PFPs are measured at ESRL.

4. Climatology Methods

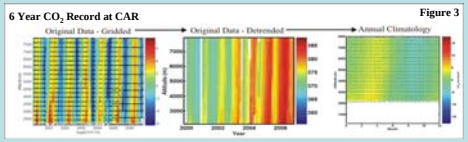

Figure 3

This study uses CO_2, CO, and SF_6 data collected from the NOAA/ESRL Aircraft Network. All of the data used in this study were binned by month, detrended, and then adjusted to one common year (2004) by normalizing them with the Mauna Loa surface site measurements. Figure 3 shows this process for the sampling record at CAR. The black dots show the sampling densities that have been used to create the annual climatology.

5. Sulfur Hexafluoride Data

Background:

SF_6 is an important greenhouse gas to monitor due to its increasing atmospheric burden, high global warming potential (GWP), and long lifetime. During the 1980s and 1990s surface measurements show that SF_6 was increasing at a rate of 7% per year (Geller *et al*., 1997; Maiss and Brenninkmeijer, 1998). SF_6, by mass, is 23,000 times more effective at changing radiative forcing than CO_2 over a 100 year time scale (IPCC., 2001). The lifetime of SF_6 is 3,200 years (Ravishankara *et al*., 1993). It is primarily used to insulate high voltage electrical equipment such as electrical transmission and distribution systems in gas-insulated switching gear transformers. The major sink of SF_6 is a chemical reaction in the mesosphere. There is little uptake from the ocean because SF_6 is relatively insoluble in water.

Results:

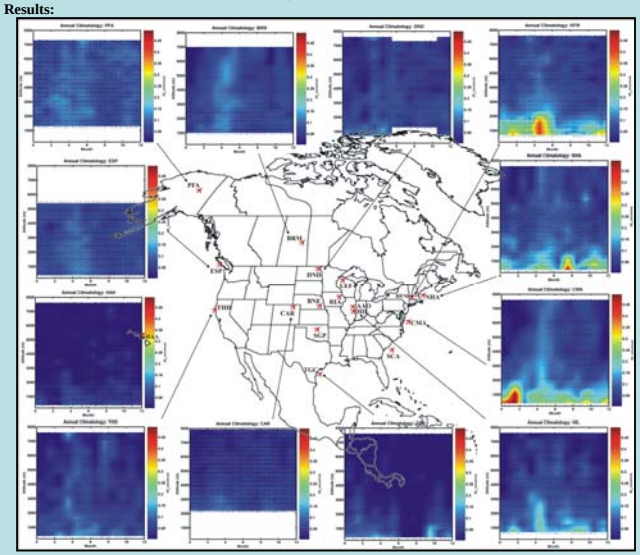

Figure 4: SF_6 values range from 0.05 to 0.45 nmol/mol

Conclusions:

Unlike CO_2 and CO gradients which reflect vertical mixing, seasonal and interannual changes in interhemispheric transport, and sources and sinks, SF_6 is relatively constant. Figure 4 shows high levels of SF_6 in three East Coast sites: Harvard Forest, Massachusetts (HFM); Worcester, Massachusetts (NHA); and Cape May, New Jersey (CMA). The elevated levels seen at the East Coast sites are likely attributed to the close proximity to SF_6 sources. During summer, enhanced vertical mixing dilutes SF_6 signals in the planetary boundary layer (PBL) with SF_6-depleted air from the free troposphere (FT). This occurs in the Northern Hemisphere (NH), reducing the quantity of SF_6 in the PBL (Gloor *et al*., 2007). In general the mixing ratio of SF_6 at most sites show a negative vertical gradient (Gloor *et al*., 2007). This is clearly shown in Figure 4, especially on the Eastern Coast where there are higher levels of background SF_6.

6. Carbon Dioxide Data

Background:

CO_2 is a major component of the carbon cycle. The annual mean global average growth rate of CO_2 from 1990 to 1999 was 3.2+/-0.1 PgC/yr (IPCC, 2001). CO_2 is released into the atmosphere when carbon-containing fossil fuels such as oil, natural gas, and coal are burned. This process accounts for ~75% of CO_2 emissions (IPCC, 2001). The rest of the emissions are due to changes in land use. Long-term natural sinks of CO_2 include ocean uptake as well as a build up of terrestrial biomass. In the short-term, the seasonal cycle at each site is driven primarily by summertime uptake of CO_2 (photosynthesis) and the wintertime out gassing of CO_2 (respiration).

Results:

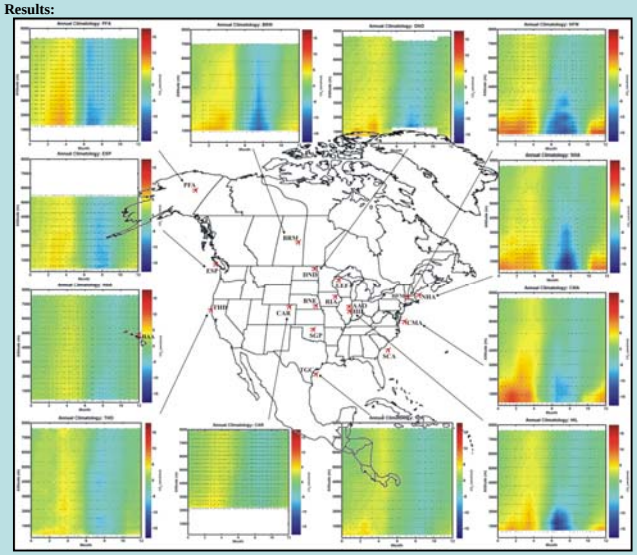

Figure 5: CO_2 values range from -15 to 15 umol/mol

Conclusions:

In the NH we see the largest uptake of CO_2 in the Midwest due to crop production and on the East Coast due to deciduous forests. During the summer months we see that vegetation uptake of CO_2 exceeds vegetation respiration and fossil fuel emissions. This leads to lower CO_2 in the PBL than in the FT. The opposite occurs in the winter when fossil fuel emissions and vegetation respiration become dominant. As air masses move from west to east the concentration differences are amplified leading to larger differences between BL and FT CO_2 concentrations. The West Coast has weaker horizontal CO_2 fluxes and exhibits a less defined seasonal cycle primarily due to the influx of marine air from westerly winds. By examining the plots in Figure 5 we can see that the sources and sinks are more defined and the vertical mixing is less defined on the East Coast.

7. Carbon Monoxide Data

Background:

CO is a very weak greenhouse gas, but its reaction with hydroxyl radical, OH, affects the lifetimes of some long-lived greenhouse gases like CH_4. The primary anthropogenic source of CO is the incomplete combustion of carbon-containing fuels. Total emissions of CO are ~2800 Tg(CO)/yr with approximately half from *in situ* production and half from direct emissions (IPCC, 2001). Through natural processes in the atmosphere, CO is eventually oxidized to CO_2.

Results:

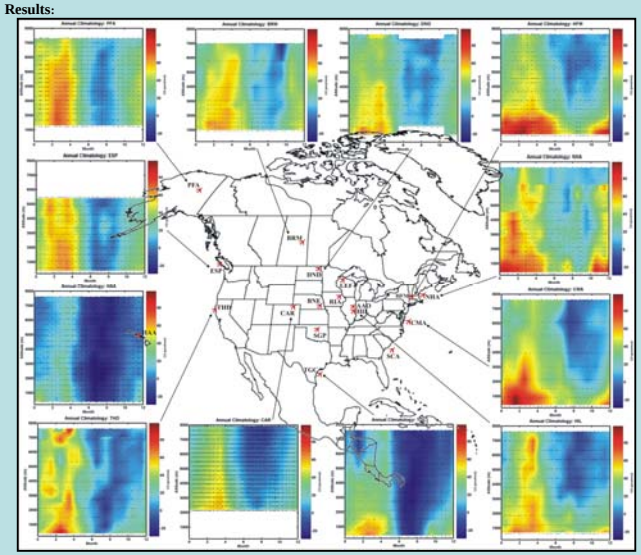

Figure 6: CO values range from -20 to 80 pmol/mol

Conclusions:

Because CO has an atmospheric lifetime of a few months, it is spatially and temporally more variable than longer-lived gases. A combination of vertical mixing and the seasonally varying concentration of OH drives the large seasonal cycle in CO, which is seen at all of our sampling sites in North America (Figure 6). This cycle is seen throughout the atmospheric column and can be attributed to mid-latitude boundary layer build up of CO. This column is carried north and around the Arctic Circle only to be entrained in the FT and mixed down into the PBL of sites like Poker Flat, Alaska (PFA).

8. Conclusions

This study shows that there are similarities in the profiles of SF_6, CO_2, and CO. Each of these gases see enhancements throughout the atmospheric column as air masses move from west to east across North America. The largest enhancements are seen in the boundary layer at East Coast sites for all three gases. We see stronger seasonality throughout the column for CO than CO_2 because the sink for CO is active throughout the troposphere while the sink for CO_2 is only at the surface. Even without detailed information on the concentration and distribution of OH, we can make a reasonable conclusion that fossil fuel emissions are affecting the seasonal cycles of CO and CO_2. Because emissions of SF_6 are fairly well known, it is an excellent tracer for testing transport in models used to determine the fate of fossil fuel CO_2 (e.g., TM5).

9. References

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10. Acknowledgments

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