Atmos. Chem. Phys. Discuss., 10, C7119–C7125, 2010
www.atmos-chem-phys-discuss.net/10/C7119/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "CO_2 and its correlation with CO at a rural site near Beijing: implications for combustion efficiency in China" *by* Y. Wang et al.

Y. Wang et al.

yxw@tsinghua.edu.cn

Received and published: 1 September 2010

We thank Dr. Andrews for helpful comments which have significant improved the manuscript.

Reviewer 2: AE Andrews (Referee)

This is a very good paper that presents an important new dataset from China. The paper is clearly written and represents thoughtful analysis. I have the benefit of having read Reviewer #1's comments. I agree that it would be useful to see the CO timeseries from Miyun. Specifically, I recommend expanding Figure 3 to include the corresponding CO data. CO data is also available for the NOAA/Globalview sites presented in that

figure, and it would be useful to see how the background CO values estimated as the 5th percentile from the Miyun observations compare.

Response: We added a new Figure 6 to show the CO time series at Miyun compared with NOAA sites, and Figure 9b to present the background CO.

One especially relevant recent study that should be referenced is by S. Han and coauthors: JGR, Vol. 114, D23202, doi:10.1029/2009JD012027, 2009

Response: We referenced this paper in the revised manuscript.

My other major comment has to do with interpreting the dCO2/dCO ratio without accounting for biological fluxes of CO2. Positive biological fluxes of CO2 in winter can be comparable in magnitude to fossil fuel fluxes and compact correlations between CO2 and CO can exist in winter even when fossil and biological sources are comingled only over large spatial scales. This assertion is based on analysis of continental data from the US (manuscript in preparation), where RËĘ2 for CO2/CO correlations can be > 0.9, but biological fluxes are thought to be of the same approximate magnitude as fossil fuel fluxes (based on CarbonTracker fluxes convolved with lagrangian particle dispersion footprints). At a minimum, I think the authors should include a discussion of the likely magnitude of biological fluxes integrated over the landscape and how consideration of biological fluxes would impact the interpretation of CO2-CO correlation slopes.

In the absences of ocean and fire sources: CO2(obs) $\sim$$\Delta$CO2(fossil) + ΔCO2(bio) + CO2 (bg) CO(obs) $\sim$ ΔCO(fossil) where Δ denotes a change in CO2 or CO resulting from upwind sources. And: CO2(obs-bg)/CO(obs-bg) = (ΔCO2(fossil) + ΔCO2(bio))/ ΔCO(fossil) Set α= fraction of total flux from fossil fuels such that: Δ CO2(fossil) = α ΔCO2(obs-bg) and: ΔCO2bio = (1-α)*CO2(obs-bg) So: CO2(obs-bg)/CO(obs-bg) = (ΔCO2(fossil) + (1-α)*CO2(obs-bg))/ ΔCO(fossil) Rearranging gives: ΔCO2(fossil)/ ΔCO(fossil) =αCO2(obs-bg)/CO(obs-bg)

So, if an estimate of α is available, then it is possible to estimate the quantity: ΔCO2(fossil)/ ΔCO(fossil)

I'm not sure accurate seasonally resolved estimates of α are available for China. I did look at CarbonTracker fluxes corresponding to the "NCN" region (100-120 degrees longitude and 40-50 degrees latitude). The fluxes are available online: ftp://ftp.cmdl.noaa.gov/ccg/co2/carbontracker/fluxes/

CarbonTracker's median values of α for January 2004-2008 range from 0.45 – 0.95, but may not be realistic. The fluxes available online are from CarbonTracker-North America, which is not optimized to solve for Asian fluxes. Data to constrain Asian fluxes in CarbonTracker-NA is very sparse. It may be possible to get more reliable estimates of α from CarbonTracker-Asia or from another source such as flux tower data or an ecosystem model.

Response: The point is well take. We revised our statement about wintertime biospheric flux. Instead of saying biosphere is dormant in winter, we emphasize on the fact that CO2 to CO correlation is strongest in winter, which implies that biospheric influence is less variable in winter and the anthropogenic signature on CO2 can be better separately using CO as a tracer.

In our correlation analysis, the CO2-CO regression intercept includes two terms: [ΔCO2 (bio) + CO2 (bg)], instead of this one CO2 (bg) term. We stated this explicitly in the ACPD paper: pg 12677, lines 20-25. In the case of large biospheric fluxes of CO2 in winter, these fluxes are implicit in the intercept of CO2-CO regression. However, as the reviewer indicates, given the large footprint of CO2 measurement, we cannot exclude contributions from biospheric fluxes on the CO2/CO correlation in winter. But without credible estimates of the biospheric fluxes over North China (the CarbonTracker estimate is subject to large uncertainty for China), we cannot evaluate the impact of biosphere on the correlation analysis. We added some discussion on this in the text:

"In addition, non-combustion (biospheric) sources of CO2 that are comingled with com-

bustion sources over large spatial scales will contribute to observed CO2-CO correlations because their variances are affected by the same atmospheric transport (Gloor et al., 2001; Folini et al., 2009). In particular, the overall respiration component from a densely populated urban area may be significant relative to combustion." "Respiratory CO2 from soils and livestock is likely an important source of CO2, but because these sources are spatially distinct from densely populated urban areas we expect the correlation between this CO2 and CO to have a weaker correlation. Soil respiration in winter is likely minimized by cold temperatures and arid climate."

This paper makes good use of data from NOAA's Earth System Research Laboratory and the Globalview-CO2 dataset, which has many contributors. I hope that data from Miyun will also be made freely available. I would be happy to discuss any of these comments via email.

Response:- We thank NOAA ESRL for providing convenient access of CO2 and CO data. The references to these data are updated in the revised manuscript.

Other specific comments:

Pg 12667 lines 10-15 and pg 12668 line 1-5: NOAA "baseline stations" has a specific meaning within NOAA. It refers to the fully staffed observatories (e.g., Mauna Loa, Barrow, South Pole, Samoa). The sites used here are part of NOAA ESRL's Global Cooperative Flask Sampling Network.

Response:- We made corrections.

Pg 12671: Please describe how standards are traceable to NOAA.

Response: We added the text: "The three working and surveillance standards were obtained from Scott-Marrin Inc. and calibrated to 0.1 ppm accuracy against a suite of NOAA primary standards maintained as part of a dedicated calibration system in our laboratory at Harvard that supports several other ground-based measurement sites and airborne instruments [Daube et al., 2002]."

Pg 12672: It should be mentioned that diurnal variations of CO2 are strongly dependent on intake height (see e.g., P Bakwin et al., Tellus B, 50(5): 401-415, 1998). Also, the absence of a strong diurnal cycle does not require that biological fluxes are neglible, only that the fluxes do not have a strong diurnal cycle.

Response: The point well taken. The sentence is revised as: “The day/night shift between photosynthesis and respiration, boundary layer dynamics, and pollution transport all contribute to observed diel cycles in CO2 mixing ratio, with the magnitude dependent on sampling height above the surface (Bakwin et al., 1998).”

Response: Discussion of the winter diel cycle is revised to: “The diel cycles are relatively flat in winter indicating that biological fluxes are more constant throughout the day.”

Pg 12673: Given that nighttime data is strongly dependent upon boundary layer height, it might be better to use afternoon only data for the trends/seasonal cycle analysis.

Response: We agree with Dr. Andrew’s concern about the nighttime data and that is why we focused on daytime only data in analyzing the changes in wintertime CO2/CO correlations. To compute the seasonal amplitude, however, afternoon only data will deliberately exclude respiration fluxes in summer, causing a bias in the seasonal amplitude. Therefore, we still chose to show the 24-hr data in this session, but added the following text to discuss the differences if afternoon only data are used: “If only the afternoon observations (noon – 6 pm) are considered, the average seasonal amplitude (detrended) increased to 16.5 ppmv and the average rate of growth is 1.9 ppmv.”

Pg 12674: It would be interesting to include some discussion of how the seasonal cycle is likely affected by the monsoonal circulation.

Response: We agree with the reviewer that it is an interesting and potentially important topic to analyze in general the impact of monsoon on the seasonal cycle of CO2. At our site, under the influence of monsoon circulation, the prevailing winds switch from

northwesterly in winter to southwesterly in summer. The high CO2 levels frequently sampled in June were attributed to the summertime monsoon with prevailing southwesterly winds. As the northwesterly is generally clean continental air masses and the southwesterly brings regional pollution, we would expect the monsoonal circulation to reduce the seasonal amplitude of CO2 at our site. However, it is the biosphere that dominates the seasonal cycle of CO2 rather than anthropogenic emissions, the question then arises as to how the monsoonal circulation affects biospheric activity over China. This is a complicated problem and deserves in depth discussion and analysis. As our paper is already too long as suggested by the editor, we opt not to discuss this question in the text.

Pg 12675: Respiration from large-area vegetation sources is likely to be larger than the human component even in winter. I think this would be especially true for the North China region analyzed in section 4.3.

Response: The climate for North China region in winter is cold (below freezing point) and dry. North China is not a major forested region in China, but dominated by cropland which is left idle in winter given the climate. We discussed this impact in the text: “Respiratory CO2 from soils and livestock is likely an important source of CO2, but because these sources are spatially distinct from densely populated urban areas we expect the correlation between this CO2 and CO to have a weaker correlation. Soil respiration in winter is likely minimized by cold temperatures and arid climate.”

Footprints for continental boundary layer observations are of order 100kmËĘ2 (see e.g., Folini, D. et al., JGR-D, 114(D8), 27 April 2009; Gloor, M., et al. (2001). "What is the concentration footprint of a tall tower?" Journal of Geophysical Research-Atmospheres 106(D16): 17831-17840.) So, sources far upwind can contribute to observed CO2-CO ratios and even coarsely co-located sources can produce compact correlations.

Response: We added the references in the text. See our response above.

Pg 12677: I recommend adding CO and background CO to figure 3, as suggested above.

Response: New figure 6 and figure 9b are added for CO and background CO.

Pg 12679: I don't agree with the assertion that vegetation does not affect CO2-CO correlations in winter. If some reasonable estimates of the wintertime biological fluxes for North China can be acquired and taken into account, then I think this section would be much improved.

Response: We revised our statement in the text. Instead of saying biosphere is dormant in winter, we emphasize on the fact that CO2 to CO correlation is strongest in winter, which implies that biospheric influence is less variable in winter and the anthropogenic signature on CO2 can be better separately using CO as a tracer.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 12665, 2010.