Response to anonymous Referee #1

We thank the reviewer for their comments and specific suggestions to improve our manuscript. The reviewer comments are shown in italic font, and our response is shown below, in plain text.

This is an excellent paper, shedding new light on an important issue: how to relate vertical tracer gradients in the lower troposphere to transport and mixing processes. The presentation is rigorous, involving analytical modeling, quantitative analyses employing both observations and numerical models. The paper also recognizes most of the limitations of the presented framework.

I have a few points and comments that I would like the authors to address before the paper is published:

1) Pg. 11462, First paragraph: The authors assert (using Fig.1) that the variations in mixed-layer concentrations decrease as the averaging times increase. Isn't this a simple consequence of averaging over a longer time window? Or am I missing something here? This point needs to be clarified.

We agree that the presentation of Fig. (1) was confusing, and have therefore moved the figure and its discussion to supplemental materials. We focus instead of making the same point about the timescale dependence of storage and entrainment in Section 3.4, where we conduct a formal scale analysis of the mixed-layer conservation equation (note Section 3.4 became section 3.1 in the revised manuscript).

Our point was that the storage and entrainment terms represent rates of change, as calculated by changes in concentrations or mixed-layer depths divided by a time interval. Concentrations that fluctuate by 10 ppm from day to day would have to fluctuate by 90 ppm over the course of a 90-day season for the storage term to remain as large in seasonal averages as in daily averages. Such large fluctuations would show up in timeseries of concentrations, but they do not, as indicated by comparing the 90-day and 1-day running averages in Fig. 1a,b. In fact the seasonal changes are the same order of magnitude as daily changes, which means the storage term becomes an order of magnitude smaller at seasonal timescales than at daily timescales.

The same argument applies to the entrainment term, by examining the mixed-layer depth timeseries in Fig. 1c. These observations suggest that neglecting the vertical and horizontal advection terms will result in large errors at seasonal timescales, when storage and entrainment account for a smaller portion of the CO2 budget relative to advection.

We can motivate the same result in terms of length scales. Fluctuations in mixed layer depth can move a particle in the mixed-layer over a distance on the order of the mixed layer depth (~1000 m), but this distance cannot exceed the maximum mixed-layer depth. However advection can move a particle by a distance on the order of the wind speed multiplied by the time over which the wind blows. Therefore, over longer periods of time, advection becomes much more efficient than fluctuations in boundary layer depth at moving mass into and out of the mixed-layer.

2) Fig. 2: Are the SGP results based on observations or were they model-generated? I wasn't sure when reading it.

The results are based on the CT/TM5 model at all three sites (SGP, LEF, HFM) in Fig. 2, which we noted in the revised manuscript.

3) I am inferring from the results in this paper that the "diurnal rectifier effect" is insignificant. If so, please state so explicitly. Only the "seasonal rectifier effect" was mentioned.

The diurnal rectifier effect is still important for understanding concentration gradients at diurnal timescales. We show that the mechanisms responsible for the diurnal rectifier effect, primarily diurnal fluctuations in mixed-layer depth, are not as important for transport and mixing at seasonal timescales. The diurnal rectifier effect may still be important for understanding errors in transport model inversions because the diurnal cycle influences the measurements assimilated into those inversions. Our paper focuses on mechanisms of transport and mixing and does not discuss these data assimilation issues.

We added a statement to the introduction to clarify the difference between diurnal and seasonal rectifier effects:

> Comparisons of transport models and observations have not taken into account the timescale dependence of transport and mixing, which has led to hypotheses of both overestimated and underestimated vertical mixing in transport models, corresponding to both overestimated and underestimated northern terrestrial carbon sinks in the inversions. The dilution by transient boundary layer growth approximately balances surface CO2 fluxes when analyzed during the daytime or over a few days (Raupach, 1991; Raupach et al., 1992; Levy et al., 1999; Lloyd et al., 2001; Styles et al., 2002), and can successfully explain the diurnal rectifier effect (Yi et al., 2004). However over longer periods of time boundary layer depth reflects a statistical equilibrium between surface heat fluxes acting to increase, and radiative cooling acting to decrease boundary layer depth, with clouds coupled through their effect on radiative cooling (Betts et al., 2004). Radiative cooling in turn balances adiabatic warming in subsiding branches of the circulation, bringing boundary layer trace gas concentrations into equilibrium between surface fluxes and transport by the subsiding flow (Helliker et al., 2004; Betts et al., 2004).

4) Pg. 11471, Line 18: "horizontal" is a typo. The phrase should read "zonal and meridional advection"

Yes, it includes both zonal and meridional advection. We clarified our definition of horizontal advection where it is first introduced, after equation (1) in Section 2.1. Note that the revised manuscript writes horizontal advection more formally, as the dot product of the horizontal gradient operator and horizontal wind velocity vector.

5) Fig. 4c,f: It appears that the points plot on an "universal line" when scaling with t is carried out. This is pretty interesting (and amazing, to say the least!). What is the slope of this universal linear relationship? What is the physical significance of the value of this slope?*

The slope is around +2. If our scaling were perfect the slope would be +1. The subsidence rate has a strong seasonal component, which slightly increases the advection magnitude as the timescale increases toward 90-days, resulting in a slope steeper than 1.

We agree with the reviewer that the scaling result is interesting, however these results only guide us to approximate solutions to the mixed-layer conservation equation, and could distract other readers from the important implications of our paper for rectifier effects and transport model errors. We therefore moved the scaling plots (Fig. 4) to supplemental materials, to decrease the burden on the average reader, and in response to the issue of length raised by the second reviewer.

6) Pg. 11475, Line 20: SF6 is referred to as “an ideal tracer due to its well known...emissions”. However, SF6 emissions are still subject to non-negligible errors. For instance, Hurst et al. [2006] found from aircraft observations that the EPA estimate for the U.S. may be overestimated by 50%.

Hurst, D., et al., Continuing global significance of emissions of Montreal Protocol-restricted halocarbons in the United States and Canada, J. Geophys. Res., 111 (D15302), doi:10.1029/2005JD006785, 2006.

We agree with the reviewer that SF6 is still not “ideal” as a tracer, and removed this wording from the revised paper.

7) Section 5 (Discussion): Here mixing strength is used to refer to the subsidence strength w. This is a little confusing to the reader, as I usually associate mixing to turbulent eddies, and not to w. Make sure that this point is clarified to the reader up front.

We thank the reviewer for bringing this issue to our attention. We added clarification of these points in the introduction:

> It helps to keep in mind that the terms involving dh/dt (first and second left-hand side terms of equation 1) and the vertical advection (third left-hand side terms) represent two separate physical processes (Betts, 1992). The former accounts for turbulent entrainment of free-troposphere CO2 by the mixed-layer growing into the free-troposphere, whereas the latter implicitly accounts for the turbulent entrainment of free-troposphere CO2 that has descended to the top of the mixed-layer through vertical CO2 advection. We refer to these terms separately as entrainment and vertical advection, respectively, but use the term vertical mixing when referring to the combination of both processes.

References:

Betts, A. K.: FIFE atmospheric boundary-layer budget methods, Journal of Geophysical Research-Atmospheres, 97, 18523-18531, 1992.

Betts, A. K., B. Helliker, et al.: Coupling between CO2, water vapor, temperature, and radon and their fluxes in an idealized equilibrium boundary layer over land, J. Geophys. Res.-Atmos. 109, 2004.