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Interactive Comment

Interactive comment on "Representativeness and climatology of carbon monoxide and ozone at the global GAW station Mt. Kenya in equatorial Africa" *by* S. Henne et al.

Anonymous Referee #2

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General Comments

This manuscript provides a good overview of the Mount Kenya observatory, in terms of the first 5 years of CO and O3 measurements there, transport pathways to the station, and initial interpretations of those measurements in the context of seasonal transport variations. This work indicates significant potential for the MKN station to become a significant long-term resource. Of the results presented here, the transport analyses and the assessment of the degree to which nighttime measurements are characteristic of the regional free troposphere will be most valuable for future work at that station. These are the strongest aspects of this work. The measurements are also used to investigate seasonal trends and impacts of biomass burning and the presence or absence of ozone formation and destruction upwind of the station under various transport conditions. These aspects of the paper were frequently not supported to the degree necessary to draw strong conclusions. It is probably not possible to adequately support them without significant additional analyses, including for example modeling analyses, which would be beyond the scope of this paper. I recommend condensing, or perhaps in some cases removing, several of these portions.

Specific Comments

Introduction

page 73 (meaning 17773), lines 25-26: Is it reasonable to refer to the station as a baseline site for "tropical Africa"? Tropical Africa is a large region. With so little known about spatial variations in atmospheric composition across Africa, as is emphasized in this Introduction, it would be more appropriate to refer to MKN as a baseline site for eastern tropical Africa.

Methods

page 75, lines 24-25. The 16 ppb confidence interval for 1-hour-average CO seems high. It is hard to judge this value, as precision and accuracy are mixed into once uncertainty value. However, the 48C-TL should be able to give a precision of $\pm$8 ppbv for 2-minute average measurements if using a method similar to that of Parrish [1993]. Please add a brief discussion of what the main contributor(s) to the specified uncertainty were, and what the measurement precision was.

page 78, line 18. O3-CO relationships can only be used to estimate upwind ozone production, not ozone production potential. Ozone that will be formed downwind from remaining NOx or from NOx released from remaining PAN does not affect the ozone observations.

page 78, lines 22 to 27. This paragraph is confusing and apparently erroneous, because the Parrish comment that is noted concluded that regression techniques that include errors in both x and y are needed, and ordinary least squares (with errors assumed to be present only in the y variable) was not adequate. However, after noting this, the text states that the method used here, with errors in both x and y, was compared with the reduced major axis technique, and that this “confirmed this tendency” noted by Parrish. But RMA is a regression technique for systems with error in both x and y, so this comparison was between two alternative techniques for regression when there are errors in both x and y.

Results.

Section 3.2. Diurnal Cycles

Page 80, lines 18–20+. This seems to imply that the thermal versus synoptic classification distinguishes between times with, and without boundary layer influence. More information on the criteria used to identify these days is needed. (The Henne et al. paper that is cited is not available). In addition, more information is needed to explain why the “syn” days should not have BL influence. In addition, ALL of the categories exhibit significant diurnal cycles in Figure 2. If there were no ABL influence in any of these categories, then that category should not show a diurnal cycle in Figure 2.

Page 81, lines 16–20. The time period selected as “FT observations” (2100–0400) needs to be supported in terms of the specific start and end time selected. In particular, the diurnal cycles of CO and O3 are not flat during 2100–2300, so it appears that measurements during those hours are not characteristic of FT conditions (as FT conditions are indicated during 2300–0400).

Section 3.3. Annual cycle.

The conclusion on page 81, lines 26–28 (that the secondary CO maximum in the overall annual cycle is due to one year, 2003) is inconsistent with the assertion in the abstract (page 70, line 19) that there is a secondary CO minimum in November. This secondary

CO minimum does not appear to be present during most years.

The discussion of the role of the ITCZ location and large-scale circulation as a determinant of the annual cycles of CO is convincing. The discussion for ozone is less so, because it is not clear in this section why transport from the northeast during Jan-early March (with elevated pollution CO) is not depleted in ozone, while the return of northeasterly flow in October to December brings ozone-depleted air. Similarly, why is there not similar loss in March–June when air flow off the ocean occurs? The answers to these questions may relate to differences in the transport altitude, which are discussed in the next section. However, this section needs to be modified so that these apparent inconsistencies are removed. It may make sense also to reverse the order of sections 3.3 and 3.4, discussing all transport seasonality first, then the mixing ratios' seasonality.

Section 3.4. Cluster results

Given the large interannual variations in the frequency of occurrence of several clusters, including the near absence of some clusters during some seasons, have the authors conducted any cluster analyses using one season at a time? It is possible that the number of clusters and the cluster locations could differ in such an analysis. For example, flow in the AP cluster may be associated with different weather systems during summer than during winter, leading to differences in transport pathways that could only be identified using season-specific cluster analysis. At a minimum, it would be worth plotting the AP cluster (and others) separately for each season to find out whether the members in each season are similar to those in other seasons. (If no significant differences are found, then these plots would not be needed in the paper.)

Section 3.5 Interannual variability and biomass-burning

Most of the first half or more of this section seems out of place, because it presents an analysis that is motivated (in this text) by the seasonality of CO levels measured in previous studies, not this study. This entire section could be shortened considerably.

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The motivation for this section is given on page 87, lines 11–14: pollution concentrations peak in August to October while maximum fire activity from fire counts peaks in June–July. But the CO levels in the SA cluster peak in July to August, so does this motivation apply to the MKN analyses?

A major point is also made of the fact that FRP is high later in the year than is area burned (Fig 8b versus 8a and page 88, lines 20–21). But it is fire radiative *energy*, not fire radiative power that is most closely related to fuel consumption. Does Fig 8b show FRP averaged over each full month and all locations (including non-fire pixels) or averaged over fire pixels only? The latter averaging would not give a result that is proportional to emissions, since area burned changes seasonally.

Section 3.6: Ozone-CO correlations

Page 91, lines 1–8. Trajectories cannot be used in this way to derive upwind emissions for individual events. Each trajectory indicates just a midpoint of the transport pathway, or one of a range of true pathways, and emissions in a region around each trajectory line can contribute to levels at MKN. Since the authors use FLEXTRA trajectories, can FLEXPART simulations be used to really estimate the upwind fire impacts quantitatively? If this is not possible, then at least all fires in the region bounded by all seven trajectories should be counted. Beyond this, additional support for a significant influence of fire emissions during the selected periods is needed. Are there any other measurements at the station that can corroborate the significance of fire impacts during these four periods?

Page 91 line 19 to page 92 line 29. With only four events, and with ages and emission regions determined using trajectories, rather than a transport model, the conclusions drawn here (regarding a significant relationship between ozone/CO slope and fire emissions age) are carried much too far. Either a better estimate of fire emissions and age should be obtained (e.g. using FLEXPART) or the discussion should be shortened significantly.

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Section 3.7.1 CO comparison with MOPITT

There are several issues that limit the degree to which this comparison and the results of Fig 13 can be used. (In addition, the specific technique used should be described.) First, to interpret Fig 13 as showing mixing ratios in the upwind regions, CO must be assumed to be inert. However, the previous section concluded that significant loss of both CO and ozone occurred over the southern Indian Ocean. Second, Figure 13 does not show values “at 600 hPa” as indicated in the caption. In fact, the mean vertical location varies with location in Fig 13, because earlier discussion in the paper emphasized differences in transport altitude among transport pathways. Some discussion of how altitude and chemistry affect the interpretation Fig 13 and the conclusions drawn regarding latitudinal CO gradients is needed..

Page 94, lines 7+. The text states that “MKN trajectory statistics that were more representative for the FT” only were used. But how was that done? Earlier in section 3.7.1 it was stated that all trajectory locatinos between the surface and 600 hPa were used.

Given the uncertainties in the interpretation of Figure 13, plus the limitations of MOPITT for sensing the lower troposphere, it is difficult to interpret a comparison between figures 13 and 14. In fact, in the end (page 95, line 1+) the authors conclude that “large disagreement between MOPITT and MKN likely can be attributed to uncertainties connected to the trajectory statistics.” If the method is this uncertain, it should not be used here at all. I suggest removing this section.

Section 3.7.2. Ozone versus SHADOZ

This section is critical to the evaluation of the representativeness of the MKN station for measurements characteristic of the regional FT—a main objective of this paper. The conclusion here is that surface deposition of ozone may occur, leading to ozone measurements lower than the true FT values, even at night.

This finding must be considered in other locations in the paper, where the reader is

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currently left with a different (and apparently misleading) impression.

- Page 81, lines 16–17: “measurements at MKN were representative of free tropospheric conditions during nighttime . . . ”
- Page 70, lines 11–12: “nighttime measurements were in general representative of FT conditions.”
- Page 96, lines 18–20: “nighttime measurements were in general representative of FT conditions”

(The authors should also consider whether seasonal variations in transport or vegetation, in combination with surface deposition, could contribute to any of the paper’s other conclusions regarding ozone.)

Technical Corrections

Abstract: Please note the period of measurements in the abstract.

Figure 1. The ozone data for hours 20 to 24 are not visible, because they are covered up by the legend.

page 85, line 24; page 86, line 5. It is confusing to refer to boreal summer for the southern hemisphere flow discussion—austral winter would be more appropriate.

Fig 9 shows FRP but the text (page 89, line 9) states that it shows fire counts.

Page 89. Lines 20–22 are out of place.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 17769, 2007.