**Atmospheric
Chemistry
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Discussions**

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.

S. Henne et al.

Received and published: 3 March 2008

We would like to thank the anonymous referee for his/her comments that improve the presentation of our results.

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

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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.

Answers to the individual concerns raised in these general remarks are answered below when dealing with each specific comment.

page 73 (meaning 17773), lines 20-21: 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.

We agree with this confinement of the area represented by the station and changed this in the revised manuscript.

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 ś8 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.

Uncertainties were calculated for each individual 1 minute measurement. The following components were considered: instrument noise (random part of the uncertainty) 8 ppb for 5 minute averages, non-random part of uncertainty (zero drift, span drift, nonlinearity) $\sqrt{7^2 + 0.006^2 \cdot C^2}$ ppb, where C is the measured concentration. The random part

of the uncertainty can be understood as the precision of the instrument and is in the same range as mentioned by the referee for the Parrish (1993) study. The non-random part of the uncertainty can be understood as the accuracy of the instrument. For the aggregation to 1 hour averages the combined uncertainty was estimated applying the suggestions from ISO11222, additionally considering the uncertainty due to missing data. The average combined uncertainty for 1 hour values was 8 ppb. The expanded uncertainty was then derived by expanding with k=2. We added a more detailed description of these individual uncertainties in the revised manuscript.

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.

This was already remarked by referee 1. We changed the term "ozone production potential" to "ozone export".

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.

We are sorry for the confusion. Actual comparisons were done between the Press et al. method, RMA and ordinary least square regression. The comparison actually showed that usually slopes were largest for Press et al. followed by RMA and ordinary least square regression. We corrected the erroneous parts of this section.

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*Page 80, lines 18-20+. This seems to imply that the thermal versus synoptic classifi-
cation 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.*

The two criteria were intended to identify days without ABL influence. It was thought
that the main driver for ABL influence is the thermally induced circulation. The first
criterion was designed to identify days that show no thermally induced flow systems,
category syn. The second criterion based on specific humidity variations was intended
as an independent control using humidity as boundary layer tracer. As stated, all cat-
egories show a diurnal cycle of CO, which means that also on days without direct
up-slope flow during the day, ABL air is able to reach the site. As a second mechanism
responsible for this transport convective mixing in the CBL and the rise of the CBL top
above the station altitude were identified by Henne et al. 2007. See also the answer to
reviewer 1 concerning the different categories.

Since the Henne et al. 2007 article is still in the review process, we repeat some of the
concepts and findings explained above in more detail in the revised manuscript

*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 par-
ticular, 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).*

The end time of the FT conditions is well defined by the fact that at this time the down-
slope flow (not shown) weakens and shortly after concentrations of water vapor and
CO start to rise. The start of the FT conditions is less well defined. However, using

2100 as start of the FT period can be supported by the following considerations. Firstly, the down-slope flow is most stable during the period 2100 - 0400. Secondly, variation in CO are smaller than 3 ppb which is small compered to the full diurnal amplitude of about 16 ppb. The same can be said about the O3 concentrations (1 ppb compared to 4 ppb diurnal amplitude). We agree that the selection of 2100 is a little subjective and we already mentioned in the manuscript that this selection might need to be restricted further to the period 0100-0400. We now put more emphasise on the fact that this restriction might be necessary under certain conditions.

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.

We agree that our abstract is a little misleading. We rephrased the summary of the annual cycle in the abstract.

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 to 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

We are in agreement with the referee that the influence of the ITCZ on the O3 annual cycle is much less convincing than its influence on CO. We would like to point out two

considerations that might explain this. First, compared to CO inter-hemispheric gradients for O3 are smaller than for CO, therefore a distinct change from north hemispheric to south hemispheric air has less influence on O3 than on CO. Second, the northern hemispheric scale annual cycle of CO peaks in March when advection towards Kenya is clearly from the northern hemisphere. In contrast O3 in the northern hemisphere peaks around May. Therefore, it's not the peak concentrations of northern hemispheric O3 that are advected towards Kenya but considerable lower O3 concentrations. Both points make it more difficult to simply explain the O3 annual cycle simply by the position of the ITCZ and therefore the general flow direction towards Kenya. It is exactly this more complex behavior that makes it necessary to conduct a more detailed transport study using air mass trajectories. So to us section 3.4 can be seen as a consequence of section 3.3. With the additional insights gained from the trajectory study it is then possible to explain, that O3 was higher from Jan-Mar and again in May because of the origin at higher altitudes in the NA cluster.

We addressed the discrepancies in identified by the referee in section 3.3 by adding the considerations given above to section 3.3.

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.)

When clustering is performed on any subset of trajectories it would obviously yield a different categorization. Clustering techniques cannot avoid a certain degree of sub-

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jectivity by the user. It was our intention to analyze the most important groups on an annual basis. We performed clustering analysis for individual years and with and without uncertainty trajectories. These qualitatively yielded the same results as if clustering was applied to all trajectories. However, the cluster means slightly differed for each year, but always six significantly different clusters were detected.

As suggested by the referee we re-analyzed our clustering results on a monthly basis, generating a cluster footprint plot like Fig. 5 for each month separately. The month-to-month changes in the cluster footprints were not very pronounced. The major results can be summarized as follows:

- EA: Apr, Nov (rain seasons) intensified local, easterly contribution

- AP: May - Sep, more central Arabian origin; Oct - Mar, more coastal Arabian origin

- NA: Feb extends further into Indian Ocean, only at higher altitudes

- SIO: No significant seasonal variation

- SA: No significant seasonal variation

- NIO: May - Oct, only upper air contribution

We added these findings in the cluster description.

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. 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.

We agree that the motivation for Fig 8 is a little lengthy. Still, we think that the result presented in Fig 8 is rather interesting for other researchers focussing on biomass burning in southern Africa. Fig 8b gives the average (of different years) monthly sum of FRP in the specific region. The sum of FRP can directly be converted to FRE by multiplying with "time = 1 month" and FRP is therefore equally related to fuel consumption. The so far noted temporal offset between fire counts and pollution levels can be explained with this figure. In conclusion, we agree to remove large parts of the motivation in section 3.5 but will retain Fig 8. and the discussion of inter-annual variability.

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?

Certainly we would expect to derive more quantitative results using a Lagrangian Particle Dispersion Model (LPDM). However, we expect our trajectory method, which already considers some "dispersion" by using uncertainty trajectories, to produce quali-

tatively similar results with the benefit of highly reduced CPU and data handling costs. For the period 2003-07 to 2004-12 we have FLEXPART calculations for MKN available ($\sim$3300 cases). An extension of these simulations for the full 5 year period of this study would be time consuming and is not intended at the moment. We compared FRP time series generated with the simpler trajectory approach with that generated using FLEX-PART calculations. We chose FRP instead of fire counts because it is closer related to real emissions. Moving from fire counts, which were used in the original manuscript, to FRP did not change the events detected with the trajectory method. The FRP time series were well correlated (r=0.83). There are no other major CO sources than biomass burning within Eastern Africa. Observed periods of increased CO and increased simulated fire influence (both conditions had to be fulfilled) can therefore be expected to be of biomass burning origin. Summarizing, we are convinced that our trajectory method is able to accurately identify biomass burning events. However, the correlation between simulated age of the "collected" FRP from the trajectory and the LPDM approach was weaker (r=0.6). Using FRP along the trajectory instead of fire counts also changed the estimated age of the emissions for the detected events. This indicates that the uncertainty of the age determination might be too large to draw strong conclusions concerning the development of the O3-CO relationship with age of the emissions (see next comment).

An improvement of the trajectory approach was suggested by the referee ("count all fires in the region bounded by all seven trajectories"). However, such an approach does not yield satisfactory results when flow splitting occurs and the distance between the end points of the seven individual trajectories would be very large. This situation was often observed at MKN when individual trajectories originated in the northern and others in the southern hemisphere. In such a case integrating all fires within the area bound by all seven trajectories would yield erroneous results.

In addition to CO and O3, black carbon measurements (two wavelength aethalometer) are made at Mt. Kenya. Unfortunately, these time series are often interrupted due to

system failures and cannot be used as an additional parameter in our study.

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.

Our estimates of the slope of ozone vs. CO agree well with earlier estimates. With the restrictions given for the estimation of the age of the emissions, we however agree that the conclusion about the relationship between increasing slope as a result of increasing air mass age, which was already observed by other authors (Yokelson et al. 2003, Takegawa et al. 2003), cannot necessarily be supported by our results. We still present the 4 detected biomass burning events together with their O3-CO slope and their age in the revised manuscript, but limit our conclusions according to the concerns described above.

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.

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Given the uncertainties in the interpretation of Figure 13, plus the limitations of MO-PITT 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.

First, the assumption that CO is inert during the 10 day transport was already discussed in the manuscript and we mentioned that it might cause underestimation of CO at the edge of the fields derived from the trajectory statistics. Second, as correctly stated the CO derived with the trajectory statistics is not an average at 600 hPa but is more representative of the column form the surface to 600 hPa. We assumed that this range would best agree with the column sampled by MOPITT for the 700 hPa. However, we agree that uncertainties in both methods limit our ability to draw strong conclusions. We intended to demonstrate that a point measurement can be used to derive a aerial picture that at least qualitatively agrees with the results derived from satellite remote sensing.

Since also referee 1 criticized this section, we decided to remove it from the revised manuscript. However, we are considering to replace it with a MOPITT inter-comparison at the location of MKN.

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 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 18-0: "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.)

This subject is also emphasised by referee 1 and we repeat our reply given to referee 1 here: The differences between sounding and mountain measurements might not stem from dry deposition in the vicinity of the station alone but might result from a systematic bias of the soundings at low altitudes. The flow towards the site during night-time is in general in down-slope direction. Above the site mainly grassland dominates the slopes of the mountain. Typical values for O3 dry deposition values for night-time grassland are about 0.1 cm/s. In order to deposit 10 ppb O3 from an initial concentration of 40 ppb it would than be necessary to advect air in very shallow layers (< 20 m) along the slope for more than 1 hour. Average down-slope wind speeds at MKN are in the order of 5 m/s. Within 1 hour air traveling at this speed would be out of region of influence of the mountain. Therefore, we are not convinced that dry deposition is the main cause for the observed differences between sounding and mountain measurements. We added an extended discussion of this comparison and the potential of O3 deposition to the revised manuscript.

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.

All technical corrections were followed as suggested by the referee.

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

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