3rd review of " Characterizing tropospheric ozone and CO around Frankfurt over the 1994-2012 period based on MOZAIC-IAGOS aircraft measurements" by H. Petetin et al.

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Summary:

This paper presents a very useful analysis of the extensive data set of vertical profiles of ozone and CO over the Frankfurt region collected by the MOZAIC-IAGOS program. The paper is much improved from the previous drafts. Nearly all of my concerns have been fully addressed, but one concern remains; when this concern is addressed, I recommend that this paper be accepted for publication.

Final concern:

In their trends analysis, the authors correctly point out the importance of autocorrelation of the data, and analyze the trends by methods that account for the correlation of annual and seasonal means between successive years. However, autocorrelation at other time scales (points 1 and 2 below) and with altitude (point 3) is also important, and should be accounted for in some of the analyses. Three specific issues are identified below.

1) The shaded region in Figure 5 is now defined, but I suspect that the ±2 standard error is underestimated here. I presume that it is calculated by dividing the standard deviation of the measurements by the square root of the number of measurements. However, not all measurements are independent, since there is likely a great deal of autocorrelation between successive measurements. That is, a given 4-second average of O_3 is likely to be very similar to the immediately receding and immediately following 4-second averages. Was this autocorrelation accounted for in the calculation of the standard errors? If not, the standard errors should be recalculated based on the standard deviation of the measurements divided by the square root of the number of INDEPENDENT measurements. A short discussion of the method used should be given in the Supplement.

2) I am also concerned that the confidence limits derived in Section 3.4.1 are overly optimistic. The shift in the seasonal cycle is now based on daily data, instead of the monthly average data considered in the previous version of the manuscript. However, ozone measurements from sequential days are fairly strongly autocorrelated, because ozone changes in the troposphere are largely driven by synoptic scale transport, which has something like a 5 to 7 day time scale. The authors must examine the degree of autocorrelation, and reduce the numbers of degrees of freedom in the confidence limit calculations accordingly. For example, if they find that the autocorrelation becomes insignificant only after a 5-day lag, then the numbers of degrees of freedom should be reduced by a factor of 5 from the number of daily averages. This would increase the confidence limits in Table 2 by about a factor of the square root of 5. In this regard, if the confidence limits are calculated with proper consideration of this autocorrelation issue, I would not expect the confidence limits to be much improved by using daily averages over those obtained from using monthly averages. The reason for this expectation is that the calculation of the monthly average minimizes the sum of the squares of the deviations of the daily averages from the resulting monthly averages. Then the regression fit of the monthly averages to the sine function minimizes the sum of the squares of the deviations of the monthly averages from the fitted sine function. On the other hand, the regression fit of the daily averages to the sine function minimizes the

sum of the squares of the deviations of the daily averages from the fitted sine function. Formally, the process of fitting the sine function gives comparable results regardless of whether it is done in two steps (daily data averaged to months, and then a fit to monthly averages) or one step (a fit to daily averages). However, there may be subtleties in the data structure that actually do give more precise results from the one step process, so it is reasonable to maintain the fit to daily data, but the autocorrelation issue must be properly considered. A short discussion of these issues should be included in the Supplement.

3) Autocorrelation is also important in the vertical. If I understand correctly, the daily averages were separately calculated for three atmospheric layers: the LT (a layer 1 to 2 km thick), the MT that is (several km thick), and the UT (~1.6 km thick). Thus, a day with a single altitude profile gives three averages, one for each layer. However, especially in the MT, I suspect that a single profile actually gives two or more independent measurements representing ozone or CO in the mid-troposphere. The authors should investigate the autocorrelation of the measured concentrations as a function of altitude offset. I expect that the autocorrelation is insignificant for vertical offsets of ~2 km, thus allowing several measurements of ozone or CO to be obtained from each profile, at least in the MT. If this issue is properly considered, then the numbers of degrees of freedom would be greater, implying that the confidence limits in Table 2 should be smaller, at least in the MT. A short discussion of these issues should be included in the Supplement.