We would like to thank the reviewers for highlighting the sections in the paper that need reworking. In the response we clarify several general points, while major revision was made in the revised paper. Reviewer comments are followed by author's clarification and responses in bold text.

Response to comments to referee #1 on "Methanol-CO correlations in Mexico City pollution outflow from aircraft and satellite during MILAGRO"

I recommend that the authors redo their analysis with Version 5 of the TES methanol product when it becomes available.
The TES V005 algorithm has been operational since the fall of 2011, but does not provide methanol retrievals. The methanol retrieval results presented in the paper were obtained from the algorithm developed at AER, which is essentially identical to the algorithm that will be implemented operationally in the TES V006 algorithm. Thus the methanol results from V006 are expected to be very close to those presented here.

In the abstract,
Last sentence of the first paragraph: Since the ratios are significantly different from the two aircraft because of possible calibration issues, I believe that speculating on the cause of higher ratios during MILAGRO is premature.

Not sure what exactly the reviewer means. If the sentence in question is "$\Delta CH_3OH/\Delta CO$ ratios from the two in-situ aircraft measurements are far higher than previously reported CH_3OH emission ratios relative to CO from US cities", then it is actually based on the literature as shown in Table 1 and no speculation is involved.

2) Second sentence in the second paragraph: I think the word, similar, is not the correct word. The TES ratios are lower than the ratios from either aircraft.

The sentences in question is changed to: The TES derived CH_3OH/CO ratios during MILAGRO are 19±4 ppt ppb^{-1}, as compared to those observed from the DC-8 (26–39 ppt ppb^{-1}) and C-130 (41–55 ppt ppb^{-1}) observations.

3) You did not demonstrate that TES can clearly distinguish differences in the ratio due to different source categories in CH3OH.

The slopes of methanol/CO ratios measured by TES are regionally very different: ~0.023 ppb $ppb^{-1}$for Mexican region and 0.042 ppb ppb^{-1} for Amazonia. Given that the sources of methanol in these two distinct regions are very different, that TES can detect this is an interesting result in itself.

4) The fact that the MILAGRO data do not allow for a validation of the TES data does not support the conclusion in the last sentence of the second paragraph.

The fact that the aircraft ratios in the MILAGRO data do not agree closely with the corresponding TES data could be explained by accuracy of the aircraft measurements (discussed in section 2.1), or the spatial and temporal consistency between aircraft and satellite measurements (section 3.2.3). The motivation of this study is to gain better source information of CH_3OH in terms of its magnitude, seasonality and spatial distribution by any potential of using the CH_3OH-CO correlations from TES observations with high spatial/temporal coverage. Through the analysis of Mexico City and Amazon Basin, we show the potential of utilizing TES derived CH_3OH-CO ratios globally for better understanding of emission sources of CH_3OH.

Section 2 & figures: a) You discuss errors in both the aircraft and satellite data. Please plot these uncertainties on your figures 4-7. Are the vertical lines in Figure 4, the range of data or uncertainties? b) Please state whether the correlations are statistically significant as I'm concerned that the number of points, particularly in figure 6, are too few. c) In Section 2.2 (line 28), I believe that the first sentence of the fourth paragraph is an exaggeration because of the word, extensively.

a)The vertical lines in Figure 4 are the standard deviation of the data. In the correlation plots (Figure 6 and 7), we show the errors for the fitting of regression lines.
b)In the revised paper, we combine all MILAGRO TES data into one panel. Increased number of points should be able to enhance the statistical significance.
c)Removed the word extensively.

Section 3: a) I don't understand the last sentence of the second paragraph (line 17). What other major sources of methanol are there? Anthropogenic and biogenic really cover the vast majority, right? b) I don't understand why you continued your analysis knowing the issues that the TES CO product has with higher surface altitudes. The implication of figure 4c is a red flag for me and the discussion in the second paragraph of Section 3.2 is disturbing. Why not redo this analysis with Version 5? c) The second sentence of the second paragraph of Section 3.4 (line 21) contradicts your conclusion that the methanol to CO ratio is of any use to distinguish between sources of methanol.

a)Yes, as indicated by the correlation with other indicator tracers, the major sources of methanol were anthropogenic and biogenic sources.

b) As we discussed in Section 3.2.3 of the revised paper –
In contrast to the aircraft observations, the TES observations of CO show no decreasing trend with distance downwind of the city. The limited spatial variability in TES CO for this area has been previously reported by Shim et al. [2009] but the reasons causing the lack of spatial variability are not clear. There was a concern that the handling of the TES CO prior constraints associated with the high altitude of Mexico City contributes to the inadequate spatial variability in TES CO during MILAGRO. However, shifting the CO constraints to account for the high surface elevation regions in Mexico City showed little difference in CO retrieval results,

which indicates the currently unshifted CO priori constraints at elevated surface regions are unlikely to be the reason for the lack of spatial variability in TES CO. Possible reasons for the lack of spatial variability in TES CO during MILAGRO could include under-sampling of TES locations and coarse vertical resolution of the TES measurements. In situ outflow profiles measured during MILAGRO show concentrations that peak in a relatively narrow range. The TES retrievals are unable to distinguish between profiles with a sharp, strongly enhanced peak, and profiles where the trace gas enhancement has lower peak values but is spread over a wider vertical range. Therefore, TES does not reproduce the extreme high peak values observed by the aircraft, and would be expected to show lower average values than the plume-chasing aircraft, particularly in the region closest to the city center.

However, the vertical resolution of the TES measurements is not expected to bias the derived $\Delta CH_3OH/\Delta CO$ ratio, as discussed in Section 3.2.1. Under-sampling, which could contribute to the lack of spatial variability in TES CO during MILAGRO, is expected to have limited impact on the CH_3OH-CO correlation.

c)The sentence in question - "$\Delta CH_3OH/\Delta CO$ ratios for air masses with significant source influence vary little with geographical location." Is only limited to the Mexico City outflow region.

Anonymous Referee #2
Received and published: 19 April 2012

General Comments:
The authors compare determinations of methanol (CH3OH) and carbon monoxide (CO) retrieved from the Tropospheric Emission Spectrometer (TES), with observations of CH3OH and CO made in the outflow of Mexico City during the 2006 Milagro Campaign. The paper argues that the ratio of CH3OH/CO can be used as an indicator of the sources, and source strengths of CH3OH. The authors state that satellite observations can provide unique perspective where in situ measurements are lacking. The ideas proposed in the paper are interesting and if determined would be of value to the community, however issues with the TES determinations of CO and CH3OH over high surface elevation sites (such as that studied here) seriously compromise the validity of the conclusions. While this study is of value, it should not be published until the issues with the retrieval over high surface elevation sites are resolved. As I result, I recommend the paper be rejected, and resubmitted when the TES retrievals in these complex regions can be validated.

We address this issue in detail in Section 3.2.3.
There was a concern that the handling of the TES CO prior constraints associated with the high altitude of Mexico City contributes to the inadequate spatial variability in TES CO during MILAGRO. However, the experiment with shifted CO constraints in altitude for high surface elevation regions in Mexico City shows little difference in CO retrieval results, which indicates the currently unshifted CO priori constraints at elevated surface regions are less likely to be the reason for the lack of spatial variability in TES CO. Possible reasons for the lack of spatial variability in TES CO during MILAGRO could include under-sampling of TES locations and coarse vertical resolution of the TES measurements. However, the vertical resolution of the TES measurements is not expected to bias the derived $\Delta CH_3OH/\Delta CO$ ratio, as discussed in Section 3.2.1. Another possible reason of under-sampling, which could contribute to the lack of spatial variability in TES CO during MILAGRO, is expected to have limited impact on the CH_3OH-CO correlation.

Specific Comments:
P5706, L15: What do the ranges represent? Is this simply the min-max? Why was this not reported as a mean+/-stdev as in all other references cited? What is the uncertainty associated with these ranges?

In the revised paper, we use "mean+/-stdev" format as suggested.

P5707, L15: What are the sink specific lifetimes? Which process is more important?

The major sink is due to the oxidation by OH, leading to CH_3OH lifetime of around 1-2 weeks. The dry and wet depositions are minor sinks, with CH_3OH lifetime

ranging from a few weeks to a few months. Jacob et al. (2005) has more detailed information.

P5706, L28: Given all of the previous work on this topic (cited above), what specific new insight is this paper bringing? How exactly will it reduce the uncertainty in the source strengths, seasonality (was this discussed), spatial distribution?

By taking advantage of generally better-constrained CO sources and the CH_3OH-CO correlations from TES observations with high spatial/temporal coverage, we could gain better source information of CH_3OH in terms of its magnitude, seasonality and spatial distribution. This is exactly the motivation of this study. Through the analysis of Mexico City and Amazon Basin, we show the potential of utilizing TES derived CH_3OH-CO ratios globally for better understanding of emission sources of CH_3OH.

P5708, L15: Is there a reference for the validation of the CO or CH3OH product with ambient measurements?

We have detailed discussion in Section 2.2.

P5709, L17: What is the ratio of CH3OH/CO for various other aircraft studies?

We have detailed discussion in Section 3.5 and Table 1.

P5711, L10: The data should only be used on averaging times greater than the sampling time of the instrument with the longest time resolution (2.8min).

That is exactly why we merge the data to 3 min interval, which is longer than the time resolution of 2.8 min.

P5715, L18: “TES does not resolve the CO pollution over the Mexico City Basin” This is a major problem. Figure 4c, provides the reader with zero confidence that the TES determinations in this region are meaningful. The concentration values are well over a factor of 2 different from the observations and the lack of any spatial differences in CO in the outflow of Mexico City make this analysis impossible to conduct. Until these issues are resolved this paper should not be published.

In Section 3 of the revised paper, we have detailed discussion of this issue.

Anonymous Referee #3
This paper uses methanol (CH3OH)-carbon monoxide (CO) correlations from aircraft and satellite data from the MILAGRO campaign to infer biogenic and anthropogenic sources for emissions out of Mexico City. The main goal of this paper is to demonstrate the potential for TES CH3OH and CO data to gain insight into global sources through their relationship. Although the ideas presented are interesting and satellite observations of these species could be of value to the community for source attribution, the lack of information in the TES retrievals over this elevated surface region and the substantial differences with the aircraft measurements do not support the conclusions presented by the authors. I do not recommend this paper for publication currently and suggest resubmission after improvements are made to the TES CO retrievals and/or current versions of the retrievals are presented.

My specific comments are:
In the last paragraph of Section 1:
(1) In the last sentence of the second paragraph, uncertainties are mentioned (ie. magnitude, seasonality, and spatial distribution). How will these results improve these?

By taking advantage of generally better-constrained CO sources and the CH_3OH-CO correlations from TES observations with high spatial/temporal coverage, we could gain better source information of CH_3OH in terms of its magnitude, seasonality and spatial distribution. This is exactly the motivation of this study. Through the analysis of Mexico City and Amazon Basin, we show the potential of utilizing TES derived CH_3OH-CO ratios globally for better understanding of emission sources of CH_3OH.

(2) A comment is made that satellite / aircraft comparisons "proved challenging" because of the "limited sampling coincidence within reasonable spatial and temporal criteria." There is no mention of what these "reasonable" criteria are here and should be discussed in at least one sentence and/or reference.

One related reference, (Shim et al., 2009), is added in the text.

(3) In the second to last sentence, "in order" is not needed.

Fixed.

(4) The last sentence starting with "Descriptions" should not be a new paragraph.

Fixed.

In Section 2:
(1) Cady-Pereira et al. (2012) and Wells et al. (2012) are good references for the TES CH3OH retrievals, but there should be a few more points from their papers presented in this discussion. Cady-Pereira et al. (2012) states that CH3OH retrievals are most sensitive

between 900-700 hpa (which appears to be case here too) with "good" profiles of 0.5 -1 DOF. What was the average DOF for the profiles used in this study? The statement on line 9-10 on pg. 5713 is not specific enough, especially when information content is of concern.

We added in last paragraph of Section 2: "The averaged DOF for CH_3OH is about 0.6 during MILAGRO, and 0.8 for large retrieved values, i.e., when CH_3OH RVMR > 1 ppb. This indicates TES measurements provided significant amount information of CH_3OH during MILAGRO. "

(2) In the TES CH3OH retrievals, there are a possible four different a priori profiles used. The a priori for this region should have been discussed in at least one sentence (since the lower the DOF the more the profile is based off that constraint.)

We added in 3rd paragraph of Section 2: "A priori profiles were generated from a global chemical transport model on a 2° latitude by 2.5° longitude grid, and binned into 4 categories: "clean and enhanced marine (with the threshold of mixing ratios of 1 ppb below 500 hPa), clean and enhanced continental (with the threshold of surface mixing ratios of 2 ppb) scenes."

(3) Why is it not mentioned that Wells et al. (2012) presented comparisons of DC-8, C-130, and TES CH3OH retrievals during MILAGRO? Their results (good or bad) would have provided additional validation that is missing in this discussion.

We added in 3rd paragraph of Section 3.2.3 : "Wells et al. (2012) compared both DC-8 and C-130 CH_3OH data with TES observations using a chemical transport model as an intercomparison platform; the lower CH_3OH exhibited by TES is mainly due to sampling issues. C-130 data contain a pronounced urban influence as sampling was focused over Mexico City while TES orbit did not track directly over the city. The DC-8 flight tracks focused on sampling Mexico City outflow during transport over the Gulf of Mexico. "

(4) TES version 4 CO retrievals are used here despite knowing the probable issues TES will have in the MCMA. Is there a reason, version 5 retrievals were not selected? If there is, that should have been made clear. Otherwise, I revert back to my original suggestion to using later versions of CO retrievals for this analysis, particularly after reading in Section 3.2 the trouble encountered with the CO retrievals over Mexico City as visible in Figure 4c.

The TES V005 algorithm has been operational since the fall of 2011. TES CO V005 retrievals are not significantly different from V004 retrievals over the MILAGRO region, even though the a priori and constraints are different. We suggested in our previous draft that the handling of the TES Version 4 CO prior constraints associated with the high altitude of Mexico City could contribute to the inadequate spatial variability in TES CO during MILAGRO; however our recent experiment with shifting the a priori when the surface elevation increases shows little difference

in CO retrieval results. Furthermore, prompted by comments from the reviewers, we looked again at the locations of the TES observations where we obtained good CH3OH retrievals and found that very few were at high altitude (see map in Figure 1). Therefore we believe the reported lack of TES sensitivity to CO over Mexico City has little or no impact on our analysis.

In Section 3:
(1) The statement, "TES does not clearly resolve the CO pollution over the Mexico City Basin," concerns me. Despite simulations trying to rule out vertical resolution as a problem for the CO retrieval, TES still does not show the trend observed by the aircrafts. At this point, the analysis should have been suspended until further validation could be made on the CO retrievals over elevated regions, or another polluted region should have been used as example for this method of source attribution.

We added the corresponding text in the 1st paragraph of Section 3.2.3.

(2) In Section 3.4, the authors recognize the difficulty to use this aircraft data for evaluation of TES (although Wells et al. (2012) used a similar dataset for their comparisons). Once this was realized, perhaps "coincidence" with aircraft data during this time period should not have been a priority and separate statistics should have been completed with an increased number of TES profiles for this region. The limited number of points in Figure 6 is alarming.

In the revised paper, we combine all MILAGRO TES data into one panel. Increased number of points should be able to enhance the statistical significance.

(3) Line 25 on pg. 5719 should have the word "to" not "too".

Fixed.

(4) The differences in the CH3OH and CO relationship between MCMA and the Amazon Basin show promise for global applications of this method with satellite data. Until critical retrieval errors are removed, the quantitative aspect of this ratio is not reliable in my opinion.

We added the corresponding text in the 3rd paragraph of Section 2.2.