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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Winter- and summertime continental influences on tropospheric O_3 and CO observed by TES over the western North Atlantic Ocean" *by* J. Hegarty et al.

Anonymous Referee #2

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This paper by Hegarty et al. is a worthy attempt to employ TES observations of both O3 and CO in an effort to discern the influence of continental sources from North America on the chemical composition of the free troposphere over the Western North Atlantic Ocean (WNAO). The paper is most successful in providing good evidence that TES captures realistic synoptic scale variability, and TES observations suggest transport patterns consistent with previous research based on airborne field campaigns. However, the paper seems to try too hard to make their results match every element of earlier aircraft work. In fact, it would benefit the paper if the authors remained more focused on what can actually be observed with the data they have analyzed. The paper includes one novel result, illustrating the influence of shallow convective instability,

driven by cold air outbreaks over the WNAO in the wake of cyclonic storms. The authors demonstrate that these events appear to redistribute boundary layer pollutants into the lower free troposphere, or possibly even up to 400hPa when the convection occurs over the warmer waters of the Gulf Stream. While these results are interesting, it is also important that the authors establish that there is no bias in the CO retrievals under these meteorological conditions as a result of low cold cloud fields.

I do think this paper warrants publication, but I believe it needs more than minor edits. There are two main points, scientific issues, that I raise in the next few paragraphs of this review. The first has to do with the change in the TES retrieval a priori, the second has to do with the misrepresentation of what can be reasonably inferred about the atmosphere from plots of composite synoptic patterns like average mean sea level pressure. These need to be addressed. Then I have flagged a number of more minor problems in the text. These can be fixed more quickly, with an eye toward editing for clarity in language and brevity of exposition.

Overall, this paper is extremely difficult to read in the present form. It is too long, with numerous lengthy passages of imprecise language attempting to describe in words what is shown graphically in images. The paper is poorly organized, skipping back and forth between seasons and between static maps of mean synoptic patterns and then much later introducing plots of trajectories. The paper would benefit greatly from some judicious editing. This would presumably also lead to the correction of several grammatical errors. For example the very first sentence of the Abstract has grammatical errors, and should be recast, perhaps as follows: " The distribution(s) of tropospheric O3 and CO(,) and the (synoptic) factors regulating (these distributions) over the western North Atlantic Ocean during winter and summer, were investigated using profile retrievals from the Tropospheric Emission Spectrometer (TES) from 2004-2006." The corrections are indicated with parentheses. I suggest you define winter and summer later in the paragraph, to keep the main topic sentence clear.

One of the main points, made right at the outset of this paper, is that the seasonal a priori biases the TES observations, and the authors even seem to suggest that if the a priori was not altered, they would produce unrealistic seasonal composites. This is rather confusing, and would seem to call many of the published TES results into question. Furthermore, the differences in the a priori profiles based on the Mozart model , and the universal a priori employed here are never shown. The authors should at least illustrate the range of the seasonally varying a priori versus the universal profile they used. Since the a priori was changed, this also raises the question of what happens to TES profiles if they didn't converge. This has the potential to be more likely since the extremes of any one season may be further from a universal a priori profile that from a seasonally variable mean profile. Does this lack of convergence ever happen as a result of switching to the universal a priori? Anyway, by not accepting the standard TES retrieval approach, the authors imply their own published results are in fact dependent on the approach they have chosen!

The authors establish two goals, the first seems almost trivial, to characterize the seasonal distributions of TES derived O3 and CO over the WNAO. However, the second goal is somewhat muddled. Stated more succinctly, it appears the goal is to associate chemical variability (in TES O3 and CO) with synoptic scale variability that controls transport and dispersion. The problem with this approach is that rather than group the TES observations according to transport patterns derived from trajectories, the authors are intent on identifying "synoptic circulation patterns". This is a non-standard meteorological term, and it gets used along with other terms, such as map types, and synoptic-scale circulation activity, in a way that implies the authors are talking about the three-dimensional atmospheric flow. In reality, it is misleading to refer to circulation when the authors have used spatial correlation of meteorological data to identify mean synoptic conditions, specifically, they have classified TES data into groups of days with similar mean sea level pressure.

The problem is, the authors make some very dangerous over-simplifications regarding the transport, or as they often refer to it, the circulation, represented by these composite

synoptic maps. A mean meteorological map, the average of several days that have a high degree of correlation, does not represent a "circulation". In fact, the circulation, or movement of air, associated with a synoptic pattern is a complex three dimensional description of flow. The conceptual model of airstreams in a cyclonic system, referred to in the text and figures, considers that you hold the system stationary, or that you move with the cyclone, that is, they represent storm relative motion. If you want to understand the origin of an air parcel within a specific location of an actual specific synoptic pattern, you really need to calculate trajectories. Not every cyclone will have a strong warm conveyor belt, or dry airstream, or they will not always be present in the same location. These features are a function of the dynamics of specific events. By the way, the representation of airstreams drawn over figure DJF1, even in the conceptual sense, is inaccurate. The WCB should be delivering air from well ahead of where one would analyze a surface cold front; it represents the adiabatic ascent of air in the warm sector rising up over a warm frontal boundary. Similarly, the other airstreams are off in their representation of conceptual, storm relative motion, as drawn in this figure. In addition to a mean synoptic patterns not representing circulation, neither does it describe a "storm track". These are all confusing terms that the authors use when really all they are talking about are mean synoptic patterns.

To address some of these problems of misrepresentation, Table 1a should be dropped completely. Figure 1 should simply refer to DJF mean synoptic patterns. The meteorological characteristics attributed to these patterns, which suggest knowledge of movement, or evolution of conditions, can only be determined from back trajectories. For instance, Table 1a includes a description for DJF1: "Subsiding northwest flow around back of cyclone center covering much of northeastern US and southeastern Canada". In fact, much later in the paper, in Figure 6, there is finally a presentation of back trajectories for at least one sub-region of synoptic pattern DJF1, and it clearly represents some transport that is subsiding from the northwest, however, it shows a number of other transport patterns, or air parcel origins as well. Obviously transport path, or correspondingly, source regions for material are not in fact well characterized by a single synoptic pattern. By definition, these maps represent an average snap shot. DJF1 shows a cyclone over the WNAO east of Newfoundland, with high pressure centered over the central east coast of the US. However, the temporal evolution of this pattern is not illustrated from a simple representation of mean sea level pressure. It is not even reasonable to assume a storm track from a static composite map. This might be at least partially justified if there was a corresponding mean upper-air pattern associated with the surface MSL, but this is not referred to in the discussion of these surface composites.

This is indeed why trajectories are often used to categorize chemical data, instead of synoptic maps: evolution of the pattern matters as much as the pattern. What happened in each case captured by this mean synoptic pattern, did the low push SW to NE, or more directly from W to E, or in fact did it track from NW to SE as the ridge builds in behind it? This will make a significant difference in discerning the transport, and therefore the location or type of continental influence being delivered over the WNAO. Using only mean surface pressure, there is simply no way to consider the evolution of the pattern, or the speed of transition, or effectively, the storm track. This information can be identified , to some extent, by looking at the three dimensional motion of back trajectories. These can help to capture the actual evolution of the synoptic pattern. This is why trajectories were used in previous work (cited by the authors), for example the papers of Cooper, Parrish and Kiley. Depending where you sample a cyclone (where you fly an aircraft for instance), your relative position within the cyclone may determine the type of airstream you sample, but the specific transport is a function of the evolution of conditions, as well as the relative location of the cyclone. Therefore, it is very dangerous to refer to a mean synoptic pattern and use it to say where the air on a given day is originating, or where it goes next. The strength and precise location of a descending dry air stream, or the adiabatic ascent of air in a warm convenyor belt is not predetermined by a general synoptic condition, but is determined by the specifics of the wind and temperature fields and gradient magnitudes on a specific day.

By grouping the events using synoptic patterns in MSL, it is apparent that the trajectories will not all be the same, and therefore the inferred source region or continental influence is not some distinct location, but a composite of various influences. Again, this is why it doesn't really make sense to refer to the "meteorological characteristics" that are listed in Tables 1 a and 1b. Furthermore, since these synoptic patterns represent a composition of many individual days, it is also clear that the composite of associated chemical observations may result in the cancellation of high and low values over some parts of the domain of consideration, the WNAO. This may confound the interpretation of the composite chemical fields. The authors oversimplify or ignore this issue as well. Previous work, as cited, has shown that trace gas signatures within different parts of a cyclone may vary according to the origin and the evolution of the component airstreams. However, while the composite mean sea level may be used to represent the synoptic conditions, only trajectories can be used to discern the three dimensional motion of air parcels. This is much more clearly illustrated when the trajectories shown in figure 6 are finally introduced.

The authors should not refer to circulation patterns, but to synoptic patterns. For example Section 3.1 would become Winter Synoptic Patterns and 3.2 Summer Synoptic Patterns. Dropping Table 1 (a and b), the frequency of occurrence of days described by each mean pattern should be included as a label on Figures 1 and 2. The authors should also explain why in each season, over 30% of the days are not classified (the frequency figures do not total 100%).

I have a real problem with Tables 2 and 3. For each season, the authors calculate the slope of the O3-CO relationship under different synoptic patterns. However, of the 18 slopes calculated during summer, only 5 are statistically significant at 0.01, and another 3 are significant at 0.05. Even though the other 10 values are not significant, the authors go on an compare these values, and discuss the relative relationship of O3 to CO between regions, or under different synoptic patterns. If a slope is not significant, it should probably not be reported, and its magnitude should certainly should not be compared with other slopes. Similar issues arise with Table 3 where only 5 out of 15 slopes are statistically significant.

Section 5 begins to address the variation within a season, the goal is to correlate the composition with the synoptic conditions. Again, I would remove the word circulation. What is really being considered is: "Transport influences on O3 and CO distributions in winter". This absolutely depends on the use of back trajectories. However, as is obvious from the spaghetti plots of trajectories shown, classifying days according to their mean sea level pressure does not capture one dominant source region or transport pattern. The composite distributions of chemistry, again may have confounding cancellations of high and low values averaged over the same location, and it gets attributed as a lack of transport to this location. You might address this by calculating the variance or standard deviation of the chemical pattern, this would highlight regions that observed a lot of variability in composition, variability that has been averaged away in the production of the composite.

On page 23222, its is noted that O3 and CO distributions were different between different synoptic patterns. The meaning of synoptic pattern is clearer than referring to map type. In general, the synoptic patterns (not circulation types) could be grouped into two main types, one representing the relative position and strength of a cyclone, which seemed to enhance continental export, and the other representing an off shore anticyclone, which seemed less conducive to continental export. (Remove references to storm track). It is quite difficult to follow the main idea of the text on pages 23223. Which seems to be trying to address the origin of enhanced CO.

The authors cite evidence of enhanced CO at 681hPa as evidence that warm conveyor belt transport is lofting boundary layer air into the free troposphere. However, when one looks at the trajectories plotted in figure 6 there is no strong evidence of the WCB which should provide lift from around 800 to 300 hPa. The authors describe a warm conveyor belt "transports air masses within an ascending stream". In fact, it would be better to suggest that air parcels are transported within an ascending stream. The works of

Cooper et al., cited in the manuscript, report the movement of airstreams based on backward and forward trajectories from the location of specific aircraft observations. It is not analogous to simply plot a composite synoptic map based on correlations of mean sea level pressure, and then assume that there is a corresponding mean vertical structure. Again, if all the trajectories for each TES overpass have been calculated, why not identify specific airstreams, defining each observation based on its location relative to each real (specific) cyclone for each date, and the representative forward and backward trajectories. This would be a more analogous approach to the published work by Cooper et al, as well as Kiley et al. If this extra effort is not made, at the minimum, the current paper should be reorganized such that the trajectory results for winter synoptic patterns are included with the introduction of the synoptic pattern (Section 3.1), and not folded in after the discussion of summer patterns. Find a way to combine sections 3.1 and all of section 5, and then combine section 3.1 and all of section 6. This seems like it would make more sense, and it's the only way it makes any sense to talk about transport or circulation.

For the discussion of summer synoptic patterns, their variability and associated chemistry, why establish five distinct synoptic patterns , when later you collapse them into only three unique groups? Just introduce the three main patterns from the outset. Again, there are at over 30% of the days that appear to remain unclassified, (based on frequencies reported in Tables 1a and 1b), and yet this never seems to be mentioned in the text! By combining data into larger groups, you may find that statistical tests of the slope between O3 and CO observations may result in more significant results.

Suggested changes to figures:

Figures 1 and 2, plot the maps (continental and state outlines) in grey, this will allow the isobars to stand out more clearly. Since only three summer patterns are discussed, simply collapse the patterns at the outset, and describe three main JJA synoptic conditions. As noted in the discussion, this figure appears to misrepresent the location of conceptual airstreams.

Figure 4. In the discussion of the spatial correlation of highest CO and O3, to my review of Figure 4, these do NOT correlate, the highest O3 is south of Kentucky and Virginia, while the highest CO is north of Kentucky and Virginia, peaking in Ohio?!

Figure 5. Again, it might be helpful to overlay the synoptic MSL pattern in light grey, since you are trying to relate the spatial pattern of the chemical data to the spatial pattern of the high and low pressure centers. I constantly found myself flipping back to Figure 1. Since it was stated at the outset that TES profiles that were flagged as cloudy were removed, it is not at all clear how representative these plots of chemical observations may be. Is there any artifact of cloud cover in these images?

Figure 11. Again, is it possible that the patterns in seasonal ozone are influenced by where TES retrieved cloud free pixels? This would result in lower frequency of reporting at the center of the mean cyclone for example (please address this issue).

Figures 7-10. Although I think this is a very nice analysis of the influence of a cold air outbreak on the mixing of the lower troposphere, it makes good meteorological sense, I would still like the authors to address the role of these low level clouds on the TES retrieval. In addition, I have suggested soundings that could better illustrate the conditions indicative of the hypothesized convection. In both cases, I believe there is a better sounding, based on time and/or location. You should use the closest soundings to the TES over pass, this would be Yarmouth, Nova Scotia rather than Grey Maine. For the Wallops case, the 00UTC sounding is more conducive to the observed CO profiles reaching nearly 400hPa. If one lifted saturated air from near the warm ocean surface (see red line above), and it followed a moist adiabatic lapse rate, the parcel would be warmer than the environment, the air blowing off the continent, as represented by Wallops Island, all the way up to about 450hPa. The area between the red parcel profile and the ambient environment temperature is a measure of the convective available potential energy (CAPE) and there is no convective inhibition to overcome. This seems quite plausible. I have included these figures for reference.

The discussion on page 23255 has a few confusing statements, perhaps they include typos? First, the authors state that "the CO level decreased dramatically over . . .Rhode Island. . .This may be evidence that subsidence inhibited any lofting of pollutants to altitudes to which thermal instruments such as TES are less sensitive to CO". The authors then go on to note: "To the south the profile showed no CO enhancement. . .possibly due to the dispersion of the pollutant plume since surface winds near the Florida coast were backing to the east and while further to the northeast they remained westerly." Again, this is very poorly worded, and leads to confusion. Perhaps you mean to say "winds near the Florida coast were becoming on shore, or northeasterly, although further north along the east coast, they still had a northwesterly component"? However, this may not be the most relevant piece of information regarding the origin of the air that TES sampled. For these individual profiles, why not just look at the back trajectories for this specific case, and not infer the different source region for different parts of this individual TES orbit, particularly since transport differences are what you seem to be trying to explain.

The following is a list of grammatical and typographical errors, and or suggested additions to text. Parentheses, (), represent additions, {}represent deletions. This is not an exhaustive list of all errors, but a sampling of the kinds of changes that should be made, for example, in addition to removing all references to synoptic circulations, I would suggest that all references to ozone or CO "levels", be changed to refer specifically to mixing ratios (or volume mixing ratios). I cite specific examples below, but other instances can be found throughout the document.

Page 23214, line 16...more frequent(ly); line 18, ozone {levels} (mixing ratios); line 21 further(more); end of paragraph, you should consider a reference to the role of lightning production of ozone (add Cooper et al., 2007, reference included below).

Page 23215, line 5, . . .grouped TES observations by synoptic (weather patterns) {circulation type} and created composite O3 and CO distributions . . . .to identify the average chemistry associated with each synoptic pattern to postulate factors regulating the composition. {the salient characteristics and their causal mechanisms}.

Page 23216, line 11 . . .we used. . .which (removed) {removes}

Page 23216, line 17, of {the} each element of the true state vector

Page 23215, line 15, which is a post-processing diagnostic (that) {which} defines Page 23218, remove references to “circulation”, replaced with synoptic pattern, or composite mean sea level pattern

Page 23218, line 21, the warm conveyor belt transports air {masses} (parcels in the warm sector, lifting air from) the boundary layer. . .

Page 23219, line 15, from (the) continental boundary layer . . .was possibly restricted; the authors have the trajectories to evaluate whether large scale subsidence under the high was restricting vertical transport.

Page 23220, line 17, they typically developed as migrating upper level {synoptic waves} (troughs or short waves) approached the coastline.

Page 23221, line 16, This is not an “export pathway”! (The northward shift of the highest composite CO mixing ratios relative to the winter chemical pattern suggests a correlation with the northward shift in the mean sea level pressure pattern.)

Page 23221, line 27, To identify possible sources for {types of} the {higher} (enhanced) O_3 and CO {levels} (mixing ratios) at {the} 681hPa we {applied the correlation} calculated the correlation between O_3 and CO

Page 23222, line 6, Slopes do not “take place”, similar slopes were derived from observations in the lower free troposphere

Page 23227, line 29, one of the striking features was the {high levels} (enhanced mixing ratio) of CO

Page 23229, line 17, the five original {map types} (synoptic patterns) for summer could

be grouped in three more general {flow patterns} sets of synoptic conditions.

Page 23230, These passages make very misleading analogies of flow, referring the the synoptic pattern as if it was a stationary, persistent feature, and not simply a average of several days of similar mean sea level pattern. Again, this is very dangerous and wrongheaded. All of this discussion, and on page 23231, 23232, makes these same interchangeable statements about transport, derived as if the synoptic pattern was fixed in space.

Once these recommended changes have been made, I am confident the paper will be much stronger, and ready for publication.

Additional Reference suggested: Cooper, O. R., M. Trainer, A. M. Thompson, S. J. Oltmans, D. W. Tarasick, J. C. Witte, A. Stohl, S. Eckhardt, J. Lelieveld, M. J. Newchurch, B. J. Johnson, R. W. Portmann, L. Kalnajs, M. K. Dubey, T. Leblanc, I. S. McDermid, G. Forbes, D. Wolfe, T. Carey-Smith, G. A. Morris, B. Lefer, B. Rappenglück, E. Joseph, F. Schmidlin, J. Meagher, F. C. Fehsenfeld, T. J. Keating, R. A. Van Curen and K. Minschwaner (2007), Evidence for a recurring eastern North America upper tropospheric ozone maximum during summer, J. Geophys. Res., 112, D23304, doi:10.1029/2007JD008710.

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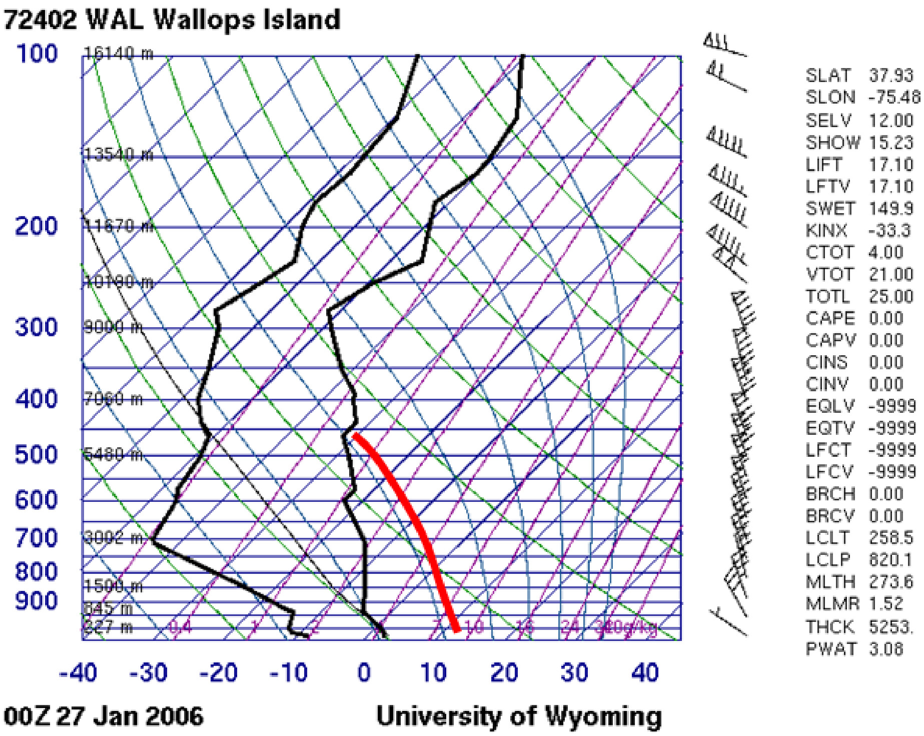

Fig. 1. A parcel lifted from the surface in this environment, but along a moist adiabat starting around 12C would have significant CAPE and would mix to an equilibrium level around 450 hPa.

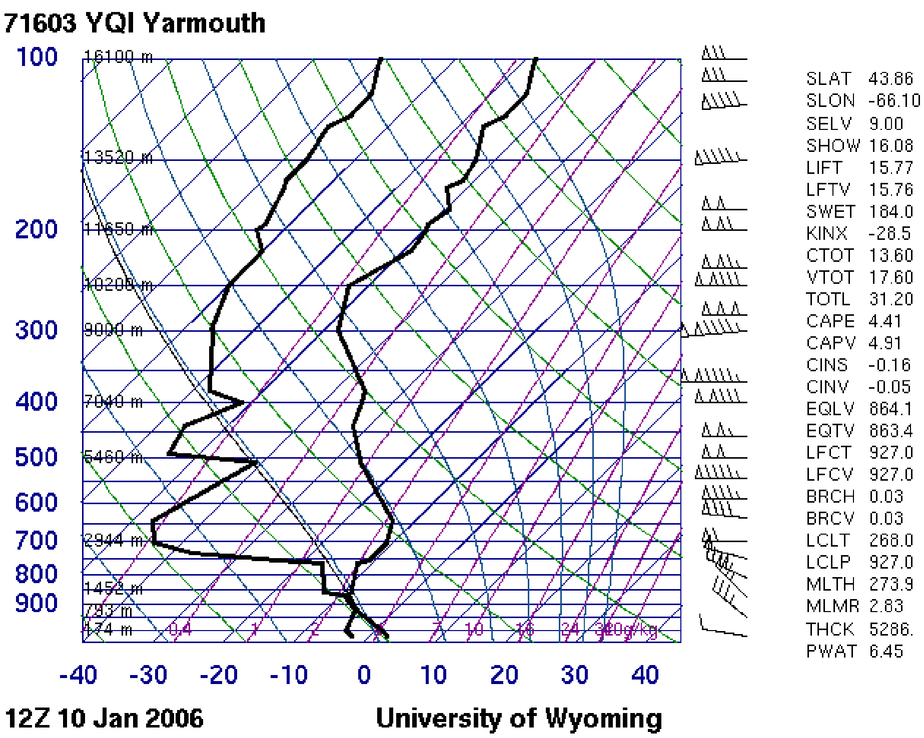

Fig. 2. This sounding from Yarmouth shows the potential instability, and is closer in space and time to the TES orbit.