We would like to thank both referees for their helpful and insightful comments. Our responses to both reviews are in a single document. The referee comments are in bold and our response is given directly following the comment.

Referee #1

My biggest complaint is that Section 5.1 lacks any comprehensive error analysis. For instance, Table 3 presents no uncertainty estimates for the canister-derived emission estimates. On p.1104, L10 it is stated that "a boundary layer height of 125 m was used", but no assessment of uncertainty or variability in that parameter is given.

Thank you for pointing out our oversight of not including an error analysis for some of the results presented in Tables 3 and 4 and for not providing a thorough discussion of some of our assumptions made in estimating emission rates. Clearly, there are many potential sources of uncertainty in our calculations, including the boundary layer height, the assumption of local emissions (i.e., minimal horizontal and vertical mixing), and that the NMHC/ethyne ratios are representative emission ratios. We have added text to explain our choice of a nocturnal boundary layer height (H = 125 m) and to explicitly state that our TF GC emission rate estimates, and hence the daily canister emission rates, are directly proportional to the assumed value of the boundary layer height:

"Stable nocturnal boundary layer heights typically range from ~50-200 m at midlatitude continental locations (e.g., Hastie et al., 1993; Gusten et al., 1998; McKendry and Lundgren, 2000; White et al., 2003); thus we chose 125 m as a representative value for the TF site (e.g., Talbot et al., 2005; Sive et al., 2007; Mao et al., 2008; White et al., 2008; Zhou et al., 2005). If we use H = 50 m or 200 m, our emission rate estimates discussed below vary by ± 60%. It must be kept in mind that the resulting emission rate estimates are directly proportional to the boundary layer height which varies with meteorological conditions, time of day, and season (Talbot et al., 2005)."

We originally included the standard error (SE) of the TF GC emission rates in Table 3 which was the SE of the linear regression between the change in NMHC concentration (dC) versus the change in time (dt). We have recalculated the TF GC emission rate error estimate by propagating the error of the SE of the regression and the assumed variability in boundary layer height (± 60% or 75 m). The results of this calculation are included in column 2 of Table 3 (column title: Winter 2006 TF GC Emission Rate). For the winter 2006 daily canister emission ratios, we have included the SE of the linear regression between ethyne and the respective NMHC in column 4 of Table 3. Additionally, the winter 2006 daily canister emission rate errors are propagated from the error in the TF GC ethyne emission rate and the SE of the daily canister emission ratios. These results are included in column 6 of Table 3. The New Hampshire and New England emission rate errors were propagated from the winter 2006 TF GC emission rate (columns 7 and 8 of Table 3 and column 2 of Table 4).

In the abstract it is stated that "toluene emissions were overestimated (20-35%) in both versions of the NEI". I'd be hard pressed to believe that the overall uncertainty on the emission rate calculation presented here is better than 20-35%, to allow such a claim.

We agree that the overall uncertainty of both our emission rate estimates and the NEI is likely rather large and that we didn't properly address the potential uncertainty in our calculations (see our response to the first comment above). To the best of our knowledge, an overall assessment of the uncertainty in the NEI estimates is not available, but it can be assumed the uncertainty is large. There are numerous possible sources of error in the NEI, such as incomplete or inaccurate emission reporting, different emission rate calculation methods, and changes in emission rates due to new regulations. Based on the results obtained from the propagation of error calculations, we have changed the text in the abstract and in section 5 to include the percent difference between the TF and NEI emission rates but do not refer to differences as overestimates or underestimates.

Abstract:
"Emission rates of benzene, toluene, ethylbenzene, xylenes, and ethyne in the 2002 and 2005 EPA National Emissions Inventories were within ± 50% of the TF emission rates."

Section 5:
"The emission rates of benzene, toluene, ethylbenzene, xylenes, and ethyne in both the 2002 and 2005 NEI agreed within the error limits and were the same order of magnitude as the TF emission rates (Table 4). The emission rates of benzene, ethylbenzene, and xylenes were fairly similar (within ± 15%) in the two versions of the NEI. The relative distribution of emissions in the NEI and at TF was the same in 2002 with toluene > xylenes and ethyne > benzene > ethylbenzene….."
"A notable difference between the two versions of the NEI is the higher toluene emissions from nonroad sources (specifically recreational, lawn, and garden equipment) in 2005 than in 2002 (Table 4). While still within the error limits, the largest difference between the central TF emission rate and the NEI was for toluene. The 2002 and 2005 total NEI toluene emissions were 30% and 50%, respectively, higher than the TF estimates……"
"The total 2002 NEI and TF ethyne emission rates agreed remarkably well (~2300 Mg/year) (Table 4). However, the total 2005 NEI ethyne emission rate was ~40% lower than the central TF value, but was still within the error limits of the TF emission rate."

The nighttime calculation (wind speed <1 m/s) seems like it will provide very local emission estimates representative of the immediate area, as opposed to the daily canister approach. Please comment on this discrepancy.

That is an interesting comment which we have considered before. As the reviewer states, the emission rates shown in Table 3 are presumably representative of source regions of different sizes. The TF GC emission rates, which were calculated using nighttime data when transport was minimal (wind speed < 1 m/s), are assumed to reflect local emissions. Assuming an upper limit of 1 m/s for 5 hours, transport from sources within an 18 km radius around TF is possible. The daily canister emission rates reflect daytime data when the boundary layer is deeper and mixing and transport are occurring. Thus these emission rates can be assumed to reflect sources from a larger area.

Despite the different areas potentially represented by the two sets of emission rates, both approaches yield similar results (within the same order of magnitude). Possible explanations for this similarity include: (1) the two estimates are not completely independent because the daily canister emissions are calculated using the TF GC ethyne emission rate; (2) local NMHC emission rates are similar to rates throughout New England; (3) the midday daily canister emission rates retain a signature from the nighttime local emissions. The air masses observed at TF during midday may reflect a combination of the local emissions under the NBL which are mixed with remnant air from the previous day's mixed layer in the morning.

One of the ongoing objectives of the trace gas analysis from the TF site is to evaluate how representative the TF data is of the rest of the region (i.e., New England). As mentioned in the introduction of this manuscript, previous research has observed that the chemical composition of air masses encountered at TF is representative of the region (e.g., Talbot et al., 2005). The similarity (i.e., same order of magnitude) between the TF GC and daily canister emission rates may be an additional example of the regionally representative characteristics of the TF measurements.

p. 1105, L29: suggest "do not appear to be varying detectably" in place of "do not appear to be varying significantly"

We rephrased this sentence to now read:

"The consistency between the estimates for different winters and for winter and summer suggests that emission rates do not appear to be varying detectably with season or year."

p. 1106, L5- agreement to within a factor of 4 (compared to White 2008) and 80% (between the two methods) does not necessarily "indicate the robustness of the emission rate estimates"…please provide additional reasoning or detail to buttress this argument.

We agree that the emission rates for some compounds calculated by both methods in this work and in White et al. (2008) do not agree as well as others and that this can be indicative of problems with the calculation methods. White et al. (2008) and this work provide the first emission rates for these compounds from this region. In addition to providing regional emission rate estimates, one of our objectives was to evaluate the potential utility of using the continuous TF GC data and the daily canister samples to determine the emissions of VOCs from the Northeast U.S. and to develop a regional VOC budget. Considering all of the potential sources of uncertainty and that the measurements represent different time periods, we consider the fact that the two sets of emission rates reported in this work and in White et al. (2008) agree within the same order of magnitude to illustrate that our approaches have great potential for providing reasonable estimates of regional emission rates. Using different calculation methods will provide a range of emission rate estimates which can hopefully be used to constrain the regional VOC budget. We are currently examining the applicability of these emission rate calculation methods to other time periods. We have rephrased the text in section 5.1:

"We consider the fact that the winter 2006 TF GC and daily canister emission rates and the summer 2003 and 2004 emission rates from White et al. (2008) agree within an order of magnitude (with the exception of m+p-xylene) to be good agreement. While the emission rates calculated using the box model approach and the emission ratio method are not completely

independent, this analysis does provide promising results that the daytime canister samples provide representative information on regional emission rates."

Section 3. You should discuss whether there is a regular seasonal change in wind direction that could influence the observed seasonality.

Thank you for pointing out that we did not include this information. We have added the following text to section 3.1:

"It should also be mentioned that there is a variation in dominant air mass transport pathways throughout the year which may contribute to the seasonal variation in NMHC mixing ratios. For example, in the winter, the transport of clean, Canadian air masses to New England which are representative of background conditions is more frequent (Munger et al., 1996; Moody et al., 1998; Shipham et al., 1998). In contrast, transport from the south and west is more frequent during the summer (Moody et al., 1998; Fischer et al., 2004; Mao and Talbot, 2004b)."

p. 1100, L11-14. These statements don't seem to hold water. Presumably both LPG and FF use correlate to some extent with population, and any fine-scale differences between the two have been integrated/mixed during transport to your site. I don't see any basis for the statement "suggests that non-vehicular exhaust emissions, such as residential use of natural gas or LPG, were important sources of ethyne and benzene."

We agree with the reviewer that the chemical composition of the air masses measured at TF is a mix of emissions from various sources, which usually can not be distinguished, and that population definitely plays a role in the spatial variation of emission sources; we have rephrased the text accordingly. The point we were trying to make in the sentence in question (page 1100, lines 11-14) is along the same lines as the reviewers comment. The strong correlations between propane, ethyne, and benzene (Figure 9), which have similar atmospheric lifetimes, suggests that these compounds originated from either the same source, collocated sources, or various sources at similar distances from TF. We suggested that combustion emissions related to residential natural gas or LPG use are a possible contribution to the strong correlation between ethyne and benzene with propane because of the widespread use of these gases in New England (Environment Northeast, 2009; Energy Information Administration State Energy Consumption Estimates, 2009). While we do not have conclusive evidence that this is true, we also do not have evidence that it isn't; the correlations may just be a reflection of pervasive fossil fuel use emissions from vehicle exhaust, power generation, and/or various gasoline related sources (e.g., whole gasoline in vehicles, natural gas, LPG). Furthermore, Goldan et al. (2004) illustrated that (1) propane and benzene were correlated and (2) the relationship between propane and benzene was different in air masses observed off the New England coast compared to other U.S. locations. This supports our suggestion of a regional propane source influencing the benzene distribution over New England. We have rephrased the text to:

"The strong correlations between propane and ethyne (Figure 9a), propane and benzene (r^2=0.71-0.76, not shown), and benzene and ethyne (Figure 9b) suggest that emissions from natural gas, LPG, and fossil fuel/incomplete combustion (e.g., vehicles) were concurrent and/or

collocated. Furthermore, this illustrates a pervasive and continuous influence of emissions from natural gas and petroleum sources on the composition of air masses observed at TF."

You may want to consider moving parts of the methods sections to Supplemental.

We appreciate and understand the reviewer's suggestion to move parts of the methods section to a supplemental section, and we are aware that not all readers are interested in this information. However, we prefer to keep the majority of this information in the manuscript so that it is readily available to those who are interested, and those who are not, may simply skip this section. We did remove some details from sections 2.1 and 2.3 which were not specific to the compounds discussed in this manuscript in order to reduce the overall length of this section.

Section 3.1 Previous VOC surface measurements in this region from other campaigns would also provide useful context for this section.

Our original main objective for this section was to compare the TF NMHC trends with previous studies of the seasonal variation of NMHCs over North America (with the exception of the Baker et al. (2008) and Gilman et al. (2009) references) which is why we did not include measurements made over the ocean (e.g., NEAQS 2002) or specifically focused on the summer (i.e., NARE, NEAQS 2002, and ICARTT). We appreciate the reviewer's suggestion and acknowledge that these should be referenced and will be included in the manuscript. We have added the following text to section 3.1:

"In summer, the C_2-C_6 alkanes, ethene, and ethyne were similar to or higher than values reported in Canada the previous decade, including the downwind site at Chebogue Point, Nova Scotia (Jobson et al., 1998). With the exception of HF, summertime mixing ratios of propane, ethene, and ethyne at TF were consistently higher than or similar to values reported for rural sites throughout the continental U.S. and Canada. In contrast to the 1993 NARE campaign, the median mixing ratios of C_3-C_8 NMHCs were factors of 2-3 (factor of 6 for toluene) lower at Chebogue Point during summer 2004 (Millet et al., 2006) than at TF."

We have also added the following text to section 5.1 to indicate that the impact from NMHC emissions on the continent have also been documented off the NH coast, and hence, in continental outflow:

"It is worth noting that the influence from a large propane source in southeastern NH has also been documented off the New England coast (Goldan et al., 2004)."

Section 3.3. Is there a high-NO_x assumption here?

Thank you for pointing out that we had not addressed the role of NO_x in summer O_3 production in this section. We are, in fact, assuming that there are sufficient levels of NO_x available for O_3 production to occur. Daytime summer NO mixing ratios at TF range from ~0.2-1.3 ppbv (hourly average from ~06:00-18:00 (Talbot et al., 2005); this corresponds to when the daily canister was collected). During the summer 2004 ICARTT campaign, NO_x mixing ratios ranged from 0.21-17.5 ppbv (average 2.3 ppbv) (Griffin et al., 2007). These mixing ratios are sufficient for the

$RO_2+NO \rightarrow RO+NO_2$ reaction to be dominant over peroxy radical self reactions or the RO_2+HO_2 reaction (e.g., Flocke et al., 1991; Roberts et al., 1998). These conditions are favorable for O_3 production. We have added the following text to section 3.3:

"For these calculations, we are assuming that conditions are favorable for O_3 production. The atmosphere over New England in summer is generally NO_x-limited because of the abundance of biogenic VOCs. Nonetheless, summer NO_x mixing ratios at TF have been observed to range from 0.21-17.5 ppbv (average 2.3 ppbv) (Griffin et al., 2007). These NO_x mixing ratios are sufficient for the NO + peroxy radical reaction to be preferred over peroxy radical self reactions (e.g., Flocke et al., 1991; Roberts et al., 1998)."

p. 1087, L1. 10:00-15:00- are these integrated samples over this entire time frame, or merely filled at some point during this window? Please clarify.

The canister sample was filled at some point during the 5 hour interval ranging from 10:00-15:00 EST. We have clarified the text to read:

"An ambient canister sample has been collected at some point between 10:00-15:00 (EST; UTC-5 hours) each day at the top of the 15 m tower next to the manifold inlet for all the instruments housed in the TF trailer."

p. 1093, L4. Based on Table 1 and Fig 3d, I would characterize this as a summer minimum, not late spring, for these 3 compounds.

We agree with the reviewer that ethene, ethyne, and benzene mixing ratios are low throughout the summer. These three compounds first reach their annual minimum in late spring (May-June). The intention of the original phrasing of this sentence was that ethene, ethyne, and benzene reached their annual minimum in late spring and remained at minimum levels through the entire summer. In order to clarify our point, we have rephrased this sentence to:

"Ethene, ethyne, and benzene remained at their annual minimum mixing ratios from late spring to late summer (Figure 3d)."

p. 1094, L19. suggest "(total range 2.4-5.7 ppbv)"

We agree that inserting the word 'total' will help clarify that the O_3 mixing ratio range listed in the parentheses is the sum of contributions from several precursor NMHCs. We have changed the text accordingly to read:

"These calculations indicate that 0.5-2.4 ppbv, 0.46-0.65 ppbv, 0.15-0.24 ppbv, 0.08-0.1 ppbv, and 1.2-2.3 ppbv (total range 2.4-5.7 ppbv) of additional O_3 could be formed as a result of the enhanced propene, i-pentane, n-pentane, n-hexane, and toluene mixing ratios, respectively, in summer at TF."

p. 1097, L15-16. also VOC oxidation in the case of CO, cf. Hudman et al., GRL, "Biogenic vs. anthropogenic sources of CO over the United States", 2008.

Thank you for pointing out that we had neglected to mention that CO also has a secondary source from the oxidation of VOCs. We have added the following text to the first paragraph of section 4.1:

"The oxidation of anthropogenic and biogenic VOCs is also a potential source of CO, particularly in summer when biogenic NMHC mixing ratios are high (section 3.4) (Hudman et al., 2008). Model analysis of a pollution episode during August 2004 in New England suggested that photochemical sources contributed less than 10% of the CO (Griffin et al., 2007). Moreover, CO over New England and the Gulf of Maine during summer 2004 could largely be explained by urban/industrial and biomass burning sources (Warneke et al., 2006). Thus, in our analysis, we neglect the possible secondary contribution to CO."

p. 1099, L2. "strong enough to PARTIALLY counteract OH chemistry"

We agree with the reviewers' suggestion to insert the word 'partially' in this sentence and have added it to the text.

"Our measurements demonstrate that fuel evaporation and headspace vapor emissions of the C_5*-*C_6 *alkanes were still strong enough to partially counteract OH chemistry throughout the entire summer each year."*

Referee #2
Page 1086 beginning line 3. The authors may wish to make clear exactly what new data are presented here and what (if any) has been published previously.

We appreciate the reviewer's comment and think it is a good idea to indicate which data from the TF site has been published before, specifically for the VOCs, and to make it clear that this manuscript includes data for compounds that have not been presented before. We have added the following text to the last paragraph of the introduction.

"We have been conducting VOC measurements at Thompson Farm since 2002. Multi-year VOC data published thus far includes measurements of C_1*-*C_5 *alkyl nitrates (Russo et al., 2010), methyl iodide (Sive et al., 2007), and oxygenated VOCs (OVOCs) and select NMHCs (Jordan et al., 2009). Summertime measurements (2002-2004) of NMHCs, OVOCs, and marine-derived halocarbons at TF were discussed in Talbot et al. (2005), Chen et al. (2007), White et al. (2008), and Zhou et al. (2005, 2008). In this work, four years (2004-2008) of ambient* C_2*-*C_8 *NMHC and anthropogenic halocarbon data from daily canister samples collected at the Thompson Farm site in southeastern New Hampshire are presented and discussed."*

There are many instances where LPG emissions are referred to in the text, but it seems that most (all?) of the references used for comparison of emission ratios for LPG comprise studies of LPG from cities outside the USA (Mexico City, Santiago and China). Are there any data for LPG source emission ratios for New England?

We agree that it would be useful, interesting, and perhaps more relevant, to include emission ratios from the U.S. Unfortunately, and somewhat surprisingly, we are not aware of any emission ratios specific to LPG for New England. There have been several source profile and apportionment studies, primarily for mobile sources (exhaust, headspace vapor, whole gasoline), conducted in the U.S. over the past two decades, but none (that we are aware of) that were focused on New England. We added some references (Scheff and Wadden, 1993; Goldan et al., 1995; Fujita et al., 2001; Watson et al., 2001; Mukerjee et al., 2004) for LPG ratios obtained from the weight percent of LPG/petroleum refining source profiles for the U.S. The sentence comparing propane/n-butane and propane/i-butane ratios in section 4.1 now reads:

"The slopes of the correlation between propane and n-butane (2.2-2.7, Figure 7a) and i-butane (3.8-5.5, not shown) agree with LPG emission ratios (2-4 and 3-7, respectively) (e.g., Scheff and Wadden, 1993; Blake and Rowland, 1995; Goldan et al., 1995; Chen et al., 2001; Fujita et al., 2001; Watson et al., 2001; Mukerjee et al., 2004; Barletta et al., 2008) indicating that LPG emissions are widespread and prevalent in New England."

A few of the added references listed above were originally included with the propane/ethyne and propane/benzene ratios for U.S. sources given in section 4.2. We have added a few more references to this section as well:

"The propane/ethyne, propane/benzene, and benzene/ethyne vehicular exhaust and whole gasoline emission ratios are typically < 1 while ratios from natural gas, LPG, or gasoline vapor

are ≥~1 (Conner et al., 1995; Fujita et al., 1995; Mukund et al., 1996; Lawrimore et al., 1997; Watson et al., 2001, Choi and Ehrman, 2004; Mukerjee et al., 2004; White et al., 2008).”

I am confused by the statement that the ethyne/benzene ratio at TF is consistent with measurements throughout the US, and if it is, what does this imply for LPG (etc) being “important sources” of regional ethyne and benzene? Moreover, I find the discussion of decreasing benzene/ethyne ratios is awkwardly worded-especially in light of the conclusion that there has been no change in the ratio, either observed or estimated. Do the authors mean that the reduction of benzene emissions observed in the mid-90s has now stabilized?

We agree that stating the benzene/ethyne ratio at TF is consistent with other measurements made in the U.S., which implies a predominantly vehicular source, and also suggesting that combustion related to natural gas/LPG is a possible source of ethyne and benzene could be confusing. Our suggestion that natural gas or LPG, in addition to vehicular emissions, could also contribute to the benzene and ethyne observed at TF was proposed as an explanation for the strong correlations between benzene, ethyne, and propane. The same point can be made by simply stating that the propane, benzene, and ethyne emission sources are concurrent or collocated. Furthermore, based on measurements made during the NEAQS 2002 campaign, Goldan et al. (2004) illustrated that (1) propane and benzene were correlated and (2) the relationship between propane and benzene was different in air masses observed off the New England coast compared to other U.S. locations. This supports our suggestion of a regional propane source influencing the benzene distribution over New England.

According to the EPA, the benzene content of gasoline has remained within a similar range (~0.6-0.8%) since the late 1990’s (http://www.epa.gov/otaq/rfg.htm). It is reasonable to assume that the reduction in benzene emissions has stabilized. The point we were trying to make is that the benzene/ethyne ratio was higher in the early-mid 1990’s (>0.3) and before reformulated gasoline regulations began in 1995. These regulations reduced and limited the benzene content of gasoline. The benzene/ethyne ratio has been observed to be lower and fairly constant (~0.2) throughout the U.S over the past decade. Since we began collecting the daily canister samples (January 2004), it does not appear that the ambient benzene/ethyne ratio has changed.

As a result of comments/suggestions by both reviewers regarding the interpretation and explanation of propane/ethyne, propane/benzene, and benzene/ethyne ratios, we have reorganized and rewritten some of section 4.2:

“The propane/ethyne, propane/benzene, and benzene/ethyne vehicular exhaust and whole gasoline emission ratios are typically <1 while ratios from natural gas, LPG, or gasoline vapor are ≥~1 (e.g., Conner et al., 1995; Fujita et al., 1995, 2001; Mukund et al., 1996; Lawrimore et al., 1997; Watson et al., 2001, Choi and Ehrman, 2004; Mukerjee et al., 2004; White et al., 2008). Throughout 2004-2008, the propane/ethyne and propane/benzene ratios ranged from 1-5 and 3-25, respectively, demonstrating the stronger influence of natural gas or LPG relative to incomplete combustion as a source of propane throughout the entire year. This corroborates previous work at TF and Appledore Island (10 km off the NH coast) during summer 2004 which concluded that LPG was the dominant source of propane throughout the entire day in southern NH (White et al., 2008). Moreover, during the NEAQS 2002 campaign, Goldan et al. (2004)

illustrated that the relationship between propane and benzene was unique to the northeast U.S. and different from other U.S. locations. This supports our observations of a significant influence from LPG leakage at TF. Furthermore, the slope of the benzene vs. ethyne correlation was the same in each season of every year (slope of all data=0.21, r^2*=0.91) (Figure 9b). This ratio value is indicative of a vehicular source and is consistent with observations of ambient benzene/ethyne ratios measured during several spring-summer field campaigns conducted throughout the U.S. (Fortin et al., 2005; Harley et al., 2006; Parrish, 2006; Sistla and Aleksic, 2007; Warneke et al., 2007) and in major cities (Parrish et al., 2009). The strong correlations between propane and ethyne (Figure 9a), propane and benzene (*r^2*=0.71-0.76, not shown), and benzene and ethyne (Figure 9b) suggest that emissions from natural gas, petroleum, and fossil fuel/incomplete combustion (e.g., vehicles) sources were concurrent and/or collocated. Moreover, this illustrates a pervasive and continuous influence of emissions from several anthropogenic sources on the chemical composition of air masses observed at TF."*

Going back to the impact of LPG on the TF measurements, the authors state that the propane emission rate is 1-2 orders of magnitude larger than the other NMHCs (page 1105, line 7). These emission rates are again compared to those for Mexico City and Santiago, but not for other regions of the US. I am very interested in whether this high emission rate for propane is a local phenomenon (right around the TF site?), or is representative of S NH or S New England? Since it has already been stated that the TF site has been characterized as representative of the region, some of these previous findings maybe could be outlined here to support this question for the TF site? It would also be interesting if the NEI comparison in section 5.2 included a comparison of the estimated and observed emissions for propane. Is this information available?

We completely agree with the reviewer that comparing our results to other studies conducted in the U.S. would be useful. We have conducted exhaustive searches, but have only found very limited quantitative information on regional emission rates. Clearly, there is an urgent need for such information. Other than work conducted by our research group in New Hampshire (White et al., 2008), we are not aware of any regional emission rate estimates of >C_3 NMHCs in the U.S., with the exception of some work focused on aromatics (e.g., Mukund et al., 1996). White et al. (2008) presented results from concurrent hourly canister samples collected at 5 sites throughout southeast NH, as well as hourly data from the continuous in situ GC at TF, during August 18-19, 2003. This study demonstrated that the NMHC, specifically propane, trends at TF are similar throughout southeast NH.

A comparison with the NEI emissions of propane is beyond the scope of this work; we are currently working on a regional propane paper which specifically addresses the points brought up by the reviewer. We have conducted several spatial surveys which involved collecting canister samples concurrently throughout each New England state during different seasons. The main objectives of these studies were to characterize the regional distribution of VOCs and to compare the TF data to the rest of the region. We have included a figure to highlight the widespread propane leakage throughout New England from two of the spatial surveys conducted in 2006 (Figure 1). The different spatial distribution of ethane and propane (Figure 1c, d) is indicative of a propane source other than natural gas. We are currently analyzing the regional VOC survey data, the continuous in situ TF GC data, and the daily canister sample data in order to characterize the regional nature of VOC emissions, to further evaluate the emission rate estimation approaches presented in this work, and to constrain the estimates. In addition, the work in progress includes comparisons of the observed emission rates for VOCs not presented in this work, such as propane, with the NEI. These results will be presented in a forthcoming paper.

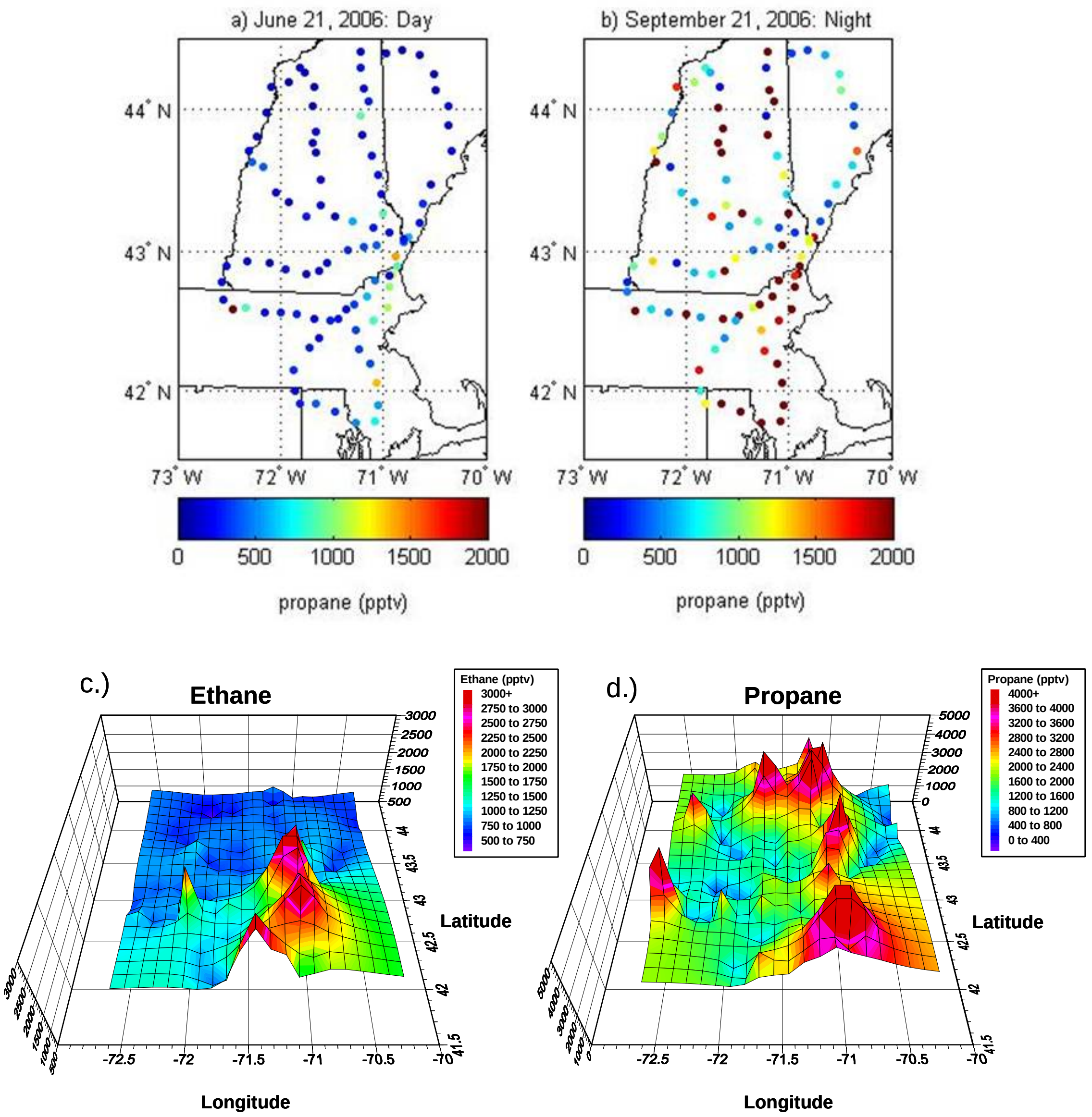

Figure 1. Propane mixing ratios along the regional sampling routes on a) June 21, 2006 and b) September 21-22, 2006. The June samples were collected during the day while the September samples were collected at night. Spatial distribution of c) ethane and d) propane on the night of September 21, 2006.

Section 5.2 Page 1106 paragraph 2. I suggest the authors clarify why ±20% is "good" agreement for benzene, ethylbenzene and xylenes, while 20-30% is an "overestimate" for toluene. I recommend that the authors address emission uncertainties.

We agree with the reviewer that we did not sufficiently address the uncertainty associated with our emission rate calculations. There are many potential sources of uncertainty in our calculations, including the linear regressions, the boundary layer height, the assumption of local emissions (i.e., minimal horizontal and vertical mixing), and that the NMHC/ethyne ratios are representative emission ratios. We originally included the standard error (SE) of the TF GC emission rates in Table 3 which was the SE of the linear regression between the change in NMHC concentration (dC) versus the change in time (dt). We have recalculated the TF GC emission rate error estimate by propagating the error of the SE of the regression and the assumed variability in boundary layer height (± 60% or 125 ± 75 m). The results of this calculation are included in column 2 of Table 3 (column title: Winter 2006 TF GC Emission Rate). For the winter 2006 daily canister emission ratios, we have included the SE of the linear regression between ethyne and the respective NMHC in column 4 of Table 3. Additionally, the winter 2006 daily canister emission rate errors are propagated from the error in the TF GC ethyne emission rate and the SE of the daily canister emission ratios (column 6 of Table 3). The New Hampshire and New England emission rate errors were propagated from the winter 2006 TF GC emission rate (columns 7 and 8 of Table 3 and column 2 of Table 4).

We have removed references to the NEI "overestimating" or "underestimating" emission rates compared to the TF emission estimates in section 5.2. Instead, we interpret the differences between the TF and NEI emissions as percent differences. For example, we have rephrased the last sentence of the abstract to:

"Emission rates of benzene, toluene, ethylbenzene, xylenes, and ethyne in the 2002 and 2005 EPA National Emissions Inventories were within ± 50% of the TF emission rates."

The discussion of emissions from fireplaces and woodstoves being reduced by 70% in the NEI is awkwardly worded (to my mind). Do the authors mean to imply that wood and fireplace emissions actually were reduced from 2002 to 2005? (I do not think so.) And I assume that most VOC emissions from fireplaces and woodstoves are assumed to be in the form of ethyne? (since the other nonpoint emission estimates in Table 4 are not reduced nearly so much from 2002 to 2005). Please clarify this paragraph.

We did not intend to imply that actual VOC emissions from woodstoves and fireplaces in New Hampshire were reduced by 70% from 2002 to 2005. According to the EPA 2005 NEI website (http://www.epa.gov/ttn/chief/net/2005inventory.html), the total VOC emissions in the fireplace and woodstoves source category in the 2005 NEI were reduced, compared to the 2002 NEI, because of a change in the emission factor used to calculate the emissions. We calculated that the total VOC emission rate for New Hampshire in the residential woodstoves and fireplaces source category in 2005 was ~70% lower than in 2002 using the emission rates which we obtained from the NEI database.

The nonpoint source categories for which there was an EPA recommended source profile containing ethyne were open burning of land clearing debris and household wastes and residential woodstove and fireplace combustion. The VOC emissions from open burning were minor compared to the residential combustion emissions in both the 2002 and 2005 NEI. Consequently, the nonpoint ethyne emission rate we calculated predominantly reflects emissions from residential woodstoves and fireplaces. Benzene, toluene, ethylbenzene, and xylene emissions from fireplaces and woodstoves were essentially the same in both versions of the NEI which is why their emission rates are similar in 2002 and 2005. We have rephrased this paragraph to:

"The total 2002 NEI and TF ethyne emission rates agreed remarkably well (~2300 Mg/year) (Table 4). However, the total 2005 NEI ethyne emission rate was ~40% lower than the central TF value, but was still within the error limits of the TF emission rate. Ethyne emissions in the nonpoint source category reflect residential stationary source fuel combustion with a minor contribution from open burning (yard and household waste). Residential wood and fireplace combustion was the largest nonpoint source of VOC emissions in northern New England (New Hampshire, Maine, Vermont (second highest in Massachusetts and Connecticut)) in both versions of the NEI. According to the EPA, the emission factor used to calculate emissions for the woodstove and fireplace source category was reduced for the 2005 NEI (www.epa.gov/ttn/chief/net/2005inventory). Consequently, the VOC emission rate from the woodstove and fireplace source category was ~70% lower in the 2005 NEI than in the 2002 NEI. This resulted in the lower ethyne emissions in 2005 and the larger discrepancy with the TF emission rates. This illustrates the necessity of additional studies to accurately quantify the VOC emissions from residential combustion in the northeast U.S. (e.g., Jordan et al., 2009)."

Page 1107, line 15- Is the lower benzene/ethyne ratio for the 2002 NEI compared to the 1996 and 1999 NEI estimates a reflection of the trend reported by Parrish, or maybe a reflection of it? Please clarify how the NEI values are arrived at. Also reconciliation of the NEI comparison of the ethyne/benzene ratio in the context of the LPG source mentioned earlier would be useful.

We are not quite sure what the reviewer is asking. The point we were trying to make is that Parrish (2006) reported that the benzene/ethyne ratio value decreased from the 1985 NAPAP inventory to the 1996 NEI to the 1999 NEI. Even though the 2005 NEI benzene/ethyne ratio is higher than in 2002, the ratio values we obtained for the 2002 and 2005 NEIs were lower than in the 1999 NEI. This may suggest that the benzene/ethyne ratio has continued to decrease. However, this does not indicate whether the benzene or ethyne emissions are correctly apportioned in the different source categories. We have rephrased the text to read:

"The total benzene/ethyne ratios in the 2002 and 2005 NEIs were lower than the ratios given in Parrish (2006) for the 1996 and 1999 NEIs (0.9 and 0.7, respectively). The benzene/ethyne ratio in the onroad exhaust source category (~0.5) was higher than ambient ratios (~0.2). Our results are consistent with Parrish (2006) who reported that there was a downward trend in NEI benzene/ethyne ratio values but that there are still problems with the apportioning of benzene and ethyne emissions in the onroad source category."

The TF and NEI ratio values were calculated by converting the emission rates (Mg/year) to molar units and then calculating the ratio. We added text to the Table 4 caption explaining how the NEI ratio values were calculated. The Table 4 caption now reads:

"The toluene/benzene and benzene/ethyne ratios were calculated by converting the emission rates for the respective source category listed in this table to molar concentrations. The TF toluene/benzene and benzene/ethyne ratio errors were propagated from the error in the TF emission rates."

Typos: Page 1096, line 15 "used" should be "use"

Thank you for pointing out this typo. It has been corrected in the text.

Page 1100, line 6 should be source of

This error occurred during typesetting. We will make sure it does not appear in the final publication.

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