<u>Responses to Evaluation by Anonymous Reviewer 1.</u>

The authors thank the reviewer for their very thorough examination of the paper and thoughtful suggestions for its improvement. The final revision will incorporate these changes in addition to those proposed by the additional reviewers. Responses to the reviewer's comments and further details of the planned changes are presented below.

General Remarks:

As the reviewer discerned, the motivation in this study was to employ unique in-situ measurements in a physical-chemical analysis of a low altitude plume's evolution from its origins(the NYC source region), through its passage across the Gulf of Maine, and its eventual contribution to North American outflow across the North Atlantic Ocean. Particular interest was devoted to characterizing the lower tropospheric mechanisms which facilitated and shaped the plume's transit as well as its impact on surface conditions across northern New England. The final revision will feature key changes in accordance with both reviewers' suggestions along with new contributions from an additional co-author, Dr. Andy Neumann of NOAA's Earth System Research Laboratory. Key points that will be addressed include:

1) The removal of *section 4.2 and corresponding figure 12* as suggested by the reviewer. This will tighten the focus of this study around the physical and chemical transformation of the plume during the three days it traversed Long Island Sound and the Gulf of Maine. References will be made to the related works by Methven et al. (2006) and Real et al. (2008) in which the intercontinental transit of the plume has been addressed more rigorously.
2) An expansion of the inland impact analysis, with discussion of selected trace gas relationships to validate the influence of the NYC plume on surface mixing ratios at the AIRMAP locations and the possible use of coastal wind profiler observations to discuss the periodicity nature of the mixing of the plume inland and the influence of cross-shore flow on the plume's transit.
3) A revision of the data description, with description of the aircraft measurements and uncertainty thresholds detailed more extensively and current errors in their description corrected and an additional acknowledgement to Thomas Ryerson from NOAA for generating some of the data used in the paper.

Major Points:

1a) *Figures: All of the figures are too small making it difficult to read axis labels, contour labels and location names.*

- Agreed, the figures will certainly be resized (full page) for their final edited form.

1b) *In addition, much of the data plotted in the figures, and even individual figures themselves, are not referred to in the text. The failure to select relevant data means that much unnecessary time is spent working out which line and figure is being discussed in the text. Better selection of data is essential to make this paper suitable for publication.*

- The reviewer's point is taken concerning the extensive data presented in the analysis. Filtering of the figure content as well as improved referencing to particular data presented therein the will be addressed.

2a) *As the authors state, internal boundary layers (IBL) usually form due to discontinuities in the 'surface' properties. In this paper a plume of polluted air is advected across the coast above a marine IBL. This polluted residual layer then adjusts to the new lower boundary properties – now the top of the marine IBL and not the surface – thus forming a second IBL above the marine IBL. It is not obvious from the paper that this is the structure the authors are describing.*

- The confusion regarding the IBL description is noted and the discussion of IBL formation will be expanded beyond the classical definition (Stull, 1988) currently employed in the text to address the scenario observed in this study. If the reviewer is referring to the marine atmospheric boundary layer (MABL) with the use of the term "marine IBL" then yes, that was the structure observed and described. Specifically, an IBL formed as the NYC plume was advected over the coastal waters of Long Island Sound, overrunning the MABL in the process. Distinct stages of plume layer's evolution towards the IBL structure were clearly discernable during the aircraft intercepts, as shown in figure 3. From these, it was seen that, the plume was initially encountered in a residual layer of the continental atmospheric boundary layer near the NYC source region (figure 3 a)-d), black lines). A simplified schematic this atmospheric structure is presented in figure R1 below. Subsequent transformations were evident in the downwind encounters (moving from northeastward from NYC to the GOM). As seen in the mixing ratio profiles of O_3, CO, SO_2, and HNO_3 presented in figures 3a)-d), adjustment of the plume airmass to its new surroundings (green lines and red lines) was manifested in its subsidence, and coalescence towards the surface.

  These structural changes observed in the NYC plume on 7/20 closely resemble the IBL formation described by Skyllingstad et al. (2005) with its evolution into a stable mixed layer over time and distance being similar to the observations of Smedman et al. (1997). Over the North Atlantic, this scenario has been detailed in several closely related studies. Despite the absence of significant forcing, Owen et al. (2006) observed the transport of North American plumes from the Eastern U.S. to the Azores in detached layers just above the MABL. In this, decoupling of the continental airmass, usually through the formation of a residual layer following the diurnal fluctuation of the daytime boundary layer, is the ventilation mechanism for the plume's release. This was recently detailed in the coastal atmospheric study by Dacre et al. (2008) and observed during previous field campaigns in the North Atlantic, as reported by Gong et al. (2000) and Angevine et al. (2004).

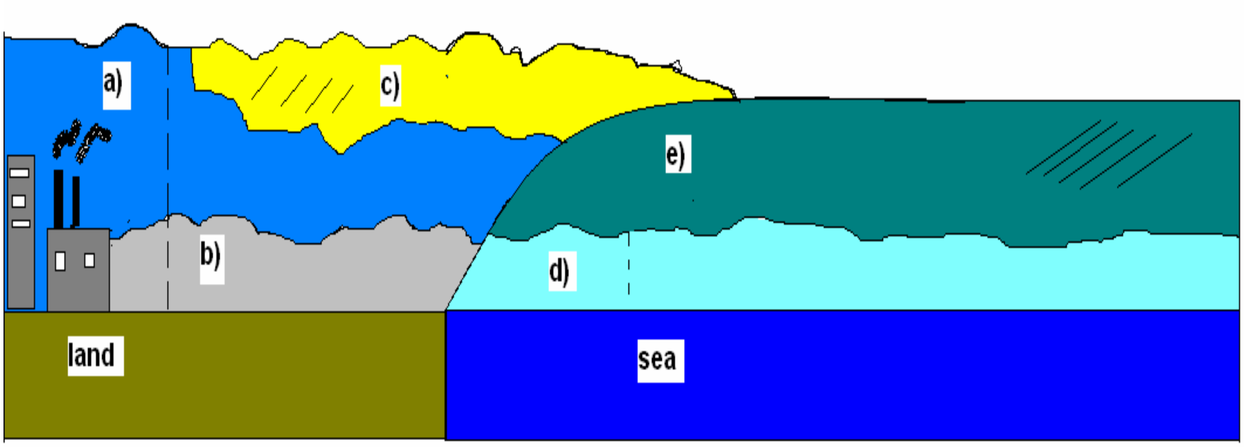

Figure R1. A simplified schematic of atmospheric features in coastal plume transport as observed in the NYC plume study. Shown are the continental boundary layer in which pollutants accumulate. This boundary layer fluctuates between daytime height a) and nocturnal height b), with boundary layer pollutants periodically becoming trapped in a residual layer c) during this diurnal cycle. Airmasses within the residual layer may become entrained or advected over the ocean in the presence of offshore flow, overrunning the MABL d) in the process. This leads to the development of an internal boundary layer e), in which the plume is detached from the surface and conveyed over extensive distances.

2b) *Also, it is not clear how the authors relate the meteorological characteristics of this layer to the chemical measurements.*

- The analysis presented has placed emphasis on characterizing the lower atmospheric structure inducing/facilitating the NYC plume transformation and transit. The author's believe the close relationship between the chemical and physical transformation of the plume are established in the paper. However, the addition of further quantitative expressions such as covariance's between physical and chemical parameters or calculation of lateral and vertical fluxes within the plume layer could be added, if that was what the reviewer was seeking in this comment.

Minor Points

Specific/Technical points: The authors thank the reviewer for their thorough evaluation and the final version will incorporate all of the technical corrections they have advanced. Responses to other particular minor points are then discussed further here.

1. Abstract: There is no explanation for why this study was performed or motivation for the work. This needs to be included in the abstract.

- Understood. A statement of motivation will be included in the abstract for the final revision.

5. P2401, line 21: What process is responsible for the ventilation into the shallow tropospheric layer? Weak synoptic forcing isn't a ventilation mechanism.

- Decoupling is the ventilation mechanism of the NYC plume on 7/20. This is discussed in the major point response section, 2b). In the final revision, weak synoptic forcing and ventilation will be defined more explicitly per their use.

6. P2402, line 1: Use of the word 'off' is ambiguous here. Do you mean that the southwesterly flow is in an offshore direction, or that the southwesterly flow is observed occurring over the ocean?

- This was meant to describe Southwesterly flow over ocean.

c. The figure caption states that figures a, b and c show mean sea level pressure. However, the contours do not look like mslp. Is this correct? Perhaps they are wind speed?

- Figure 1 presents mean sea level pressure 1a)-1c), geopotential heights at 850m in 1d)-f) , geopotential heights at 500mb in 1g)-i) for the three days of the study (7/20, 7/21, and 7/22) and were obtained from the NCEP Reanalysis as available on the NCDC website.

d. The figure caption states that a, b and c show surface wind vectors. Are the wind vectors scaled? If so, include a reference vector.

- The wind vectors are scaled and a representative vector will be added to the legend. It is expected that this figure will be adjusted with an incorporation of brief day by day synoptic analyses suggested by the second reviewer (see that response).

10.a. Do the SO_2 concentrations for 19:20 UTC go off the scale below 500m? If so, extend the axis to include all values.

- The cutoff on the axes was made based on the lack of data below the altitude shown in the case of the 19:20UTC profile and the text will be adjusted to address this ambiguity.

11. P2403, line 4: Shipping is also likely to be another large source of SO2 in this region.

- This is a good point and adjustment of the statement will be made for the final version.

12. p2403 line 4 and fig 4: Why did the authors choose to perform the Lagrangian back trajectories from 1900m, 1200m and 500m? Are these related to the layers of pollutants seen in figure 3? Similarly, what is the justification for the heights of the forward trajectories? Why are the heights chosen for the forward and back trajectories different?

- The figure, text, and the Lagrangian back trajectory computations will be made uniform for the final version. The different heights were selected to match the altitude of the plume's peak as well as its upper and lower bounds near the source region.

13. P2403, line 26: The correlation coefficient '0.83' does not appear in figure 5a. What does this value refer to?

- This is a typographical error. The 0.83 value is the correlation coefficient that should have appeared in Figure 5a and the text.

15. P2404, line 15: The relationship between O_3 and NO_y is described as being 'weaker'. Weaker than what? Also the correlation coefficient is 0.63 in the text but 0.56 in the figure.

- Weaker was used to refer the slope differences in the *O_3/ NO_y* relationship when it was apparent airmasses of different ages (with lesser/weaker slopes corresponding to aged airmasses) may have been mixed into the plume. The text value is accurate and the figure will be adjusted accordingly.

18. P2406, line 1: Where has this detachment of the plume from adjacent vertical layers been shown?

- This statement was made to establish the detachment detailed in the subsequent text and also a connection to related works where similar detachment was observed or simulated with regards to transport over the North Atlantic .

19. P2406, line 5: 'Entrainment' implies mixing of two air masses. The process referred to however, could be explained by advection of the plume into the coastal residual layer.

- The use of entrainment was deliberate, yet advection is another possibility and this statement will be reworded accordingly.

20. P2406, line 26: 'The wind has shifted by 30_o'. Over what time period and from which direction?

- A partial typo here. There was a 20° shift in wind direction between across the vertical extent of the plume layer as shown in figure 3g. This shift from 240° to 260° or from southwesterly to westerly in direction.

23. P2408, lines 1-15. Why are the authors convinced that the peaks seen at 18:30, 19:20 and 20:00 are emitted from the same source, and are subsiding from 1500m to 250m?

- The chemical ratio analysis and the evaluation of the whole air samples (WAS's) was performed to establish the identity of the plume throughout the study period, including the profiles made at 18:30, 19:20 and 20:00 UTC on 7/20.

24. P2410, line 25: What heights were the aircraft transects performed at?

- The transects were between 300 and 500m in altitude which will be detailed in the final version.

31. P2416, line 2: Define SIBL. Stable/stratified/surface internal boundary layer?

- An SIBL is a stable internal boundary layer and will be define in the text.