Responding to Referee #1

1) Figures are too many, they are currently not well edited, and they focus on individual MDEs while summary figures showing general patterns are missing.

We agree that the figures could be improved. For Figure 3; We deleted the detailed chemical characteristics for each case and we modified the vertical distribution of Hg°, O_3, C_2H_6, and Br_2 for all inside and outside MDEs. C_2H_6 and Br_2 are the representative hydrocarbon and halogen compounds. For Figures 4-7; Five rows were reduced to 4 rows and the y-axis is now in mixing ratios. The lines were changed from all solid lines to different line type options.

2) My main criticism on the manuscript refers to the modeling component: in my view, there is a lack of comparison of model results with observational results, and the model component is almost fully detached from observations. The use of box model simulations may not be the best analysis tool for the observations: box models mainly reproduce temporal patterns of individual air masses, while aircraft observations have strong spatial characteristics. A more suitable modeling analysis may hence include a model with vertical and /or areal outputs which might allow much better comparison of observations.

We understand that you point out a lack of comparison of box model simulation results with aircraft observational results and the difference view between box model simulation and aircraft data (temporal vs. spatial). However, the purpose of a box model is to understand mechanisms and rates instead of duplicating observations. Furthermore, regional and global models lack the detailed chemical reactions that are contained in our box model. Those models are employed mainly to simulate large-scale dynamics and limited chemistry. Our goal of this study was not to see how the model captured observational characteristics but how varying chemical environments affected MDEs. The dynamical environment in the Arctic at low altitude is isolated, stable (capped by an inversion), and changing slowly compared to the other environments. We hypothesized that MDEs could therefore be influenced considerably by chemical mechanisms with local source impacts (possibly halogen molecules from the ocean). We utilized the box model to suggest important chemical mechanism that occurs in MDEs by considering comprehensive chemistries that other models can not resolve specifically. The spatial distribution from the aircraft measurement, as you pointed out, was utilized to drive general chemical characteristics in MDEs environment. The general characteristics of the MDEs were two sensitivity conditions; high photolysis rate constant and high Br_2 concentrations, while an interesting characteristic in some of the MDEs was a high NO_x regime. Therefore, we did sensitivity experiments with these conditions by utilizing a comprehensive chemical box model to investigate the varying conditions and chemistries in the Arctic. In other words, we probed various conditions to better understand the primary chemistry driving MDEs.

3) Finally, the discussion and conclusions of the study re-iterates many previously reported results from observational and modeling studies, and does not seem to focus enough on specific novelties (e.g. vertical and areal aspects and implications, effects of pollution) gained from this aircraft campaign.

This is the first time that MDEs have been sampled from an aircraft, and with such comprehensive chemical observations including Br_2, Cl_2, and BrO. The general characteristics including the vertical extent, their spatial scale, and spatial distribution of MDEs is the focus of another paper by Mao et al., JGR, 2010. That paper showed using observations the importance of halogens in MDEs. This paper was a follow-up using modeling to confirm the observational implications. We added several sentences on p.9 to clarify this point. We also cite the Mao paper in our manuscript.

Our findings show that "the results suggest a close relation between O_3 and Hg°", that simulations suggest "negligible effect of Hg° reaction with BrO on the occurrence of MDEs", and that "emissions of halogen compounds is imperative to the occurrence of MDEs and ODEs in the Arctic springtime". These results are not particularly novel. Interesting novel aspects, however, include effects of NO_x regimes and photolysis rate constants. We explained where MDEs were observed in Figure 1 and 2 and section 3. We have added more citations to the Mao et al. paper to make it clearer that the general features are described there but here we focused primarily on the chemical mechanisms. Moreover, we added more discussion of the high NO_x regime from the flight measurement with utilizing simulation results in lines 5-18 on p.19 as your suggestion.

4) No summary or systematic comparisons of these cases are made in order to elucidate common patterns causing these MDE.

We originally summarized 8 MDEs in the first paragraph in Section 3 and we moved the summary to last paragraph in Section 3. The MDEs are quite varied in their physical and chemical characteristics so it is difficult to draw specific general conclusions beyond what we have included in the manuscript. This is also covered more thoroughly in the Mao paper which we added on p.9.

5) On page 10081, line 4-10: the authors state that 14 MDE were observed with Hg^0 mixing rations depleted below 50 ppqv. However, the discussion of MDE events as well as box model analyses only focus on 8 cases that "exhibited generally distinct features of MDEs, i.e., the concurrence of high Br_2, low O_3, and low Hg^0 mixing rations". This selection seems highly subjective and causes bias since the authors only consider cases with high bromine levels– what about the other cases? It seems to me that there is as much to learn from the cases where high Br2 and low O_3 are not present? In the abstract, the authors state that "MDEs occurred near the surface and always over the Arctic Ocean accompanied by concurrent ozone (O_3) depletions, enhancement in Br_2 mixing rations….". This seems incorrect because the authors only selected the cases where they observed O_3 depletions and Br_2 increases in the first place. What about the other 6 cases where this was not the case?

In Figure 1, 14 MDE cases occurred near the surface over the Arctic, so we utilized the word "always". As you mentioned, we changed the sentence to "Measurements showed that MDEs occurred always near the surface over the Arctic Ocean and were accompanied by concurrent ozone (O_3) depletion, enhancement in Br_2 mixing ratios, and decreases in ethyne and light weight

alkanes" on p. 2. The following sentences (line 23 on p.5 – line 4 on p.6 of manuscript) explained the other six cases and the reason why we could not compare 8 concurrent cases and 6 non-concurrent cases because of a lack of sufficient measurement data. Non-concurrent cases occurred over very short distances (e.g. 1-3 data points of flight measurement) in figure 1. In addition, gaseous elemental mercury (Hg°) ranged 31-49 ppqv in non-concurrent cases and most cases showed that the MDEs (<50 ppqv) were included in relatively lower values (51-100 ppqv) of Hg° for several minutes measurement (7-26 minutes) except one case. Therefore we did not consider those 6 cases due to a lack of sufficient data for analysis. We agree that it would be interesting to examine the 6 cases.

6) It is not mentioned how these calculated lifetimes compare to observational results, so what is the purpose of these sensitivity analyses? I suggest that modeling results are directly compared to observations, e.g., in common figures showing both observations and model results.

As you mentioned concerning the flight measurements and box model simulations, it is very hard to compare model results to measurement data. The observations do not show the lifetimes of the various processes. This is why the box model was employed too augment the observational data. Box model simulations were utilized to determine how fast mercury depletion occurs in some controlled chemical environments and we used the concept of lifetime of Hg° to represent how fast mercury depletion occurs in each environment. To check if our model simulations were realistic, we did comparisons of selected chemical species deletion amounts from observations and model output. These are presented on lines 7-15 on pp.14 and they were quite similar.

7) A similar problem relates to the model results showing the different products of Hg oxidation (e.g., HgO, HgCl, HgBr, $HgBr_2$, etc) and how the composition of oxidized Hg changes as rate constants and photolysis constants change– while this seems interesting, it is not entirely clear what the value of these results are and no clear discussions and conclusions are made in regards to these results.

Other RGM compounds (e.g. HgCl, $HgCl_2$, HgBr, and $Hg(OH)_2$) were not described in the text because of very low values compared to $HgBr_2$ as shown in Figures 4-7. Speciation of RGM (mainly $HgBr_2$ and HgO) was mentioned in lines 21-23 on p.13, lines 20-21 on p.16, and lines 8-11 on p.17. Differing rate constants of Hg° reaction with Br radical led to different compositional mixes of RGM as described in lines 7-15 on p.15.

8) I suggest to make a clear table with all sensitivity analyses runs, clearly name each simulation run and have consistent names/description of these in the text, in the methods section, and in the figure legends.

We added the table 4 in the manuscript and the table is shown below.

Table 4. Summary of principal sensitivity experiments.

	Different rate constants									Different halogen conc.					Different photolysis			Different NO_x regime		
	Br radical			Br_2		Cl racical		OH radical		Br_2 (pptv)			Cl_2 (pptv)							
	G	K	D2	A	B	K	D1	G	P	1	3	5	1	5	High	mid	low	high	mid	low
S1	X			X		X			X	X			X			X			X	
S2	X			X		X		X		X			X			X			X	
S3		X		X		X			X	X			X			X			X	
S4			X	X		X			X	X			X			X			X	
S5	X				X	X			X	X			X			X			X	
S6	X			X			X		X	X			X			X			X	
S7	X			X		X			X		X		X			X			X	
S8	X			X		X			X			X	X			X			X	
S9	X			X		X			X	X				X		X			X	
S10	X			X		X			X	X			X		X				X	
S11	X			X		X			X	X			X				X		X	
S12	X			X		X			X	X			X			X		X		
S13	X			X		X			X	X			X			X				X
S14	X			X		X			X	X			X			X			X	

* The capital alphabet in different rate constants categories were described that G for Goodsite et al. (2004), K for Khalizov et al. (2003), D1 for Donohoue et al. (2005), D2 for Donohoue et al. (2006), A for Ariya et al. (2002), B for Balabanov et al. (2005), and P for Pal and Ariya (2004).
* S14 is the case of only considering initial conditions of halogen concentration

9) It is unclear to me why the authors only consider gas-phase reactions – shouldn't they add all known reactions with Hg0 in order to assess likely mechanisms for MDE are in the Arctic environment? Also, I was surprised that sea-salt aerosol uptake and deposition isn't included in the models given that in the marine boundary layer this is the main removal pathway for oxidized Hg?

We mentioned the reason why we did not consider heterogeneous chemistry in this study in line 19 on p.7 – line 7 on p.8 and we added the sentence; "These indicated that the environment in the springtime over the Arctic Ocean was too dry and cold with very little sea salt" in lines 8-9 on p. 8.

10) The final abstract in regards to climate change seems speculative, is not supported by measurements nor by the model runs, and in my view should be deleted.

We disagree. It is thinking based on implications of our results. We think that with the extensive attention being paid to effects of climate change in the Arctic, one should begin to think about its affect on chemical cycling. It is more to stimulate future modeling studies with global models, as the outcome is highly uncertain.