acp-2012-452 (Cole et al.), Response to Anonymous Referee #1

Thank you for your comments and suggestions. Please see below for our responses.

1. It seems that the authors are apt to suggest that anthropogenic and natural emissions in the North America as well as atmospheric transformations played a more important role here. In my opinion, the authors should differentiate the regional effect of locations of the six sites. As we can see from the map, several sites in mainland areas exhibited more pronounced decreasing trend compared to the site in coastal areas. This may indicate the levels of Hg at coastal sites might be mainly regulated by natural sources, which should maintain at the consistent levels during the years. For the Zeppelin site, additional effect may be also originated from emissions of Asian developing countries, the anthropogenic inventory of Hg was not well understood and may have increased significantly recently, which could compensate the effect of decreasing anthropogenic Hg emissions in North America.

- We looked at two mainland sites (Egbert, St. Anicet) that can be compared with the two mid-latitude coastal sites (Kejimkujik, Mace Head). From this limited number of sites we were not able to discern a significant difference in trends and thus it was not discussed. The role of oceanic emissions (in terms of changes in Arctic ice cover) is discussed, but oceanic influence at lower latitudes is something we will monitor in years to come. We touched on the possibility of a greater contribution from Asian source regions to the Arctic sites by indicating that the GRAHM model doesn't support this. Based on your comments, we have expanded on this topic a bit more in the revised paper by including some back-of-the-envelope calculations. The revised section reads: "A source attribution study using Environment Canada's Global/Regional Atmospheric Heavy Metals (GRAHM) model found that Asia contributed about 30-35% of surface GEM at Arctic, sub-Arctic and mid-latitude sites, with relative contributions varying by site and season but no large difference between the Arctic and lower latitudes (Durnford et al., 2010). Even if we assume that Zeppelin Station (designated Ny-Alesund in Durnford et al.) receives 35% of surface-level GEM from Asian sources and mid-latitude sites receive 30%, in order to explain a difference in the trends of 2% yr^{-1} emissions from Asia (natural and anthropogenic) would have to have increased by 26% yr^{-1} and the rest of the global emissions decreased by 14% yr^{-1} for the decade, which is a larger change than can be explained by current inventories. Therefore we conclude that some of the difference in trends may be explained by a difference in source regions, but not the majority."

2. It is better to show the annual mean Hg concentrations at all the sites in a new table.

- Overall mean, SD, and median have been added to a new table 2, with trend results also moved to Table 2.

3. In line 10 on page 2, the time period of the study should be introduced before.
- We have changed the previous sentence to read: "Ten-year records of total gaseous mercury (TGM) from 2000 to 2009 were analyzed…"

4. In the section of method description. Firstly, I have no doubt of the data quality in this study. However, for this ten-year continuous monitoring, do you have some protocols to make the observation data be comparable throughout the 10 years?
- Yes, there have been established protocols for both data collection and post-processing (QC) for the Canadian sites. We have revised the methods to include the following: "For the Canadian sites, consistent TGM sampling protocols were followed during the measurement period (Steffen and Schroeder, 1999) and the data were treated by an objective quality control (QC) process, the Environment Canada-developed Research Data Management and Quality Assurance System (RDMQ) (McMillan et al., 2000). Specifics of the QC criteria for the Canadian Atmospheric Mercury Measurements Network (CAMNet) are discussed in depth elsewhere (Steffen et al., 2012). At Zeppelin, protocols for the Norwegian mercury monitoring program were used, much of which are similar to the Canadian protocols. The data from Zeppelin have been quality controlled by hand, using many of the same QC criteria as the RDMQ system."

5. line 14-30 on page 11, are there some studies with respect to the long-term trend of Ozone or bromine at the polar site? There may be relationship between the oxidants and RGM and PBM productions.
- We are not aware of long-term measurements of bromine species, but yes, ground-level ozone has been measured at Alert since 1992. A seasonal trend analysis of these data showed very similar trends to TGM, particularly in the spring. We have added this result to the indicated paragraph as follows: "Since the springtime oxidation of GEM is linked to high levels of bromine radicals that also catalytically remove ozone from the boundary layer in spring (Simpson et al., 2007), a decline in these radicals should also lead to an increase in spring tropospheric ozone concentrations. Analysis of ozone measured at Alert from 2000 to 2009 (NAPS, 2013) revealed trends in March, April and May that were very similar to the trends in TGM with a decrease in March and increases in April and May, though only the May trend was significant (not shown). This agreement was not unexpected given the tight correlation between TGM and O_3 in the polar spring (Schroeder et al., 1998) and supports the hypothesis of a contribution from bromine chemistry to the observed spring TGM trends."

6. line 15 on page 15, the 'Svalbard' should be changed to Zeppelin station.
- We have revised to "the island of Spitsbergen, where Zeppelin Station is located…"