Response to Referee #3

We greatly appreciate all of the comments, which have improved the paper. Our point-by-point responses are provided below.

Anonymous Referee #3

General comments

The manuscript by Cheng et al. provides an overview of the source apportionment studies for speciated atmospheric mercury using a receptor-based approach. Related methodologies are described, study examples are given, and future research directions are recommended. In general the manuscript is well organized and clearly written, and its subject is relevant to the scope of ACP. Therefore, I recommend that the manuscript be accepted for publication if these following comments are sufficiently addressed.

One general comment is to add a summarizing table for the comparison of different methodologies, including input and output parameters, advantages and disadvantages, etc. Such a table would facilitate the readers' understanding of the similarities and differences among receptor-based methodologies.

Response: We have added in the revised paper a summary table (Table 1) comparing the various receptor-based source apportionment methodologies used to analyze speciated atmospheric Hg data, in terms of the type of model, data required, model parameters, potential Hg sources identified and major advantages and disadvantages.

Specific comments

P5499, L11: A reference for "the PMF model" is needed here.

Response: References for the PMF model have been inserted: "The PMF model (Paatero and Tapper, 1994; USEPA 2014b) is accessible from the USEPA website." (1st paragraph of 2.1.2)

Paatero, P. and Tapper, U.: Positive Matrix Factorization: a Non-Negative Factor Model with Optimal Utilization of Error Estimates of Data Values, *Enviromet*rics, *5,* 111-126, 1994.
USEPA: EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, 2014b. http://www.epa.gov/heasd/research/pmf.html

P5500, L4: An explanation for "Delta-C" is needed when it is mentioned for the first time in the manuscript.

Response: An explanation for Delta-C has been added in the revised paper as follows, "For atmospheric Hg source apportionment, the input variables have included speciated atmospheric Hg (GEM, GOM, PBM) and trace gases (CO, NO_x, O_3, SO_2), trace metals, $PM_{2.5}$, particle number concentrations, and/or carbon (black carbon, Delta-C) measured at the receptor site (Liu et al., 2003; Cheng et al., 2009; Wang et al., 2013). Delta-C is the difference in black carbon measured at two wavelengths, 370 nm and 880 nm, which is indicative of wood combustion (Wang et al., 2013)." (2nd paragraph of 2.1.2)

P5502, L6-7: I think the uncertainties of GOM and PBM concentration measurements are not only 40% and 70%. Gustin et al. (2013) suggested that GOM and PBM concentrations "could be 2-to-3 fold higher than that reported in the literature".

Response: Gustin et al. (2013) reported "the precision between collocated instruments is 0.4 to 20%, 15 to 40%, and up to 70% for GEM, GOM, and PBM, respectively (Ebinghaus et al., 1999; Aspmo et al., 2005; Lyman et al., 2007; Brown et al., 2008; Peterson et al., 2009; Steffen et al., 2012)." These uncertainties relate to comparisons between Tekran instruments. The study also concludes that "Collectively, the data showed that RM concentrations could be 2-to-3-fold higher than that reported in the literature." This is based on a comparison of the oxidized Hg data between other mercury instruments and the Tekran instruments.

In the revised paper, we have replaced the measurement uncertainty estimates with those reported from a more recent study by Gustin et al. (2015), who suggested GOM concentrations are underestimated by a factor of 1.6 to 12 depending on the chemical composition of GOM based on comparison of data between various mercury instruments. The extent of the GOM measurement uncertainties have not been widely accepted by the scientific community according to online peer-review discussions for this study (http://www.atmos-chem-phys-discuss.net/15/3777/2015/acpd-15-3777-2015-discussion.html); however, research on this important issue is progressing. For PBM measurements, it is still unclear whether it is underestimated or overestimated and how large the uncertainties are (Gustin et al., 2015). Nevertheless, the uncertainties for GOM and PBM are large compared to GEM because the exact chemical composition is unknown and thus calibration standards have not been developed to determine the accuracy of the measurements. Our main point is that the PMF model data quality screening features would be useful for uncertain parameters, like GOM and PBM. (last paragraph of 2.1.2)

P5507, L12: A full name for "the FLEXPART model" and a related reference are needed here.

Response: The sentence has been revised as follows, "The FLEXPART-WRF (FLEXible PARTicle-Weather Research and Forecasting) model simulates the transport and dispersion of air pollutants (Stohl et al., 2005; Fast and Easter, 2006). In CFA studies for speciated atmospheric Hg, FLEXPART-WRF simulated the path of 100–1000 particles released from the receptor location (Rutter et al., 2009; de Foy et al., 2012)." (2nd paragraph of 2.2.3)

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, doi:10.5194/acp-5-2461-2005, 2005.
Fast, J. D. and Easter, R.: A Lagrangian Particle Dispersion Model Compatible with WRF, in: 7th WRF User's Workshop, Boulder, CO, USA, 2006.

P5517, L12-14: What chemical species is(are) the marker(s) of sewage treatment?

Response: Pollutant emission ratios (e.g. NO_2/Hg, $PM_{2.5}$/Hg, and SO_2/Hg) were calculated for sewage treatment plants, cement production, chemical manufacturing, fossil fuel power generation, and metal/steel production sources using emissions data in Cheng et al. (2009). The study found the pollutant emission ratios from sewage treatment plants were at least 10 times smaller than other Hg point sources. The pollutant ratios for one of the PMF model factors were found to be similar to those of sewage treatment and were therefore assigned to this source. We added in the revised paper that "If

trace metals or aerosol chemical composition data were available at this receptor location, Zn, Pb, Cu, Cl, V, and Ni can be used as chemical species markers for municipal waste disposal/incineration (Graney et al., 2004; Keeler et al., 2006; Watson et al., 2008)." (1st paragraph of 3.2)

P5518, L16-24: The authors may consider moving these several sentences about the goodness and evaluation of the PMF methodology to Section 2.

Response: The sentences related to how the goodness of fit of the PMF model was assessed have been moved to the second paragraph of 2.1.2. We kept the other sentences in the last paragraph of section 3.2 to emphasize the lack of evaluation of the PMF results for speciated atmospheric mercury.

P5526, L18-26: "PSCF and GFD are also more likely to report high probability source areas near the receptor location because ..." It is not clear how this reason is different from the two uncertainty sources mentioned above: "the trailing effect and high distribution of trajectory endpoints near the receptor region".

Response: The revised paper has clarified these sentences (section 4, back trajectory receptor models, 2nd summary point). The increasing number of trajectory endpoints (also the same as the higher distribution of trajectory segment endpoints) near the receptor location in PSCF and GFD models is different from the trailing effect issue, which may identify false source areas downwind or upwind of actual sources. This is because an equal weight is applied to all trajectory segments along a trajectory, while actual sources are often concentrated in specific areas (Stohl, 2006). Thus, these models perform better at identifying the direction of source areas than the distance of the source areas to the receptor location (Rutter et al., 2009; de Foy et al., 2012).

The increasing number of trajectory endpoints approaching the receptor location leads to the potential false identification of sources near the receptor location since a longer residence time indicates a greater likelihood of contributing to the receptor site. The model results need to be verified with emissions inventory sources. For PSCF, the higher residence time for grid cells near the receptor location compared to grid cells further away from the receptor location affects the PSCF calculation in terms of a larger denominator value in Eq. 4.