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Interactive comment on "**Potential sources and processes affecting speciated atmospheric mercury at Kejimkujik National Park, Canada**" *by* **Xiaohong Xu et al.**

Anonymous Referee #1

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In this work, Authors aimed to identify the factors affecting ambient Hg concentrations (in the form of gaseous-GEM, oxidised-GOM and particulate-bonded-PBM) at a receptor site using PMF and PCA approaches. They also tried to summarize the similarity and differences in PMF factors and PCA components, to evaluate the PMF model performances for Hg forms, investigate the impact of meteorological parameters on PCA results, assess the sensitivity of PMF results and performance to different treatment of missing data and low concentration values of speciated Hg.

Anyway, despite the attention paid in missing/<MDL data treatments, lacks are present in describing how the PMF model was performed. In this case, a rigorous categorization of the variables is important along with the definition of the Total Variable (which

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allows estimating the contribution of a chemical species to a reference variable, PM for example). In addition, a preliminary analysis of some parameters like Q, IM and IS versus the number of factors, allows the user to obtain useful information on the correct solution for PMF. All these information are not reported in the work. Finally, the authors performed an analysis of the performance of the PMF model, considering the observed/predicted forms of Hg. In this case, PMF outputs clearly shown that the model was not able to well reconstruct the variables GEM, GOM and PBM. Then in my opinion, further elaborations need in order to improve the solution and the relative stability. In conclusion, I believe that the manuscript could not be considered matured for publication in its present form considering the reasons indicated above. It could be improved with further PMF analysis. In addition, even if in this work a different data treatments have been done, the datasets analysed have already been published and this limits the novelty of the results (Chen et al., 2013). I also give some specific comments that could be useful for eventual re-writing of the paper.

Specific comments Page 2, line 64: Author could cite some example regarding works that assessed model performances of RM, such as Cesari et al., 2016 Environ Sci Pollut Res 23:15133–15148 and Belis et al. 2015 Atm Environ 123:240-250.

Page 3, line 88. A map could be useful in order to understand the sampling site position together with the sources, listed in the paper, affecting that area.

Page 4, line 111. Authors should indicate the amount of aerosol mass characterized.

Page 6, line 130. Authors should better indicate in the text the dimensions of the datasets analysed and if these dimensions respect the conditions requested in order to obtain statistically stable SA analysis. From literature, we have that these conditions are: the minimum required number N of samples N>30+0.5*(V+3) where V is the number of species considered (Henry et al., 1984 Atmos Environ18:1507-1515), and the more restrictive condition N>50+V (Thurston and Spengler, 1985 Atmos Environ19:9-25.).

Page 6, lines 176-180. Authors wrote that one of the objectives of this work is to identify the factors affecting ambient Hg concentrations using PMF model. In this sense, authors should explicit what chemical specie they used as Total Variable in PMF analysis: PM or Hg (GEM or GOM or PBM)?

Page 6, line 185: Please, give some examples of stability indexes for model runs.

Page 7, line 194. Please, indicate the dimensions of the analysed dataset and if these datasets are the same considered for PMF analysis.

Page 7, line 209. Authors should explicate why the 4-factors solution is the best solution. I am wondering if they have analysed the trend of some parameters (such as dQ, IM and IS, see Lee et al, 1999 Atmos Environ 33: 3201-3212; Viana et al., 2008 Atmos Environ 42:3820-3832; Brown et al., 2015, Sci Total Environ 518-519: 626-635) with the number of factors, from 4 to 8 for example, in order to obtain some "objective" information about the best solution. If not, please consider to perform this analysis in order to justify the choice of 4-factors solution.

Page 9, line 267. Again, what species, PM or Hg-form, has been considered as Total Variable? If the T.V. is the PM, how is possible to obtain a factor contribution for 2010?

Page 10, lines 302-307. The coefficients of determination together with Figures S1-S2 show that the model in this case is not able to reconstruct the Hg – concentrations. The reason could be different, depending on, for example, the reduced number of samples (a solution could be to merge the two datasets), or the high percentage of missing values/data lower than MDL. In my opinion, Authors should check the categorization of the variables, performed considering both the S/N value and the percentage of missing values or lower than MDL: for example, in my opinion GOM could be considered as a BAD variable. Again, the choice of a different number of factors could help in obtaining a better reconstruction. Authors should perform other runs with the aim to improve the output of the model. The same observations are for the dataset Case 2010.

Page 14, line 440. Referring to PC3 in table 8, how can you explain the opposite load values of GOM and Temperature? Photochemical production of GOM happens with high temperature and solar radiation, so I would imagine this variable having the same sign.