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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An assessment of atmospheric mercury in the Community Multiscale Air Quality (CMAQ) model" *by* T. Holloway et al.

Anonymous Referee #2

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General comments:

In this manuscript, author evaluated the performance of CMAQ-Hg model by using observed mercury concentration and wet deposition data obtained at two sites (DL and MKE) in the Great Lake Region. CMAQ-Hg is capable of capturing the monthly mean and seasonal variations of wet deposition. The model is less capable of predicting the mean of measured GEM concentrations at the remote site (DL) while underestimating the mean at the urban site (MKE). Also, CMAQ-Hg overestimates the observed reactive gaseous mercury (RGHg) and particulate mercury (PHg), with a mean bias of greater than 250%.

One of the highlights of this paper is that the authors used divalent mercury concentration as model evaluation criterion for model performance evaluation. Mercury wet

deposition or total mercury concentration are usually used as the primary factors in earlier studies (Bash 2010, JGR; Bullock et al., 2008, 2009, JGR; Lin et al., 2007, AE; Pongprueksa et al., 2008, AE; Sunderland et al., 2008, EP). The author also point out the potential deficiency in models. It is crucial for authors to cautiously fully address the uncertainties in model results and measurements.

1. Uncertainties in CMAQ model inputs:

Page 2135 line 6 "using the default boundary conditions (Table A1) for CONUS simulations", it may be a appropriate to use fixed boundary conditions to drive CMAQ-Hg and to compare with observation in the time series plots (Figure 2a and 3a) as we know how importance of model boundary conditions dominate simulated Hg0. The authors acknowledge the error of using fixed boundary conditions (page 2147 line 13-15). I don't see any reason that fixed boundary conditions should be used in this study.

Page 2136 line 20-24: It seems that mercury emissions from nature sources or reemission are not included in the inventory, but previous researches have indicated its importance to model simulated results (Lin et al., 2012, AWMA; Gbor et al., 2006, 2007, AE);

2. Uncertainties in measurements

Page 2138 line 2-4: "As described in Manolopoulos et al. (2007), samples were taken every two hours using a Tekran ambient mercury analyzer .....", Lyman et al., in his recently published paper of "release of mercury halides from KCl denuders in the presence of ozone" (ACP 2010) mentioned that the standard method for measurement of gaseous oxidized mercury may bring 29-55% lost of total RGHg compounds and "this method may not be as robust as previously thought". Therefore authors couldn't avoid addressing this issue in the section 2.4 and should carefully analyze its influences in the discussion.

3. Uncertainties in data comparisons

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Page 2134 line 26-27: “Thus, model simulations were limited to 2003 for comparison with both sites” - using model results in 2003 to compare with measured data in 2004 is not well justified although the authors claim that meteorology is not a dominant effecting factor (Page 2134 line 25-26). This assumption is questionable because mercury wet deposition strongly depends on precipitation rate and it could vary year by year. The authors discussed the influences of wind direction on observed mercury concentrations at DL (figure 4 and Page 2145 line 9-16). Why didn’t the authors run CMAQ-Hg for 2004, which will eliminate the uncertainties caused by time mismatch between model results and observations?

Page 2138 line 6-9: “The MKE site is . . . . . as the ambient concentration sampler”, for MKE site, mercury concentration and wet deposition were measured at different locations. The question is whether or not it will cause uncertainty when comparing the observations made at two locations with model results that were extracted at one location.

Page 2141 line 1-4: “Simulated wet deposition . . . . . where Fig. 1 shows the locations of the GLR measurement sites included in these calculations”, does it mean mercury wet deposition data used in comparison were derived from all the MDN sites covered by GLR domain and were not just limited to DL and MKE?

Page 2141-2142 section 3.2 “wet deposition”, earlier studies have suggested precipitation bias in CMAQ should be handled carefully before comparing wet deposition between model and observation. It would be more appropriated if author also consider this issue and address the influence. The authors investigated the model components such as emission, chemistry and removal processes, etc., and tried to interpret the discrepancies between model results and observations. Such efforts are useful in future model development. However, some conclusions made by the authors are not supported by the data. For examples:

Page 2144 line 14-17: “This pattern might be explained by a compensating error in

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the wet deposition rate . . ... too-high dry deposition is explored below.", true only if the authors can justify precipitation bias and data mismatch between model and measurement.

Page 2145 line 19-23: "This evidence points to a modeled RGHg lifetime in CMAQ-Hg that is too short . . .. . . we hypothesize that dry deposition rates are unrealistically high in the model", one alternative explanation is the uncertainty of mercury emission inventory. Generally speaking, both wet and dry deposition are equally important to RGHg concentration in CMAQ-Hg model.

Other comments

Page 2137 line 11-14: "For DL . . .. . . the same model grid cell (WI site #26)", mercury and other air pollutant observations are not made at the same site. I don't know how those data can help in the explanations of mercury observations.

Page 2138-2140 section 3.1 "Comparison with ozone, nitrogen dioxide, and sulfur dioxide", why did the authors spend three pages to discuss model predictions in those species? If they are important to mercury, more discussions regarding their relationships with mercury should be provided, not just using a few sentences in Page 2140 line 19-29.

Page 2147 line 3-6 : "At DL . . .. . .suggesting that boundary inflow alone contributes the majority of simulated mercury at the rural site . . .. . . 99% of the BC-simulated Hg0 is captured by the ZE scenario, and 91% of BC-simulated RHg is captured", the claims somewhat contradict the findings by Manolopoulos et al., 2007. In his paper, they concluded that "Plumes reaching the Devil's Lake site from a nearby coal-fired power plant significantly impacted SO2 and RGM concentrations at Devil's Lake, but had little impact on Hg0".

In summary, this paper deserves to be published if the authors can carefully revise the discussion.

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