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Interactive comment on “Modeling natural emissions in the Community Multiscale Air Quality (CMAQ) model – Part 1: Building an emissions data base” by S. N. Smith and S. F. Mueller

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We greatly appreciate your review and comments. Here we have provided our replies along with your comments/questions.

SC C1154 COMMENT: p. 1770-1771: Section 3.4 describes gaseous HCl and ClNO₂ and sea-salt aerosol emissions from oceans. Knipping and Dabdub (2003) suggested that interaction of sea-salt aerosols with gaseous species can also release molecular chlorine (Cl₂) emissions from coastal areas. If authors think that Cl₂ can also be emitted from oceans, then such emissions can be estimated and included in the database. AUTHOR REPLY: Chlorine in the form of HCl and ClNO₂ were more readily available

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for our modeling purposes than were factors for direct Cl₂; therefore, due to project/time constraints we only included this pathway of chlorine. Perhaps Cl₂ could be included in future work.

SC C1154 COMMENT: Chlorine emissions from oceans are estimated; however, the article does not include any discussion on possible chlorine emissions from the Great Salt Lake. If authors think that chlorine can be emitted from the Great Salt Lake, then such emissions can be estimated and included in the database. If authors think that chlorine emissions are not released from the Great Salt Lake, an explanation can be included in the article. AUTHOR REPLY: Efforts were made to acquire suitable emission factors/flux for chlorine from the Great Salt Lake (GSL); however, none of our inquiries on the subject returned plausible factors within the time constraints of this project; therefore, natural chlorine emissions from the GSL will simply have to be pursued in future work.

SC C1154 COMMENT: Annual total HCl and ClNO₂ emissions from oceans were taken from the GEIA Reactive Chlorine Emissions Inventory. It would benefit readers to explain how these emissions were spatially allocated (spatial distribution) into grid-cells for the CMAQ model. Similarly, the procedure used for spatial allocation of reduced sulfur from oceans can also be described. AUTHOR REPLY: A basic explanation of the process is provided here; however, we feel that to provide further details on our geoprocessing methods is outside the scope of this document. We could provide further details upon request and as schedules permit. The GEIA RCEI provided annual totals for the GEIA 1° x 1° domain that were then assigned to our 36 km resolution modeling domain grid cells using spatial area intersection/overlap geoprocessing tools in ArcGIS. Regarding reduced sulfur, DMS; DMS from the ocean area within our 36 km resolution modeling domain was taken from the annual mean flux of Sulfur (S) from the oceans as delineated in Figure 5 of Kloster et al. (2006). The global spatial distribution graphic/raster of this annual flux of S was georeferenced and digitized into ArcGIS shapefile formatted polygons for each flux class. Then these flux class polygons were

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allocated via spatial intersections/overlap with ocean-containing-36km grid cells in ArcGIS. Essentially, if multiple flux values were assigned to a given 36 km grid cell, the total flux was determined by the percent area of the 36km grid cell each flux polygon consumed. An aggregate of these flux values per cell was then calculated based on this area consumed percentage.

SC C1154 COMMENT: p. 1779: “There is a need to update existing aerosol modules in models like CMAQ to more accurately simulate the dechlorination of sea salt particles and the subsequent release of reactive chlorine to the atmosphere. Changes are also required in CMAQ so that its chemistry modules can simulate oxidation reactions involving reactive chlorine”. The chemistry module in the released version of CMAQ_v47 already includes oxidation reactions involving reactive chlorine. AUTHOR REPLY: We used version 4.6 because that was the most current version available when the project started.

SC C1154 COMMENT: Figure 10. Relative proportions of anthropogenic and natural emissions are compared for July. It would be informative to include such a comparison for January also. It is not clear about the anthropogenic chlorine emissions used for the comparison for July. The 2002 NEI includes emissions of HCl and Cl₂; total HCl emissions 563,568 tons and Cl₂ emissions = 12, 568 tons, (Table – 4.4 in ftp.epa.gov/EmisInventory/2002v3CAPHAP/documentation/volumeiii_hap_main_07may2008.pdf). Anthropogenic chlorine emissions should include these emissions. AUTHOR REPLY: We have provided a plot similar to Fig 10 for January in the supplemental pdf file included in our response to reviewer comment RC C970 (See Fig. A. in [RC_C970_SupplementalAC.pdf](#)). Regarding anthropogenic chlorine used in the comparison for July, this refers to the anthropogenic HCl and ClNO₂ emissions we discuss in Section A3 of Appendix A. Our work derives from and expands on the VISTAS RPO 2002 emissions inventory modeling platform to include additional “natural” sources.

SC C1154 COMMENT: Figure A7. Temporal profile shows ClNO₂ is zero during most of the nighttime. ClNO₂ is released from the interaction of N₂O₅ and sea-salt aerosols

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both of which can be present at night. Thus, ClNO₂ can be present during nighttime. It will benefit readers to explain reasons for using zero ClNO₂ emissions at night. Recent study of Thornton et al. (A large atomic chlorine source inferred from mid-continental reactive nitrogen chemistry - vol 464, 11 March 2010, doi:10.1038/nature08905) also suggests that ClNO₂ can be emitted at night (Figure 3). AUTHOR REPLY: You are absolutely correct and I am glad you noticed this; I'm afraid I somehow misinterpreted the Osthoff et al. figure we cite by overlooking the timescale assuming a full day while it was only a half day. Fortunately the peak flux we retain in this interpretation is in agreement with Thornton et al. and fortunately this only impacts diel variation in the emissions and not diel totals; however, there absolutely should be some allocation of these ClNO₂ emissions into the night hours. I simply will have to note this as a caveat in the final revision of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 1755, 2010.

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