PREPARATION OF VEGETATION MERCURY EMISSION USING BEIS3 - PROTOTYPE DEVELOPMENT AND PRELIMINARY PROCESSING

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1. INTRODUCTION

Emission inventory is one of the required input fields for the comprehensive chemical-transport modeling of atmospheric mercury. Accurate emission inventory is critical to reduce model uncertainties[1]. However, mercury emission from vegetation (natural) sources has not been treated rigorously in previous modeling efforts of atmospheric mercury [2-3].

It is commonly accepted in mercury research community that mercury emission from vegetation is caused by the deposition from earlier anthropogenic emission followed by the transpiration of vegetation. Several studies have indicated that the vegetation emission of mercury may dominate that from anthropogenic sources, and there is a need to re-assess the this diffuse emission contribution[4]. More recently, it is reported that the mercury emission from vegetation exhibits a strong diurnal variation[5]. In the atmosphere, this emission source may play an important role in the concentration and deposition of mercury due to its significant mercury input at ambient level, especially in summer. However, there is not a modeling tool for mercury emission from vegetation to address the modeling need and emission estimates.

The objective of this study is to develop a prototype processor to estimate and process mercury emission inventory from vegetation within the framework of Biogenic Emission Inventory System Version 3 (BEIS3)[6]. BEIS3 is a flexible modeling system for estimating the emission inventory of volatile organic compounds (VOCs) and NOx from biogenic source for comprehensive air quality models that incorporate the simulations of transport, reactions and deposition for various atmospheric pollutants.

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2. METHODS

The version of BEIS3 used in this development is V3.11, a stand-along research version on UNIX/Linux platform. The 1-km-resolution, 230-category USGS Biogenic Emissions Landcover Data Version 3 (BELD3) were utilized to generate the normalized vegetation-specific mercury emission in a 36-km Lambert Conformal grid covering the entire continental United States. The surface temperature and cloud-cover corrected solar radiation from a mesoscale meteorological model (MM5) were retrieved and converted into model-ready format using a Meteorology-Chemistry Interface Processor (MCIP2). The converted data were then used for temperature and irradiation corrections to calculate the diurnal variation of vegetation Hg emission for each landuse category, i.e.,

$$\text{Emission Flux (ng h}^{-2}\text{ m}^{-2}\text{)} = \text{Standard Emission (ng h}^{-2}\text{ m}^{-2}\text{)} \times C_T \times C_L$$

where C_T is the temperature correction factor and C_L is the irradiation correction factor. In the calculation of C_T and C_L, formulation similar to Guenther Algorithm[7] for biogenic VOC emission was assumed since a the emission-temperature-irradiation relationship is not yet available for mercury: The constants in the algorithm were evaluated from regression using the Hg flux measurements made by Lindberg *et al*.[5]

A mercury emission factor table was created to represent the standard emission intensity. Table 1 shows the implemented standard Hg emission in the model, which represent the average daytime emission of gaseous elemental mercury. The output from the model is temporally (hourly) and spatially resolved gridded emission in netCDF format ready for applications in Eulerian-based chemical transport models including CMAQ-Hg and CAMx. Figure 1 shows the data flow of the

model processing. The source codes of BEIS3 were modified to include gaseous elemental mercury (Hg^0) as one of the emitted species. To compare emission quantity of the vegetation mercury emission to the anthropogenic mercury emission, annual simulations were performed using 2001 meteorological data in the 36-km CONUS domain, and the month-by-month Hg emission from the two source categories were evaluated and compared.

Table 1. The standard emission used in the calculation of vegetation Hg emission

Tree Species and/or Land Cover	Water	Wetland	Maple	Oak	Pine	Others(1)
Summer (ng hr^{-1} m^2)	1.0	40	4.0	5.3	4.0	4.0
Winter(2) (ng hr^{-1} m^2)	0	20	0	0	4.0	Model(2)

(1) These include all other forest species and agricultural land use.
(2) Based on the winter/summer emission ratio for VOC in BEIS3.

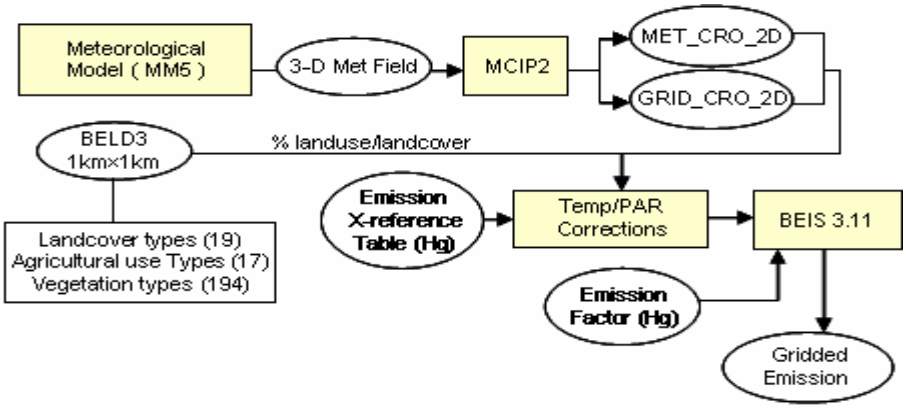

Figure 1. Data flow for developing biogenic mercury emission processor

3. RESULTS AND DISCUSSIONS

Figure 2 shows the diurnal cycle of elemental mercury emission from Florida wetland by flux measurement and model estimate. The model fluxes were calculated from the standard Hg emission flux, and the irradiation and ground-level temperature at the flux measurement site. As seen, the implemented algorithm depicts the diurnal cycle of Hg^0 emission reasonably well. Similar implementation was also carried out for the other tree species and land cover in the modeling domain.

Figure 3 shows the typical monthly sum of vegetation Hg^0 emission in summer (July) and in winter (January). In Figure 3, the modeled hourly flux was summed for the entire month in each model grid cell. In the summer time, the greatest emission occurs in the Florida wetland due to the high standard emission (40 ng hr^{-1} m^{-2}), temperature and irradiation. The southern central states also contribute significant vegetation Hg^0 emission due to the relatively high temperature and irradiation in the region. In the winter time, the Hg^0 emission greatly decreases due to the lower (zero for many species) standard emission implemented in the model and much lower irradiation and temperature during the winter season. As a result, significant emission occurs only in the deep south of the domain, and the emission quantity is much smaller than that in summer.

The Hg^0 emission quantity from vegetation is also compared to that of anthropogenic emission. The anthropogenic mercury emission is based on the USEPA NEI99 Final Version 3 and the temporal allocation is based on the standard temporal profiles published in USEPA CHIEF (Clearinghouse for Inventories & Emission Factors) website (http://www.epa.gov/ttn/chief/). The primary emission characteristics differences between the vegetation and anthropogenic Hg

emission are (1) the vegetation emission has strong diurnal and seasonal variations while anthropogenic emission doe not, (2) the vegetation emission constitutes only elemental mercury (Hg^0) while the anthropogenic emission releases Hg^0, reactive gaseous mercury (HgII) and particulate mercury (PHg), and (3) the vegetation emission releases mercury only in the surface layer of the domain while anthropogenic emission of mercury from point source is subject to the plume rise of the emission units.

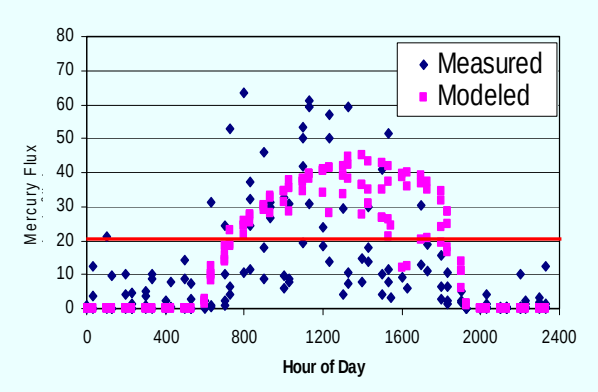

Figure 2. Diurnal cycle of Hg^0 emission

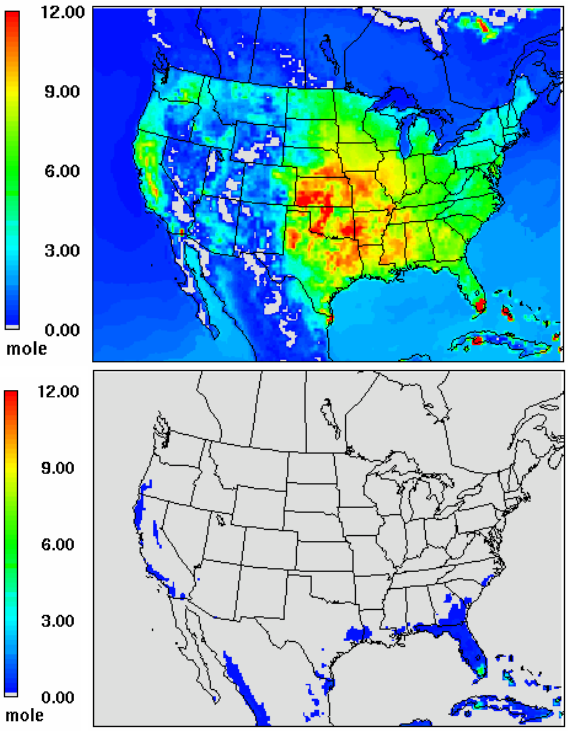

Figure 3. Typical summer (top) and winter (bottom) sum of Hg^0 emission from vegetation

Figure 4 shows the month-by-month mercury emission quantity by vegetation and anthropogenic sources. These results were generated by annual emission processing of both vegetation and anthropogenic Hg emission using 2001 meteorological data in BEIS3 and SMOKE Version 2.0. In Figure 4, the strong seasonal variation of vegetation Hg emission can be seen. The anthropogenic mercury emission does not show much seasonal variation. It can also be clearly seen that mercury emission quantity from vegetation and anthropogenic sources are comparable in summer when the temperature and irradiation are relatively high. For example, in the month of July, vegetation emission constitutes 47 % of total mercury emission into the atmosphere. However, in the winter time, anthropogenic source dominates the Hg emission due to the much lower standard emission of the vegetation, surface temperature and irradiation. Summing up the total emission for the entire modeling domain, vegetation emits 44.1 tons of mercury while anthropogenic (point and area) sources emit 143 tons of mercury.

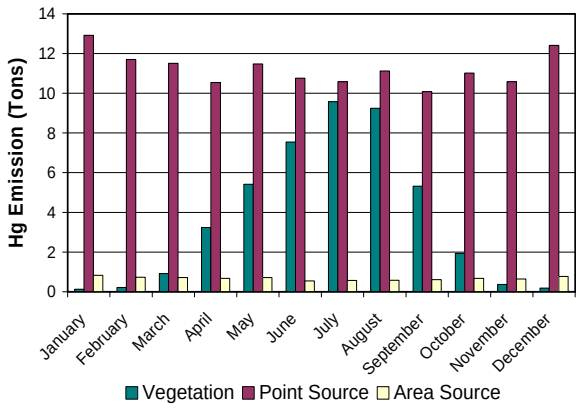

Figure 4. Comparison of vegetation Hg emission to anthropogenic Hg emission

A prototype vegetation mercury emission processor in BEIS3 framework has been developed. The developed processor can be used for estimating vegetation mercury emission at high temporal and spatial resolutions along with biogenic VOC and NOx to support the chemical transport modeling of atmospheric mercury. In this development, the standard emission factors were implemented for a limited number of vegetation species only. New flux data can be easily implemented in the model once data become available.

4. ACKNOWLEDGEMENT

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