

Methylmercury Risk Assessment for National Forest Lands

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Background

The Air Program of the Forest Service works to understand how air pollutants are affecting resources on the national forests. This information is then shared with the air regulatory community through various avenues to help reduce emissions and impacts to natural resources. To support these efforts, the Air Program is interested in identifying the areas of national forest land at greatest risk to the negative impacts of mercury and to the extent possible understand more about the probable sources of pollution. A national-scale risk assessment for potential impacts from mercury will assist the Air Program in decisions about mercury deposition monitoring as well as other mercury-related studies. Information now exists to conduct this assessment with the use of Geographic Information Systems.

It is well established that methyl-mercury (MeHg) contamination within aquatic systems is a serious environmental health problem, resulting in physical and neurological damage to humans from the consumption of fish. Most states have established fish consumption advisories related to methyl-mercury contamination. Sulfur enhances mercury methylation and therefore increases potential impacts to the environment. The majority of sulfur and mercury input to aquatic systems is from atmospheric deposition, and the majority of that input originates from the burning of fossil fuels. Recent research results show that higher concentrations of total mercury in fish are found in watersheds with gold or mercury mining, forested wetlands and evergreen forests (Scudder et al. 2009.). The current project was conceived to combine these factors and develop a national risk assessment for mercury deposition on national forest lands.

Objective

The original plan for this project was to conduct a mercury deposition risk assessment for the National Forest System lands in the lower 48 states using information from the Mercury Deposition Network (<http://nadp.sws.uiuc.edu/mdn/>), EPA's Atmospheric Mercury Deposition Project for Watershed Planning¹ (<http://www.epa.gov/owow/tmdl/techsupp.html>), sulfur deposition monitoring, and newly published information on land use/land cover influences on ecosystem sensitivity to mercury. However further investigation caused us to redefine the focus of the project to the risk of methylmercury forming within the watershed.

Recent USGS research summarized in Lubick (2009) has shown that mercury deposition, primarily atmospheric deposition from industrial emissions, is just one influence on the levels of bioavailable methylmercury. "Variations in ecosystem properties that govern methylmercury production in an ecosystem are probably much more important [in determining which ecosystems have high methylmercury in fish] than the variation of mercury deposition across the country," said Mark E.

¹ EPA's Mercury Deposition Project "contains the results of a fine, 12-km grid cell modeling analysis for the lower 48 States including roughly 300 of the top mercury emitters in the US that were "tagged" in order to allow source attribution analysis for individual areas of concern, such as waterbodies, watersheds, or catchments". In order to provide States, Tribes, and Regions with the ability to derive deposition analyses for specific watersheds or other areas of interest, a companion Geographic Information System (GIS) tool was developed by ESRI. Named the *AggreGATOR*, this tool, which will be available to ESRI licensees at no cost, allows a user to overlay the 12km gridded deposition modeling output onto any polygon, e.g., the counties that border a waterbody of interest, and obtain mercury deposition loading information for that polygon along with attribution graphics derived from the tagged sources. EPA conducted this analysis for the FS and provided the information in a geodatabase.

Brigham (from Lubick 2009). Wetland abundance within a watershed, carbon levels within soils, dissolved organic carbon in streams, suspended sediment concentrations in streams and streamflow have been shown to be key factors in determining the levels of methylmercury in waters, and mercury concentrations in aquatic fauna, within a given watershed (Brigham et al. 2009; Chasar et al. 2009; Marvin-DiPasquale et al. 2009).

While these recent studies focused on mercury and methylmercury in aquatic ecosystems, past studies have looked at soils as a sink for atmospherically deposited mercury (Grigal *et al.* 2000, Kolka *et al.* 2001). These studies linked mercury retention in soils with organic carbon content and climatic factors which can influence microbial activity. Mercury concentrations found in soils of the central United States did not altogether match the patterns of deposition (Perry et al. 2009). This highlights the importance of watershed characteristics and soil properties on determining the levels of mercury and methylmercury even when there is good mercury deposition data available.

Once we shifted to the objective of the study to focus on predicting areas with the highest chance or risk of having bioavailable methylmercury, we found that we needed to modify our plan for the GIS analysis. Some additional key points that went into selection of the parameters used in the analysis are captured in the following paragraphs.

The percent coverage of wetlands in a watershed continues to be a primary driver in the risk assessment based on the publication by Scudder et al., 2009. The findings reported by USGS and summarized in the following list, from the National Water Quality Assessment (NAWQA) and Toxic Substances Hydrology Programs in April 2009 also supports the use of percent wetlands in the analysis:

- MeHg is formed in wetlands and other seasonally inundated areas in watersheds and then transported to streams by runoff.
- Watershed properties, particularly wetlands which contribute elevated dissolved organic carbon, drive variations in methylmercury concentrations in surface waters.
- The most important influence on mercury levels in fish is the amount of MeHg available for uptake into the food web.
- Highest levels of mercury in game fish occur in forested watersheds with high densities of wetlands and DOC, such as coastal plain streams of the eastern and SE US.
- Methylmercury contamination in aquatic ecosystems can be predicted using watershed characteristics (wetland density, and surface water DOC, pH and sulfate).
- Recently deposited mercury is more efficiently converted to methylmercury and bioaccumulated in aquatic environments than historically deposited mercury.

Stream density and percent soil organic matter were added as risk factors based on recent publication of papers addressing mercury mobilization and the role of hydrologic flow paths, and the role of soil organic carbon in cycling of mercury.

- Demers et al. 2010. Mercury mobilization and episodic stream acidification during snowmelt: Role of hydrologic flow paths, source areas, and supply of dissolved organic carbon. *Water Resources Research*, Vol. 46, W01511, doi:10.1029/2008WR007021.
- Schelker et al. 2011. Hydrological mobilization of mercury and dissolved organic carbon in a snow-dominated, forested watershed: Conceptualization and modeling. *Journal of Geophysical Research*, Vol. 116, G01002, doi: 10.1029/2010JG01330.

Sulfur deposition was included as a risk factor because it is known that mercury in the soil can be methylated by sulfate reducing bacteria under anaerobic conditions.

Mercury deposition was excluded as a risk factor primarily because we learned that the spatial variation in MeHg typically exceeds the variation in atmospheric Hg deposition (per discussion with Dave

Krabbenhoft (USGS) who has done extensive work with methylmercury and developed a risk assessment for the National Park Service based on surface water chemistry).

Methods

Two groups of regions were analyzed: 1) features for the 6th-level (12 digit) HUCs that contain US Forest Service lands within the continental United States, and 2) features for the wilderness areas that contain US Forest Service lands within the continental United States. A geodatabase was produced that contains the following information by either 12 digit HUC (hydrologic unit code) or wilderness area: percent land cover in wetlands, average percent organic carbon in soil, stream density, and total modeled sulfur deposition. All of these cover national forests in the lower 48 states. The geodatabase also includes interpolated wet sulfate deposition for the eastern states only.

For each parameter, or “layer”, the regions were classified by quartiles; the upper quartile (4th) representing the highest risk, the middle two quartiles (2nd and 3rd) representing moderate risk and the lower quartile (1st) the least risk. For example, watersheds that contain the highest percentage of wetland land cover (the 4th quartile) fall into the high “risk” category. The parameters were then analyzed in various combinations and mapped to show three levels of risk.

A summary of the analyses can be found in Appendix 1.

Results

Figures 1-5 show the classification of 12 digit HUCs into quartiles by risk factor: stream density, organic carbon in soil, percent wetlands, wet sulfate deposition (east only), and sulfur deposition.

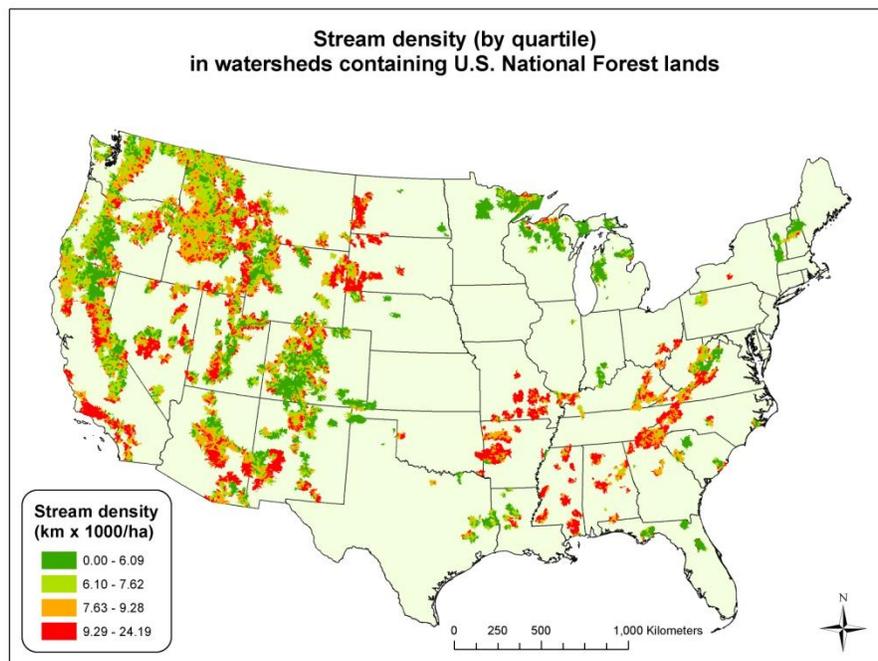


Figure 1. Stream density (km x 1000/ha) in the HUC6 watersheds, symbolized by quartile.

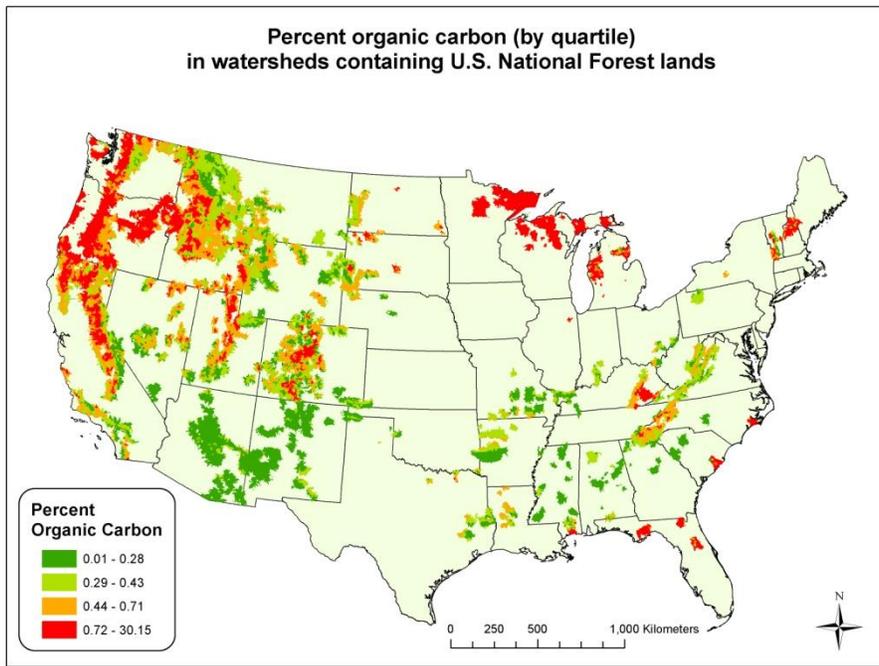


Figure 2. Percent organic carbon in the HUC6 watersheds, symbolized by quartile.

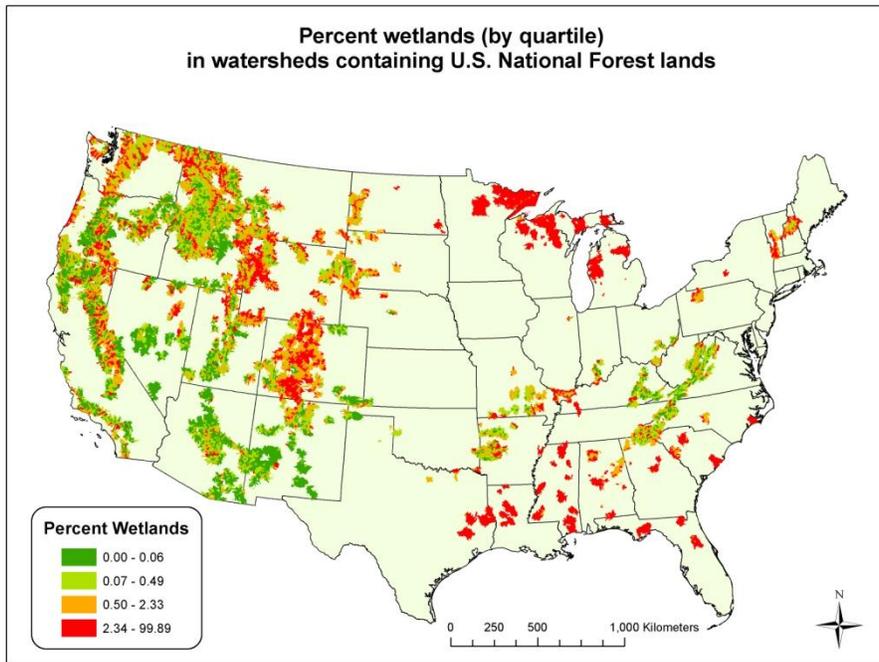


Figure 3. Percent wetlands in the HUC6 watersheds, symbolized by quartile.

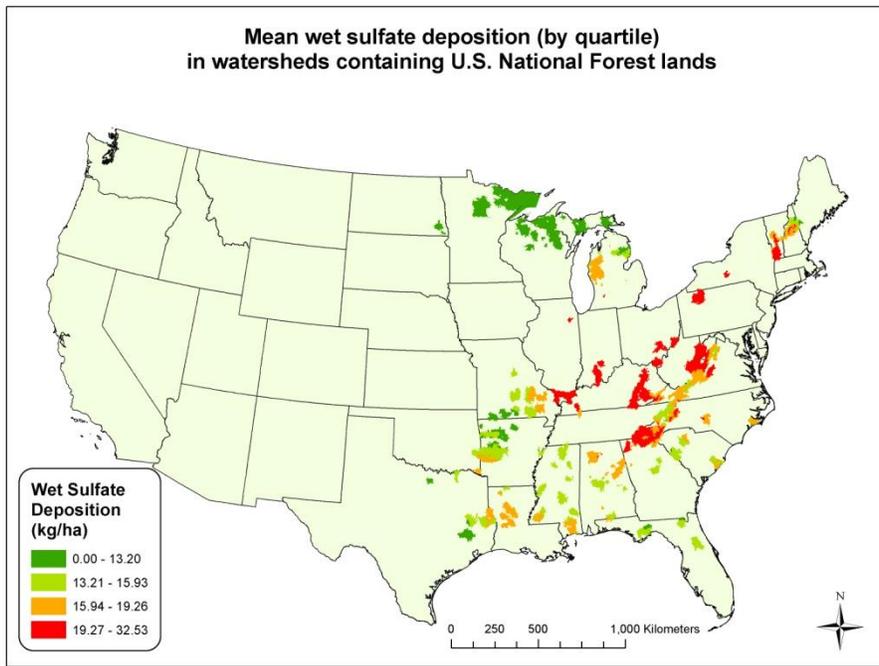


Figure 4. Mean wet sulfate deposition (kg/ha) in the HUC6 watersheds, symbolized by quartile.

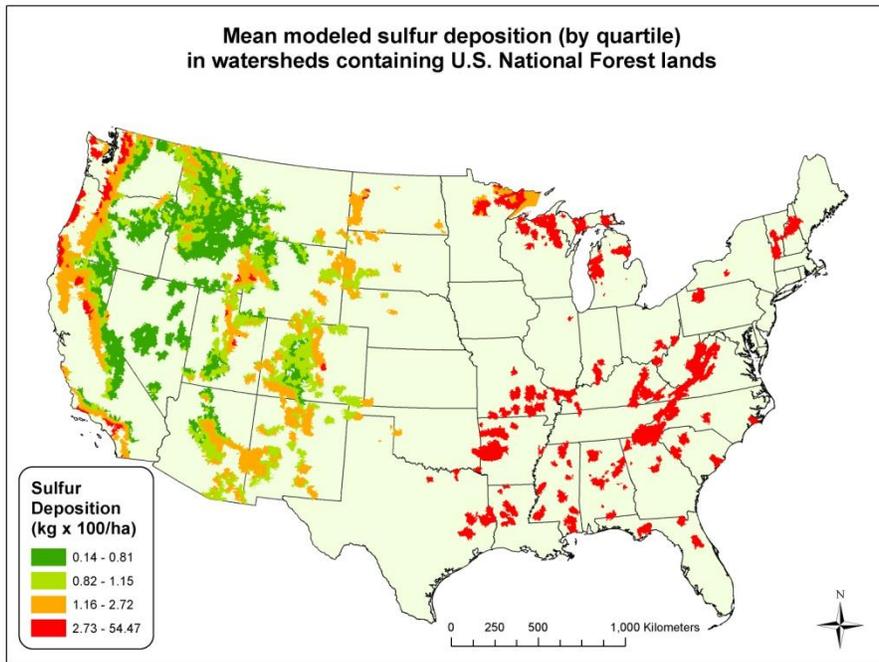


Figure 5. Mean modeled sulfur deposition (kg/ha) in the HUC6 watersheds, symbolized by quartile.

Figures 6-10 depict five different combinations of risk factors for the 12 digit HUCs. The red areas are considered at highest risk of having methylmercury formation, however these predictions have not yet been validated.

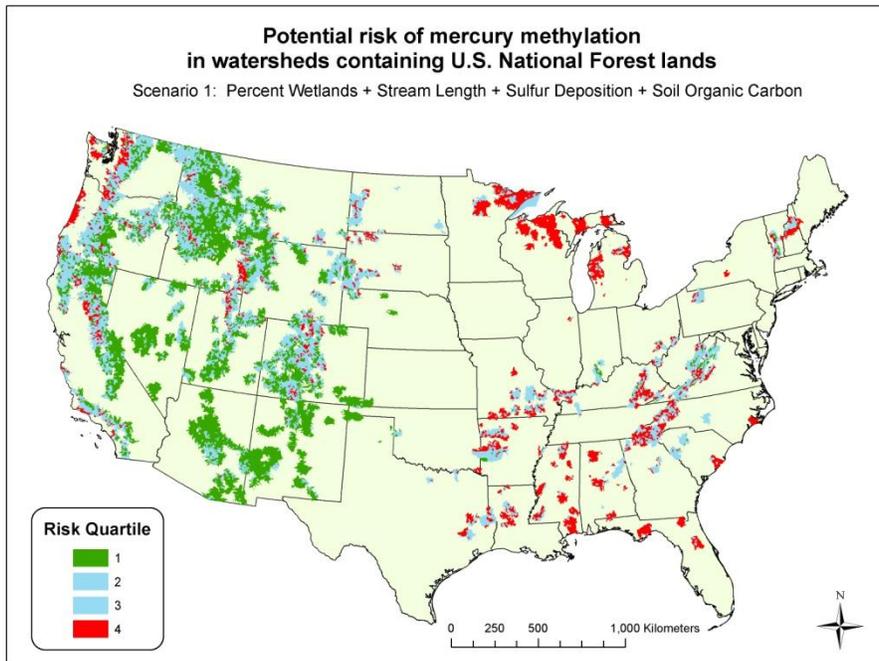


Figure 6. Scenario 1: wetlands + streams + sulfur + organic carbon.

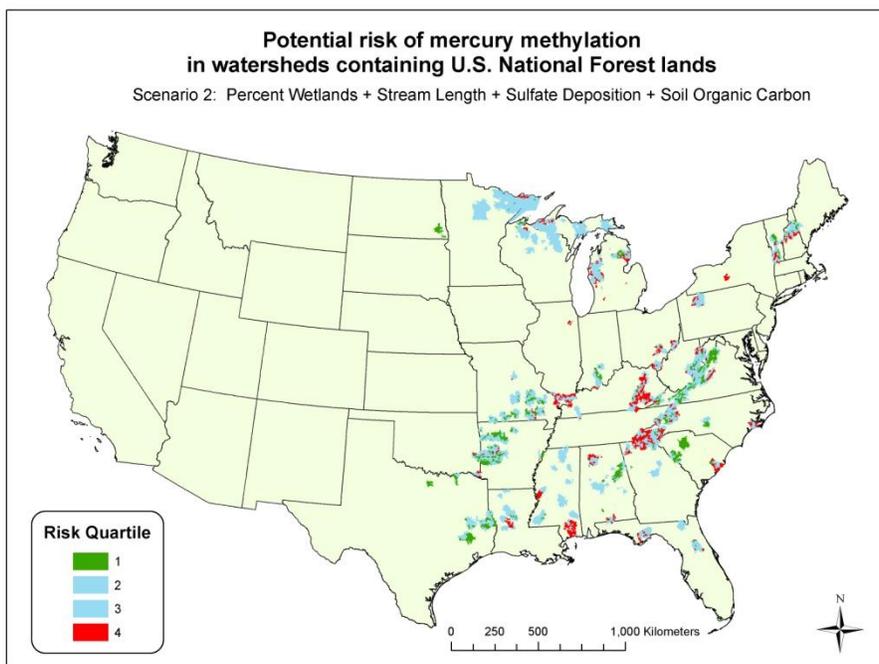


Figure 7. Scenario 2: wetlands + streams + sulfate + organic carbon.

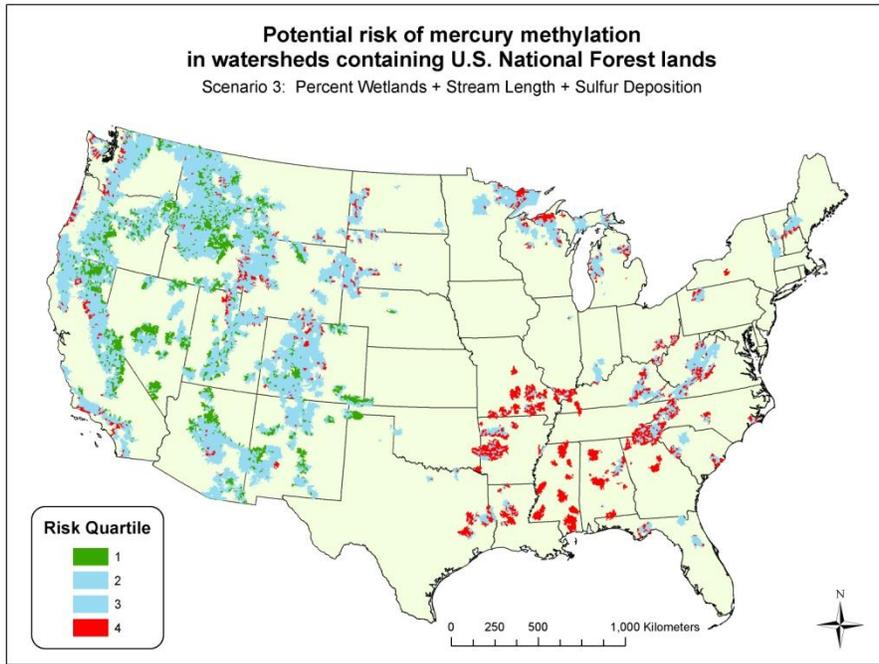


Figure 8. Scenario 3: wetlands + streams + sulfur.

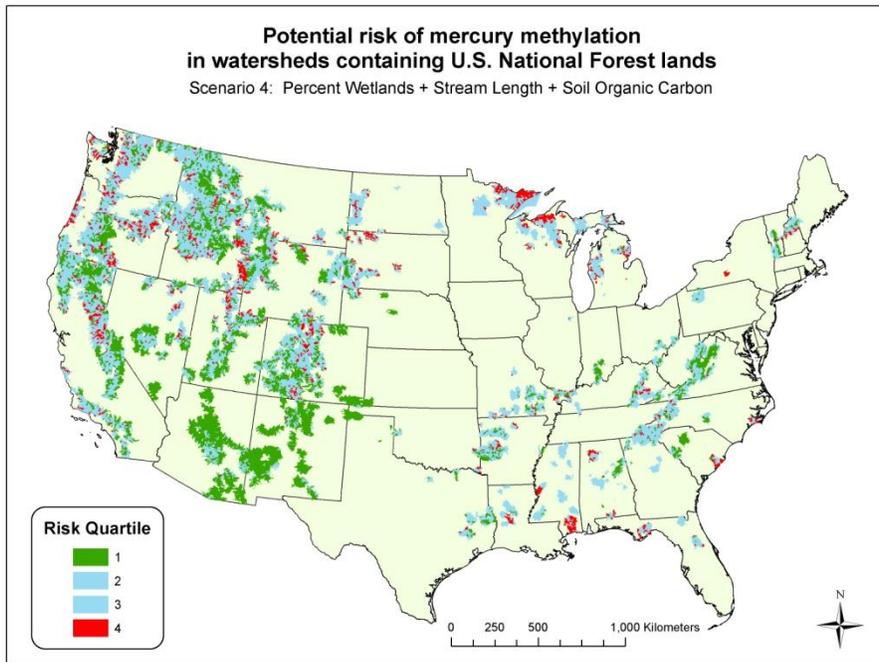


Figure 9. Scenario 4: wetlands + streams + organic carbon.

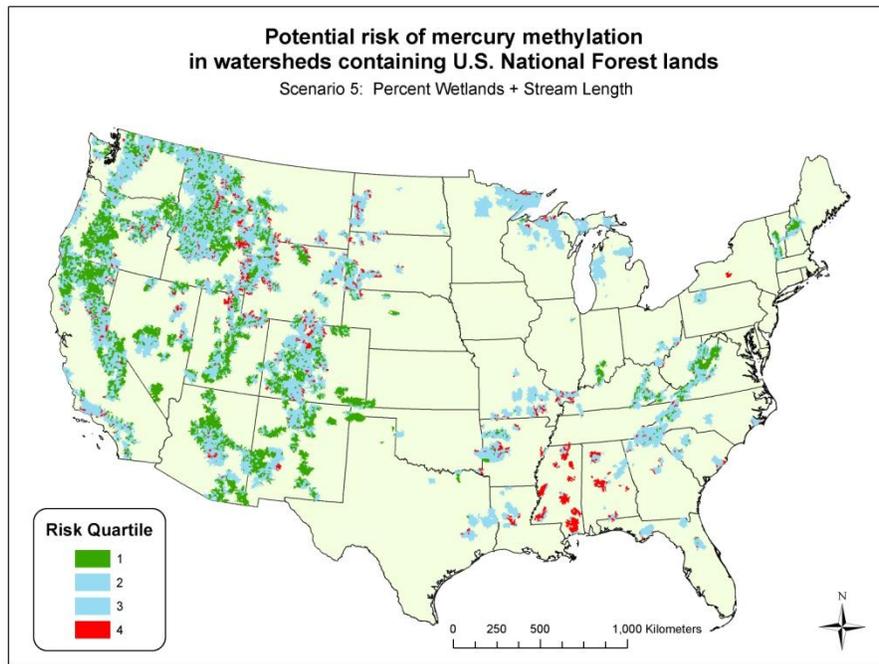


Figure 10. Scenario 5: wetlands + streams.

Recommendations

1. Obtain data that can be used to evaluate the risk assessment scenarios to determine validity of the results. This could be done nationally or regionally, depending on availability of data. Appendix 2 provides a list of potential data sources and contacts.
2. Combine the modeled mercury deposition (and emissions source) information aggregated at the 12 digit HUC with the MeHg risk assessment results. Idea: create a GIS tool that allows you to navigate to a particular HUC and view the MeHg risk rating, mercury deposition estimate, and emissions source info. EPA provided the mercury deposition and emission sources summary by HUC in a geodatabase.
3. Share these results with the WO groundwater program manager, Chris Carlton, who is interested in this work.

References

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Appendix 1 – GIS Analysis and metadata

Analysis regions

Two groups of regions were analyzed: 1) features for the 6th-level (12 digit) HUCs that contain US Forest Service lands within the continental United States, and 2) features for the wilderness areas that contain US Forest Service lands within the continental United States. Each feature has a set of calculated metrics to assess the potential risk of methyl mercury formation. The base feature set for the 6th-level (12 digit) HUCs was the *HUC6_select* layer (from Dwight Atkinson, EPA). The base feature set for the wilderness areas was obtained from www.wilderness.net. From this, features that were in the continental United States and had the value “FS” in the ‘AGBUR’ field were selected.

Risk Factor Fields.

Values for factors important in methylation risk were calculated as follows:

Stream: Stream Density. The sum of the length of the NHDPlus flowline segments in each region divided by the region area (units: km/ha). Data used: NHDPlus flowline data for each region.

Organic Carbon: Mean percent organic carbon; the average of percent organic carbon in each region (units: %). The estimates of organic carbon were provided by Erika Cohen (USDA Forest Service, Southern Research Station). Organic carbon was estimated from organic matter using the following relationship; organic carbon = organic matter / 1.724 (U.S. Department of Agriculture, Natural Resources Conservation Service. National Soil Survey Handbook, title 430-VI. <http://soils.usda.gov/technical/handbook/> Accessed April 8, 2009). The units for organic matter and organic carbon are percent. Organic matter data were obtained from the STATSGO soils database and processed originally for the critical loads assessment conducted by McNulty et al (Steven G. McNulty, Erika C. Cohen, Jennifer A. Moore Myers, Timothy J. Sullivan, Harbin Li, 2007, Estimates of critical acid loads and exceedances for forest soils across the conterminous United States, Environmental Pollution, Volume 149, Issue 3, October 2007, Pages 281-292).

Wetlands: Percent wetland land use. The percentage of each region that has a land use classified as wetlands (units: %). Data used: the 2001 NLCD landuse raster (value = 11-open water, or 90-woody wetlands, or 95-emergent herbaceous wetlands).

Sulfate: Mean wet sulfate deposition. The average amount of wet sulfate deposition (from 1983-2008) in each region in the eastern United States (units: kg/ha). Data used: annual wet deposition grids for the Eastern U.S. Provided by: Bill Jackson, Air Resource Specialist, USDA Forest Service, National Forests in North Carolina, Asheville, NC. The procedures utilized to produce the data are described by: Grimm, J.W., and J.A. Lynch. 1997. Enhanced Wet Deposition Estimates Using Modeled Precipitation Inputs. Final Report to the USDA Forest Service, Northeast Forest Experiment Station, Northern Global Change Research Program (23-721).

Sulfur: Mean modeled sulfur deposition. The average amount of modeled annual sulfur deposition in each region for the continental United States (units: kg x 100/ha). Based on the ‘S_1A’ field (average sulfur deposition (kg S/ha) which includes both wet and dry deposition. Community Multiscale Air Quality Modeling System (CMAQ) data were obtained from the Environmental Protection Agency by Rick Graw, Air Resource Program Manager, USDA Forest Service, Region 6, Portland, OR. The meteorological data and emissions inventory used in the modeling run are described in detail below.

2. Input Data and Model Configuration

2.1. Meteorology

The CMAQ model requires gridded meteorological data to provide estimates of various meteorological parameters such as temperature, wind speed and direction, relative humidity and planetary boundary layer (PBL) height. The 5th generation Mesoscale Model (MM5; Grell et al., 1994) is an Eulerian meteorological model that provides estimates of the meteorological parameters required by the CMAQ model, and has been used and tested extensively with the CMAQ model over the past 15 years. For this work, the MM5 version 3.7.4 was used for both the 36-km and 12-km domains. The 36-km MM5 domain consists of 165 by 129 grid cells covering the entire CONUS, and includes portions of Canada and Mexico. The 12-km domain consists of 290 by 251 grid cells covering the eastern two-thirds of the U.S..

Boundary conditions for the 2002 – 2005 36-km and 12-km MM5 simulations were provided by the 40-km Eta Data Assimilation System (EDAS) data; while the 12-km North American Model (NAM) data were used as boundary conditions for the 2006 36-km and 12-km MM5 simulations, with any missing data filled in using the 32-km (North American Regional Reanalysis (NARR) data. The MM5 simulations utilized the Kain-Fritsch 2 (KF2; Kain, 2004) cumulus parameterization; the asymmetric convective model version 2 (ACM2; Pleim, 2007a,b) PBL scheme; the Reisner 2 (Reisner et al., 1998) explicit microphysics scheme; the Dudhia shortwave radiation scheme (Dudhia, 1989); the RRTM longwave radiation scheme (Mlawer et al., 1997); and the Pleim-Xiu (PX; Xiu and Pleim, 2001; Pleim and Xiu, 1995) land surface model (LSM). Both the 36-km and 12-km MM5 simulations utilized 34 vertical layers, with a surface layer of approximately 36 meters. The meteorological outputs from both sets of MM5 simulations were processed to create model-ready inputs for CMAQ using the Meteorology-Chemistry Interface Processor (MCIP; Otte et al., 2005) version 3.4.

2.2. Emissions

The 2002 National Emissions Inventory (NEI) version 3 was used as the primary basis for the 2002 – 2006 emissions inputs. Version 3 of the 2002 NEI is documented at <http://www.epa.gov/ttn/chief/net/2002inventory.html#documentation>. For the major point sources, namely electric generating units (EGUs), year specific continuous emission monitoring (CEM) data were used. Year specific updates to mobile emissions were done using the MOBILE6 model, and daily estimates of fire emissions based on satellite detection of fires were included as well. Monthly NH₃ emissions from livestock were adjusted according to the inverse-modeling recommendations of Gilliland et al. (2006), while emissions from fertilized crops are from Goebes et al. (2003). For inventories outside of the U.S., which include Canada, Mexico and offshore emissions, the latest available base year inventories were used. The CMAQ model-ready emissions were created using the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system.

2.3. CMAQ Model Configuration

The CMAQ simulations were performed at the 36-km horizontal grid spacing for the CONUS, while for the eastern two-thirds of the U.S. a CMAQ simulation using 12-km horizontal grid spacing was performed. Chemical boundary conditions for the 12-km simulation were provided by the 36-km simulation and boundary conditions for the 36-km CMAQ simulation were provided by a non-year specific GEOS-Chem (Bey et al., 2001) simulation. The boundary data were created by taking the median value of a 2.0 degree by 2.5 degree (latitude-longitude) 24-vertical layer 2002 GEOS-Chem simulation and averaging the three-hourly data to monthly values. These monthly averages were then used as boundary conditions for all five years of the 36-km CMAQ model simulations.

The CMAQ simulations utilized version 4.7 (Foley et al., 2010) of the model, the latest version of the model available at the time. The simulations included a 10-day spin-up period for the 36-km simulations, while a 3-day spin-up period was used for the 12-km simulations. The CMAQ simulations were performed using the same horizontal dimensions as their respective meteorology simulation except that the horizontal

dimensions were reduced by five grid cells on each of the four lateral boundaries to avoid artifacts that can appear along the domain boundaries in the meteorological simulations. However, unlike the meteorological simulations which utilized 34-vertical layers, the CMAQ model simulations were performed using 24-vertical layers. The CMAQ model simulations used the AERO5 aerosol module (Carlton et al., 2010), the Carbon-Bond 05 (CB05) chemical mechanism with chlorine chemistry extensions (Yarwood et al., 2005) and the ACM2 PBL scheme (Pleim, 2007a,b).

2.4. Precipitation Bias Adjustment

At least some portion of the error present in the CMAQ estimated wet deposition is due to errors in the precipitation estimates from the meteorological model. Since both the NTN observed and MM5 estimated precipitation are available for each NTN site, the modeled wet deposition can be adjusted to account for the error present in the model estimated precipitation. This adjustment is accomplished here by linearly adjusting the CMAQ estimated wet deposition by the ratio of the observed to estimated precipitation. For example, in the case where the observed precipitation is greater than the model estimated precipitation, the ratio is greater than one, and therefore the model estimated wet deposition is increased.

Note that this precipitation adjustment technique assumes that the observed to modeled precipitation ratio is well correlated with the observed to modeled deposition ratio. In other words, it is not assumed that the wet deposition scales linearly with precipitation, but only that the relationship between the errors in the model precipitation estimates and the error in the CMAQ model deposition estimates is linear. Since the bias adjustment was applied over the aggregated seasonal and annual totals, there were no instances in which the observed precipitation was greater than zero while the model estimated precipitation was zero. However, in such instances where there is observed precipitation but no model predicted precipitation, the current method of bias adjustment would keep the model estimated wet deposition zero for all species. The impact of this precipitation bias adjustment will be presented for each of the wet deposition species.

Quartile Fields.

Quartile cutoffs were calculated for each risk factor field, as shown below. In each quartile field, a value of “1” was assigned if it was in the 1st quartile, a value of “2” was assigned if it was in the 2nd or 3rd quartile, and a value of “3” was assigned if it was in the 4th quartile.

Quartiles for 6th-level (12 digit) HUCs:

Stream_Q: Quartile values for Stream Density. Quartile cutoffs—1st: 0.006090,
2nd: 0.007620, 3rd: 0.009284, 4th: 0.024191.

Organic_Q: Quartile values for Organic Matter. Quartile cutoffs—1st: 0.278873,
2nd: 0.433037, 3rd: 0.710013, 4th: 30.148529.

Wetlands_Q: Quartile values for Wetlands. Quartile cutoffs—1st: 0.064117,
2nd: 0.487311, 3rd: 2.327118, 4th: 99.894379.

Sulfate_Q: Quartile values for Sulfate Deposition. Quartile cutoffs—1st: 13.195808,
2nd: 15.933795, 3rd: 19.261171, 4th: 32.528809.

Sulfur_Q: Quartile values for Sulfur Deposition. Quartile cutoffs—1st: 0.807059,
2nd: 1.147595, 3rd: 2.717642, 4th: 54.470001.

Quartiles for wilderness areas:

Stream_Q: Quartile values for Stream Density. Quartile cutoffs—1st : 0.004837,
2nd : 0.006654, 3rd : 0.008409, 4th : 0.025525.

Organic_Q: Quartile values for Organic Matter. Quartile cutoffs—1st : 0.296424,
2nd : 0.459687, 3rd : 0.855821, 4th : 18.486264.

Wetlands_Q: Quartile values for Wetlands. Quartile cutoffs—1st : 0.006619,
2nd : 0.136215, 3rd : 0.778624, 4th : 98.860085.

Sulfate_Q: Quartile values for Sulfate Deposition. Quartile cutoffs—1st : 14.201461,
2nd : 17.260344, 3rd : 20.969616, 4th : 32.171677.

Sulfur_Q: Quartile values for Sulfur Deposition. Quartile cutoffs—1st : 0.994405,
2nd : 1.597799, 3rd : 5.437564, 4th : 24.575716.

Scenario Fields.

Five risk assessment scenarios were performed by adding various combinations of quartile fields.

Scenario1: [Wetlands_Q] + [Stream_Q] + [Sulfur_Q] + [Organic_Q]

Scenario2: [Wetlands_Q] + [Stream_Q] + [Sulfate_Q] + [Organic_Q]

Scenario3: [Wetlands_Q] + [Stream_Q] + [Sulfur_Q]

Scenario4: [Wetlands_Q] + [Stream_Q] + [Organic_Q]

Scenario5: [Wetlands_Q] + [Stream_Q].

Appendix 2 – Data sources and contacts for evaluating the risk assessment products

Potential National Data

Fish tissue data from Fish and Wildlife Service. My initial contact was Karen Nelson (via Ann Mebane). Karen indicated they could pull data from ECDMS. Bruce is the person who would actually do the database query for us.

- Karen Nelson (Environmental Contaminants Program). Phone: 406-449-5225, ext 210. Email: Karen_nelson@fws.gov
- Bruce Nierwienski (IT Database “guru”). Phone: 304-876-7744. Email: bruce_nierwienski@fws.gov

Fish and sediment data from USGS. My initial contact was David Donato, computer scientist with the Eastern Geographic Service Center in Reston, VA. I originally started with investigating the EMMMA dataset (<http://emmma.usgs.gov>), but didn’t get too far. They have MeHg data for fish tissue, but I believe you would have to go through the process of standardizing that to species and weight. Probably better to go with a regional approach with scientists who are familiar with standardizing for their area. (See MN ideas below). David suggested that I also look at <http://tim.er.usgs.gov/geochem/>. This is the topical site index site which is more informative about available stream data.

- David Donato. Phone: 703-648-5772. Email: didonato@usgs.gov

Fish data from Great Lakes Region. Trent Wickman led me to Bruce Monson (Minnesota Pollution Control Agency, Environmental Analysis and Outcomes Division). Bruce has done a lot of work in Minnesota and throughout the Great Lakes with mercury, and particularly with standardizing the mercury data by year, season of collection, size of fish, etc. He was actively working on a publication that is relevant to our risk assessment early in 2011. This might be the best data to use for a regional validation of our risk assessment scenarios. He recommended looking at the MercNet inventory for data: <http://mercnet.briloon.org/>

- Bruce.Monson@state.mn.us