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Contact Information

209 Penn. Ave., SE
Washington, DC 20003

Tel: 202-454-5249

Fax: 202-454-5223

Robert Ferguson

Executive Director

bferguson@ff.org

Misuse of Rainwater Mercury Data In National Wildlife Federation's Cycle of Harm Report

Summary and Conclusion

The National Wildlife Federation (NWF) appears to have misinterpreted scientific data and presented both inappropriate and incorrect data analysis in support of arguments presented in their "National Recommendations for Eliminating Mercury Pollution" (pp 7-8 of Cycle of Harm). Detailed issues with NWF's Cycle of Harm report are outlined in this paper.

Also of serious concern, NWF's conclusions seem to start from a *preconceived* concern with exposures to mercury rather than being based on an impartial scientific evaluation of available measurements, as well as on the state of current scientific knowledge on the natural cycling of mercury in the environment. For these reasons, the recommendations in Cycle of Harm are not scientifically credible.

Owing to its highly public release to the popular media, the report may misdirect resources and concerns of the sport fishing industry, the health care industry, policy-makers and regulators, and members of the public. To date, the mercury concentrations measured in rainwater from all over the U.S. are neither unexpected nor at harmful enough levels to warrant the dramatic and extreme conclusions given in NWF's Cycle of Harm report.

CSPP's Analysis and Commentary

In a May 2003 National Wildlife Federation (NWF) report ("Cycle of Harm: Mercury's Pathway From Rain to Fish in The Environment"[1]), it was stated that rainwater in many U.S. states contains "ominous levels of mercury and threatens the health of people and wildlife." This report is the third in the NWF's "Clean the Rain" series[2] "to educate Americans about risk mercury poses and give concerned citizens the information and tools to protect themselves and their environment."[3]

But a careful reading confirms that the Cycle of Harm report contains significant misleading information concerning not only the role of rainfall in the natural cycling of mercury (Hg) in the environment but also on the potential health threats from man-made sources of Hg emission[4]. The report raises concerns for its analytical methods for the following four reasons:

Figure 1

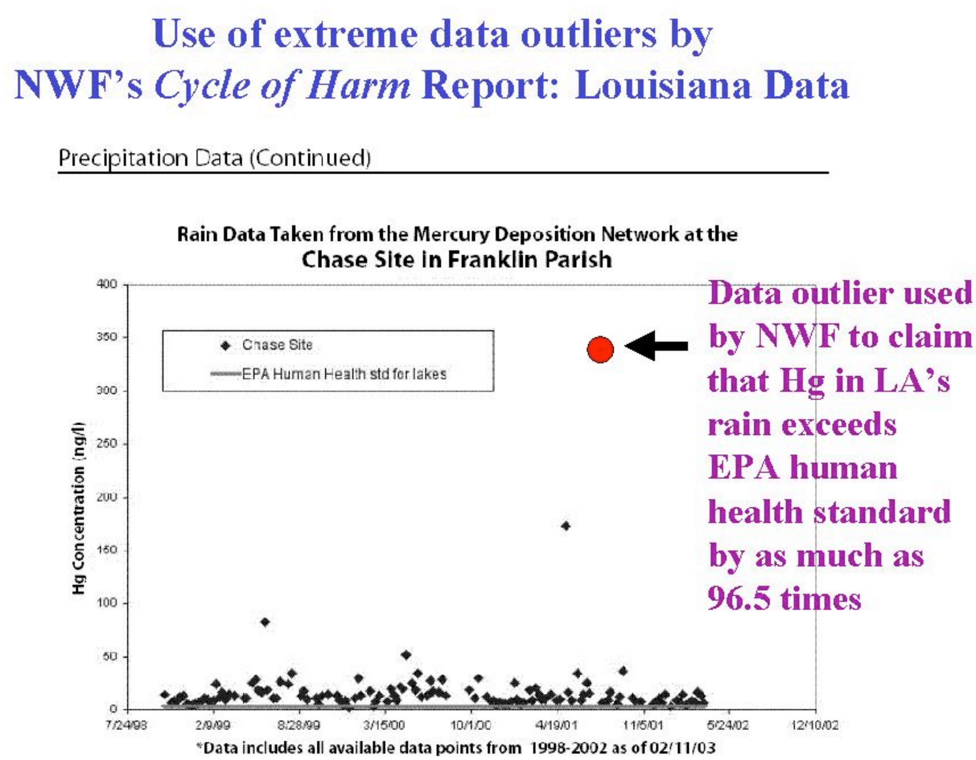


Figure 1: Hg concentration in rainwater at Chase, Franklin Parish, Louisiana; Page 62 of Cycle of Harm — note the data outlier (marked by the red dot for emphasis) used by NWF in their analysis for Louisiana. The EPA human health standard level is set at 3.5 ng/L (parts per trillion) for mercury in lakes by NWF while the average Hg concentration in rainwater collected at Chase, Louisiana is 9.9 ng/L. But see point 3 of this commentary for the explanation why a direct comparison between the two numbers is misleading.

Reason 1: The report consistently adopted the highest data outliers as representative of high data values. These outliers are the ones that are most likely explained by sample contamination[5] or unusual meteorological conditions (i.e., early phases of storm events with low volume of rain water or high concentrations of dissolved mercury) to highlight that these rainwater Hg measurements exceed EPA "human health standard level for mercury in lakes [water]" by e.g., a factor of 10 for the state of New York, 23 for the state of Florida, 77 for the state of Maryland and up to about 96 for the state of Louisiana.

Figures 1 and 2 show the examples of Hg concentration data from rainwater collected at Chase, Franklin Parish, LA and at the Chesapeake Biological Laboratory (CBL), MD, respectively. But *NWF has failed to consult the proper scientific literature* (e.g., Mason et al. 2000[6] for the Chesapeake Biological Laboratory data series) that actually recommended removal of those outliers from the published record[7] on the ground that *these data points are either contaminated or statistically unrepresentative*. It is clear that NWF's report has misunderstood and misused the rainwater Hg data collected by the scientific community. Some of the CBL data are obtained by specific event sampling while data from the Mercury Deposition Network represents a weekly integrated rainfall sample. Thus even a direct comparison of CBL data with the MDN data may not be appropriate.

Figure 2

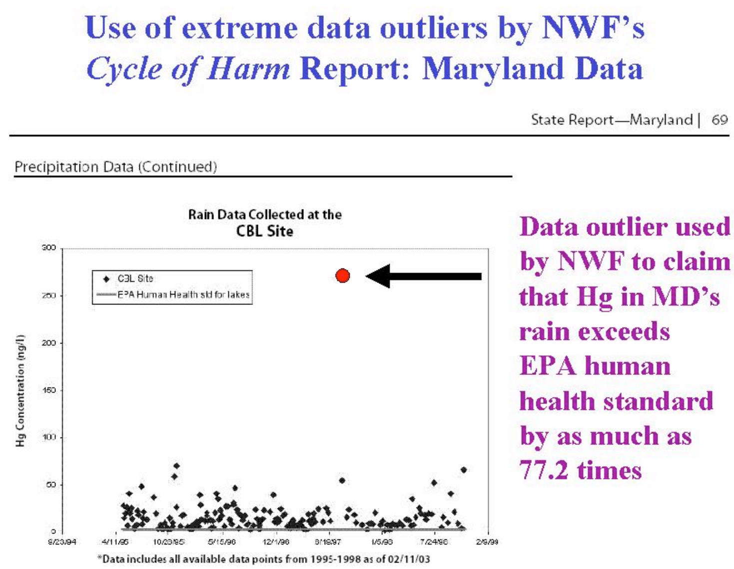


Figure 2: Hg concentration in rainwater at Chesapeake Biological Laboratory (CBL), Maryland; Page 69 of Cycle of Harm note the data outlier (marked by the red dot for emphasis) used by NWF in their analysis for Maryland. The EPA human health standard level is set at 3.5 ng/L (parts per trillion) for mercury in lakes by NWF while the average Hg concentration in rainwater collected at CBL, Maryland is 17.7 ng/L. But see point 3 of this commentary for the explanation why a direct comparison between the two numbers is misleading.

It should also be noted that published scientific literature has long documented an *inverse relation* between Hg concentration in rainwater and the actual volume of the rain sample. **Figure 3** shows that inverse relation for the weekly rainwater collected at Little Rock Lake, north central Wisconsin from 1993 through 1999 (Watras et al. 2000[8]). It is clear from **Figure 3** that higher Hg concentrations can simply be associated with very low rainfall volume events[9] that are a function of the meteorological conditions, rather than having any ties to enhanced exposure to emission sources. There is a "washout" effect whereby the first rain at the end of a dry spell or season has higher values, basically "cleansing" the atmosphere of ionic mercury. Thus it is neither surprising nor alarming to find occasional rainwater Hg data points that are above 100 ng/L.

Figure 3

Inverse relation of Hg concentration in rainwater and actual amount of rain: The highest Hg concentration is not because of increased exposure to emission sources but simply a result of low precipitation volume.

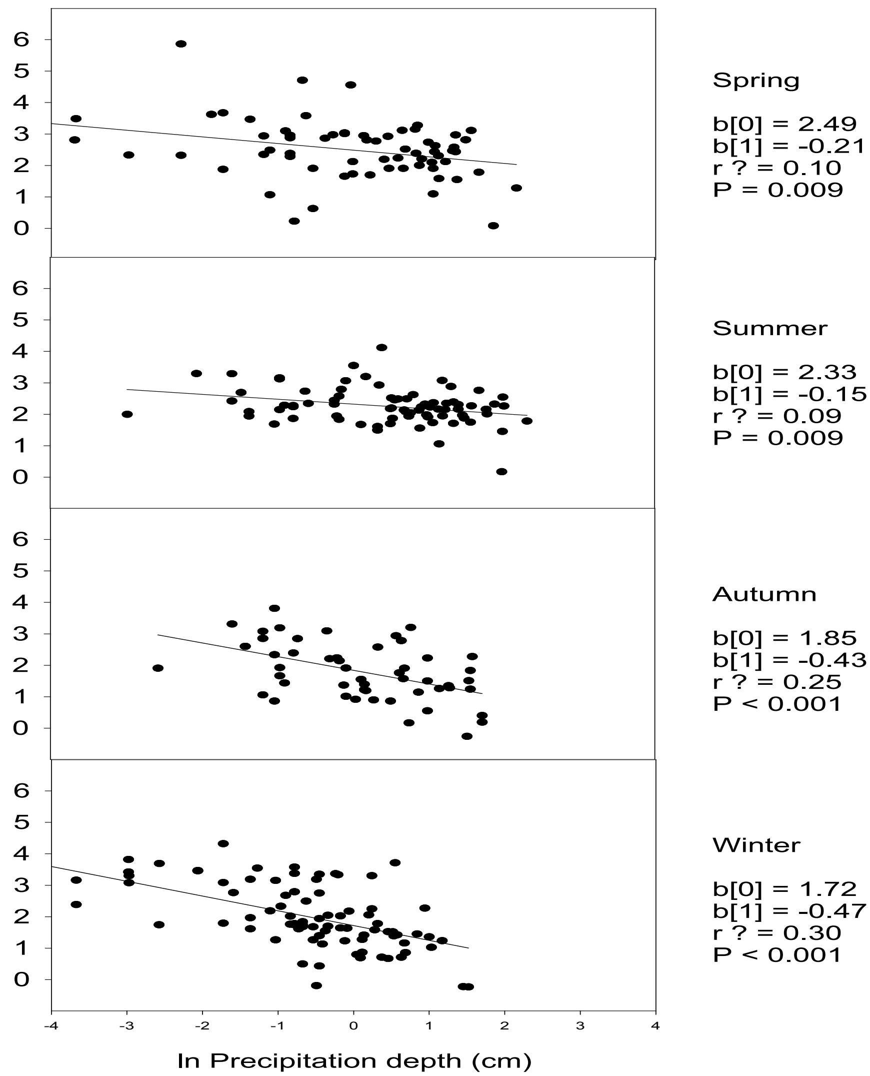


Figure 3: Inverse relation between Hg concentration in rain water and the amount of rain for data collected at Little Rock Lake, north central Wisconsin from 1993 through 1999 (Watras et al. 2000). Both axes are expressed in units of natural logarithm of Hg concentration in rainwater and the amount of rain.

<u>Reason 2:</u> NWF's comparisons of the mercury content in rainwater to those in lake, river or ocean waters on which the EPA based its health recommendation are *neither compatible nor correct* because of the large differences in the sampling volume for the two types of water involved.[10] All mercury in rainwater will ultimately be diluted by the enormous volume of background waters contained in our lakes, rivers and the oceans. For example, the amount of rainwater that falls globally each year is only about 0.0004 times that of the volume of water in the world oceans. In other words, comparing the Hg content in rainwater with the Hg content in all natural water bodies is inappropriate *because* it is an incompatible comparison of "an apple to an orange." Furthermore, the "EPA water quality criterion for human health" cited by NWF is actually for aquatic systems with mercury concentrations in water that will result in a particular mercury concentration in fish in those same waters. So the human health threshold concentration should not really be compared to the rainwater concentration as reported in Cycle of Harm.

<u>Reason 3:</u> If the actual mercury concern is health related through human exposure to methylmercury via fish consumption, then the research focus should be on the actual mercury content in the U.S. aquatic ecosystems instead of measurements of a subsidiary variable such as the amounts (slightly concentrated) in rainwater.[11]

Figure 4

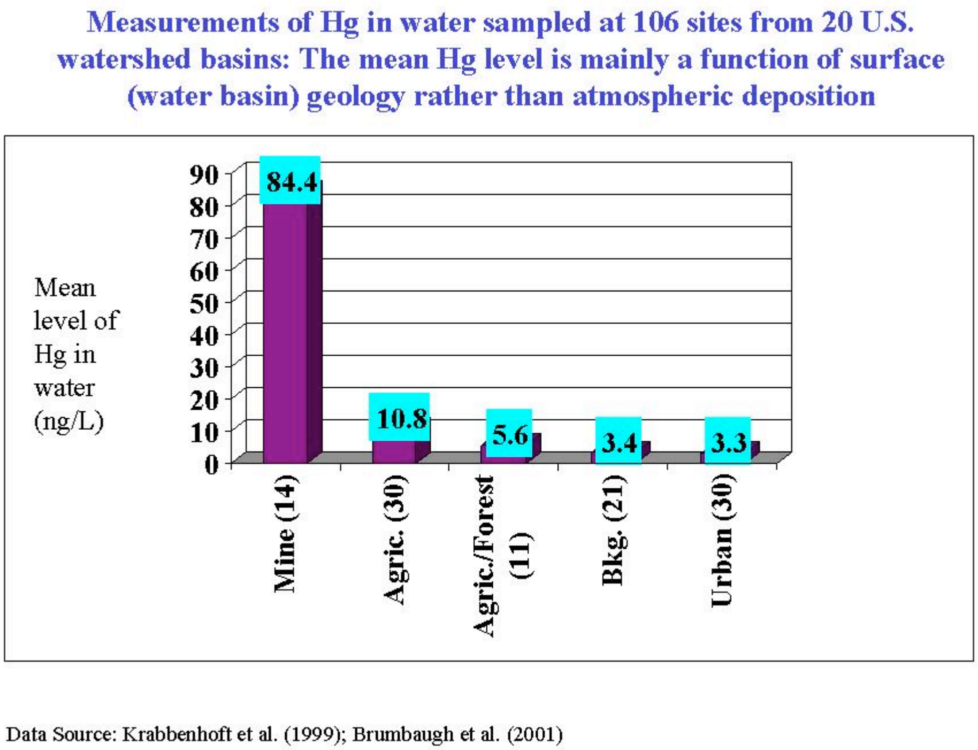


Figure 4: Mean level of Hg in water sampled at 106 sites from 20 U.S. watershed basins studied by the U.S. Geological Survey.[12]

Such a focus reveals that the total mercury contents in lake waters sampled from 20 U.S. watershed basins range from about 0.3 to 5 ng/L (parts per trillion) for background/reference and urban sites, increases to relatively higher levels for agriculture and forested sites and then reaching as high as 1100 ng/L for sites with current or prior mining activities.[13] **Figure 4** shows the mean level of Hg in water from the 106 sites of these 20 U.S. watershed basins measured by scientists from the U.S. Geological Survey. It is most important to recognize that the mean level of Hg concentration in these natural watersheds is *dominated by pre-existing conditions* of surface and sedimentary geology[14] or water discharges *rather than atmospheric deposition*. This explains why mercury levels in lakes and rivers at urban sites, which are *believed* more directly prone to Hg contamination by nearby emissions such as from power plants, are *not* guaranteed to have higher Hg levels than those at aquatic ecosystems from remote background or agricultural sites.

Figure 5

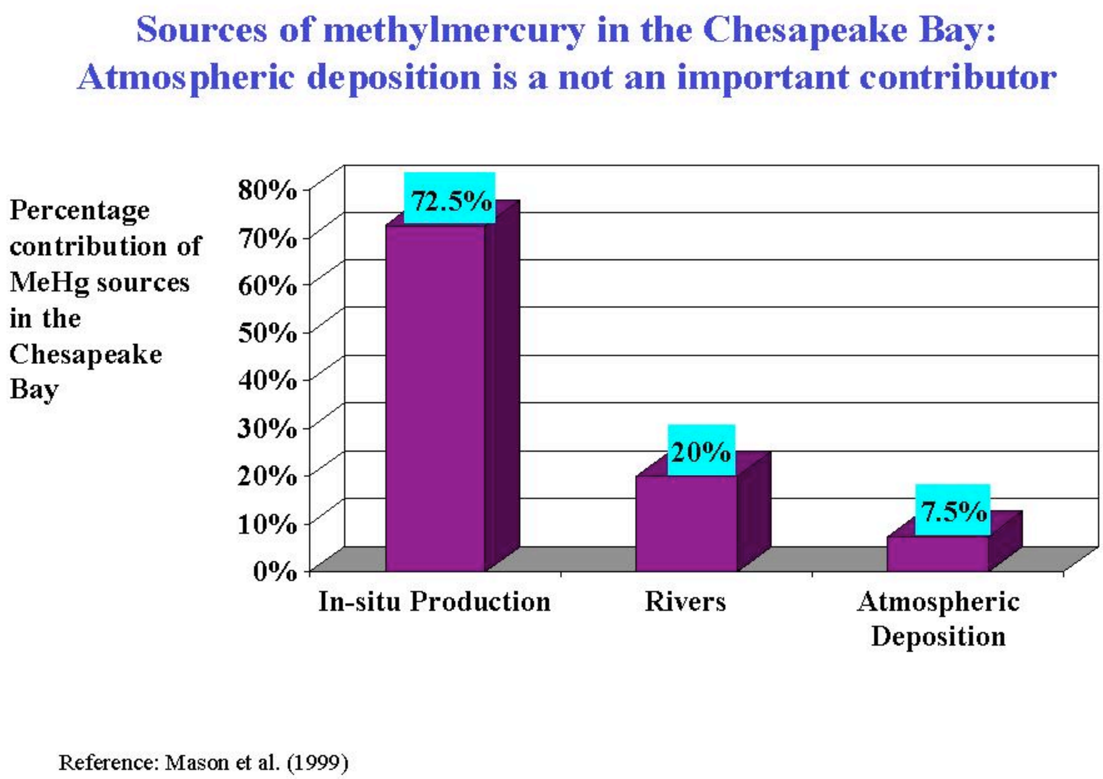


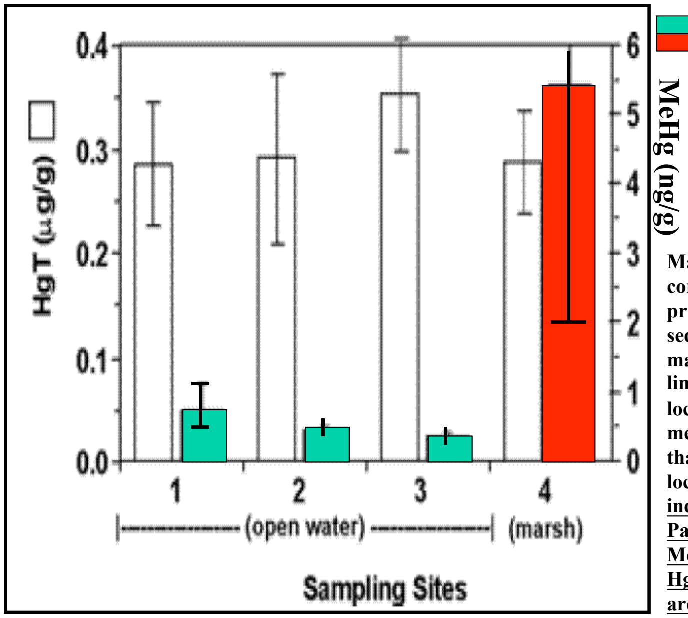
Figure 5: Sources of methyl mercury (MeHg) in the Chesapeake Bay.

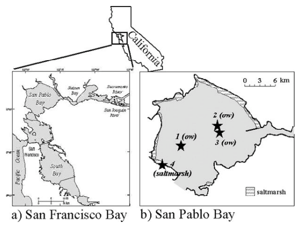
Thus, the data presented in **Figure 4** suggest that the levels of mercury in rain waters, ranging from 0.2 ng/L to 350 ng/L (even including the extreme data outliers) as collected by the Mercury Deposition Network[15] and in turn badly re-interpreted in NWF's Cycle of Harm report, are *neither unusual nor alarming*. The attempt by NWF to tie the rainwater Hg strictly to man-made emissions from U.S. power plants remains *unproven and unconvincing*.

Reason 4: It is significant to point out that the mercury in rainwater exists mostly in the dissolved ionic form of mercury (Hg^{2+}) rather than methylmercury (MeHg) — the biologically active form of mercury believed to have relevance for the well-being of both humans and wildlife. For example, Mason et al. (1999) found — through careful budgeting of the biogeochemical cycling of mercury and the tracking of various species of Hg in the Chesapeake Bay[16] — that most (72.5%) of the methylmercury in the system has to come from in-situ production annually, while remote transport from rivers contributes another 20% and atmospheric deposition supplies only about 7.5% of the total (**Figure 5**).

Figure 6

Mircobial methylmercury (MeHg) production in marsh wetlands are 25-50 times more than in open-water locations around San Pablo Bay area





Marvin-DiPasquale et al. (2003) concluded that "Microbial MeHg production ... in 0-4 [cm] surface sediments was also the highest in the marsh [3.1 ng/g/day] and below detection limit [< 0.06 ng/g/day] in open-water locations. The marsh exhibited a methylation/demthylation ratio more than 25X than of all open-water locations. ... These preliminary data indicate that wetlands surrounding San Pablo Bay represent important zones of MeHg production, more so than similarly Hg-contaminated adjacent open-water areas. " (p. 260)

Reference: Marvin-DiPasquale et al. (2003) Environmental Geology, vol. 43, 260-267

Figure 6: A contrast of total mercury (HgT in _g/g) and methyl mercury (MeHg in ng/g) concentrations for surface sediments from four different sampling sites (3 open-water versus 1 salt-marshland sites) highlighting the significant boost in the production of methyl mercury (scale on the right) in the biologically and organically richer marsh wetland site despite the almost-equal amount of total mercury in all four sites. This evidence confirms the fact that the production of methyl mercury is not limited by the amount of total mercury. [Readapted from Figure 2 in Marvin DiPasquale et al. 2003]

Again, it needs to be re-emphasized that the biomethylation and bioaccumulation processes in the food web that ultimately convert inorganic Hg into MeHg in fish (the biologically toxic form of Hg) *are not* directly related to, nor limited by, the amount of inorganic mercury (as emitted from U.S. power plants) available. Instead, methylmercury production appears limited by ecosystem dynamics, water quality variables like dissolved sulfate, parameters like the population of algae and/or zooplankton,[17] the availability of nutrients and/or sunlight and so on.

In a recent study of microbial cycling of mercury in sediments of San Pablo Bay, California, Marvin-DiPasquale et al. (2003) noted that "sediment geochemistry (redox, sulfide, pH, organic content, etc.) is a much more important control on MeHg production than is the absolute total mercury concentration."[18] It was found that despite the relatively constant level of total mercury in all four sampling sites in the study (3 open-water and 1 salt-marshland sites), the production and concentration of methylmercury are significantly enhanced at the biologically active and organically rich marsh wetland site (the red bar in **Figure 6**).[19]

Figure 7

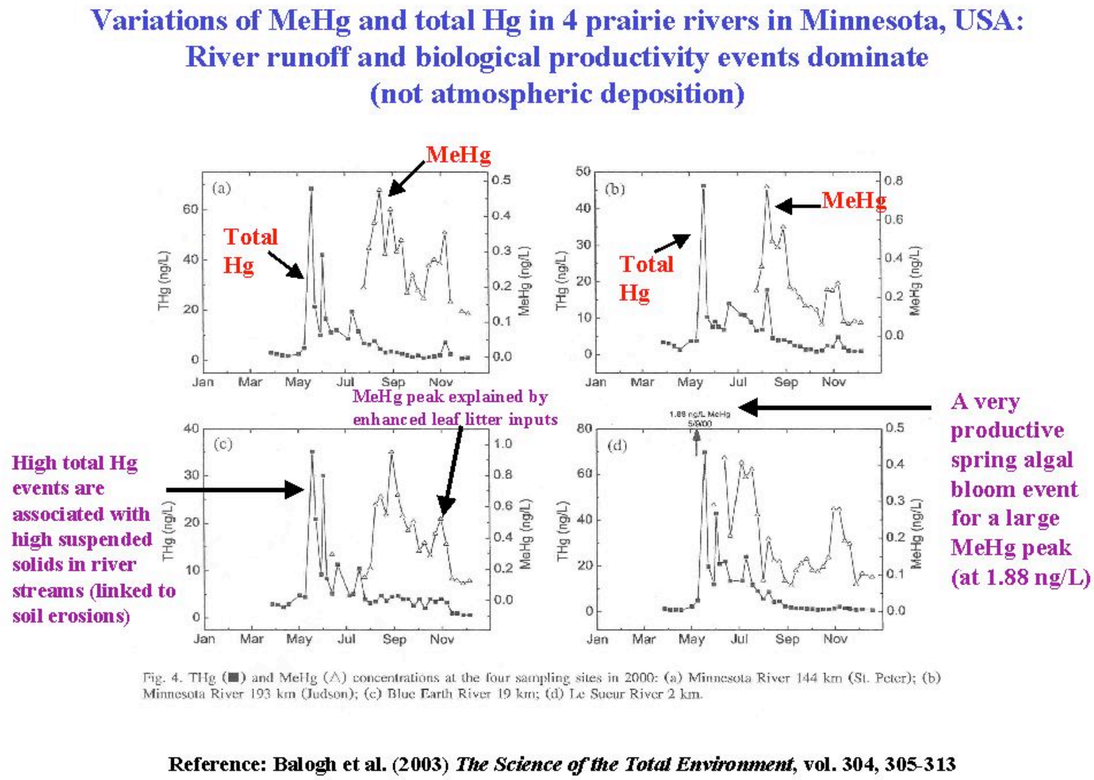


Figure 7: Ecosystem factors (the population of algae and zooplankton, the availability of nutrients and sunlight, suspended solids, leaf litter inputs and so on) of change for both total mercury (THg) and methyl mercury (MeHg) concentration in 4 river streams (St. Peter portion of the Minnesota River; Judson portion of the Minnesota River; Blue Earth River, Le Sueur River) in Minnesota, USA

Figure 7 gives another example of the changes in both the total mercury (Hg) content and methylmercury (MeHg) content for 4 prairie river systems in Minnesota (Balogh et al. 2003[20]). The authors concluded that the level of MeHg in these river systems "varied in responses to a number of diverse influences" that are linked to biological activity and river runoff events.

To date, *there is no clear evidence for linking the spatial variability of MeHg in rivers and other aquatic ecosystems to U.S. power plant emissions*. This is why it is still highly premature to conclude that even a severe reduction of mercury emissions from U.S. power plants must necessarily lead to a significant decline in the apparently *natural* levels of methylmerucry in fish. In other words, even a full 100% reduction of the current 40-50 tons of Hg from US power plants would not be able to account for the remaining 99% of annual Hg emissions from other natural and man-made sources.

Conclusion

The NWF report employs faulty data analysis practices and deficient scientific understanding of the natural cycle of mercury. Thus, its findings are *not scientifically credible nor its conclusions warranted*. Any public policy prescriptions reliant upon it would likely provide no benefits while producing the unintended consequence of putting at unnecessary risk both economic and human health, especially for minorities, the elderly, the poor, women and children (see WHITE

PAPERS: *EPA Mercury MACT Rulemaking not Justified by Science* and *Analysis of the Sierra Club's Alarmist Claims About the Health Impacts of Mercury* at www.scienceandpolicy.org).

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[1] The full report is available at: http://www.nwf.org/nwfWebAdmin/binaryVault/cycleofharm.pdf

[2] The first two reports for the NWF's "Clean the Rain" campaign series are available at http://www.nwf.org/cleantherain/CTRexec.html and http://www.nwf.org/cleantherain/ctriiexec.html, respectively. Both reports adopted a similar approach, as in *Cycle of Harm*, of comparing measurements of mercury content in rainwater for various locations in the U.S. to "health standards" established for both human and wildlife that were based on Hg content in aquatic systems, without the critical explanation of the large dilution -- because of the large collecting volumes in our lakes, rivers and oceans -- of the Hg content in rain water as it enters the aquatic system.

[3] From page 3 of *Cycle of Harm* report, foreword by Mark Van Putten, President and CEO of NWF.

[4] See addition important health information on mercury at www.scienceandpoliyc.org

[5] A scientist (Steven A. Claas) studying environmental mercury pollution has this informative description on what is involved in making measurements of mercury concentration in various samples: "I actually spend most of my time in an inside laboratory. We work in very special labs because accidental contamination of our samples with mercury is a real problem. Mercury is everywhere, even if in small amounts. ... Unfortunately for those of us making the measurements, the concentration of mercury in many of our environmental samples is incredibly low. Lake water often has only 0.000000001g of mercury in one liter! [nanogram or ng of Hg per liter of water, or ng/L] With levels this low, just breathing on a sample (with our silver-mercury dental fillings) can double the concentration. Our "clean labs" are equipped with special filters that remove microscopic sized particles that may contaminate samples. Air is scrubbed to remove gaseous mercury before it enters the lab. We wear special lab garments - coats or jumpsuits, booties, and hats - to avoid shaking dust from our clothes into samples. We can't touch any of our equipment with our bare hands; I feel naked without my plastic gloves on. We also dress like this when we collect samples in the field. Sometimes it's tough to convince gawking tourists that the strange garb is to protect our samples, not us." (http://www.dist214.k12.il.us/users/asanders/Scientists/Claas.html)

[6] R. P. Mason, N. M. Lawson, G. R. Sheu (2000) *Atmospheric Environment*, vol. 34, 1691-1701.

[7] See Figure 3 in p. 1695 of Mason et al. (2000) where the 270.4 ng/L outlier for CBL was not considered publishable hence not reported in the scientific paper.

[8] C. J. Watras et al. (2000) *Environmental Science & Technology*, vol. 34, 4051-4057. See additional careful discussion on the different methods of measuring Hg content in rainwater in K. A. Morrison, E. S. Kuhn, C. J. Watras (1995) *Environmental Science & Technology*, vol. 29, 571-576.

[9] With rain gauge precipitation amount of the order of 0.1 to 0.5 mm or net bottle catch volume of less than 10 mL---which are "extremely light rainfall events at best" (D. Legates, July 3, 2003, private communication)

[10] We note that National Wildlife Federation may be fully aware of this false comparison because they admitted in their report that "In this comparison, we assume that as long as mercury concentrations in rain are higher than water quality standards, it will be difficult to reduce the mercury concentrations in a lake or river to levels that don't pose risks to people and wildlife." (p. 19 in Chapter 3 of *Cycle of Harm*). This assumption is clearly simplistic because it ignored both the importance of the background level of mercury and the dissociation of the biomethylation and demethylation processes from the availability of mercury from anthropogenic sources.

[11] Furthermore, wet deposition through rainwater is not necessarily the most important or the most dominant way for the delivery of Hg to terrestrial systems [R. P. Mason et al. (1994) *Geochimica et Cosmochimica Acta*, vol. 58, 3191-3198; C. H. Lamborg et al. (2002) *Geochimica et Cosmochimica Acta*, vol. 66, 1105-1118].

[12] D. P. Kranbbenhoft et al. (1999) U.S.G.S. Toxic Substances Hydrology Program Water Resources Investigation Report 99-4018B, volume 2. Available online at http://toxics.usgs.gov/pubs/wri99-4018/Volume2/sectionB/2301_Krabbenhoft/. See also W. G. Brumbaugh et al. (2001) A national pilot study of mercury contamination of aquatic ecosystems along multiple gradients: Bioaccumulation in fish, USGS/BRD/BSR-2001-0009, iii + 25 pp.

[13] D. P. Kranbbenhoft et al. (1999) U.S.G.S. Toxic Substances Hydrology Program Water Resources Investigation Report 99-4018B, volume 2. Available online at http://toxics.usgs.gov/pubs/wri99-4018/Volume2/sectionB/2301_Krabbenhoft/

[14] J. J. Rytuba (2003) *Environmental Geology*, vol. 43, 326-338; M. S. Gustin (2003) *The Science of the Total Environment*, vol. 304, 153-167. The geological factor is not only predominant for the contamination level of mercury in the natural watersheds, but the geological sources can also emit significantly to contribute to atmospheric mercury. Gustin (2003) found that "[a]rea emissions [based on careful geological field data collected for Nevada] were estimated to be ~3 times higher than the value used in global models, and annual emissions are ~1/5th of the total coal fired utility broiler emissions for the US (Chu, 2000). Natural source emissions in Nevada are approximately equal to the current anthropogenic emission estimate for the state, which are derived from gold ore processing and coal combustion." (p. 164)

[15] http://nadp.sws.uiuc.edu/nadpdata/mdnsites.asp

[16] R. P. Mason et al. (1999) *Marine Chemistry*, vol. 65, 77-96.

[17] P. Pickhardt et al. (2002) *Proceedings of the National Academy of Sciences*, vol. 99, 4419-4423.
[18] From p. 266 of M. C. Marvin-DiPasquale et al. (2003) *Environmental Geology*, vol. 43, 260-267.
[19] Marvin DiPasquale et al. (2003) said: "marsh sediments located around the periphery of the bay appear to be the most active zone for net MeHg production, presumably because of both the organic-rich nature of these zones, and the ability of rooted macrophytes [bottom water's large plant life] to supply molecular oxygen at depth in the sediment, thus keeping reduced-S levels low." They continued, "This has important implications for the wetland reconstruction proposed for this area. Without sufficient safeguards, substantial wetland reclamation efforts may result in an overall increase in MeHg production in these newly constructed wetlands, with net MeHg export to the larger bay." In other words, *reclamation of wetlands may have to contend with potential complications of enhanced MeHg production and the ultimate transport elsewhere.*
[20] S. J. Balogh et al. (2003) *The Science of the Total Environment*, vol. 304, 305-313.