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How mercury flows downstream

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Abstract
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Researchers from the U.S. Geological Survey (USGS) have published a suite of papers in *ES&T* that elucidate the availability of mercury in streams and how it makes its way into fish and other life-forms in these dynamic ecosystems. The three papers (DOI 10.1021/es802694n, 10.1021/es802698v, and 10.1021/es8027567) represent one of the most comprehensive studies of stream mercury dynamics, and they hint at answers to many of the questions raised by previous research.

Although other research has looked at mercury in stream systems, most studies of the formation of methylmercury and how it gets into fish have been done on lakes, a much more placid environment. This new suite of papers summarizes results from a multiyear study of eight U.S. streams in three states representing different geographic zones Florida, Oregon, and Wisconsin as well as urban and less developed settings.

Led by Mark Brigham, a geochemist at USGS who is lead author of one of the papers, the researchers tracked proxies for mercury availability and methylation, such as dissolved organic carbon and sulfur levels in sediments. They also looked at mercury concentrations in different predator fish by moving up the food chain from algae to the insects and small fish the predators eat.

Over a 4-year period, the researchers sampled one urban stream and one or two less developed streams in each state. They took water samples near stream gauges to assess the variability of flow and water inputs, and they took sediment samples throughout each stream’s course. They

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also collected enough fish and insects of different species and in various seasons to represent an entire food web for each stream. For all of these samples, the researchers measured total mercury, methylmercury, dissolved organic carbon, and other markers. For biota, they tracked C and N isotopes to find the life-forms' places in the food chain.

Each of the papers points to three key findings. First, the entire catchment (all the spots that supply water to the stream) is critical in determining mercury and methylmercury flux to the streams; this is particularly true in catchments with more wetlands. Second, the availability of carbon and mercury and the setting in which they are available to bacteria that methylate mercury are important. However, methylmercury concentrations in sediments may not match concentrations present in the water flowing above, which highlights the importance of upstream inputs. And third, what's in the water seems to have the biggest impact on the levels of mercury that appear in higher trophic levels, from invertebrates to midlevel and predator fish.

"That supply of methylmercury to the base of the food web is going to be the driver to what you see in the top predator fish," says Lia Chasar of USGS, a coauthor of one of the papers. And "it's not just supply, but how available it is to the critters." That availability varies with the different watershed characteristics and settings that the team studied. For example, "not all DOC [dissolved organic carbon] is the same," she says, and carbon availability in a stream in Florida will be different from that of a stream in Oregon.

The findings do not change the impact of mercury deposited in an ecosystem: more mercury delivered to a stream or lake means that more mercury ends up in fish. To clarify that process, this project determined which variables control the differences in the mercury levels in fish across systems that receive their mercury via atmospheric deposition, says Edward Swain of the Minnesota Pollution Control Agency. The team "identifies landscapes that are sensitive, or more vulnerable, to mercury deposition," Swain adds.

"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," comments Brigham. "Wetlands abundance in a watershed keeps on popping up as an important factor," he says. The team found that although mercury concentrations are magnified with each step up the food web, the transfer of mercury through each trophic level occurs at a fairly consistent rate across streams.

"Not one of these [pieces of the system] is the dominating factor, but each one of these is important," says Christopher Knightes, a researcher and mercury modeler at the U.S. EPA's National Exposure Research Laboratory in Athens, Ga. "What we're ultimately concerned about is methylmercury in the fish."

The project is “an empirical correlation study, with the strength of covering many different systems and doing all the statistical work,” Knightes continues. “It doesn’t tell you the cause, but tells you the correlations” of where and how mercury will be present and where it will bioaccumulate in fish. It sets the stage, he says, for future mechanistic work to explain exactly what happens in this relatively new arena. These data and correlations also will inform the models that EPA continues to develop for the total maximum daily load program, related to mercury in streams.

“The strength of this study is that they’ve got a lot of data from a lot of sites, and the data are integrated” across those streams, comments Charles Driscoll of Syracuse University. The team also took care to choose streams that did not receive mercury directly, for example, from mining waste. Only mercury carried by air or rain enters these streams or is deposited in their catchment areas. The study confirms what has been reported in bits and pieces before: “Deposition is important, but it’s not an overriding factor. The landscape characteristics are really determining mercury transport and chemistry” in streams, Driscoll says.

To control mercury levels in fish, ideally there must be a way to control mercury emissions coming from coal-fired power plants, smelters, and other sources, Driscoll says. Yet several mitigating factors are also at work, as shown by the differences the USGS team found in urban versus nonurban streams. Most people would expect more mercury in fish in urban streams, but the new research indicates that that’s not the case. For example, in urban streams, mercury isn’t available to biota because of the relatively high sulfur and sulfate levels, which seem to bind up the available mercury, as indicated in the paper by Mark Marvin-DiPasquale et al.

By pinpointing the conditions of an ecosystem that could make it sensitive to mercury cycles, researchers could influence policy making related to mercury sources such as power plants. In February, the U.S. Supreme Court declined to hear a case about EPA’s Clean Air Mercury Rule, which was finalized in 2005. This decision allows EPA Administrator Lisa Jackson to consider new mercury-control strategies and standards to protect the environment. “The results from this project are timely and relevant for policy makers at U.S. EPA,” points out USGS team member David Krabbenhoft, a coauthor of two of the papers.

The USGS scientists took a truly “biogeochemical approach to seeing why certain streams have higher levels of mercury in them, where it is being produced, and how it’s being accumulated in the food web,” says Vincent St. Louis, a biologist at the University of Alberta (Canada). “This kind of [research] has long been needed. The focus has been on lakes all these years and on oceans, and not a lot of good broad-scale studies have been done on streams. [The three papers are] a strong body of evidence that people will be referencing for a long time.”

St. Mary’s River in Florida is one of eight streams across the U.S. included in a sweeping study of the biogeochemistry of mercury in streams.
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