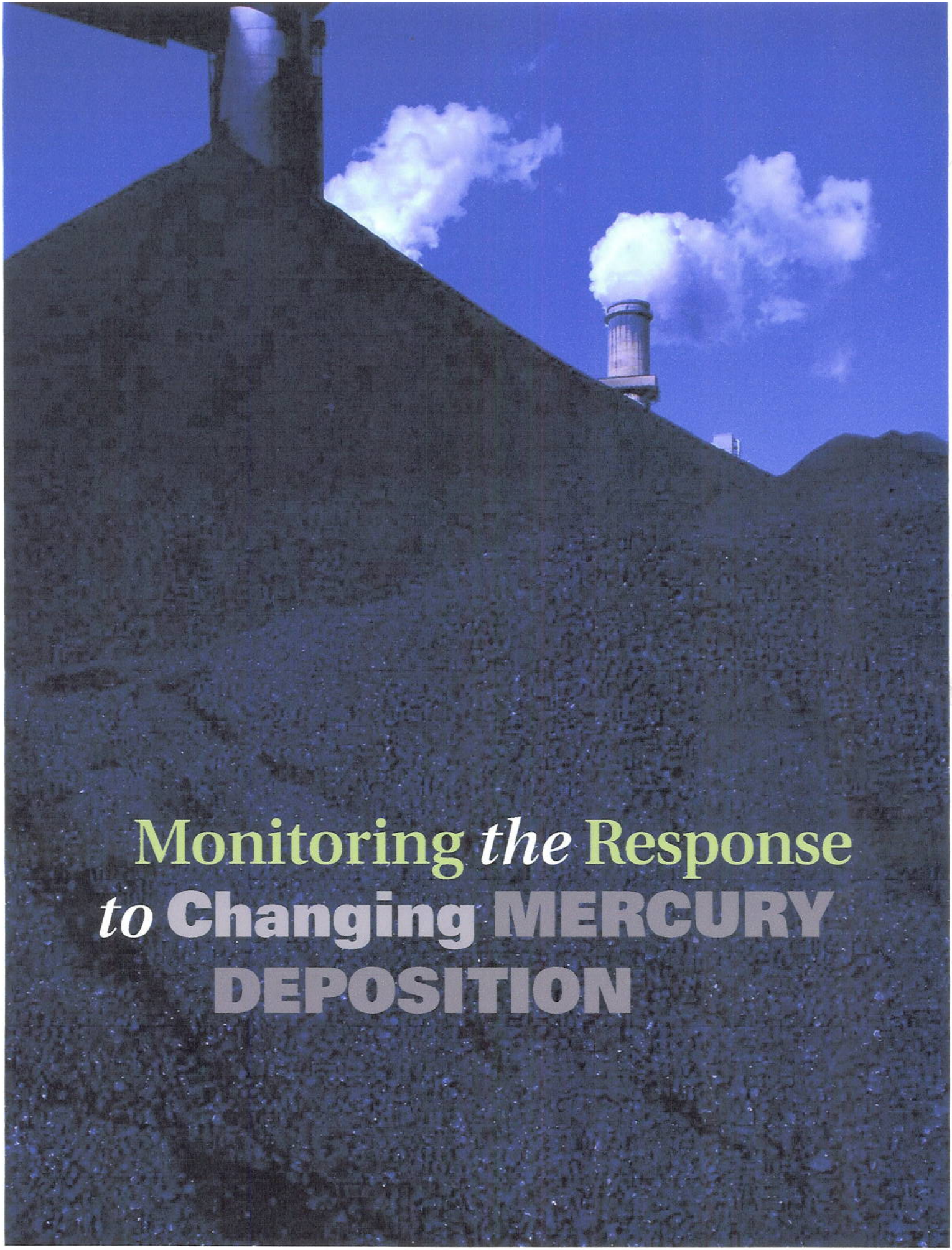

Monitoring *the* Response *to* Changing MERCURY DEPOSITION

Scientists and policy makers need a cohesive framework to evaluate the effectiveness of regulations on mercury emissions in the United States and Canada.

Because advisories have been posted for so many water bodies against consumption of fish with elevated concentrations of potentially dangerous methylmercury (MeHg), regulations limiting mercury emissions have been promulgated in many countries or are likely to be put forward in the near future (*1*–*5*). Yet, many questions about the environmental benefits of emissions reductions remain unanswered. Current computer models and other assessment tools provide widely divergent estimates for the effectiveness of emissions controls at reducing MeHg levels in fish (*6*–*8*). In addition, no broad-scale data sets are available to test model predictions. Some intensive studies and syntheses of regional databases have been conducted, but their overall applicability to different ecosystems or at the continental scale is uncertain.

The problem is that the terrestrial–aquatic mercury cycle is complex, with many nonlinear processes that link atmospheric mercury emissions and MeHg bioaccumulation in fish and wildlife (*7*; Figure 1). As a result, how effective emissions reductions will be in decreasing biotic MeHg levels in freshwater, estuarine, and coastal ecosystems is not clear. Thus, any changes in the MeHg levels in aquatic ecosystems, particularly in fish and wildlife populations, should be documented and compared with reductions in mercury emissions and deposition. Although a significant effort has been made over the past decade to understand the causal link between mercury emissions and MeHg bioaccumulation into aquatic food chains, currently no coherent monitoring or assessment framework exists that can quantitatively document the temporal environmental changes in mercury levels across ecosystems. Clearly, it is crucial for scientists and policy makers to develop a monitoring framework that can accurately evaluate the effectiveness of current and pending regulation. This paper proposes such a framework.

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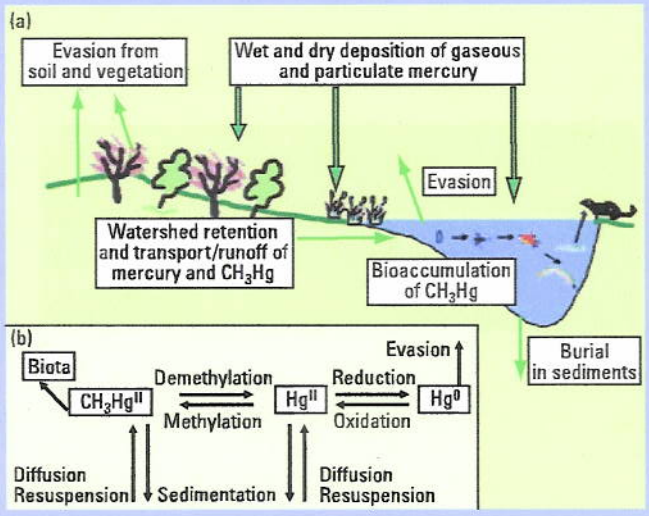
The monitoring strategy outlined here was developed in September 2003, by 32 mercury scientists from academia, industry, government, and nonprofit organizations in the United States, Canada, and Europe who were gathered in Pensacola, Fla., for a Society of Environmental Toxicology and Chemistry (SETAC)-sponsored workshop. The workshop was charged with identifying suitable chemical and biological indicators of mercury in the environment and proposing a network for measuring and documenting changes resulting from reductions in mercury emissions in the United States (*10*). The design of that kind of program is complicated by uncertainties, such as the sources of mercury deposition at any specific location and the sensitivity of different watersheds and water bodies to mercury input, specifically in terms of its rate of conversion to MeHg and its subsequent bioaccumulation. A holistic, multimedia, long-term monitoring approach is needed to detect change across such a diverse and complex system. Ideally, the program would begin immediately to establish a baseline and would continue for 15–20 years.

FIGURE 1

Major routes into the environment

(a) Mercury can take several routes into ecosystems, including wet and dry deposition. (b) Most methylation of mercury takes place in aquatic systems.



Primer on mercury biogeochemical cycling

Figure 1 indicates the links between mercury deposition and MeHg bioaccumulation in aquatic food chains. Atmospheric mercury occurs in two forms in the gas phase: elemental mercury (Hg^0), which is the dominant form, and ionic Hg^{II} species, which are collectively termed reactive gaseous mercury (RGHg). This is an operational definition because of the limitations of currently available measurement techniques (*11*, *12*). The dominant component of aerosol mercury (HgP) and of both dissolved and particulate matter in the aqueous phase is Hg^{II}. With a lifetime of hours to days, RGHg is rapidly removed from the atmosphere, whereas less-reactive Hg^0 has a residence time of up to a year (*7*, *13*). Uptake of Hg^0 by vegetation is now considered an important deposition mechanism.

The different forms of mercury have varying reactivity following deposition. Dissolved Hg^{II} species can be reduced to Hg^0 in surface waters. If waters become saturated with Hg^0, then a substantial fraction is returned to the atmosphere via evasion (*13*). Reduction and re-emission also occur in terrestrial ecosystems.

Although the time scale is not well determined, the route of mercury through watersheds to the aquatic system is thought to be slow and convoluted (*14*). It has been estimated that only a relatively small amount of the total input of mercury to most watersheds, typically <20%, is transported to the associated aquatic system. However, for aquatic systems with large watersheds, this input is greater than the mercury directly deposited to the water surface. The bioavailability of mercury supplied from the watershed to methylating bacteria and its potential for reduction to Hg^0 are poorly understood (*15*).

The mechanics of mercury transport through terrestrial watersheds and the cycling of the metal within freshwater, estuarine, and coastal systems determine the extent to which it is transported to sites of mercury methylation (*15*, *16*). Such sites are typically in the upper reaches of water-saturated but anoxic zones, such as sediments and wetlands. In both marine and freshwater ecosystems, the microbial communities responsible for mercury methylation are primarily sulfate-reducing bacteria.

Many chemical, biological, and physical factors, such as bacterial community structure, pH, redox status, and nutrient and sulfate concentrations, influence MeHg production, but mercury supply is obviously an important variable (*16*). Atmospheric deposition of sulfate from anthropogenic sources to sulfate-depleted environments, such as temperate lakes, has most likely exacerbated the degree of mercury methylation and bioaccumulation (*17*). Eutrophication and other disturbances have also affected the extent of mercury methylation (*15*). Observed changes in MeHg concentration and net mercury methylation may be among the most sensitive indicators of change within aquatic ecosystems, but their relationship to mercury deposition is complex (*16*). Because MeHg can be demethylated to inorganic mercury biotically in sediments and abiotically in surface waters, rapid cycling occurs between the mercury and MeHg pools. Measured concentrations therefore represent short-term, steady-state standing stocks.

It is not clear whether changes in mercury input will result in a linear change in mercury methylation. Computer models, such as one developed for the Florida Everglades (*8*), tend to predict a linear response, but there are little data to support the predictions. On the other hand, preliminary results from an ongoing study in Canada called METAALICUS demonstrate that mercury entering the surface water directly from the atmosphere is more likely to be methylated, with the probability decreasing over time

(*18*). Unfortunately, this study has not been under way for sufficient time to assess the long-term methylation potential of mercury deposited to the terrestrial watershed and its subsequent bioaccumulation.

Given the complexities in mercury cycling, no simulation models currently exist that can accurately predict the response of MeHg concentrations in the various compartments of terrestrial and aquatic ecosystems to changes in mercury loading rates (*6*, *8*, *18*). Simulation models, developed from earlier conceptual models, have only been calibrated and tested against field data from a few sites and are therefore of limited use in predicting the response in more diverse ecosystems. Further model development requires collecting specific field data for validation. Clearly, a critical need is the development of models that can reasonably simulate mercury cycling and concentrations at diverse sites on the basis of site characteristics and external mercury loads.

Considerations in study design

Tracking and detecting changes in atmospheric mercury deposition relative to reductions in emissions from anthropogenic sources can be complex; lifetimes, source profiles, and transformations of the various mercury species differ, and measuring these components is difficult (*6–8*). The concentration and reactivity of atmospheric oxidants influence atmospheric transformations (*6–8*, *13*, *19*). Also, detection of the response to changing anthropogenic mercury emissions in the continental United States will be confounded by variations in natural emissions, the rate of re-emission of previously deposited mercury, and mercury emissions from other countries (*6*, *7*, *13*). Thus, any framework for monitoring atmospheric wet and dry deposition must account for the contribution of all forms of atmospheric mercury from all sources.

The time scale of the ecosystem response to mercury emissions reductions is unknown. As a result, the metrics used to evaluate change must include indicators that respond on varying time scales. For example, whole-body MeHg concentrations in upper-trophic-level organisms change relatively slowly—over time scales on the order of months to years, as MeHg is depurated and, in some organisms, demethylated. Most MeHg resides in muscle tissue. Lower-trophic-level organisms, such as zooplankton, respond rapidly to changes in water-column MeHg concentration (*20*). However, such changes may be transient or responsive to short-term variability of other factors rather than to longer-term changes in mercury input (*20*).

Clearly, mercury monitoring indicators must be chosen to reduce the confounding impacts of short-term variability while integrating the signal so that any change can be ascertained. To assess longer-term trends, the monitoring program will need to be maintained for 15–20 years. Additionally, baseline data are needed so that future changes can be detected; therefore, the monitoring program must be instituted as soon as possible to ensure adequate background information.

However, decision makers need more than mercury concentrations to be able to ensure the defensible interpretation of the indicators, such as MeHg concentrations in fish. Other necessary information includes land use; food-web structure; the introduction of exotic species; point-source discharges; changes in climate, atmospheric chemistry, and acidic deposition; in situ chemical and physical properties; and hydrological regimes (e.g., retention time and water level fluctuation). These factors could affect freshwater differently than coastal environments. A broad-based strategy is also needed to select sampling locations that quantify the effects of local point-source and regional atmospheric mercury emissions as well as provide a generalized assessment for the whole continent. Clearly, while the environmental settings (e.g., water-body type, geographic location) closest to mercury emission sources need to be examined, less-responsive, remote environments also should be monitored. Ecosystem response will depend on the relative amounts of wet versus dry mercury deposition and direct versus watershed mercury sources. Each ecosystem is unique, and our ability to predict and document trends in mercury concentration in indicators will depend on our understanding of the factors that influence the metal's biogeochemical cycling.

Although the proposed design is primarily a data-collection program, researchers also need models to extrapolate among monitoring sites, interpret data, and critically examine the response of indicators to changes in atmospheric mercury deposition. Aquatic ecosystem models will be used to test anticipated changes against the observed response and to ascertain the magnitude of the response due to variations in mercury deposition compared with other confounding factors. In addition, models can predict the spatial and temporal patterns of mercury concentrations and fluxes under various future scenarios. The models would determine the relative contributions of various mercury sources over time and estimate the likely mercury attenuation trajectory and time to recovery under different environmental scenarios. Other factors, such as sulfate and organic matter that impact bacterial activity, could also possibly cause an increase in fish mercury concentration even as atmospheric deposition decreases (*16*). Such scenarios could be tested with the aid of a mechanistic model.

The proposed network design should be at least national and preferably continental in scale.

The proposed network design should be at least national and preferably continental in scale and include a range of long-term monitoring locations (cluster sites) across different ecosystems as well as selected study sites that are more intensively monitored (intensive sites). Although the United States

TABLE 1

Types of indicators for cluster or intensive sites

Indicator	Site	Frequency
Air and watershed		
Atmospheric mercury speciation; wet and dry deposition flux[a]	IN	C
Weekly wet deposition and flux	CL	W
Hg^0 evasion/flux[a]	IN	M
Watershed yield (surface-water and groundwater flux)	IN	M
Chemical characterization		
Historic sediment depth profile[b]	IN	I
Hg^0, MeHg, and %MeHg in surface (0–2 cm) sediment	CL	S
Hg^0 and MeHg in surface water	CL	S
Hg^0 and MeHg water-column profiles	IN	S
Aquatic biota		
Phytoplankton and algae	IN	M
Zooplankton/benthic invertebrates	IN	M
Yearling fish	CL	S
Piscivorous/commercial fish	CL	A
Wildlife[c]	CL	A

Site: IN = intensive sites only; CL = cluster sites and intensive sites.
Frequency of sampling: C = continuously; W = weekly; M = monthly; S = every 6 months; A = annually; I = every 3–5 years.
[a]Event-based wet-deposition collection at intensive sites, weekly integrated sampling at cluster sites. At intensive sites, flux estimates would include wet, dry, gaseous, and particulate deposition; throughfall and litterfall; and snowpack sampling, as appropriate. Hg^0 concentration and evasion fluxes would be for both aquatic and terrestrial environments.
[b]Intensive sites and a subset of cluster sites would be sampled to determine historic mercury trends.
[c]Birds, small and larger mammals; both short-term and integrative sampling.

consists of 4 ecosystem domains (polar, desert, humid temperate, and humid tropical) and 14 divisions (*21*), the continent could be subdivided into <10 "ecoregions" for this proposed monitoring program. That means placing up to 20 cluster sites per ecoregion; sites are grouped based on similar atmospheric loads (or load reductions) but are randomly chosen within each ecoregion.

At these sites, the primary indicators would be measured over a prolonged period of time (Table 1). Individual sites within a cluster would have similar ecological characteristics (e.g., southeastern coastal-plain streams) but probably different site characteristics (e.g., pH, dissolved organic carbon [DOC] concentration, acid-neutralizing capacity, and watershed/water-body ratio). Selection criteria for cluster sites would be based on multiple factors, such as watershed and water-body type, and would represent remote and impacted sites, dry regions, and saline waters, as well as a wide range of ecosystem types, potential exposure "hot spots", and mercury loading rates.

More continuous, multimedia monitoring of both changes in mercury loading and MeHg assimilation into biota would be conducted at fewer (≤10) intensive sites. Sites where change is expected would be emphasized, although background sites must also be monitored. To take advantage of existing resources, these sites should be established quickly in conjunction with current wet-deposition stations and/or ecosystem study sites and should include detailed atmospheric, watershed, aquatic, and biota sampling. Where possible, priority should be given to sites with intensive ongoing monitoring programs that currently do not collect mercury data, such as the Long Term Ecological Research Network. Ongoing collaborations with other efforts that meet multiple needs (e.g., global change, urban sprawl, and changing land-use issues) should also be sought.

Indicators

What are suitable criteria for choosing indicators? They must be comparable across ecosystems; able to integrate variability in space and time; relatively simple to interpret; either easy to sample, process, and quantify analytically or already measured or part of an existing database; responsive to mercury loading on a relatively short time scale; able to be tied to changes in MeHg production; and theoretically and empirically sound. Appropriate indicators should reflect changes in exposure to humans and wildlife as well. Such criteria determine the relative value of each metric within any study design, given the need for a balance between financial resources and scientific rationale.

Airshed and watershed. Wet-deposition measurements are relatively easy to accomplish and are currently monitored at the national level through the Mercury Deposition Network, which is shown in Figure 2 (*22*). However, this weekly collection program has limitations in its geographical coverage and does not provide data suitable for some computer model simulations. Even given these concerns, the workshop's consensus was to recommend a widely distributed weekly mercury wet-deposition monitoring program at the cluster sites (Table 1). At intensive sites, event-based wet-deposition collection was the consensus recommendation.

The intensive sites would measure mercury atmospheric speciation. Atmospheric Hg^0 concentration strongly reflects the global atmospheric mercury pool and does not necessarily provide a sensitive local indicator of short-term regional change (*23*). In contrast, the concentrations of RGHg and HgP show a higher regional variability and will respond rapidly to changes in emissions because these species have a relatively short residence time in the atmosphere, are easier to control at the emission source, and have a strong anthropogenic signal (*6, 7, 19, 24*). They are, however, relatively difficult to measure, and they form in situ via atmospheric chemical reactions (*6, 11–13, 24*). Current estimates of mercury dry deposition remain highly uncertain (*6, 7*), but given the importance of the dry deposition flux, methods for the measurement of atmospheric mercury speciation and dry deposition will need to be standardized and rigorously calibrated for this program.

Measurements of atmospheric mercury speciation at intensive sites would be coupled with estimates of deposition and ecosystem fluxes, including litterfall and throughfall, as well as measurements of atmospheric ozone and nitrogen oxides, sulfate, major ions

in precipitation, and other atmospheric compounds. Such sites would generate data in support of regional- and global-scale atmospheric modeling efforts and specifically provide information for estimates of dry deposition to complex surfaces, such as forests. Soil, groundwater, and surface-water measurements of mercury speciation would also be made to examine the role of air–surface exchange of Hg^0 in impacting mercury transport to methylation sites.

Well-chosen aquatic systems can accurately determine changes in mercury deposition over time.

Mercury export from watersheds is typically a small fraction of the yearly input from the atmosphere (*14*). Export is also influenced to some extent by changes in mercury input, although the response time is very slow. Other disturbances, such as changes in land use, can create larger responses in a shorter time. Rainfall amount and other climatic variables also influence export, especially with respect to sporadic and extreme events (*13*). Numerous variables not directly related to short-term changes in atmospheric mercury input similarly influence export of MeHg (*14*). Thus, export fluxes are not good indicators for monitoring short-term changes in atmospheric mercury, but they should be examined at intensive sites. Given the complications associated with the interpretation of data on export from land, both intensive and cluster sites should include water bodies with little or no watershed.

In-lake chemistry. Changes in atmospheric deposition are recorded by the mercury concentration gradient in sediments, peat bogs, and glacial ice. Therefore, carefully selected cores are appropriate trend indicators because they smooth short-term variations in mercury deposition and integrate spatial variability (*25*). A large body of experimental and observational evidence vouches for their reliability, and well-established protocols exist for collecting, processing, and interpreting sediment-core records (*25, 26*). Well-chosen aquatic systems can accurately determine changes in mercury deposition over time, despite the influence of watershed input, sediment mixing, and other potentially confounding factors. However, because of the rate of surface sediment mixing relative to sediment accumulation, sediment cores cannot resolve changes at intervals <5 yr. Estimated accumulation rates could be matched with information from atmospheric deposition monitoring at co-located sites. Cores should be collected at the intensive sites and a subset of the cluster sites.

Although the relationship between biota and sediment mercury and MeHg levels is difficult to con-

FIGURE 2

National Atmospheric Deposition Program

In total, the Mercury Deposition Network includes 97 wet-deposition monitoring sites in the United States and Canada.

struct (*27*), sediment MeHg data provide an integrative measure of the impact of changes in mercury input and other factors on net MeHg production (*16*). Thus, total mercury and MeHg should be measured in surface sediments (0–2 cm) at all sites (Table 1). Because of the relationship between short-term, assay-determined methylation rates and the in situ MeHg sediment concentration in numerous ecosystems (*16*), these difficult assays are only recommended for the intensive sites. Researchers have proposed that sediment MeHg concentration and %MeHg are the best indicators of in-lake changes of bulk MeHg concentration (*16*). The %MeHg measurement will determine whether change is directly or indirectly related to variations in atmospheric mercury input. Because of large pools of sediment mercury and other factors, the total sediment value integrates the signal of mercury deposition over a period of several years in most ecosystems.

Total mercury and MeHg measurements in water or in the dissolved and particulate fractions have been made in many ecosystems to date, and these indicators are recommended for all sites; however, interpreting the response of these measurements to changes in mercury input may be difficult (*9, 16, 27, 28*). Water concentrations can be influenced by factors unrelated to mercury inputs, such as the variation in DOC, particulate matter, and particulate organic carbon concentrations (*17, 28, 29*). However, in a number of locations, primarily those with a dominantly pelagic food web, studies have shown a reasonable correlation between MeHg in water and MeHg in fish; this reflects the changes occurring at the base of the food chain (*27, 30*). In addition, recent studies in the northeastern United States have demonstrated that water mercury levels can be related to population-level impacts in wildlife (*31*).

Thus, measuring total mercury and MeHg in water at the cluster sites is recommended, because samples can easily be collected and the analytical techniques are well established. Water concentrations vary seasonally and with depth, so these indicators must be measured seasonally at the cluster sites (including during summer stratification) to characterize the anticipated spatial variability. Depth-integrated sampling of the water column would only be done at the intensive sites.

Aquatic biota. Yearling fish are the best indicator of short-term MeHg change in the food chain (*30*). Most yearlings feed on invertebrates and have a relatively limited dietary range, thus they provide a comparatively consistent, interannual indicator. Despite seasonal variation, a strong relationship exists between the MeHg concentrations in yearlings and in piscivorous fish. All the yearling fish should be sampled in the same season to avoid any seasonal variation. These smaller fish are easily sampled, and this practice affects the ecosystem less than collecting larger fish.

Monitoring of piscivorous fish, especially those that are recreationally or commercially important, is also recommended, even though these organisms may take 3–5 years to respond to changes in MeHg bioavailability (*30*). Because fish MeHg concentration increases with age, data must be normalized. Ancillary information on fish length, weight, sex, and age is required to provide a more statistically defensible, normalized MeHg value. Other factors, such as nutrient input, watershed land use change, fluctuations in water levels in shallow ecosystems, overfishing, changes in food chain structure, and variations in species competition, can also alter fish MeHg concentration (*9, 27, 30*). Because MeHg is the dominant form in fish, measurement of total mercury is an adequate metric.

Substantial information is already available on piscivorous fish muscle mercury concentrations across ecosystems because of fish consumption advisory programs. Large and growing databases are available, for example, the U.S. EPA's National Fish Tissue Study, which involves a coordinated random sampling strategy for mercury and other chemicals in fish (*32*). Such studies increasingly record both mercury concentration and the necessary ancillary information, such as weight and length, and are therefore useful benchmarks for assessing long-term changes in fish concentration (*30*).

Sampling phytoplankton, periphyton, or zooplankton is not recommended for the cluster sites. Although zooplankton are an important trophic link (*30*), they respond to changes within days to months, and the population consists of a complex mix of organisms that varies spatially and temporally within and across ecosystems (*33*). Additionally, MeHg concentrations vary seasonally, and the fraction of the total mercury as MeHg varies between species. Some benthic invertebrates within freshwater systems, such as crayfish, are potential indicators because they are ubiquitous, live for multiple years, and have a small home range (*34*). In estuarine and coastal environments, crustaceans and bivalves are good candidates for monitoring. Here too, other factors, such as organic matter content, may obscure the relationship between sediment MeHg and MeHg in benthic invertebrates (*27*), reducing their effectiveness as monitors.

Periodic sampling of selected species would be critical for evaluating long-term bioaccumulation.

Wildlife. These indicators should be chosen on the basis of the criteria previously outlined as well as how well they describe pathways of MeHg biomagnification to different trophic levels and within individuals over time. Difficulties in field sampling, narrow distribution of some species, and the lack of data limit the use of some potential indicators (*35*). For birds and herpetofauna, nonlethal sampling strategies (e.g., feathers, scales, blood, and abandoned eggs [*36*]) are common, whereas in mammals both lethal and nonlethal sampling strategies (e.g., organs and blood, respectively) are used.

Although researchers have conducted fewer mercury exposure studies in amphibians, controlled approaches have included whole-body analysis (*37*). Blood is the best matrix for understanding short-term mercury dietary uptake, whereas keratinous materials are generally good indicators of longer-term dietary uptake (*35, 38*). Therefore, the sampling of one individual can provide both short- and long-term information on mercury uptake. The mercury in blood, keratinous material, and eggs is nearly all (>95%) MeHg, and thus analysis of total mercury is again an adequate metric.

Because the primary objective for this monitoring framework is to track an aquatic-based MeHg signal, the preferred species will have a strong aquatic link, maintain small home ranges, and be ubiquitous across ecosystems. Typically, these are species that forage primarily on aquatic prey. Spatiotemporal comparisons of wildlife MeHg concentration require standardization of species, age, and sometimes sex, season, tissue, and habitat types (*35, 38*). Although most field efforts have concentrated on piscivorous wildlife, recent research indicates that insectivorous birds can also bioaccumulate MeHg (*38*).

Many piscivorous species have been chosen as indicators because of their well-accepted attributes, such as logistical feasibility, high public value, and conservation needs. For example, significant research has been conducted on continental patterns of MeHg availability using the common loon (*36, 38*); terns, gulls, and other fish-eating species were used for regional studies (*4, 39, 40*). Current evaluations of indicator candidate species also target insectivorous birds, including swallow and sparrow species in freshwater wetlands and salt-marsh sparrows and rails in estuaries, and freshwater mammals such as river otter and mink. Seabirds and marine mammals have been used as larger-scale spatial integrators and provide insight into local, regional, and global mercury signals (*41, 42*).

Migration or dispersal are not necessarily confounding factors for wildlife because blood and prey mercury levels reach a rapid equilibrium. However, intraseasonal movements can make a difference in site-specific mercury characterizations; therefore, species that tend to use multiple water bodies within their territory are poor indicators. Although the analysis of certain tissues of long-lived biota is unsuitable for assessing short-term impacts, periodic sampling of selected species would be critical for evaluating long-term bioaccumulation (*42*).

Change is already occurring, so this program should be initiated as soon as possible. Results from monitoring wet deposition and fish concentration are considered the primary indicators for detecting change. Implementing the proposed framework would reveal whether change is occurring in atmospheric mercury input and how this change is reflected in biota MeHg concentrations across various aquatic ecosystems.

Change is already occurring, so this program should be initiated as soon as possible.

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