DRAFT

A Long-term Passive Gaseous Mercury Monitoring Network: A New Initiative for the National Atmospheric Deposition Program

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1.0 Initiative Description

1.1 Advocates

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1.2 Objectives

The singular goal of this initiative is to begin a new network, to work alongside the MDN, AMNet, and Litterfall networks that measures monthly average Gaseous Mercury (GM) using a passive sampler which can be easily operated across the U.S. and international locations. This network would provide a base network for AMNet and companion to MDN, along with the potential to provide average dry deposition estimates for all site locations.

1.3 Duration

We propose that this network would operate indefinitely, as long and the information is needed and necessary.

1.4 Justification

This initiative can be justified in a number of ways and directly supports the mission of the NADP.

Relevance to Mission: The mission of the NADP, in summary, is:

The NADP monitors the nation's precipitation and the atmosphere for a range of chemical constituents, to determine the rate or flux of atmospheric pollutants to the biosphere. Ultimately, with continued measurement, these networks will provide the data necessary to determine whether spatial

and temporal trends in concentration and wet/dry deposition exist, understand the changes in direction and magnitude of chemical deposition, and understand the impacts on agricultural systems.

From the most recent National Institute of Food and Agriculture annual report, the specific goals of NADP are:

- 1. make the best measurements of atmospheric and precipitation chemical condition possible;
- 2. use standard methods and procedures to ensure that the measurements are made with the utmost *quality;*
- 3. make these measurements freely available to all users, with a particular emphasis on the research community and educators; and
- 4. strive to advance environmental measurement science through discussion, testing of new methods, assisting others making similar measurements, general outreach, and data accessibility.

We feel that this initiative directly supports the mission of making scientifically accurate measurements of Hg gas and can be used to estimate dry deposition. Therefore, this network supports the AMNet, MDN, and Litterfall networks, and makes this type of measurement available to more locations due to its low cost. It expands upon the goals of the AMNet and supports the NADP through additional site locations and more stations, etc. We feel that the methods proposed are proven through scientific review and approval, are readily available now, and will stimulate measurement and discussion. It will also fill a global need. With all of this, this initiation meets the mission of NADP and all of its goals.

This new initiative, we feel, can easily be justified in many different ways. A few of those follow here.

- This initiative supports the goals of MELD to extend the monitoring of mercury deposition (along with MDN, AMNet, and Litterfall) in new ways. Recently, a MELD workshop facilitated the evaluation of available gaseous Hg methods, beyond the AMNet Tekran system. The MMET recommended MerPAS for a long-term large network and its readiness.
- This initiative supports the budgetary needs of NADP, by providing alternative measurements to the expensive AMNet equipment, and allowing more groups without this equipment and technical expertise to monitor gaseous mercury.
- If it is scientifically valid to estimate dry deposition of mercury using longer-term measurements (monthly), then this network will allow for additional dry

deposition estimates to be made at more than the 18 AMNet site locations, which supports the goals of TDEP.

• This initiative would support the United States' response to the Minamata Convention and its call for more atmospheric Hg monitoring in a coordinated manner. This method is being used by Canadian researchers (early in the program) for a trial global network. It is also being considered for use in the APMMN, and likely other parts of the globe. Consistent measurement would go directly to the Minamata call.

2.0 Operating Protocols

2.1 Field Operations

We propose a MerPAS-based network for NADP, operating on a monthly time scale.

<u>The Sampling Device</u>: The sampling device is a passive measurement for

GM, developed by a research group at the University of Toronto^{1,2,3,4,5} with support from Environment and Climate Change Canada. The device is also produced commercially by Tekran Instruments Corp, and sold as the "MerPAS" (see figure). The passive device is very similar to the Radiello sampling device used in the AMON. The MerPAS device includes three major parts; 1) a polyethylene terephthalate (PET) jar shield, 2) a



diffusive surface

adsorbing

surface

Radiello sample body/housing¹, and 3) a sample core. Like most passive air samplers, the sample body is porous and allows gases to move from free air outside the body to the inside the body. It also provides a quiescent space for GM to diffuse to the core over this space. The core material is housed in a metal mesh core container (60 mm length, 5.8 mm diameter) which is filled with a mixture of commercially available, bituminous coal-derived, sulfur-impregnated activated carbon (AC, Calgon Carbon Corp, Product #2300). The GM adsorbs to the AC inside the sampler body, and is stable over time. The Radiello core and body slip-fit together and include an endcap to make the sampling device. The device screws into the precipitation shield which keeps the device dry. The

¹ The sampler body is a Radiello white sampler body, where AMON uses the Radiello blue body.

shield comes with a wall mount to attach to a wall, fencepost, etc. for sampling. The shield is closed with a PET jar lid for storage and transport to and from the sites.

Justification of the MerPAS use, rather than other samplers in the literature is based on four reasons.

- the McLagan et al/MerPAS has been well reviewed in the literature (references 1-5), and this method has shown to be accurate in many different environments and at the atmospheric concentration levels that we need.
- the method is currently used in a small but global effort by several Canadian scientists (personal communication, Dr. A. Steffen, no published results yet) and has been considered for addition to the APMMN networks. This suggests some global acceptance as a standard measurement that can be incorporated into national/global networks and for support of Minamata goals.
- MELD has gone through a thorough review of the MerPAS system, and per the report is a valid method now, that received a good bit of support by the MELD reviewers as a viable, inexpensive, and accurate measurement system.
- the MerPAS is a commercial product (acceptance again) by Tekran Instruments, our current partner in AMNet. After discussions with Tekran (i.e. Dr. Eric Prestbo, personal communication), they are willing to help the network in a variety of ways, and particularly with initial startup and quality assurance. After this startup phase, Tekran is willing to let NADP operate our network as we see fit.

McLagan et al (2016) reports the following basic sampler information:

- detection limit of 0.26 ng Hg (3x field blank concentration standard deviation $\{0.51 \pm 0.13 \text{ ng g}^{-1}\}$);
- sampling rate of $0.121 \pm 0.005 \text{ m}^3 \text{ day}^{-12}$;
- precision equal to $2 \pm 1.3\%$; and
- deployment time a week to at least one year.

Based on these reported values, a monthly sample at an estimated atmospheric concentration of 1 ng/m³ would easily be well above the MDL for the instrument (3.63 ng Hg for a 1 month sample).

² Tekran Instruments suggests a slightly different flow rate in this configuration of 0.111 m³/day.

<u>Sampling Locations</u>: Samples could be sited at any number of locations, including rural (expected concentrations higher than the example above), suburban and urban. We would assume that many of the likely sites would be collocated with MDN samples, and perhaps with and around AMNet sites. It is not expected that AMNet sites will close with passive MerPAS being used as an alternate monitoring strategy. Standard site location images, latitude and longitude information, and rural/urban designations would be included in a normal NADP site file. Sampling locations would be open to all, with siting restrictions developed.

<u>Sampling Time and Change Out:</u> At least initially, we would suggest a sampling time of 1 month (actual calendar months) with varying number of days. Samples would go on the first day of the month (8-10 local time) and come off on the first day of the next month (8-10 am local time). At this point, the operator would cap the sampling jar, and mail the sampler back in to the NADP labs. The new sample would then be attached to the same hanger. The exposed sample would be put back into the network shipping box and returned. Sample shipping would be two-way ground UPS service and included in the overall sampling cost. Each sample would be provided with a prepaid "airbill" for return shipping by UPS.

<u>Sample Setup</u>: The network would need to purchase and weigh out the appropriate AC (0.7 g), fill the sample chambers, wash/soak/sonicate the bodies and housing jars, and box and ship samplers. The NADP would utilize a small room for sample preparation within the Henry Mall NADP labs. All cleaning of supplies and filling of sample containers would occur in this room (much like the AMON procedures). Per the AMON Radiello bodies, sample bodies would be used 5 times (actually to be determined) and then discarded. Samples would be shipped from the HM location and received at the HM location. Samples would be transferred to the Ag Drive location for analysis.

2.2 Lab Operations

<u>Analysis</u>: Sample analysis would use the Nippon Model MA-3000 instrument, which is already operating in the HAL laboratories (<u>https://www.hg-nic.com/ma-series/ma-3000/</u>). Analysis is through Direct Thermal Decomposition-Gold Amalgamation-CVAAS. Essentially, the mercury compounds are collected using elemental carbon, and the carbon is combusted in the MA-3000. Mercury is volatilized as elemental Hg and collected by the instrument's gold traps. Analysis is by Cold Vapor Atomic Fluorescence (the same method as the MDN and AMNet).

The instrument is capable of one analysis per 5 minute period, so a 20 site network would require about 4 hours to complete, with sample set up, quality assurance samples, and sample analysis. Therefore, a very large network could be supported with the current instrument and a minimal amount of an analyst's time. The instrument is currently located within the Agricultural Drive facilities (Clean Room), but could be moved to the Henry Mall facilities if appropriate. Some instrument costs would be associated with the analysis, including cost of a catalyst, and miscellaneous analysis supplies (boats, etc.).

Assuming a 20-site network for startup, the analysis would be run monthly, once all samples have been received. New sampler bodies would be assembled and mailed out towards the end of the month. All assembly would be done at Henry Mall within the MDN/AMNet clean laboratories. Analyses would be done at Ag Drive.

This network startup is proposed to occur in two phases, which affects the sampler handling. These phases are discussed later.

<u>Sample Supply Storage</u>: sampling supply storage should not require a large area (sample protective jars, bodies, carbon, boxes). At this point, we feel these supplies could easily be incorporated into the current storage areas of the NADP.

2.3 Data Management

The data load will be fairly light. For each sample and location, these data will be captured:.

- Site location;
- Start and end times (monthly, on first day of month, off, first day of next month);
- Total mercury mass measured (ng);
- Total estimate of sample volume (m³, based on Radiello body);
- Total concentration of gaseous mercury (ng/m3);
- QR Code (A/B/C); and
- Sample notes and flags (to be determined, following AMNet);

3.0 Products

3.1 Data

This same data (from the previous section) will be presented on the web for unrestricted download. Download will be made available by site and in total, as with all networks.

Archival for the samples is not possible, because the analysis is by combustion, so there is no additional sample or remaining sample for reanalysis.

Dry Deposition estimates. Much has been planned for annual estimates of dry deposition with the AMNet data. The model of dry deposition estimates is currently being built. Dry deposition from monthly gaseous concentrations can be made, but the estimates will include more error (larger error bars).

3.2 Reports

It is envisioned that this new network and its results would be added to the mercury map summary at the end of each calendar year, would be presented much like the AMON sample results are presented (average concentration per year, perhaps seasonal map products). No additional reports are planned

4.0 QA/QC

<u>Planned Network Quality Assurance:</u> following from the AMON, this network would provide for ongoing travel blanks and duplicate sampling, along with laboratory atmospheric concentrations. This is above and beyond the basic analytical instrumentation quality assurance. This table details the specifics of these ongoing tests. This increased QA sample load is figured into the budgetary information (last section).

Sample Type	Location	Frequency	Warning Limit (ng/m3)
Travel Blank	in packaging, rotating sites	1 of 10 samples mailed	0.1
Ambient Concentration	sampler assembly area	monthly concentrations, continuously	0.5
Duplicate Concentrations	all sites randomly	1 of 10 samples mailed	+-0.2

4.1 Field Operations

The quality assurance impact to the field operation would be minimal. Every tenth sample, the operator would receive a field blank which would only need to be stored. Additionally, every tenth month, the operator would need to put on a second sampler for a QA duplicate. Shipping boxes and shipping cost would be provided.

4.2 Lab Operations

The laboratory would continue its routine measurement of mercury in the laboratory, as is currently practiced. This new sample type would also allow further laboratory monitoring for all Hg networks, if this would be of interest. This network would require additional analysis and QA load on the instrument, such as additional zero and span concentrations on the instrument (i.e. more replication of this standard operation).

4.3 Data Management

With addition of this initiative/network, an additional workload would be added to the laboratory data reviewers, and another final data review by the PO. The laboratory would need to add in an additional review, much like AMON data review. However, it is expected that the same software system could be used to automate this process.

The final PO data review programs currently used for AMON could easily be adopted for a gaseous mercury passive measurement, and would only add about 4 hours of final review.

4.4 Additional QA Planned

This network is being proposed as the "backbone" to the expansion of network capabilities for Hg measurement being considered by MELD, and detailed in their activities over the past several years. This passive network approach is well-researched and documented, and the applied techniques have been shown to be accurate and inexpensive. Therefore, we feel this network is a logical first step in the pursuit of the MELD goals as detailed in their recent summary of methods report (May 2021).

We are also proposing that an ongoing monitoring effort be added to the network QA measurements currently made at the Program Office. Specifically, we would begin routine measurement of all proposed mercury measurement methods: the speciating Tekran, MerPAS samplers (two types), the University of Nevada Reno membrane system, the Japan MOE gold trap method, the USGS goldtrap/isotope method, and the

NOAA TGM Differences method (see nearby figure). This approach will support the MELD Group goals by providing the network with experience using each system, and more information that could be used in any future network choices.



The instruments are planned to run at the Henry Mall location (5th Floor Roof, 6th floor laboratory) for ease of operation. Each of the original method supporters will be invited to provide equipment and support for their method, with daily/weekly field support provided by the NADP. Any analysis will be provided by the method supporter. NADP will be responsible for any specific QA applications and data records and analysis. A common time period for operation (or periods) will need to be established.

As an additional QA step, Tekran has offered to run a "pre-Phase 1" intercomparison. Essentially, they would supply us with ~20 prepackaged MerPAS samplers to be run at the HM location. After exposure, they are willing to run half of the sample analyses at their laboratories, while NADP runs half with our instruments. This type of QA study should show laboratory comparison directly. This additional QA step will show consistency of the NADP as it begins our Phase 1 network. Unfavorable results will guide us towards additional steps necessary to improve our capabilities. For each method, the mercury fraction collected and time periods possible are listed in the nearby table³. With all methods present, this arrangement will provide and abundance of GEM intercomparisons, along with GOM comparison with the Tekran system. Although the general 1 or 2-week intercomparison is assumed, additional testing and methods could be added as the needs of NADP change or at the request of MELD.

	Fraction Measured				
Method	TGM	GEM	GOM	PBM _{2.5}	
Speciating Tekran (2537, 1130, 1135)		5 min	hour/2-hour	hour/2-hour	
MerPAS samplers (network)	monthly				
MerPAS samplers (QA)	weekly to				
Men AS samplers (QA)	monthly				
Un. of Nevada Reno membrane system			weekly	weekly	
Japan MOE gold trap method		24 hr - monthly			
USGS goldtrap/isotope method		24 hr - monthly			
NOAA TGM Differences method		hourly	hou	ırly	

5.0 Budget and Staff Support 5.1 Field operator

Costs to the field operations will be minimal and similar to the AMON change out.

Time onsite should be approximately 5 minutes to change out the sampler and fill out the simplified field sheet. With a 1st day of the month start, 6 of 7 months will require an additional trip to the site (non-Tuesday start). This should be 10 added site visits per year. Travel time for some sites could be significant, but for most sites, this should be minimal.

	ice site POU	Printname N.S. Liaison	N S L Intials
S. SAME START AND END Image: Start And End	0001-2400 res No Cd 1 0 0 1 1 1 0 0 2 2 2 1 1 0 0 2 3 3 HLTER REMOVAL 7. REMARK 5.737 6.707 R.4in	esse check any and all conditions that apply. mment an any other site conditions in Block 7. Significant smokeor fre Fertilizer use nearby Farm animal activity nearby	

The network will provide shipping boxes and

two-way shipping. Therefore, the added shipping responsibility should be minimal. We also should be able to ship the box with other boxes being returned to the PO (MDN cooler, supply box, etc.).

³ As of this writing, we have been approached by Nippon Instruments Corp. They have a new instrument designed to measure gaseous mercury (the "AM6F"). It is possible that this instrument will be added here .

As with AMON, there is little maintenance needed. Site startup and finding a location will take some time for the operator.

With a 20-site network, sample set up should take on the order of one chemistry technician about 1 day to set up the samplers, and mail all supplies.

Estimates of the time for analysis includes both sample receipt and analysis are an additional day. Instrument analysis time is minimal (5 min/sample run). The NADP's MA-3000 is only occasionally used for Litterfall. At most, we expect the instrument to be used for this project for 2 days/month, which should be easily accommodated.

Site support from both the laboratory and the Site Liaison should be minimal, and in keeping with the amount of effort for the AMON sites. Given that no electricity or moving parts are involved, few issues are expected with the monthly sampling. Data management/Program Office

For this network, sampling load would be simple. With a 20-site network, the PO would receive 240 data points, along with the appropriate quality assurance information. As of now, the PO would have three broad activities; 1) producing average concentrations, 2) some type of average atmosphere concentration map or figure (likely similar to the AMON mapping product), and 3) average dry deposition estimates for areas around the measurement sites (after approval). We would need a new network page, add these observations to the databases, house site information and pictures, add the map and network to the annual map summary, and routine marketing documents, etc.

Financial contracting of this new initiative/network would be relatively straightforward and would be folded into the current operation with little impact.

5.2 Timetable of Phases 1 and 2

As noted, Tekran Instruments currently sells the MerPAS (Mercury Passive Air Sampler). Based upon our conversations, we are planning a phased approach to startup and operation of this network. Phase 1 will be used at network start, and a migration to Phase 2 or the final operation method.

For <u>Phase 1</u>, NADP would purchase fully ready MerPAS samplers (Radiello white bodies, packed with charcoal/sulfate core, in stainless cases, in protective shipping jars) ready to be shipped out to sites. The NADP HAL will do the analysis. After analysis, NADP will keep the used equipment, etc. After a 4–8-month transition, where the

network will operate in Phase 1 (start the sampling, work out any start up issues, learn to efficiently complete the analysis, etc.), the network will be moved to Phase 2.

<u>Phase 2</u> is where the network will purchase/make/clean all needed Radiello bodies, stainless steel cores, activated charcoal and sulfur addition, housing, miscellaneous hardware, etc. to operate the network completely. We are likely to continue to purchase the activated carbon mix for consistent particle size and carbon /sulfur mixing ratio for consistent mercury adsorption and flow rates.

The initial set up of the intercomparison would be set up within the first months after approval.

Phase 1, using Tekran products, would occur over the first 3-6 months of operation. Phase Two would operate at least 1 year as a trial network. After this period, we would expect Executive Committee to make a determination of continued trial years, or conversion to an independent NADP network.

5.3 Overall Costs of Site/Network Operation

An estimate of Phase 1 and Phase 2 costs for the network are shown in the following tables. The estimates are in line with each other, (\$2,300/1,500 per site per year, Phase 1&2) in a 20-site network. There is a significant savings with the Phase 2 approach. Much of the additional cost is with the Program Office charge (\$500), and a modest cost for personnel time for the analysis (both phases) and for sampler build (Phase 2). The cost of the passive device purchase from Tekran is small in comparison.

				Total Costs		Cost per year		notes
	Program Office Charge					\$	500	
		perc	ent time		Salary			
	Chem Tech		10.00%	\$	45,000	\$	4,500.00	25% time to 50 site network
	Fringe			\$	15,300	\$	1,530.00	
	Totals			\$	60,300	\$	6,030.00	
	Per Site Costs	Cost	t per	Cos	t per monti	Ь		
	Per Radiello, set up and ready	\$	69.00	\$	69.00	\$	828.00	
		- T		-		+		
	instrument deprecipation					\$	-	
	instrument supplies-catalyst	\$	1.06	\$	1.06	\$	12.67	
	instrument supplies-misc	\$	1.00	\$	1.00	\$	12.00	
	sample boat	\$	0.50	\$	0.50	\$	6.00	
	shipping out			\$	9.00	\$	108.00	
	return shipping			\$	9.00	\$	108.00	
	Cost of box, 1 year lifetime	\$	1.25	\$	0.10	\$	1.25	
Costs	per sample			\$	60,390	\$	1,075.92	
	multiplyer 1 in 5 travel blanks					\$	215.18	
	multiplyer 1 in 10 duplicates					\$	107.59	
	misc QA samples (10%)					\$	107.59	
Fully lo	aded per sampling site cost					\$	1,506.28	
, ,								
						anr	nual	monthly
-	er 20 site network					\$	2,307.78	\$ 192.32
	er 50 site network					\$	2,126.88	\$ 177.24
Costs p	er 100 site network					\$	2,066.58	\$ 172.22

				Total Costs		Cost per year		notes
	Program Office Charge					\$	500	
		perce	ent time		Salary			
	Chem Tech		15.00%	\$	45,000	\$	6,750	25% time to 50 site network
	Fringe			\$	15,300	\$	2,295	
	Totals			\$	60,300	\$	9,045	
				Ψ	00,000	Ψ	57010	
	Per Site Costs							
		Cost	per	Cos	st per mont	h		
	Field supplies-Protective Jar	\$	3.00	\$	0.25	\$	3.00	
	Radiello bodies	\$	12.50	\$	2.50	\$	30.00	
	Activated Carbon	\$	1.00	\$	1.00	\$	12.00	
	instrument deprecipation					\$	-	
	instrument supplies-catalyst	\$	1.06	\$	1.06	\$	12.67	
	instrument supplies-misc	\$	1.00	\$	1.00	\$	12.00	
	sample boat	\$	0.50	\$	0.50	\$	6.00	
	shipping out			\$	9.00	\$	108.00	
	return shipping			\$	9.00	\$	108.00	
	Cost of box, 1 year lifetime	\$	1.25	\$	0.10	\$	1.25	
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Costs	per sample			\$	60,324	\$	292.92	
	multiplyer 1 in 5 travel blanks					\$	58.58	
	multiplyer 1 in 10 duplicates					\$	29.29	
	misc QA samples (10%)					\$	29.29	
Fully lo	aded per sampling site cost					\$	410.08	
							ual +	
C					ulated		ety factor	monthly
Costs p€	er 20 site network			\$	1,362.33	Ð	1,500.00	\$ 113.5
Costs pe	er 50 site network					\$	1,090.98	\$ 90.9
*	er 100 site network					\$	1,000.53	\$ 83.3

6.0 Funding

- 6.1 Availability: Degree of self-support, sources of funding, commitments to longterm funding
- 6.2 Contributions: Equipment, funds, in-kind support

As discussed in later sections, Tekran Equipment will provide basic materials for an intercomparison and network start up as discussed in the QA section.

6.3 Transfers: Mechanisms for funding field operations, laboratory operations, Program Office

7.0 Operation within NADP

7.1 Within existing NADP structure

<u>How would this impact the other NADP networks</u>: during the Phase 1, this project should have minimal impact upon the NADP and its current networks, or NADP at large. The analysis sample load of a small network would be on the order of one to two days' time per month for a technician/analyst for the mail out of supplies, packaging of sampler (Phase 2), analysis, etc. Space is easily available at Henry Mall, and the analytical instrument is only occasionally used for Litterfall and other WLSH work. During Phase 2, more time would be needed to order supplies, pack sampling supplies, attend to the QA needs of the network and laboratory. But we don't think this would be a major increase of resource needs.

With network growth, personnel costs would increase. But, the network is planned to be self-supporting, and bring in additional funds to support additional time needed for analysis, sampler makeup and shipping, etc. For comparison, AMON uses one chemist for analysis, all sample cleaning, and preparation. We would assume about half this level of commitment would be needed for this network at 100 or so sites (monthly versus 2-weekly sampling).

The addition of the QA component/intercomparison would likely have more of an impact on the PO time allotment. Time for planning and the approval processes for the samplers through the UWM would require a block of time and effort. Overall, a significant but minor amount of money is needed for this effort, and is primarily

personnel. But overall, we do not expect that the network will have any adverse impact upon the current networks.

<u>Budgetary Impact to NADP Mercury</u>: in fact, we feel that the network will have a positive effect on the mercury programs of NADP. By adding in a low cost mercury options, increased budgetary support for the overall mercury operation of NADP is expected. As we are all well aware, the mercury networks of NADP (MDN, AMNet, and to some extent Litterfall) have all been slowing losing support over the past 8 years or so. As this occurs, each network brings in less funding for the overall support of the NADP general practices (such as PO management, map making, database work, etc.). A low-cost and easy measurement and network could be very valuable to NADP. We feel that this combination will appeal to many new partners for measuring mercury to reach many different goals, therefore, the possibility of expansion of this type of mercury network is likely.

No additional subcommittees would be needed, and we expect that NOS and MELD would oversee the network.

8.0 References

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