Comparison of Mercury Measurements in Snow with ISCST3 Model-Predicted Deposition Near an Industrial Emission Source in Southeastern Idaho

Michael L. Abbott
Idaho National Engineering and Environmental Laboratory
P.O. Box 1625, Idaho Falls, ID 83415-2107

David D. Susong
USGS-Water Resources Division
2329 West Orton Circle, West Valley City, UT 84119-2047

David P. Krabbenhoft
USGS- Wisconsin District Mercury Laboratory
8505 Research Way, Middleton, WI 53562

Abstract

Snow samples collected within 12 km of a major (9 g/hour) mercury emissions source in southeastern Idaho were evaluated for total and methylmercury concentrations (ng L^{-1}), converted to mercury deposition using the sample location snow water equivalent (SWE), and compared to predicted deposition values from the ISCST3 air dispersion/deposition model. Model runs were made for a range of scavenging coefficients for both particulate and vapor form. Mercury concentrations in snow near the source (μ = 5.3 ng L^{-1}) were similar to published values in precipitation at other remote U.S. locations and were about twice those measured in the Teton Range (μ= 2.7 ng L^{-1}) located 180 km downwind to the east. Estimated deposition was very low at the site because of the low annual precipitation (22 cm y^{-1}). ISCST3 model predicted-to-observed (P/O) ratios for deposition averaged from 0.6 to 1.9 across all sampling locations with the best model performance obtained using a scavenging coefficient for small (0.1 – 0.25 um) particulates and the assumption that all of the mercury is reactive Hg(II) and subject to local deposition. The default modeling assumptions recommended by the U.S. EPA for mercury Hg(II) vapor speciation in combustion facility risk assessments would have underpredicted local mercury deposition from this source by a factor of approximately 2.

Introduction

Hazardous waste combustion facilities (i.e. incinerators) may be required by the U.S. Environmental Protection Agency (EPA) under the Resource Conservation and Recovery Act (RCRA) to perform site-specific risk assessments to evaluate potential human health and ecological effects that may result from facility stack emissions. The recommended RCRA risk assessment methodology is specified in the three-volume document, Human Health Risk Assessment Protocol (HHRAP) for Hazardous Waste Combustion Facilities[1]. The HHRAP specifies the use of the Industrial Source Complex–Short Term (ISCST3) model to predict near-field (< 50 km) downwind air dispersion and deposition rates of

contaminants[2]. Direct inhalation and potential intakes from food chain transport are then calculated using spreadsheets.

ISCST3 model predictions of mercury deposition rates are strongly dependent upon many local-scale factors. Two important ones for which little data are usually available include: (1) the fraction of total emissions deposited locally (within 50-km), which is a function of the speciation between divalent mercury (Hg[II]), elemental mercury (Hg[0]) and the phase allocation between Hg(II) vapor and particulate[3], and (2) the precipitation scavenging rate of contaminants from the airborne plume. The HHRAP recommends the following default "recipe" for speciation and phase allocation: (1) 20% Hg(0), with 1% deposited locally, (2) 60% Hg(II) vapor, with 68% deposited locally, and (3) 20% Hg(II) particle-bound, 36% deposited locally. The net result of this recipe is that 48% of the total mercury emitted from a source is assumed to be deposited locally and modeled while the remaining 52% is assumed to enter the global cycle and be deposited elsewhere (and not modeled). For precipitation scavenging rates, the HHRAP recommends values from Jindal and Heinold[4], which is taken from generic atmospheric particulate aerosol measurement data not specific for mercury.

In this paper, we report the results of mercury sampling in snow near a large U.S. Department of Energy (DOE) mercury emission source in southeastern Idaho and compare the resulting deposition to that predicted using the ISCST3 model. The fraction of total mercury emissions deposited locally was examined, and a site-specific scavenging coefficient was determined by comparing the measured data to modeling results that used a range of scavenging coefficient input values.

Emissions Source and Study Area

The New Waste Calcining Facility (NWCF), located at the Idaho National Engineering and Environmental Laboratory (INEEL), was intermittently operated from August 1982 through May 2000. During this 18-year period it converted aqueous nuclear fuel processing wastes into stable granular "calcine" solids for interim storage. Significant quantities of mercury were present in the feed (1 – 3 g/L, est. 10 - 40 Mg total) as a result of its use as a catalyst in aluminum nuclear fuel dissolution. Recent stack tests measured a mercury emission rate of 9 g hr^{-1}, approximately 1.5% of the typical 600 g hr^{-1} feed rate. Most (about 88%) of the mercury emitted from the 76-m high stack is thought to be divalent $HgCl_2$ vapor based on results from an EPA metals sampling train and chemical equilibrium calculations. The remaining 12% is thought to be submicron particulate--either solid phase HgO or absorbed/condensed onto particles as $HgCl_2$. A negligible amount of elemental Hg(0) is present.

The INEEL is located in southeastern Idaho, 68 km west of Idaho Falls on the Eastern Snake River Plain (ESRP), a low-lying (1,500 m) volcanic area of basalt lava beds and a shrub-steppe ecosystem surrounded by desert mountain ranges (3,000 – 3,500 m). The area has a semi-arid cold desert climate with an average annual precipitation rate of 22 cm y^{-1}, mostly in the form of snow.

Methods

Sampling and Analytical Methods

Snow samples were collected on the INEEL on February 10 –11, 1999, after the NWCF had been operating for 17 days. This was the first time NWCF had been operating during the 1998/1999 winter. Ten sampling locations were selected within about 12 km of the source based on ISCST3 model predictions of the maximum area of deposition using meteorological data for the same 17 day "fallout period." At each sampling location, the snow was excavated to the ground, the snow face was cleaned using a non-metalic (lexan) shovel, and composite samples of the entire snowpack thickness were collected. Snow density measurements were made, and snow water equivalent (SWE) was calculated as the product of the snow density (kg m^{-3}) and thickness (m) which is equivalent to the mass (kg) or depth of water (mm) per unit area. SWE is needed to convert measured concentrations (ng L^{-1}) to deposition flux (ng m^{-2}) for model comparisons. The snow samples were then shipped frozen to the USGS Wisconsin District Mercury Laboratory (WDML) in Madison, Wisconsin.

Snow was collected in special 2-L teflon sampling jars that had been precleaned in the laboratory with hydrochloric acid (HCL), rinsed three times with reagent grade water, partially refilled with 1 percent Omni Trace HCL, and given a final rinse. The bottles were then air-dryed in a mercury-free class 100 laminar flow hood equipped with a mercury filter (gold imprenated cheese cloth) before double bagging in plastic bags[5].

In the laboratory samples were analyzed for total mercury (THg) and methylmercury (MeHg). THg analysis was preformed using EPA Method 1631: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry (CVAFS) with modifications. Method 1631 is a draft method that consists of three essential parts: oxidation of mercury species to reactive mercury (Hg[II]), reduction of Hg(II) to volatile mercury (Hg(0)), and detection of Hg(0) by CVAFS. MeHg analyses were preformed using EPA Method 1630: Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS with minor modifications[5].

The composite sample of the full snowpack thickness were assumed to contain 17 days (January 24 through February 9 fallout period) of THg deposition from the NWCF emissions, plus THg deposition from the global atmospheric inventory. Meteorological data for this 17-day "fallout period" were obtained from the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory, Field Research Division (FRD) in Idaho Falls, which operates a series of meteorological towers on the INEEL. There were 6 days of mostly frozen (snow) precipitation during the period with a total water equivalent of 20.6 mm. Total snow depth measured near the NWCF during the period varied from 15.9 cm on January 26 to 21.6 cm on February 1, after which a moderate thaw reduced the depth to 12.7 cm on February 8. During the snowfalls that occurred over the fallout period, wind directions were predominantly from the southwest.

Data Analysis

The measured THg concentrations consisted of contributions from both the NWCF and regional/global background fallout. To partition these contributions, a number a calculations were performed.

The regional background THg concentration was estimated using SWE-normalized THg measurements in snow from the Teton Range, 180 km to the east[6]:

$$C_B = \overline{\left[\frac{C_T}{SWE_T}\right]} SWE_I \qquad (1)$$

where:

C_B = THg background concentration at each INEEL sampling location (ng L^{-1})
C_T = THg concentration in Teton Range snow (2.7 ng L^{-1})
SWE_T = snow water equivalent in Teton Range snow (mm)
SWE_I = snow water equivalent at each INEEL sampling location (mm)

The concentration due to NWCF fallout only, averaged across the composite sampled snowpack (C_{NC}), was then calculated by subtracting C_B from the total measured concentration (C_T) at each sampling location.

$$C_{NC} = C_T - C_B \qquad (2)$$

Since snow existed on the ground prior to startup of the NWCF, C_{NC} needed to be adjusted to estimate the NWCF contribution that would have been ***in snowfall during*** NWCF operations (converted from a snowpack-averaged concentration to the concentration in only the snow that fell between NWCF startup and sampling):

$$C_N = C_{NC}\left[\frac{SWE_{Ii}}{P_F}\right] \qquad (3)$$

where:

C_N = NWCF concentration in snowfall during operations (ng L^{-1})
P_F = precipitation recorded over the 17-day fallout period from NWCF startup (Jan 24) to start of sampling (Feb 10) (mm)

The ratio of the average SWE at the INEEL sampling locations (60 mm) to the 17-day fallout period precipitation (20.6 mm) implies that the NWCF deposition existed in the top 1/3 of the sampled snowpack.

Concentrations were then converted to total deposition (load) in the snowpack (D_T, in µg m^{-2}) at each sampling location by:

$$D_T = \frac{C_T \ (10^{-3} \mu g \, ng^{-1})}{\rho_W} SWE \qquad (4)$$

where:

C	=	measured, background, or NWCF concentrations (ng L^{-1})
ρ_w	=	density of water (1 kg L^{-1})
SWE	=	snow water equivalent at the sampling location (kg m^{-2})

The deposition from background fallout (D_B) was estimated using C_B in equation (4). The deposition from the NWCF emissions only (D_N), which was used for ISCST3 model comparison, was then calculated by subtracting D_B from D_T.

Annual deposition (µg m^{-2} y^{-1}) from background fallout only (DA_B) was estimated by multiplying D_B by the ratio of the total annual precipitation recorded for that water year (P_A = 225 mm) to the sample location SWE:

$$DA_B = D_B \left[\frac{P_A}{SWE} \right] \qquad (5)$$

Annual deposition from continuous NWCF operations (excluding background) was estimated by multiplying D_N by the ratio of P_A to the precipitation recorded from facility startup to the start of sampling (P_F = 20.6 mm).

$$DA_{NWCF} = D_{NWCF} \left[\frac{P_A}{P_F} \right] \qquad (6)$$

The total annual deposition (assuming continuous NWCF operations) was then estimated by adding the two components ($DA_B + DA_N$).

Estimating annual snowpack deposition in this way introduces two potential uncertainties: 1) it does not account for the different scavenging efficiency of rain vs. snow and 2) it assumes that the ambient THg background air concentrations are constant throughout the year. Although rain is thought to be a more efficient collector of atmospheric THg than snow[7,8], published concentration data generally show little difference between the two[3,9].

Comparison with Model Predictions

The ISCST3 air dispersion model[2] is currently recommended for use in combustion facility risk assessments to assess downwind air concentrations and deposition rates of pollutants including mercury. The model was used in this study for two purposes: (1) determine optimum sampling locations around the NWCF using actual meteorological data over the time between NWCF startup and sampling and (2) compare model predictions of total deposition during the fallout period with that measured in the snow.

Wet deposition is a primary mechanism for transporting mercury from the atmosphere to surface waters or land[3]. ISCST3 calculates wet deposition by multiplying the vertically integrated plume concentration by a scavenging rate constant (Λ, in units of s^{-1}), which is defined as the fraction of plume particulate removed per unit time. The scavenging rate constant is computed as the product of a scavenging coefficient and precipitation rate:

$$\Lambda = \lambda R \qquad (7)$$

where the scavenging coefficient λ has units $(\text{s-mm/hr})^{-1}$, and the precipitation rate R has units (mm hr^{-1}). The scavenging coefficient may depend on the characteristics of the pollutant (e.g., particulate diameter, solubility and reactivity for gases) as well as the type of precipiation (e.g., rain vs. snow). Jindal and Heinold[4] have provided a best-fit relationship for λ for typical atmospheric aerosols as a funtion of particle size. Because of the lack of pollutant-specific data for gaseous pollutants, a particle scavenging coefficient for very small (e.g. 0.1 µm) particles is often assumed (1.6×10^{-4} $[\text{s-mm/hr}]^{-1}$)[1].

Most (about 88%) of the mercury emitted from the NWCF stack is thought to be divalent $HgCl_2$ vapor based on results from an EPA metals sampling train and chemical equilibrium calculations. The remaining 12% is thought to be submicron particulate--either solid phase HgO or absorbed/condensed onto particles as $HgCl_2$. A negligible amount of elemental Hg(0) is present. The HHRAP default assumption for THg partitioning in combustion facility risk assessments is 80% Hg(II) and 20% Hg(0), an 85-15% split for Hg(II) vapor vs. particulate, with a net 48% of the THg being deposited locally. In order to determine the best model perfomance for prediction of NWCF deposition, numerous ISCST3 modeling runs were made using different scavenging coefficients for particle sizes ranging from 0.1 to 1.5 µm and for a vapor form. For each of these runs, model predicted-to-sample observed (P/O) ratios were calculated and plotted against a 1:1 line to determine the best fit and whether the model under- or over-predicted the observed deposition.

Results and Discussion

THg Concentration in Snow

Measured THg concentrations in snow ranged from 0.55 to 5.09 ng L^{-1} on or near the INEEL (Table 1). When converted from composite snowpack concentration to the concentration in snowfall during NWCF operations (equation 2), concentrations were estimated to range from 0.55 to 9.16 ng L^{-1}. These concentrations are generally low and are similar with those measured in other remote locations, including arctic Alaska (0.9 – 7.5 ng L^{-1})[10], Canada (0.6 – 6.0 ng L^{-1})[11], and in Utah's Wasatch Range (4.5 – 11 ng L^{-1})[6]. In contrast to these relatively low values, concentrations in rain in eastern industrialized areas have been measured at much higher levels, ranging from 26 to 62 ng L^{-1} in a rural New Jersey area to 606 ng L^{-1} near a municipal waste combustor (MWC) in New Jersey[12].

Table 1. Total mercury (THg) concentrations, deposition, and the fraction of THg that is methyl mercury (MeHg) in snowpack near the NWCF at the INEEL.

Sample Site	THg Conc. (ng L^{-1}) Composite Snowpack	Estimated in Precip.	THg Deposition Snowpack (ug m^{-2})	Annual (ug m^{-2} y^{-1})	Fraction from NWCF	Fraction MeHg/ THg
1-1	1.90	4.33	0.10	0.97	0.92	
1-2	1.65	3.36	0.08	0.76	0.91	<DL
2-1	5.09	9.16	0.19	2.06	0.97	0.008
2-2	2.25	7.30	0.18	1.64	0.92	
2-3A	2.00	4.99	0.12	1.12	0.92	0.032
2-3B	2.61	7.57	0.18	1.70	0.94	
2-3C	1.56	3.84	0.10	0.86	0.89	
2-4	0.96	1.86	0.05	0.42	0.81	<DL
EBR-1	2.08	5.55	0.13	1.25	0.92	
BSB-1	0.55	0.55	0.04	0.12	negligible	<DL
Mean =	**2.06**	**5.33**	**0.12**	**1.09**	**0.91**	**0.020**
STD =	**1.22**	**2.66**	**0.05**	**0.60**	**0.04**	**0.017**

Annual Deposition and Methyl Mercury

Estimated annual THg deposition on the INEEL ranged from 0.12 to 2.06 µg m^{-2} y^{-1} (Table 1). These estimates are less than the estimated annual deposition determined in the Tetons 180 km to the east[6]. This is contrary to what one would initially expect given the higher snowfall concentrations measured at the INEEL (mean = 5.3 ng L^{-1}) relative to the Tetons (mean = 2.7 ng L^{-1}). Standard risk assessment protocol generally assumes that locations near an emission source (except within 1-2 km of a high stack) have higher impacts (air concentrations and deposition) than distant downwind locations. However, these results suggest that a particular location's annual precipitation rate may be a more important factor in determining total annual mercury deposition than a location's proximity to an emission source.

The calculated total annual depositon rates compare well with measured values from other regions of the U.S. as predicted by EPA in a nationwide mercury modeling effort[3]. Deposition rates at the INEEL are low compared to the EPA-modeled predictions for remote western regions of the U.S. (1 – 3 µg m^{-2} y^{-1}).

The ratio of methylmercury (MeHg) to THg on the INEEL ranged from 0.8 to 3.2% which compares favorably with other measured values reported by EPA[3] (0.1 to 6.3). The EPA HHRAP recommends net methylation default values of 2% for non-wetland soils and 15% in surface water bodies.

Comparison to Model Predictions

The ISCST3 model-predicted deposition pattern near the NWCF using a meteorological data set from the fallout period (1/24/99 – 2/9/99) is shown in Figure 1.

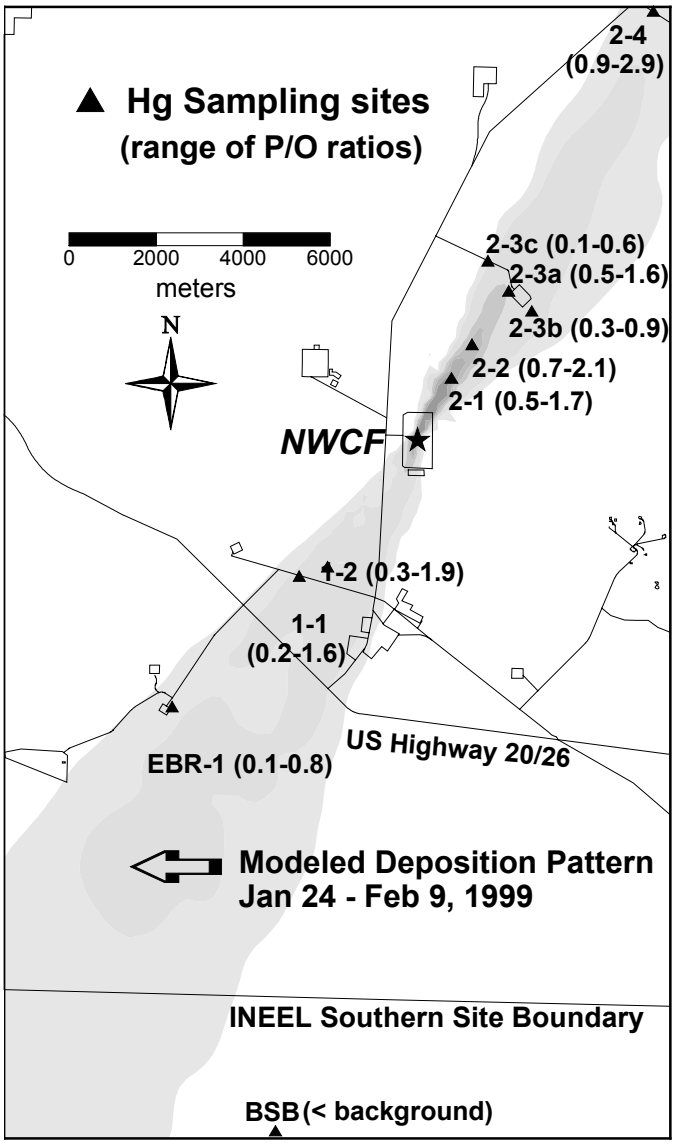

Figure 1. Modeled deposition pattern during facility operations and model predicted-to-sampling observed (P/O) ratios.

Table 2 lists the ranges of model-predicted to sample-observed (P/O) ratios at each sampling location for the different model input scavenging coefficient (SC) values evaluated. When corrected for background, the BSB sample location (17.3 km south of NWCF) showed slightly negative deposition, indicating negligible NWCF deposition at this location. At other locations, P/O ratios ranged from a low of 0.1 to a high of 2.9. The low value occurred at an off-plume centerline location (2-3c) where the predicted concentration gradient and potential for model spatial prediction error is high. The high P/O ratio (2.9) occurred at the most distant downwind location to the northeast (location 2-4, 11.8 km distance).

Table 2. Model predicted-to-sample observed ratios (SC = model scavenging coefficient with units of $[s\text{-}mm/hr]^{-1}$).

	For particlulate of diameter:			for vapor with SC =	
Site #	**0.1 µm**	**0.25 µm**	**0.5-1 µm**	**2E-04**	**1E-04**
1-1	1.6	0.9	0.7	0.5	0.2
1-2	1.9	1.1	0.8	0.6	0.3
2-1	1.7	1.0	0.5	1.6	0.8
2-2	2.1	1.2	0.7	2.0	1.0
2-3A	1.6	0.9	0.5	1.3	0.6
2-3B	0.9	0.5	0.4	0.5	0.3
2-3C	0.6	0.3	0.2	0.2	0.1
2-4	2.9	1.7	0.9	2.8	1.4
EBR-1	0.8	0.5	0.4	0.2	0.1
BSB-1	<-- not calculated (negligible NWCF deposition) -->				
Mean=	**1.6**	**0.9**	**0.6**	**1.1**	**0.5**
StdDev=	**0.72**	**0.43**	**0.23**	**0.92**	**0.47**

Validation studies for the standard Gaussian plume model used in ISCST3 have indicated optimum P/O ratios for annual average ***air*** concentrations ranging from 0.25 to 4.0[13]. Estimated P/O ratios for model deposition rates are expected to have a significantly wider range.

ISCST3 model performance using four different scavenging coefficients (SC) is shown is Figure 2. Three model runs (plots a, b, and c) assumed particulate phase emissions and used SC values for particle diameters ranging from 0.1 µm to 1.5 µm. One model run (plot d) simulated a pure vapor release and used the SC value for the smallest (0.1 µm) particle diameter ($2 \times 10^{-4}\ s^{-1}/mm\text{-}h^{-1}$) as recommended by EPA in their combustion facility risk assessment guidance[1]. This SC value most closely matches the predicted physical form of the NWCF mercury emissions (88% vapor, 12% particle phase or surface-condensed).

These plots indicate the following about mercury deposition from NWCF emissions: (1) a 0.1 µm particle size modeling assumption for the scavenging coefficient results in generally conservative predictions over the range of sample locations assessed, (2) based on the slope (< 1.0) of the regression line, the model generally overpredicts deposition at low impact locations (farther out) and underpredicts deposition at high impact locations (closer in), and (3) the assumption that all or most of the mercury is in vapor form (as specified in the EPA risk assessment guidance for combustion facilities[1]) resulted in the widest P/O value dispersion and the lowest regression coefficient.

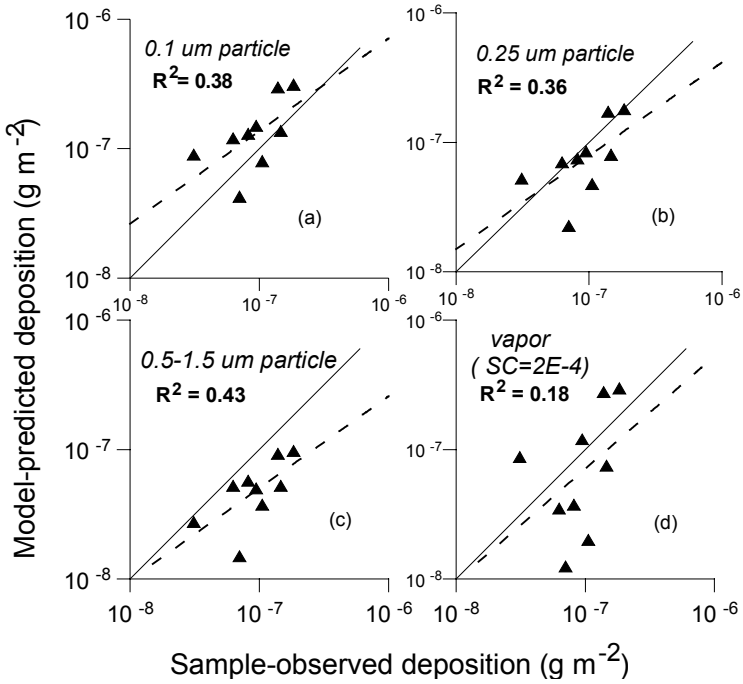

Figure 2. ISCST3 model performance using scavenging coefficients for (a) a 0.1 µm particle, (b) a 0.25 µm particle, (c) a 0.5 – 1.5 µm particle, and (d) the vapor form. Perfect model performance is indicated by the solid line.

Fraction Deposited Locally

Because of the limited number of sampling locations, an accurate estimate of the fraction of total emissions deposited locally (within 50 km) cannot be made. However, examination of the P/O ratios in Table 2 provide some insight on this parameter because they are based on the modeling assumption that all (100%) of the emissions were reactive Hg(II) and subject to local deposition (the modeling used the total measured emission rate at the stack). When averaged across all ten sampling locations, the P/O ratios ranged from 0.5 to 1.6, depending upon the assumption for scavenging coefficient, but was very close to 1 (1.1) for the most likely form (mostly vapor). This performance suggests that, for this particular source, the assumption that 100% of the emissions are available for local deposition is reasonable for risk assessment purposes. If the emissions had been scaled to account for the 48% local deposition partitioning, as recommended in the EPA HHRAP, the P/O ratios would have ranged from 0.77 to 0.24 and would significantly underpredict the deposition.

Conclusions

Mercury concentrations in snow in the immediate vicinity (within about 12 km) of the NWCF (μ= 5.3 ng L^{-1}, excluding the BSB off-site location) are relatively low and similar to those measured in other remote U.S. locations. The calculated total annual deposition (0.12 to 2.06 ug m^{-2} y^{-1}) is lower than expected and may due to the low annual precipitation rates in the area. Another possible explanation is that there may have been

unaccounted for losses in the snow between snowfall and sampling (a 1 – 2 week time period during which some snowmelt occurred). One potential mechanism for loss is reduction of the deposited Hg(II) in the snow and subsequent re-emission to the atmosphere as Hg(0).

The ISCST3 model seems to have done a good job of predicting NWCF mercury deposition rates in the areas of maximum impact (within 12 km of the NWCF). Deposition of NWCF emissions was best modeled using scavenging coefficients for small (0.1 μm) particulate and the assumption that all of the mercury is subject to local deposition. The EPA default assumptions for THg physical form and partitioning in combustion facility risk assessment modeling – mostly vapor that is 48% locally deposited -- would have underpredicted NWCF mercury fallout and resulted in the least certain predictions (greatest dispersion of P/O ratios).

This study was not intended to be a rigorous model validation but a test of methods that can be used to develop improved model input parameters for mercury. Future work at the INEEL is planned to develop larger data sets of mercury in precipitation around sources to further refine model performance and better understand the local fate and transport of mercury in this region.

Acknowledgements

We wish to thank INEEL High Level Waste Program Managers for their continued support of this project. This work was supported by the U.S. Department of Energy, Assistant Secretary for Environmental Management, under DOE Idaho Operations Office Contract DE-AC07-99ID13727.

References

1. "Human Health Risk Assessment Protocol for Hazardous Waste Combustion Facilities – Peer Review Draft," U.S. Environmental Protection Agency Office of Solid Waste, EPA530-D-98-001A, 1998.

2. "User's Guide for the Industrial Source Complex (ISC3) Dispersion Models," U.S. Environmental Protection Agency: Research Triangle Park, NC, 1995, EPA-454/B-95-003a-b.

3. "Mercury Study Report to Congress," Volumes I through VIII, U.S. Environmental Protection Agency: December 1997; EPA/452/R-97-001.

4. Jindal, M.; Heinold, M. "Development of Particulate Scavenging Coefficients to Model Wet Deposition from Industrial Combustion Sources," Paper 91-59.7, *Proceedings of the 1991 Annual Meeting and Exhibition of the Air and Waste Management Association*, Vancouver, BC, Canada, June 16-21, 1991, 1-14.

5. Olson, M. L.; DeWild, J.F. "Techniques for the Collection and Species Specific Analysis of Low Levels of Mercury in Water, Sediment and Biota", D.W. Morganwalp and H. T. Buxton (eds.), *U.S. Geological Survey Toxic Substances Hydrology Program--Proceedings of the Technical Meeting*, Charleston, SC, USA, March 8-12, 1999--Volume 2 of 3—Contamination of Hydrologic Systems and Related Ecosystems: U.S. Geological Survey Water-Resources Investigations Report, 1999, 99-4018A, 191-201.

6. Susong, D. D.; Abbott, M.L.; Krabbenhoft, D.P. "Reconnaissance of Mercury Concentrations in Snow from the Teton and Wasatch Ranges to Assess the Atmospheric Deposition of Mercury from an Urban Area", Abstract H12b-06, *Eos*, *Transactions of the American Geophysical Union* 1999, 80, p. 46.

7. Scott, B. C. "Theoretical Estimates of the Scavenging Coefficient for Soluble Aerosol Particles as a Function of Precipitation Type, Rate and Altitude," *Atmospheric Environment* 1982, **16**, 1753-1762.

8. Petersen, G.; Munthe, J.; Pleijel, K.; Bloxam, R.; Vinod Kumar, A. "A Comprehensive Eulerian Modeling Framework for Airborne Mercury Species: Development and Testing of the Tropospheric Chemistry Module (TCM)," *Atmospheric Environment* 1998, **32**, 829-843.

9. Glass, G.E.; Sorensen, J.A.; Schmidt, K.W.; Rapp, G.; Yap, D.; Fraser, D. "Mercury Sources and Distribution in Minnesota's Aquatic Resources: Deposition." Part 1 of Chapter 4 (Mercury Washout from Precipitation: Atmospheric Sources) in *Mercury in the St. Louis River, Mississippi River, Crane Lake and Sand Point Lake: Cycling, Distribution and Sources*, Report to the Legislative Commission on Minnesota Resources, Water Quality Division Minnesota Pollution Control Agency, St. Paul, MN, USA, April 1992.

10. Snyder-Conn, E.; Garbarino, J. R.; Hoffman G.L.; Oelkers, A. "Soluble Trace Elements and Total Mercury in Arctic Alaskan Snow", *Arctic* 1997, **50**, 201-215.

11. St. Louis, V.L.; Rudd, J.W.; Kelly, C.A.; Barrie, L.A. "Wet Deposition of Methylmercury in Northwestern Ontario Compared to Other Geographic Locations," *Water, Air, and Soil Pollut*. 1995 **80**, 405-414.

12. Greenberg, A.; Wojtenko, I.; Chen, H., Krivanek, S.; Butler, J.; Held, J.; Weis, P.; Reiss, N. "Mercury in Air and Rainwater in the Vicinity of a Municipal Resource Recovery Facility in Western New Jersey," *The International Symposium on Measurement of Toxic and Related Pollutants*, Durham, NC, USA, May 8, 1992.

13. Miller, W.C.; and Hively, L. M. "A Review of Validation Studies for the Gaussian Plume Atmospheric Model," *Nuclear Safety* 1987, **28**, 522-530.

Key Words List

mercury, deposition, snow, ISCST3, model, snow