

Mercury accumulation in snow on the Idaho National Engineering and Environmental Laboratory and surrounding region, southeast Idaho, USA

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Abstract Snow was sampled and analyzed for total mercury (THg) on the Idaho National Engineering and Environmental Laboratory (INEEL) and surrounding region prior to the start-up of a large (9–11 g/h) gaseous mercury emission source. The objective was to determine the effects of the source on local and regional atmospheric deposition of mercury. Snow samples collected from 48 points on a polar grid near the source had THg concentrations that ranged from 4.71 to 27.26 ng/L; snow collected from regional background sites had THg concentrations that ranged from 0.89 to 16.61 ng/L. Grid samples had higher concentrations than the regional background sites, which was unexpected because the source was not operating yet. Emission of Hg from soils is a possible source of Hg in snow on the INEEL. Evidence from Hg profiles in snow and from unfiltered/filtered split samples supports this hypothesis. Ongoing work on the INEEL is investigating Hg fluxes from soils and snow.

Keywords Mercury · Snow · Idaho

Introduction

Mercury (Hg) from anthropogenic sources has been shown to increase Hg deposition on local, regional, and global scales (Mason and others 1994; Sorensen and others 1994; Dvornich and others 1999). The atmosphere is a major pathway for transport of Hg to terrestrial and aquatic ecosystems (Bloom and Watras 1989; Mason and others 1994; St Louis and others 1995; Landers and others 1998; Glass and Sorensen 1999). A large Hg point-emissions source on the US Department of Energy (DOE), Idaho National Engineering and Environmental Laboratory (INEEL) is used in this study to investigate Hg concentrations in snow near this source and in the region. The DOE is responsible for the management of national laboratories in the USA. This responsibility includes the environmental stewardship and clean-up of these lands after decades of research and industrial use as part of the nation's nuclear energy and weapons production infrastructure. The INEEL, located in southeastern Idaho, USA (Fig. 1), is the site of research reactors, fuel processing facilities, and nuclear waste storage. Mercury is one of the US Environmental Protection Agency's (EPA) priority contaminants because of its toxicity, persistence, and bioaccumulation in the food web, and its demonstrated link to human health effects (US EPA 1997). Mercury at the INEEL is contained in liquid waste from processing spent nuclear fuel rods where it was added as a catalyst to aid fuel dissolution. Tanks on the INEEL hold about 4 million liters of waste, which contain about 2,600 kg of Hg. Liquid wastes were processed on the INEEL at the Idaho Nuclear Technology and Engineering Center (INTEC) in a low-temperature fluidized bed combustion unit that converts liquid waste to calcine solids. The calcining facilities at INTEC operated intermittently for about 37 years before being shut down in June 2000. Offgas from INTEC is composed primarily of NO₂, water vapor, fuel combustion byproducts, and Hg. The offgas air pollution control equipment consists of a quench tower, cyclone, acid scrubber, silicon-gel absorbers, and four banks of high efficiency particulate air (HEPA) filters. Recent stack tests measured an average short-term mercury emission rate of 9–11 g/h, approximately 1.5% of the feed rate (Abbott and others 2000). Most of the Hg emitted

Received: 1 January 2001 / Accepted: 30 April 2002
Published online: 3 July 2002
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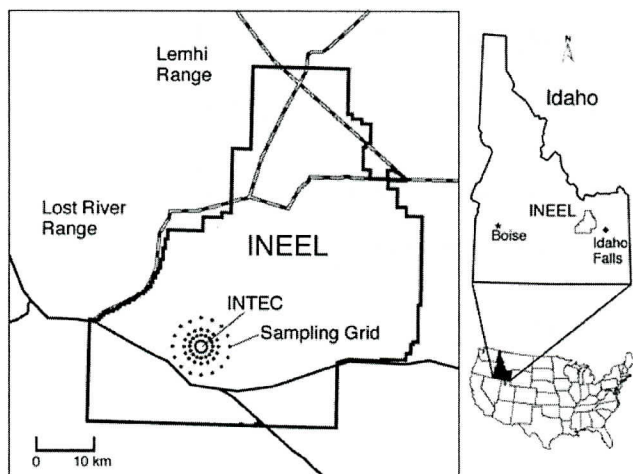


Fig. 1

Location of the Idaho National Engineering and Environmental Laboratory (INEEL) and Idaho Nuclear Technology and Engineering Center (INTEC), southeast Idaho

(88%) is reactive gaseous mercury (RGM), which is thought to be HgCl_2 . The remaining emissions are believed to be submicrometer particulates and a negligible amount of Hg^0 .

The INTEC emission source is particularly useful for the study of Hg transport and fate because (1) mercury emissions from the facility are well characterized and relatively high, (2) the source is located in a remote region that is not influenced by other significant localized or regional mercury sources, and (3) there are large climatic and ecosystem gradients (and resulting depositional processes) ranging from arid desert conditions to alpine areas with substantial snowfall. The nearest other potentially significant mercury sources (coal combustion) are located at a distance of more than 250 km in the direction of prevailing winds.

Snow is used in this study as a time-integrating sampling medium that collects dry and wet deposition. Snow is relatively easy and inexpensive to sample, and numerous samples can be collected to investigate the spatial distribution of atmospheric deposition. Although mercury in precipitation has been measured in a number of studies (Bloom and Watras 1989; Glass and others 1990; Fitzgerald and others 1991; Vandal and others 1993; Sorensen and others 1994; Burke and others 1995; St Louis and others 1995; Snyder-Conn and others 1997; Watras and others 2000), only a limited number of studies have been conducted near well-characterized mercury point sources (Dvonch and others 1999), especially in isolated areas with low background concentrations of atmospheric Hg.

The purpose of this paper is to present results of chemical analysis of snow samples collected on the INEEL and at a network of regional background sites and to compare the two sampling networks to attempt to assess the effects of the INTEC emissions on regional Hg atmospheric deposition. Total Hg (THg) concentrations in snow for the two sampling networks are presented and compared, and profiles of THg in the snow cover are compiled for selected sites.

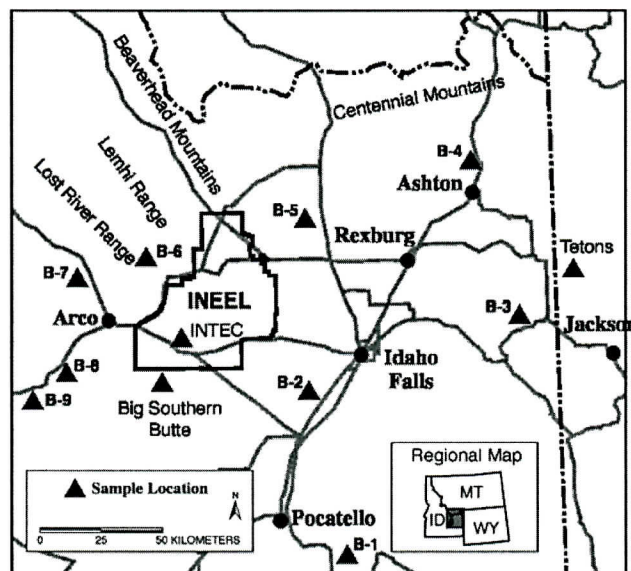


Fig. 2

Location of regional background sampling sites, southeast Idaho

Methods

Two sampling networks were used as part of this study. A network of nine regional background sites 40–150 km from the INTEC was used to assess regional Hg deposition (Fig. 2). Four sets of samples were collected from December 1999 to March 2000 at these regional sites. In addition to these regional background sites, samples were collected during the same months at five sites along an elevation transect on the west side of the Teton Range. The samples from the Teton Range are included with the regional background sites for discussion purposes.

The snow-sampling network on the INEEL was a 64-point polar grid with 16 radials and 4 circles at 1, 2, 3, and 5 km from the INTEC (Fig. 1). This network was used to assess local Hg deposition from the INTEC. The INTEC calciner shut down during the fall of 1999 for maintenance and repairs and was scheduled to restart in mid-January 2000. Samples were to be collected on the grid twice; before the INTEC started processing waste in January 2000 and again after the first major snowfall after INTEC had been in operation. However, INTEC did not start processing waste until mid-March 2000 because of technical difficulties, by which time nearly all the seasonal snow cover had melted; therefore, the INTEC grid was sampled only once in early January 2000.

Snow samples for Hg analyses were collected from snow pits as integrated samples of the entire snow cover where the snow cover was shallow (<30 cm). At sites with a deeper snow cover (>30 cm), composite samples of the snow cover were collected as well as interval samples based on the snow stratigraphy. Individual storm events were sampled where major storm cycles were visible in the snow pit. The face of the snow pit was scraped with a cleaned plastic shovel prior to sampling. The shovel was pre-washed in dilute HCl, soaked for 24 h in ultra-purified

water, and bagged before being transported to the field. Snow samples were collected in 500 mL and 2 L Teflon bottles off the face of the snow pit with the sampler wearing shoulder-length gloves and using “clean hands/dirty hands” technique (Olson and DeWild 1999). Samples were shipped frozen to the USGS Wisconsin District Mercury Laboratory. Teflon sampling jars were precleaned in a 4 N ultra-trace pure hydrochloric acid (HCl) bath followed by a minimum of three rinses with reagent grade water and then partially refilled with 1% Omni Trace HCL and rinsed again with reagent grade water. The jar surfaces were air dried in a Hg-free class 100 laminar flow hood before being double-bagged in plastic bags (Olson and DeWild 1999).

Once in the laboratory, the samples were allowed to melt. Total Hg (THg) analysis was done by using EPA Method 1631: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry (CVAFS) with modifications (US EPA 1996). Method 1631 consists of three essential parts: (1) oxidation of Hg species to reactive mercury (Hg^{II}), (2) reduction of Hg^{II} to volatile mercury (Hg^0), and (3) detection of Hg^0 by CVAFS (Olson and DeWild 1999).

Laboratory-quality assurance and quality-control methods are described by Olson and DeWild (1999). Briefly, they include duplicate analysis of all samples and a third analysis of samples that had a percentage difference of greater than 10%, spike samples at a minimum of every 10 samples, and a quality-control check sample at the beginning of each run, after every tenth sample, and at the end of the runs. Bubbler blanks were used to correct for background contamination and calculate detection limits for each run. The method detection limit for THg was determined to be 0.04 ng/L (Olson and DeWild 1999). Field blanks and duplicates were collected every ten samples.

Results and discussion

Grid total mercury concentrations

Samples from 45 of the INTEC 64 grid points were collected during January 4–6, 2000, and analyzed for THg and major ions. All samples were composite samples of the entire snow cover. Because INTEC had not been operating since August 1999, there were no emissions during the period of snow accumulation. Grid points that were not sampled were not accessible. Snow depths at the sampling sites ranged from 6 to 12 cm. Concentrations of THg in snow on the INTEC grid ranged from 4.71 to 27.26 ng/L ($\mu = 9.77$ ng/L; $\text{SD} = 4.13$) and are shown in Fig. 3. The highest observed concentration was measured at 1 km from the INTEC. Concentrations generally were highest near INTEC and decreased away from the facility. This would be the expected pattern around an operating point-emissions source. However, there were no documented emissions during the snow accumulation period from early December 1999 to January 2000. The observed elevated concentrations around INTEC and the associated

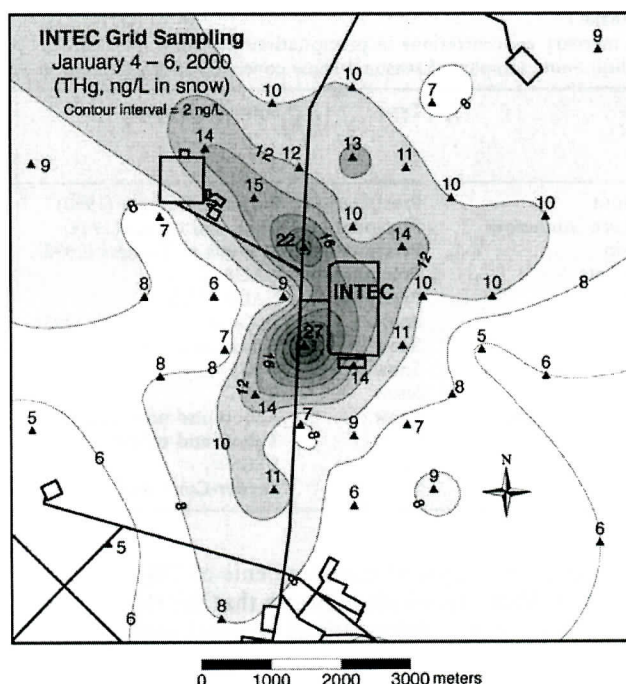


Fig. 3 Total mercury concentration in snow, INTEC grid, southeast Idaho, January 4–6, 2000

deposition pattern were unexpected and raise new questions about the source of Hg in the snow.

Background total mercury concentrations

A total of 95 snow samples were collected from background sites including the Teton Range sample sites. Not all of the regional background sites were sampled four times because of a lack of snow at some sites. This data set included composite and depth-interval samples of snow cover. The analytical results for the entire data set and for the composite samples ($n = 34$) are broken out separately because the composite samples are best used for comparison with the INTEC grid samples, which were composites of the snow cover as well. THg concentrations ranged from 0.89 to 16.61 ng/L ($\mu = 3.73$, $\text{SD} = 2.44$) for the complete background data set and from 1.24 to 8.52 ng/L ($\mu = 3.31$, $\text{SD} = 1.45$) for the background composite samples.

THg concentrations observed in snow collected on the INTEC grid are elevated as compared to THg concentrations measured in snow in other areas of North America (Table 1). St Louis and others (1995) compiled a summary of mean THg concentrations in precipitation in Scandinavia. THg concentrations ranged from 5.8 to 90 ng/L, with many of the studies reporting concentrations in the 10 to 40 ng/L range. THg concentrations from the snow samples from the INTEC grid are less than in the Scandinavian studies, in which Hg in precipitation has been attributed to anthropogenic sources. This indicates that facilities on the INEEL may be contributing to mercury deposition in the local area (<5 km). Concentrations of THg in snow from the regional background sites are

Table 1

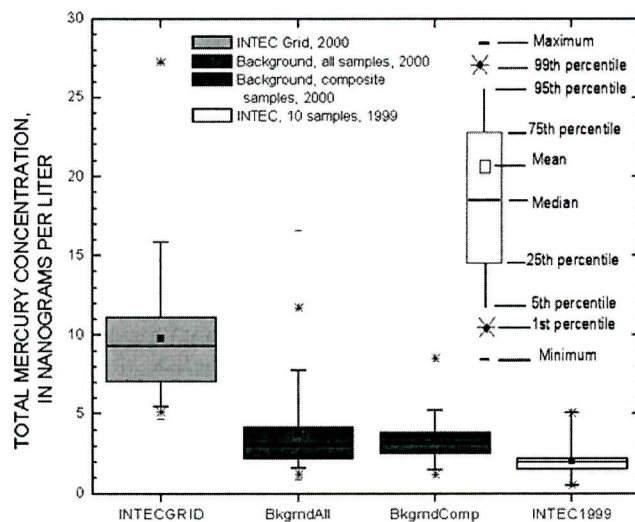
Total mercury concentrations in precipitation and snow. NADP-MDN National Atmospheric Deposition Program – Mercury Deposition Network; Snow samples of seasonal snow cover; USGS US Geological Survey unpublished data, Susong and others 2000

Site	Type	Source/reference	Year(s) AD	Average total mercury concentration (ng/L)	Average total mercury deposition (ng/m ² /year)
Vermont	Precipitation	Burke and others (1995)	1992–1993	8.3	9,260
Northern Minnesota	Precipitation	Glass and others (1999)	1990–1995	10.9	7,400
Ontario	Precipitation	St Louis and others (1995)	1992–1994	4.04	–
Minnesota	Precipitation	NADP-MDN	1997–1999	14	6,991
Colorado	Precipitation	NADP-MDN	1999	10	9,199
Ontario	Snow	St Louis and others (1995)	1994	1.08	–
Wisconsin	Snow	Bloom and Watras (1989)	1988	4.05	–
Upper Fremont Glacier	Snow	USGS	1999	4	–
Wyoming	Snow	USGS	1998–00	4	–
Idaho INTEC grid	Snow	Abbott and others (2002)	1999–00	9.77	–
Idaho background	Snow	Abbott and others (2002)	1999–00	3.73	–
Kyrgyzstan	Snow	USGS	2000	2.01	–
Alaska	Snow	Snyder-Conn and others (1997)	1994	3.9	–

similar to other published measurements of THg in snow in the USA (Table 1), which indicates that the INEEL may not be contributing substantially to regional atmospheric Hg deposition. Stack emissions from the INTEC are predominantly reactive gaseous mercury (RGM), which is highly reactive and would be expected to be scavenged out of the atmosphere quickly.

Box plots (Fig. 4) compare the THg concentrations for grid samples from January 2000 and regional background sites from December 1999 through March 2000. THg concentrations in snow from the grid are greater than THg concentrations in snow at the background sites for the entire data set and for the composite samples. This observation is confirmed statistically with the Wilcoxon rank-sum test, which indicates that grid concentrations are greater than background concentrations. INTEC was operating during the winter of 1999 and ten snow samples were collected during February 11–12, 1999, near the facility in the area of predicted maximum fallout (Abbott and others 2002). THg concentrations from these samples are shown in Fig. 4 and are less than THg concentrations from the grid samples collected in January 2000 when INTEC was shut down. This was a very unexpected result and is problematic to explain because the major source of Hg emissions was not operating when the snow fell on the grid.

Since the major source of Hg emissions was not operating, there must be another source of Hg on the INEEL. Two other possible sources were investigated. One is that another facility is emitting Hg on the INEEL. All emissions from the INEEL, however, are carefully catalogued and there are no other known sources of Hg emissions. Emission of residual Hg from the INTEC stack and pollution control equipment is another possible undocumented source, but no offgas measurements have been made when the INTEC was shut down. Another mechanism for the accumulation of Hg in snow at the INEEL could be re-emission of Hg from soils. In this hypothesis, Hg accumulates in the snow because of re-emission of Hg from soils after the snow has been deposited on the ground. Soils on the INEEL are low-organic, mineral soils

**Fig. 4**

Box plots of total mercury concentrations in snow from INTEC grid and regional background sites, southeast Idaho, 1999–2000

and thus would not readily bind Hg. The winter of 1999/2000 also was characterized by above-normal temperatures and the ground did not freeze.

Data from two of the regional background sites can be interpreted to provide evidence for possible re-emission of Hg from soils. The two sites are located at similar elevations and accumulate 1–1.5 m of snow. However, profiles of THg concentrations in snow compiled from interval samples collected from snow pits are quite different (Figs. 5, 6). At site B-4 (Ashton Hill), the highest THg concentrations were measured in the first 20 cm of snow above the ground but decrease over time during the snow accumulation season (Fig. 5). In contrast, THg concentrations in profiles from B-3 (Pine Creek Pass) are low near the ground and higher in the overlying snow cover (Fig. 6). The sampling sites are very similar with the exception that site B-4 is underlain by a 1.4–2 million-year-old rhyolite ashflow tuff and site B-3 is underlain by

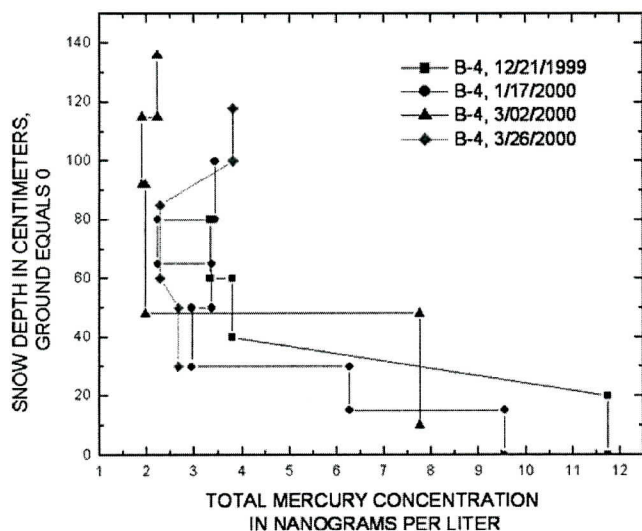


Fig. 5
Profiles of total mercury concentrations in snow at site B-4, Ashton Hill, southeast Idaho

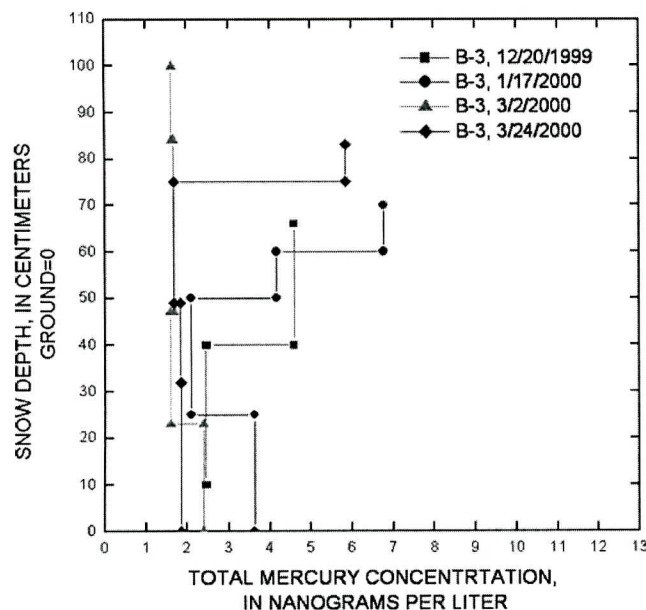


Fig. 6
Profiles of total mercury concentrations in snow at site B-3, Pine Creek Pass, southeast Idaho

Paleozoic shale. The tuff could be a source of Hg but this cannot be determined explicitly without conducting soil Hg flux measurements. An alternative explanation for the

elevated Hg at site B-4 is that there were higher concentrations of Hg in the snow when it fell. This explanation cannot be ruled out. However, the higher concentrations of THg in snow near the ground are 2–3 times those of the rest of the snow at the site, which suggests that the air mass from which this snow originated was unusual. It is interesting also to note the decrease in THg concentrations over the sampling period in the snow near the ground. This change may be the result of any number of processes from gaseous Hg fluxes through the snow to recrystallization and metamorphic changes of the snow. Recent work investigating the snow–air mercury transfer by Lalonde and others (2002) has demonstrated that a substantial fraction (54%) of Hg deposited in snow can be lost from the snow back to the atmosphere. These results indicated that Hg concentrations in snow are not stable, particularly in the first 24–48 h after deposition. Lalonde and others (2002) only present data from measurements from a shallow snow cover (<25 cm); thus it is uncertain how applicable their results are to deeper snow covers. THg concentrations at site B-3 appear to support the hypothesis of losses of Hg from new snow because of the decrease in Hg concentration with depth or age. Notably, there is a lower limit to this decrease in concentration at about 2 ng/L. This lower limit may be the particulate fraction of the THg concentration. The profile for B-4 appears to be inverted, indicating a possible soil Hg source as discussed earlier, yet there is a lower limit to the THg concentrations at this site as well.

Selected samples from the grid and background sites were split to create filtered and unfiltered samples and analyzed for THg. Filtered samples were melted, split, then filtered through a 0.45- μ m filter and processed the same as the unfiltered samples. Comparison of these split samples for the grid and background sites showed that the mean unfiltered THg concentration at the grid sites is higher than the mean for the background sites (Table 2). This also was true for the mean filtered THg concentrations; concentrations measured at the grid sites are higher than those at the background sites. However, the ratio of the mean unfiltered to filtered THg concentration (unfiltered THg/filtered THg) for the background sites is greater than that of the grid sites (Table 2) and provides additional evidence about the source of Hg in the snow at the INTEC grid sites. The majority of the mercury emissions from the INTEC are RGM, which is highly soluble and would contribute to the filterable fraction of THg in the local area. However, because INTEC was not operating when the snow accumulated, there must have been an additional unknown emissions source or the Hg may have been re-emitted from

Table 2
Mean filtered and unfiltered total mercury concentrations, grid and background sites

Sites	Number of samples	Mean unfiltered total mercury (ng/L)	Median unfiltered total mercury (ng/L)	SD unfiltered total mercury (ng/L)	Mean filtered total mercury (ng/L)	Median filtered total mercury (ng/L)	SD filtered total mercury (ng/L)	Ratio of mean unfiltered to filtered total mercury
Grid	5	8.5	8.72	1.4	3.48	3.94	0.92	1.52
Background	25	3.2	2.97	1.21	0.84	0.68	0.36	4

the soils. In this case, gaseous Hg^0 re-emitted from the soil would be either trapped in the snow or oxidized in the snow to become part of the filterable fraction. Either explanation is plausible but it is unlikely that there is an undocumented emissions source of Hg on the INEEL except perhaps residual Hg emissions from the INTEC stack.

Mercury in soils

Soils were sampled on the INTEC grid in July 2000 to determine Hg concentrations near the INTEC. The results of this work are presented by Abbott and others (2002), who found that Hg concentrations from the grid soil samples were distributed in a halo pattern around the facility with concentrations decreasing with distance away from the stack. However, the concentrations of Hg in the soils (23 ng/g maximum) were much lower than expected considering the long-term facility emissions rates and were lower than Hg concentrations reported in the literature for similar soils (Abbott and others 2002). This indicates that either Hg emissions from the facility were not deposited locally or that Hg was deposited near the facility only to be later re-emitted back to the atmosphere. The latter is more probable because previous model validation studies and sampling have shown that much of the fallout is within 15 km of the facility (Abbott and others 2000).

Conclusions

Mercury concentrations in snow were measured on two sampling networks in southeast Idaho during the winter of 1999/2000. Mean total mercury (THg) concentrations in 45 samples collected on a 64-point polar grid around a large Hg emissions source on the INEEL were higher than mean THg concentrations in samples collected at a network of regional background sites 40–150 km from the source. The snow samples from the grid accumulated on the INEEL during a period when the Hg emissions source was off line for maintenance and repair and there were no emissions. These results were unexpected and raised new questions about the source of Hg in the snow on the INEEL. Profiles of THg in snow at two of the background sites were examined to see if there were Hg gradients in the snow. The sites were similar in elevation, vegetation, and snow accumulation. One of the profiles had 2–3 times greater concentrations of Hg in snow collected from 0–20 cm above the ground. At this site the underlain bedrock is a young rhyolitic ashflow tuff, whereas the other site is underlain by Paleozoic shales. Data from these profiles could be evidence that Hg can be emitted from soils and accumulated in the snow or that the early season snow at the site contained elevated concentrations of Hg. Selected samples from the grid and background sites were split into filtered and unfiltered samples. The ratio of unfiltered THg to filtered Hg was determined. The mean ratio for the grid sites was less than the background sites. This indicates that there is a greater filterable fraction of THg in snow from the grid site than the background sites. Hg emissions from the source on the INEEL are

predominantly RGM, which would contribute to the filterable fraction in the snow. Another possible source and explanation for the unfiltered/filtered ratio is the re-emission of Hg from soils into the overlying snow cover. Decades of intermittent operation of the source may have created a pool of Hg in the soil that is re-emitted over time to the atmosphere.

Soil sampling and analysis for THg on the grid showed a halo pattern around the source with concentrations decreasing with distance. However, the mean THg concentration in the soils was below reported values for similar alkaline low organic, mineral soils. A possible explanation for the lack of THg in the soils is that much of THg deposited in the area has been re-emitted to the atmosphere. This intuitively makes sense since there is sparse vegetation, little organic material, and relatively high solar radiation fluxes.

The unexpectedly high THg concentrations in snow collected from the grid in this study have possibly identified processes that have not been considered in other mercury studies. Ancillary data from the regional background sites support the hypothesis that the re-emission of Hg from soils may contribute Hg to the overlying snow cover. Future work at the INEEL will include soil Hg flux measurements, Hg flux measurements over snow, more detailed sampling of the seasonal snow cover, and sampling of gaseous Hg within the snow.

Acknowledgements This work is supported by the US Department of Energy, Assistant Secretary of Environmental Management, under DOE Idaho Operations Office Contract DE-A107-98ID13598 A008. Special thanks to George Schneider and Joel Case, DOE Idaho Operations Office, for their support.

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