Concentration and Dry Deposition of Mercury Species in Arid South Central New Mexico (2001−2002)

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This research was initiated to characterize atmospheric deposition of reactive gaseous mercury (RGM), particulate mercury (HgP; <2.5 μm), and gaseous elemental mercury (Hg^0) in the arid lands of south central New Mexico. Two methods were field-tested to estimate dry deposition of three mercury species. A manual speciation sampling train consisting of a KCl-coated denuder, 2.5 μm quartz fiber filters, and gold-coated quartz traps and an ion-exchange membrane (as a passive surrogate surface) were deployed concurrently over 24-h intervals for an entire year. The mean 24-h atmospheric concentration for RGM was 6.8 pg m^{-3} with an estimated deposition of 0.10 ng m^{-2} h^{-1}. The estimated deposition of mercury to the passive surrogate surface was much greater (4.0 ng m^{-2} h^{-1}) but demonstrated a diurnal pattern with elevated deposition from late afternoon to late evening (1400−2200; 8.0 ng m^{-2} h^{-1}) and lowest deposition during the night just prior to sunrise (2200−0600; 1.7 ng m^{-2} h^{-1}). The mean 24-h atmospheric concentrations for HgP and Hg^0 were 1.52 pg m^{-3} and 1.59 ng m^{-3}, respectively. Diurnal patterns were observed for RGM with atmospheric levels lowest during the night prior to sunrise (3.8 pg m^{-3}) and greater during the afternoon and early evening (8.9 pg m^{-3}). Discernible diurnal patterns were not observed for either HgP or Hg^0. The total dry deposition of Hg was 5.9 μg m^{-2} year^{-1} with the contribution from the three species as follows: RGM (0.88 μg m^{-2} year^{-1}), HgP (0.025 μg m^{-2} year^{-1}), and Hg^0 (5.0 μg m^{-2} year^{-1}). The annual wet deposition for total mercury throughout the same collection duration was 4.2 μg m^{-2} year^{-1}, resulting in an estimated total deposition of 10.1 μg m^{-2} year^{-1} for Hg. On one sampling date, enhanced HgP (12 pg m^{-3}) was observed due to emissions from a wildfire approximately 250 km to the east.

Introduction

During the past decade, the persistence of mercury (Hg) concentrations in fish tissues above federal and state advisory

levels has led to increased attention to its global biogeochemical cycle. Mercury-contaminated fish stocks in areas remote from anthropogenic and natural sources reflect atmospheric deposition of Hg as the dominant source (*1, 2*). The complexity of the intermediate processes between Hg deposition and bioaccumulation results in a nonlinear relationship among transport, deposition, and concentrations in fish tissue (*3*). Accurately characterizing the input to any given region is further complicated by the different depositional properties (both wet and dry) of the three major airborne fractions (reactive gaseous, RGM; particulate bound, HgP; and gaseous elemental, Hg^0). While Hg^0 in the planetary boundary layer is generally greater than either airborne fraction, the high water solubility and deposition velocity of RGM, relative to Hg^0, implies that RGM will play a strong role in the overall deposition of Hg (*4*).

Analysis of a regional Hg transport model suggests uncertainty in the contribution of the dry deposition fraction to the total deposition in arid regions with low levels of wet deposition (*5*). Wet deposition of Hg is a reflection of the complex interaction of local atmospheric concentrations, input of background air, and wet deposition processes (*6*). As long as rain events occur and are captured, an accurate characterization of Hg in wet deposition can be obtained. The sporadic and seasonal patterns of rain events in arid regions such as the southwestern U.S. (which often receives less than 20 cm of rainfall annually) highlights the variability of the contribution of wet deposition to the total deposition and argues that dry deposition should also be measured directly.

Since 1996, the Mercury Deposition Network (MDN) has been monitoring regional wet deposition patterns of total Hg (THg) and methylmercury in rain and snow as part of the National Atmospheric Deposition Program. Weekly precipitation samples are collected at over 94 sites in the United States, Mexico, and western and eastern Canada by use of precleaned glassware and special handling procedures (http://nadp.sws.uiuc.edu/mdn/; see User's Manual). Since 1997, THg has been characterized in precipitation at an MDN site near Caballo Reservoir in south central New Mexico. This site has exhibited relatively high median THg concentrations in annual precipitation (23.8 ng L^{-1}, range 1.0−276.4 ng L^{-1}; *n* = 150) compared to a median concentration of 9.7 ng L^{-1} across the MDN program for 1996−2003 (D. Gay, personal communication).

The paucity of information on the dry deposition of Hg (direct or inferred), as well as the importance for model validation, supports the need for a widespread measurement of Hg species in dry deposition. Thus, an objective of this study was to address these gaps by characterizing the concentrations and dry deposition of three dominant atmospheric Hg species with a manual method as a cost-effective approach. The inferential determination of dry deposition of Hg was selected because this approach uses ambient concentrations with deposition inferred from deposition velocities (*4, 7, 8*). The currently accepted method for ambient Hg speciation is automated and prohibitively expensive for widespread use (*9*). We adapted the same collection techniques in a manual speciation sampling train consisting of a KCl-coated denuder, 2.5 μm quartz fiber filters, and gold-coated quartz traps. Our second objective was to employ a direct measurement of dry deposition of Hg by use of an ion exchange (IX) membrane because the medium has been reported to effectively capture RGM (*10*). A third objective was to compare dry deposition of RGM estimated from concentrations obtained from the manual speciation

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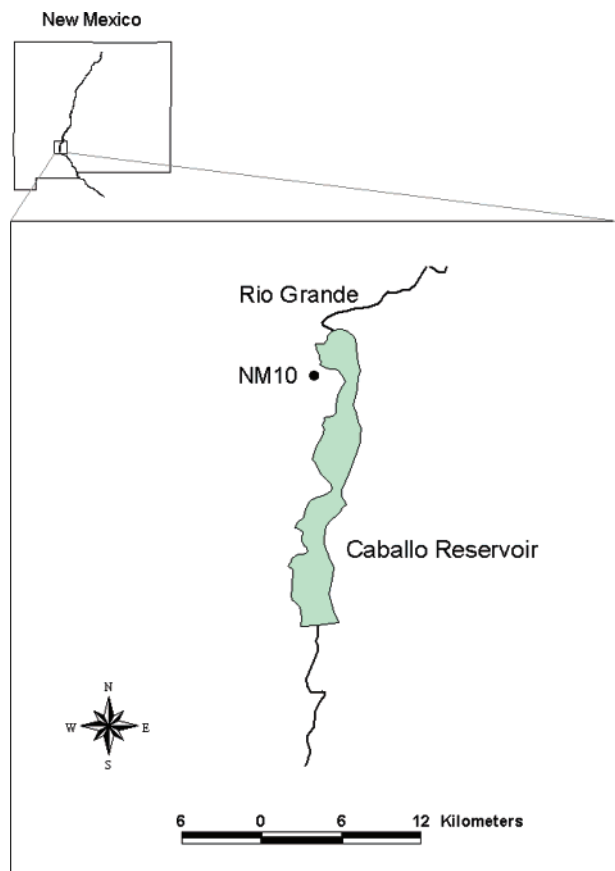
FIGURE 1. Location of the study area on the Rio Grande in south central New Mexico. The collection site was colocated with the Mercury Deposition Network collector (NM-10).

sampling train to dry deposition of RGM measured experimentally with a passive surrogate surface. A fourth objective compared the relative contribution of Hg from these dry deposition estimates to that obtained in wet deposition onto an arid landscape.

Materials and Methods

Sample Collection Site. The collection site in south central New Mexico was selected because it is distant from anthropogenic and natural sources of atmospheric Hg and home to an automated wet deposition collector (NM-10) for the MDN (Figure 1). Located approximately 96 km north of El Paso, TX, and Ciudad Juarez, Mexico, the site provided an opportunity to sample prevailing air masses from the south-southwest United States and Mexico during the summer months as well as air masses occasionally from the northwest United States throughout the remainder of the year. Deposition collection occurred on private property on the western side of Caballo Reservoir in Sierra County, New Mexico.

Manual Speciation Sampling Train. The design of the manual speciation sampling train (Figure 2) was based on

the methodology of an automated Tekran 1130/5 (9). Briefly stated, Hg species were separated in situ by sequential selective capture. As air entered the sampling train, particulates with aerodynamic diameters greater than 2.5 μm were removed by an elutriator impactor. Beyond the impactor, the etched annular surface coated with KCl selectively captured RGM. The lower diffusion constant of particulate-bound Hg and the lack of affinity of gaseous Hg^0 for KCl allow these to pass through. The denuded air stream was subsequently split into high (9.5 L min^{-1}) and low (0.5 L min^{-1}) flow-rate streams. The high-flow air stream passed through a quartz fiber filter (47 mm diameter, Whatman International Ltd., Maidstone, England) where remaining particulate matter of less than 2.5 μm in size was captured. The low-flow air stream passed through two traps consisting of gold-coated quartz chips (gold trap) to quantitatively capture Hg^0. The flow was regulated within each sampling train by a diaphragm pump, needle valve, and mass flow controller.

The manual speciation sampling trains were handled, cleaned, and assembled by ultraclean techniques. Temperature data loggers (Hobo, ONSET, Bourne, MA) were placed inside the shipping case containing the sampling trains to record temperature fluctuations to the field site and return to the analytical laboratory. Between sample collections, each sampling train was completely disassembled and all fittings and components were acid-cleaned in 10% HCl, rinsed with double-deionized water, and dried in a Class-100 clean hood. After the sampling trains were connected to the mass-flow controller in the field, the entire flow path was leak-checked. The entire sampling train, including particulate filters and gold traps, was contained within a vertically mounted 7.6 cm diameter poly(vinyl chlorine) (PVC) tube. The PVC tube was insulated by wrapping with an aluminum foil-lined plastic bubble-wrap. A silicone heat pad was mounted inside the tube and the internal temperature was maintained at approximately 40 °C. The inlet of the sample train extended 5 cm below the bottom cap of the PVC tube and was 1.5 m above ground level. Care was taken to approach or depart downwind from the sampling train.

The manual speciation sampling trains were deployed for three successive 8-h periods approximately every 2 months throughout 64 weeks (April 2001−August 2002). Each sampling train was deployed every 8 h for a 24-h period (1400−2200, 2200−0600, and 0600−1400) while a fourth was used as the field blank. Timing of deployment deviated slightly to coincide with sunrise. The first sampling train was deployed at approximately 1400 local time, removed at 2200 h, and replaced by the second sampling train. The second sampling train was removed just before sunrise at approximately 0600 and replaced with the final sampling train at the start of sunrise (official sunrise of 0.83° or 50 arc-minutes below the horizon; 11). The third sampling train was collected at approximately 1400 and followed by the deployment and immediate removal of the fourth sampling train representing the field blank. Average sample duration was 7 h and 37 min [457 min ± 34 (standard deviation), n = 21].
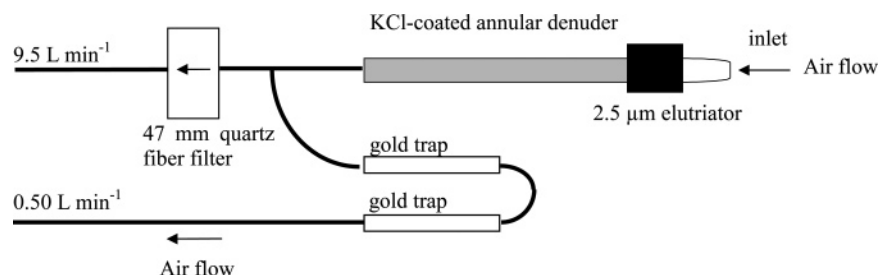
FIGURE 2. Schematic diagram of the manual speciation sampling train (not to scale). See text for details.

Surrogate Surface. An IX membrane (30 cm × 20 cm; Gelman Sciences, Ann Arbor, MI), made of polyethylsulfone and sulfonic acid groups, was selected as the surrogate surface to compare with the manual speciation sampling train. The membranes were hydrophilic and presumably efficient at adsorbing RGM and have shown some success in comparisons with annular denuders (*10, 12*). A study performed in 1998 at the same site in south central New Mexico demonstrated success using IX membranes with low method detection limits (0.95 ng m^{-2} h^{-1}) and moderate variability (mean = 4.47 ng m^{-2} h^{-1} ± 2.33, $n = 8$) (*13*).

In the laboratory under Class-100 conditions, the IX membranes were loaded onto 35.6 cm × 25.4 cm Plexiglas plates and double-bagged for shipment to the field site. At the site, clean latex gloves were worn to remove the plates from the bags and to mount them on top of a 10 cm × 10 cm untreated wood post 2.5 m above ground level. On each sampling date, an IX membrane was deployed concurrently with the manual speciation sampling trains and recovered every 8 h throughout a 24-h period. At the end of each sample period, the IX membranes were removed from the Plexiglas plates with clean latex gloves, folded and inserted into Teflon bottles (precleaned in ultraclean conditions), and returned to the analytical laboratory.

Sample Analysis. All mercury species were quantified by cold vapor atomic fluorescence spectrometry (CVAFS), which is the basis for U.S. EPA Method 1631 for the determination of ultratrace concentrations of Hg in water. The IX membranes and HgP filters from the manual speciation sampling train were subjected to BrCl digestion followed by $SnCl_2$ reduction and initially quantified by dual amalgamation CVAFS (Method 1631 equivalent). The analytical method for HgP, however, was altered part way through the study due to low ambient concentrations relative to the MDL (10 pg m^{-3}) and is described below with RGM analysis. As a result, HgP values for the first four sampling events were discarded (8 March 2001 through 30 July 2001).

Reactive gaseous mercury, HgP, and Hg^0 were analyzed by thermal desorption or conversion and quantification with a Tekran 2537A (Tekran, Inc., Toronto, Canada). For all species, Hg-free air was supplied as a carrier gas in excess to the upstream end of the thermal desorption stage and allowed to vent in a manner that would prevent ambient air mixing into the line. The Tekran 2537A was calibrated from the internal permeation source before and after every analytical set. For analysis of RGM and HgP, a pyrolyzer was installed downstream of the thermal desorption stage to ensure that any volatilized compounds were decomposed; this was followed by a high-purity soda-lime trap to protect the gold cartridges in the Tekran 2537 from attack by acidic gases. The integrity of the sampling train was tested by the injection of Hg^0 spikes with a gastight syringe (Hamilton) from a primary vapor standard (Tekran 2505).

Elemental mercury was desorbed by rapidly heating the gold traps with a Ni-chrome wire to 650 °C. The RGM fraction was desorbed and pyrolytically converted by connecting the entire sampling train, measuring the blank levels, and then rapidly heating the KCl-coated denuder in a tube furnace to 550 °C for 7.5 min. In all cases, the vast majority of the Hg was liberated in the first 2.5 min cycle. The remaining portion was detected in the second cycle with negligible quantities detected in the third cycle. Analysis of HgP required the quartz fiber filters to be loaded into a preblanked filter holder (short sections of quartz tubing) and the holder inserted into the upstream end of an oven tube, which was maintained at 650 °C. After the system blank was measured, the oven tube was tilted to allow the filter to enter the heated zone, which released Hg (presumably Hg^0) bound to the particulate matter.

Quality Control and Quality Assurance. The field blanks of the IX membranes varied greatly among sampling events at the beginning of the study despite the membranes originating from the same manufacturer's lot. The deposition measured in the first six events was not detectably different compared to the MDL (15 pg membrane^{-1}). Thus, a new lot of IX membrane was obtained which subsequently resulted in a 10-fold reduction in the MDL (1.5 pg membrane^{-1}). As a result, the deposition data generated from the first lot of IX membranes were discarded, resulting in the loss of six sample events (8 March 2001−14 December 2001).

In general, optimal weather conditions were required for the deployment of the sampling trains (i.e., no rain or wind). On one occasion (4 June 2001), windy conditions resulted in less than optimal handling procedures in the field and increased the potential for contamination and an overestimation of measured values. The data from this collection were discarded.

The MDL for ambient Hg speciation was calculated as three times the standard deviation of the method blanks. Thus, the MDL was 1.6 pg m^{-3} for RGM ($n = 6$), 0.57 pg m^{-3} for HgP ($n = 12$), 0.069 ng m^{-3} for Hg^0 ($n = 35$), and 1.5 ng m^{-2} h^{-1} ($n = 3$) for mercury collected on the IX membrane. When concentrations were at or below the MDL, then half the MDL was used to calculate means.

Inferential Calculation of Dry Deposition. The dry deposition rates can be inferred from Fick's first law of diffusion and a multiple resistance model where the overall deposition velocity (aerodynamic transport + quasilaminar + surface) is known (*14*). The mean vertical dry deposition flux, *F*, is calculated from the mean deposition velocity, v_d, and the mean local concentration, *C*, at some reference height (*14*) as $F = -v_d C$. There are currently few measurements of the deposition velocity of Hg species in any system. Therefore, we applied the annual mean dry deposition velocities used by Seigneur et al. (*8*) (0.5 cm s^{-1} for RGM, 0.1 cm s^{-1} for HgP, and 0.01 cm s^{-1} for Hg^0). The values for RGM and HgP are similar to those estimated for grasslands (*4*). Lindberg and Stratton (*4*) argued that the deposition velocity for Hg^0 was negligible below a compensation point (which is much greater than our measured concentrations); thus, we selected 0.01 cm s^{-1} as an upper limit. For RGM, both the surface resistance and quasilaminar resistances are assumed to be negligible on the basis of analogies to nitric acid. The overall deposition velocities are driven by the surface roughness length (and therefore land surface type) and the specific meteorological conditions of the boundary layer. Thus, our calculations have two important caveats. First, due to the lack of vegetation throughout the region of our study area, the surface roughness, deposition velocities, and calculated dry deposition rates, when compared to those of Lindberg and Stratton (*4*), Pai et al. (*7*), and Seigneur et al. (*8*), may be an overestimate and thus considered an upper limit. Second, due to the dependence of the deposition velocity on turbulent transport (and therefore the specific meteorological conditions of the boundary layer), the deposition velocity for any single sampling period in this study is not necessarily representative of the entire year.

Results and Discussion

Atmospheric Concentrations of Mercury Species. The concentrations of all Hg species were in agreement with other measurements made in North America and with modeled estimates (*8* and references therein). The 24-h mean concentration of RGM was 6.8 pg m^{-3} (range 1.6−25.0 pg m^{-3}) (Table 1). Despite high temporal variability, this average value was comparable to RGM obtained in two extensive field studies in Canada (ref *15*, 3.63 pg m^{-3} SD = 4.07; ref *16*, 3.0 pg m^{-3} SD = 11.0). The mean 24-h concentration of HgP was 1.52 pg m^{-3} (range 0.57−7.2 pg m^{-3}) (Table 1). These

TABLE 1. Average (24-h) Concentration and Dry Deposition of Reactive Gaseous Mercury (RGM), Mercury Deposition to a Surrogate Surface (IX Membrane), and Concentrations of Particulate-Bound Mercury (HgP) and Elemental Mercury (Hg⁰) in South Central New Mexico[a]

collection time (h)	atmospheric concn and deposition of RGM[b]			mercury deposition IX membrane[c]		atmospheric concn of HgP ($<2.5\ \mu m$)		atmospheric concn of Hg⁰	
	pg m^{-3}	n	ng m^{-2} h^{-1}	ng m^{-2} hr^{-1}	n	pg m^{-3}	n	ng m^{-3}	n
mean (24-h)	6.8	21	0.10	4.0	9	1.52	12		19
std deviation	6.59			3.42		1.99		0.37	
MDL	1.6	6	0.024	1.5	3	0.57	12	0.13	35
1400−2200	8.9 (7.97)	8	0.13	8.0 (2.48)	3	2.4 (3.28)	4	1.56 (0.48)	
2200−0600	3.8 (5.65)	7	0.056	1.7 (1.59)	3	1.5 (1.20)	4	1.60 (0.35)	8
0600−1400	7.4 (5.08)	6	0.11	2.2 (1.33)	3	0.62 (0.394)	4	1.59 (0.38)	6

[a] Sample collections were averaged within each collection time (standard deviation in parentheses) throughout the 64-week collection period from April 2001 to August 2002. Sample size is represented by n. MDL represents the method detection limit calculated as 3 times the standard deviation of the method blank. [b] Calculated by use of deposition velocity 0.5 cm s^{-1} for RGM (8). [c] Direct deposition of RGM and HgP presumably includes fine ($<2.5\ \mu m$) and coarse ($2.5-10\ \mu m$) particulates.
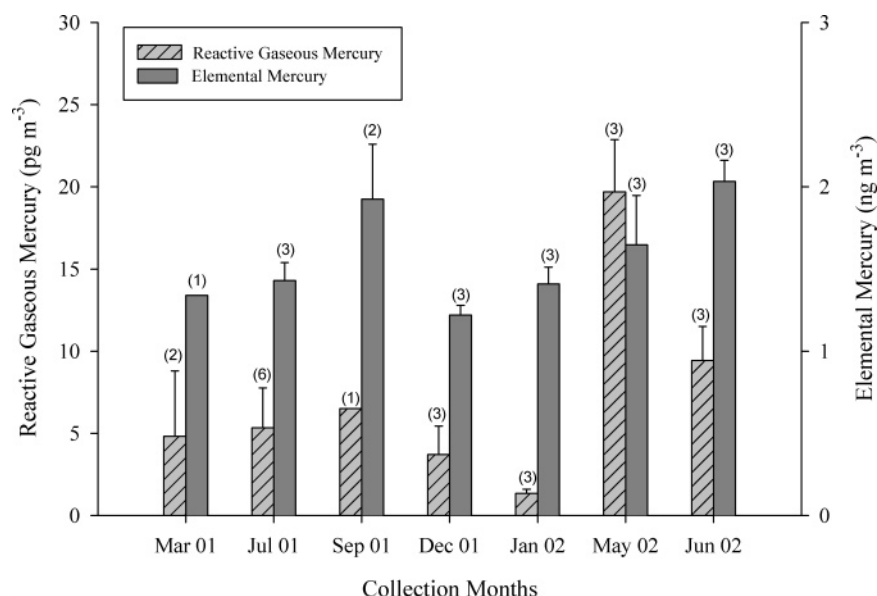

FIGURE 3. Concentrations of reactive gaseous mercury (picograms per cubic meter) and elemental mercury (nanograms per cubic meter) in ambient air throughout 24-h sample collections and averaged by month from 2001 through 2002 in south central New Mexico. Numbers in parentheses represents sample size. An asterisk indicates missing data for the collection date.

concentrations were comparable to average summertime "continental" air (2.9 pg m^{-3}) observed on the Washington coast by Weiss-Penzias et al. (17) but somewhat lower than an annual average concentration of 26 pg m^{-3} in St. Anicet, Quebec (16). One 24-h collection (27−28 June 2002) of HgP was removed due to concentrations that were elevated 12-fold from distant wildfires (range 4.5−12.0 pg m^{-3} in 24 h; see section below). The mean 24-h concentration of Hg⁰ was 1.59 ng m^{-3} (range 1.11−2.29 ng m^{-3}) (Table 1). This compares closely with "continental" air (1.43−1.67 ng m^{-3}) observed by Weiss-Penzias et al. (17) and is in general agreement with other measurements from differing land types (4, 16, 18).

Diurnal Variability and Seasonality. Despite high variability, we observed diurnal patterns for RGM concentrations, with the lowest values occurring during the night (2200−0600) (Table 1). When values were averaged for each collection period throughout the entire study, RGM concentrations were lowest at night (mean 3.8 pg m^{-3}, range 1.6−16.0 pg m^{-3}) compared to 7.4 pg m^{-3} between 0600 and 1400 (range 1.6−15.0 pg m^{-3}) and 8.9 pg m^{-3} between 1400 and 2200 (range 1.6−25.0 pg m^{-3}). The diurnal pattern suggests that RGM either was produced by local photochemistry in the boundary layers or was entrained down from the free troposphere during the daytime growth of the boundary layer (17). Similar diurnal patterns for RGM have

been observed by others (see refs 15 and 16) with concentrations greater in the daytime than nighttime and early morning. In the sample collection 1400−2200, it important to note that several hours of darkness (from 2 to 4 h) overlapped daylight sample collections and may explain the greater degree of variance associated with this sample time frame. Concentrations of HgP and Hg⁰ did not exhibit discernible diurnal patterns. Seasonal effects of RGM and Hg⁰ were not evident due to the relatively high variability associated with diurnal changes within each sample collection for the month (see Figure 3), while seasonal effects of HgP were not evident due to the small sample sizes.

Annual Estimates of Dry and Wet Deposition. From the mean 24-h measured concentrations of the three airborne Hg fractions, we estimated the dry deposition of RGM (0.88 μg m^{-2} year^{-1}; $n = 21$), HgP (0.025 μg m^{-2} year^{-1}; $n = 12$), and Hg⁰ (5.0 μg m^{-2} year^{-1}; $n = 19$) totaling 5.90 μg m^{-2} year^{-1}. The annual wet deposition for THg from NM-10 for the same time frame (2001−2002) was 4.2 μg m^{-2} year^{-1} (http://nadp.sws.uiuc.edu/mdn/), resulting in an estimated total Hg deposition of 10.1 μg m^{-2} year^{-1} in south central New Mexico. Landis and Keeler (19) obtained annual estimates of both wet (10.6 μg m^{-2}) and dry (9.7 μg m^{-2}) deposition of Hg over Lake Michigan using a hybrid model of high-resolution Eulerian-gridded meteorological data and
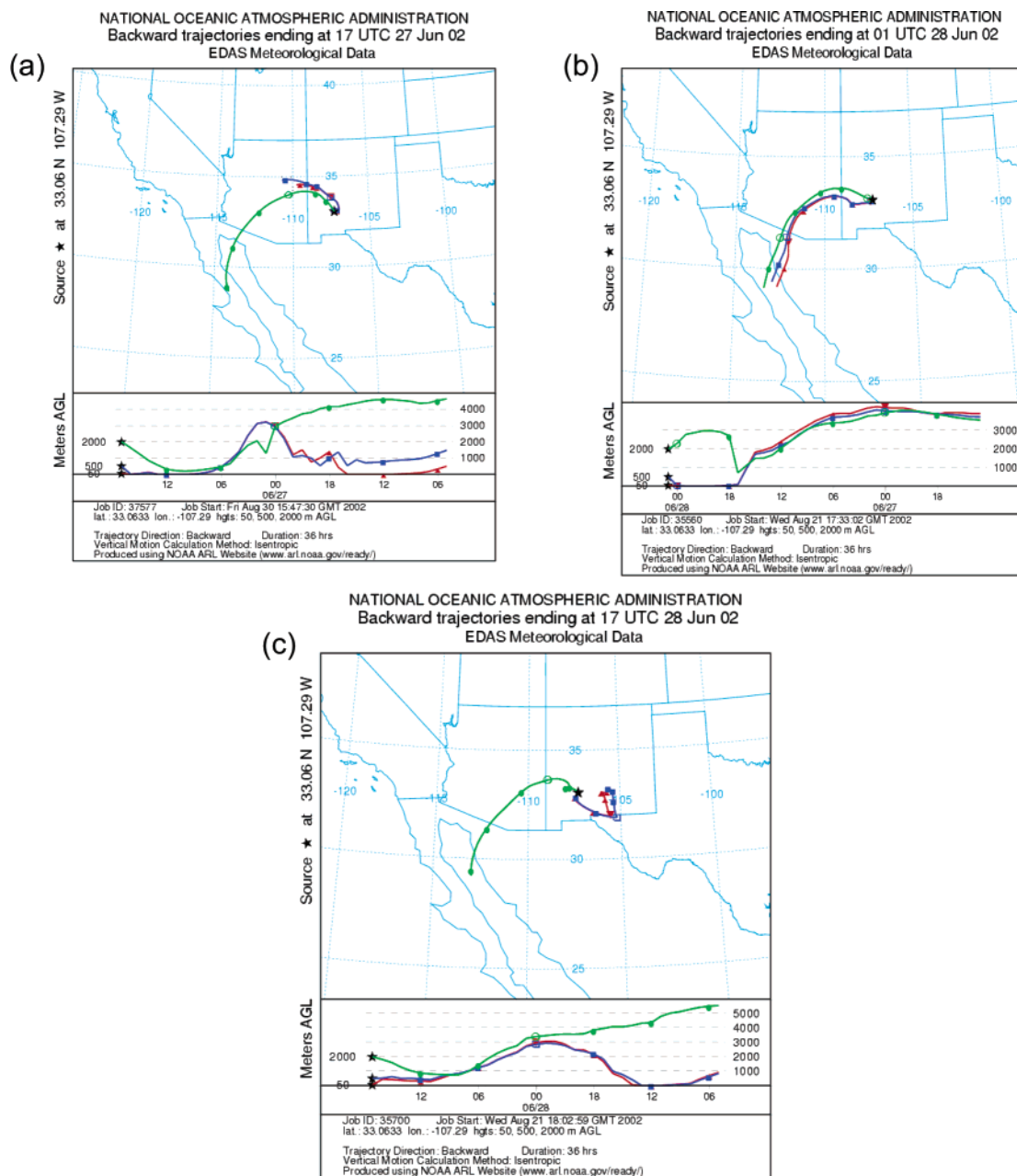
FIGURE 4. **4. HySPLIT 36-h back-trajectories for (a) 27 June 2002 at 1700 (UTC), (b) 28 June 2002 at 0100 (UTC), and (c) 28 June 2002 at 1700 (UTC). Ending air parcel height for each trajectory was 50, 500, and 2000 m above ground level.**

Hg in wet deposition. The author's elevated estimates may have been influenced by the effects of local urban areas as well as the varied land cover and climate associated with the Great Lakes region. An estimate of total mercury deposition in North America ranged from 14.3 to 19.8 μg m^{-2} year^{-1} (*20*). Our lower deposition rates are consistent with those of an arid site in a remote location in the absence of regional sources. It is important to note dry deposition of Hg^0 represented both the largest and most uncertain fraction of total deposition and suggests that future work be directed toward refining this term.

Dry Deposition of Reactive Mercury by Use of a Passive Surrogate Surface. The dry deposition of Hg by use of the IX membranes as a passive surrogate surface was much greater than for the sampling train, with an average deposition of 4.0 ng m^{-2} h^{-1} ($\pm$3.42, *n* = 9) (Table 1). The average deposition and range of values for the IX membranes were similar to values obtained with IX membranes from a previous study at the same site (*13*). A diurnal pattern was observed,

with deposition rates being much greater between 1400 and 2200 (mean 8.0 ng m^{-2} h^{-1}) compared to lower rates observed from 2200 to 0600 (mean 1.7 ng m^{-2} h^{-1}) and from 0600 to 1400 (mean 2.2 ng m^{-2} h^{-1}). The diurnal pattern in deposition may be linked to a number of factors including wind speed, turbulence, and the entrainment of reactive mercury from above the boundary layer. The particulate collection efficiency of the IX membrane is not well understood, thus we cannot predict a priori if it will collect HgP in a manner that is quantitatively related to particulates captured by manual speciation sample train. The membranes are composed of hydrophilic sulfonic acid groups that bind ionic Hg species. Presumably, the dry deposition values obtained from the IX membranes included the contribution of some fine (<2.5 μm) and coarse (2.5−10 μm) particulates.

When the mean 24-h deposition obtained from the IX membrane is extrapolated to an entire year (*n* = 9), the estimated deposition was 35.0 μg m^{-2} yr^{-1}. This annual average deposition was similar to results obtained in a

previous study with the same IX medium deployed at 72-h intervals at the same site in south central New Mexico (39.2 μg m^{-2} year^{-1}, $n = 8$) (*13*). While the annual average values from the passive surrogate surface were in disagreement with values for the manual speciation sampling train, several important issues should be noted. The direct measurements from the IX membrane sampled only a small fraction of a year and therefore may not be representative of the entire year. Despite the small number of samples, the agreement with a previous study (*13*) suggests that they are roughly representative. Finally, there is uncertainty in the dry deposition velocities of the Hg species and the collection properties of the IX membrane, resulting in comparisons that may be conjecture.

Enhancement of HgP during Wildfire-Influenced Sampling Event. One sample period in our study (27−28 June 2002) resulted in elevated HgP by nearly 12-fold when prevailing winds transported smoke to the collection site from a series of large-scale forest fires 250 km away in east-central Arizona. The Rodeo/Chediski fires (latitude 34.0986, longitude −110.433) began 18 June 2002 and burned 189 802 hectares (468 648 acres) until contained 7 July 2002. A series of 36h HySPLIT back trajectories were calculated for three collection intervals at 50, 500, and 2000 m above ground level (*21*). The trajectory for 27 June at 1700 (Figure 4a) indicated air parcel movement from directly over an area in which burning continued through 28 June 0100 (Figure 4b). Interestingly, during the third sample collection period (0600−1400), HgP concentrations decreased from 12.0 to 4.5 pg m^{-3} corresponding to a change in local wind direction and back-trajectory origin (Figure 4c). These observations support those of Friedli et al. (*22*) that forest fires are significant sources (considered to be a combination of natural and remissions) of Hg in the form of both Hg^0 and HgP.

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