

**Measurements of  
gaseous elemental  
mercury – Australia**

G. C. Edwards and  
D. A. Howard

# Air-surface exchange measurements of gaseous elemental mercury over naturally enriched and background terrestrial landscapes in Australia

G. C. Edwards and D. A. Howard

Department of Environment and Geography, Macquarie University, North Ryde, NSW, 2109, Australia

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Correspondence to: G. C. Edwards (grant.edwards@mq.edu.au)

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## Abstract

This paper presents the first gaseous elemental mercury (GEM) air-surface exchange measurements obtained over naturally enriched and background ( $< 0.1 \mu\text{g g}^{-1} \text{Hg}$ ) terrestrial landscapes in Australia. Two pilot field studies were carried out during the Australian autumn and winter periods at a copper-gold-cobalt-arsenic-mercury mineral field near Pulganbar, NSW. GEM fluxes using a dynamic flux chamber approach were measured, along with controlling environmental parameters over three naturally enriched and three background substrates. The enriched sites results showed net emission to the atmosphere and a strong correlation between flux and substrate Hg concentration, with average fluxes ranging from  $14 \pm 1 \text{ ng m}^{-2} \text{ h}^{-1}$  to  $113 \pm 6 \text{ ng m}^{-2} \text{ h}^{-1}$ . Measurements at background sites showed both emission and deposition. The average Hg flux from all background sites showed an overall net emission of  $0.36 \pm 0.06 \text{ ng m}^{-2} \text{ h}^{-1}$ . Fluxes show strong relationships with temperature, radiation, and substrate parameters. A compensation point of 2.48, representative of bare soils was determined. Comparison of the Australian data to North American data confirmed the need for Australian specific mercury air-surface exchange data representative of Australia's unique climatic conditions, vegetation types, land use patterns, and soils.

## 1 Introduction

Mercury is a global pollutant; it is ubiquitous in the environment and is a threat to the health of humans and ecosystems. At the 10th International Conference on Mercury as a Global Pollutant (ICMGP) several speakers remarked on the paucity of high quality mercury air-surface exchange data sets (UNEP, 2011). There is a general lack of representative data on the cycling of natural emissions of mercury to and from the atmosphere (UNEP, 2011; Pirrone et al., 2010; Selin, 2009; Gustin et al., 2008; Schroeder et al., 2005). The atmosphere is the central pathway for distribution of this neurotoxicant globally. Natural sources of mercury include: wildfires, soils, vegetation, geologically

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enriched terrains, volcanic, and oceanic and fresh water (Gustin, 2003). Reliable and representative air-surface exchange measurements of natural sources of mercury are needed to put into perspective the long term human and ecological risks from anthropogenic activity associated with this volatile metal. These data, along with concurrent measurements of controlling environmental parameters, are required to improve our understanding of the biogeochemical cycling of mercury thereby improving model empiricism, and better constraining estimates of natural sources inventories (Pirrone et al., 2010).

Recent efforts to establish a mercury natural sources inventory for Australia by Nelson et al. (2009) revealed there are no data on the air-surface exchange of atmospheric mercury from natural sources over Australian environs. Moreover, there is an overall dearth of flux data on natural sources and sinks of atmospheric mercury in the Southern Hemisphere (Edwards and Howard, 2011). As such, estimates of the Australian natural sources inventory are reliant on emission factors and empiricism based on North American and European data sets, leading to high uncertainty in the estimates. Estimates for mercury emissions in Australia, not including ocean, range from 117 to 567 tonnes per year (Nelson et al., 2004), and more recently 148 tonnes per year, by Cope et al. (2009).

Australia is the second driest continent on earth after Antarctica and has a diverse range of climate zones (i.e. equatorial, tropical, subtropical, desert, grassland, and temperate, based on Köppen classification). Most of Australia is covered by arid and semi-arid climates, being warm to hot with the exception of the alpine area in the southeast which has seasonal snow. In addition to wide seasonal variation the Australian climates experience long term variation under the influence of El Niño (BOM, 2008). Australian climatic conditions are unique from North America and Europe as are its vegetation types, land use patterns and soils. Australian specific mercury air-surface exchange data are therefore needed to better constrain natural sources inventories. Estimates of anthropogenic emissions for Australia suggest they account for approximately 7% of the total burden (Nelson et al., 2009, 2012). With substantially less anthropogenic

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emissions to the atmosphere, coupled with background atmospheric mercury concentrations constant over hemispheric scales and approximately 25 % less in the Southern Hemisphere (SH) than the Northern Hemisphere (NH) (Pirrone et al., 2010), Australia presents a unique opportunity to study and address uncertainties in the global biogeochemical cycle of mercury associated with land-atmosphere cycling.

To address the need for Australian specific data on natural mercury emissions, an extensive measurement program has been proposed to collect mercury air-surface exchange data and environmental correlates over natural sources in Australian landscapes. Two pilot studies have been carried out during April and June (i.e. Australian autumn and winter) 2011 over mercuriferous and background substrates near a copper-gold-cobalt-arsenic-mercury mineral field at Pulganbar, NSW. During these studies gaseous elemental mercury (GEM) air-surface exchange and environmental measurements were made.

This paper presents the first Australian specific GEM air-surface exchange measurements along with controlling climatic and substrate variables. The results are compared to results from similar studies carried out in North America. This research goes towards advancing our understanding of mercury cycling in Australian environments.

## 2 Methodology

### 2.1 Site selection and description

According to Rytuba (2003) mercuriferous mineral enrichment is concentrated in 26 mercury mineral belts globally. One of these mercury mineral belts runs along the eastern coast of Australia and extends into Tasmania (Rytuba, 2003). Generally mercury mineral belts are concentrated in geological settings associated with previous or currently active plate tectonic margins, volcanism or geothermal activity (Gustin et al., 2006). In addition to this primary mercury mineralization Australia has many pockets

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of volcanogenic massive sulphide (VMS) and sedimentary exhalitive (sedex) mineral deposits which can contain up to 1200 ppm mercury (Rytuba, 2005).

The focus of the investigation for a site to carry out the pilot studies was on the primary mercury mineralization zones. Seven potential sites were identified through information collected from the Australian Bureau of Statistics (ABS, 1912). The year book of Australia (1912) reported cinnabar had been mined or prospected around the Bingara, Clarence River and Pulganbar areas of NSW, Kilkivan and Black Snake in the Wide Bay district of Queensland, the Jamieson area of Victoria and in the vicinity of Willunga in South Australia (Fig. 1a).

Pulganbar, NSW (29°28' S and 152°40' E, elevation 81 m; see Fig. 1) was selected for the pilot studies as a known cinnabar deposit mined from 1914 to 1924. It was reported that 2000 tons of mercury ore were mined and treated from this deposit (NSW OEH, 2003). A reconnaissance survey of the area identified a natural undisturbed measurement site approximately 1 km south-east of the old mine and smelter location as shown in the mineralized zones in the sediment-acid volcanic sequence (Fig. 1c). The area selected for the flux measurements was characterised by slightly rolling terrain, was sparsely treed with eucalypt (Fig. 1b) with the forest floor principally bare soil with scant leaf litter and sparse patches of grassy understory a few cm in height.

Exploration activity at the site carried out approximately a decade earlier (CWGOLD 2008) had left excavated mineralized material from the test pits and drill holes placed on the surface. This mineralized material provided the basis for the enriched substrate measurements. In addition, several sites were selected nearby on the undisturbed overburden to facilitate background measurements.

The site is rural, located on a cattle station far from any local anthropogenic sources other than potentially from the tailings of the old mine and smelter site. The Pulganbar mine and smelter were closed in 1924 with all machinery having been removed and the mine shaft filled in. Today there is very little evidence of the old mine and smelters as the ruins are heavily overgrown with trees and grasses. In view of potential influence of the old smelter on the flux measurements an analysis of the chamber inlet concentrations

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as a function of wind direction was performed. This analysis showed no wind sector bias in the flux data presented herein. Furthermore, the measurement site is located far from any anthropogenic sources on the regional scale.

## 2.2 Dynamic flux chamber method

5 Over the past decade considerable progress has been made towards optimizing dynamic flux chamber design and sampling methodologies (i.e. materials used, configuration, flushing flows rate, sampling protocols) to minimize perturbations to the natural system being measured and systematic sampling biases (Eckley et al., 2010; Carpi et al., 2007; Zhang et al., 2002; Kemp, 2001; Gillis and Miller, 2000) The dynamic flux  
10 method used here is described in detail in Kemp (2001) and Rasmussen et al. (2005). The chamber design is of low profile, small footprint and is constructed of quartz glass. Spectroradiograph testing of materials common in mercury flux chamber construction found quartz glass to transmit the full spectrum of incoming solar radiation, in particular UV-B (Fig. 2, Kemp, 2001). Quartz glass is easy to clean and has low sorption  
15 characteristics, resulting in low blank qualities.

Based on mass balance, steady state and well mixed assumptions the dynamic chamber flux is calculated using the following equation:

$$F = Q(C_{\text{out}} - C_{\text{in}}) / A \quad (1)$$

20 Where  $F$  is the mean flux of GEM ( $\text{ng m}^{-2} \text{h}^{-1}$ ),  $C_{\text{out}}$  is the mean GEM leaving the chamber ( $\text{ng m}^{-3}$ ),  $C_{\text{in}}$  is the mean GEM concentration entering the chamber ( $\text{ng m}^{-3}$ ),  $Q$  is the volumetric flow rate ( $\text{m}^3 \text{h}^{-1}$ ) and  $A$  is the chamber areal footprint ( $\text{m}^2$ ).

A Tekran<sup>®</sup> 2537 Total Gaseous Mercury Analyser coupled with a Tekran<sup>®</sup> 1110 synchronized two port sampling system was used to measure GEM in the inlet and outlet air streams. The sampling protocol was designed to eliminate any mercury analyser  
25 cartridge bias and switching/delay effects in the mean concentration difference calculation.

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## 2.3 Experimental design and environmental measurements

Dynamic flux chamber flux measurements of gaseous elemental mercury were undertaken over enriched substrates and background soils at the Pulganbar site. Two pilot field studies were carried out during the Australian autumn (April) and winter (June).

Flux measurements were carried out over six locations during the April and June field campaigns, three mercuriferous and three backgrounds, providing a basis for representative sampling and assessing variability. The sampling design also allowed for evaluation of the role of overburden and local deposition and variation in controlling parameters.

GEM fluxes were measured continuously at each chamber location except for short periods for blank measurement, calibration checking and servicing the instrumentation. A short period at the beginning of the June study was dedicated to ambient concentration measurement.

A number of meteorological and environmental parameters were measured simultaneously during the monitoring periods using a suite of field portable instruments described below. A small caravan housed the Tekran<sup>®</sup> 2537 and computer. The site was powered by a gasoline fuelled 6 KVA portable generator (Hg emission from exhaust, at  $1.18 \text{ ng m}^{-3}$ , was not significantly different from ambient Hg concentration measured on site).

### 2.3.1 Environmental variables

Ambient measurements of air temperature, humidity, barometric pressure, wind speed and wind direction were obtained at screen height (i.e. 2 m) using a WXT520 mini weather station (Vaisala Pty Ltd.). Nearby, ten meter wind speed and direction were obtained using a propeller wind vane (R. M. Young Inc.). At the same location a tipping bucket precipitation sampler was deployed (Campbell Scientific Inc.). The four components of solar radiation, long wave incoming,  $\text{LW}\downarrow$ , and outgoing,  $\text{LW}\uparrow$ , and short wave incoming,  $\text{SW}\downarrow$  and outgoing,  $\text{SW}\uparrow$ , were measured using a Kipp and Zonen CNR1

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(Campbell Scientific Inc.) at several locations in the measurement footprint at the 1.5 m height. UV-B (280 to 315 nm) was measured near the chamber location with a UVR1-B ultraviolet pyranometer (Middleton Inc.).

Air temperature and relative humidity were measured inside the chamber and just outside the chamber at the 30 cm height using a CS215 temperature and humidity probe (Campbell Scientific Inc.). Soil temperature was measured using thermocouples placed in the soil surface at 2 cm within the chamber footprint, and at the same depth nearby outside the chamber footprint. A soil temperature profile was obtained near the chamber measurement location at the 2, 5 and 10 cm depths, complimented by soil heat flux plates at 5 cm depth. Soil moisture measurements were obtained using a CS616 water content reflectometer (Campbell Scientific Inc.). Soil moisture was also obtained gravimetrically.

Data from the sensors described above were collected using Campbell Scientific Inc. data loggers.

### 2.3.2 Substrate sampling and analysis

At the end of the measurement period, for each chamber location, bulk soil samples were collected from the chamber footprint ( $0.1 \text{ m}^2$ ). Substrate samples were taken from the surface interval (0 to 2 cm) and from a deeper interval (5 to 10 cm below the surface). Samples were dried and homogenized, and particle analysis was performed using sieves. The total mercury (THg) concentration in the soil samples was determined using a Milestone<sup>®</sup> DMA-80 mercury analyser (EPA Method 7473).

To enable comparison of Pulganbar GEM fluxes with NH flux studies where acid digestion and CV-ASS analysis was used for THg in soil, a set of Pulganbar soil sample duplicates were sent to an independent laboratory (National Measurement Institute, NSW), for analysis of total mercury using acid digestion and cold vapour atomic absorption spectrophotometry (CV-AAS). Sample means of the Pulganbar results from

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the two analytical methods were compared using a standard student *t*-test. This testing showed the means of the two methods not to be significantly different.

Elemental determinations were also carried out on the samples using XRF to obtain mineralization profiles. Soil samples were collected to determine soil bulk density. Soil organic and inorganic matter content was determined using the Loss of Ignition (LOI) method described by Nelson and Sommers (1996).

### 3 Results and discussion

GEM chamber flux measurements were obtained at six locations at the Pulganbar site. Three locations were naturally enriched substrates labelled M1 to M3 and three were background substrates (i.e.  $< 0.1 \mu\text{g g}^{-1}$ ), labelled B1 to B3. Measurements were undertaken over two periods, the first during autumn, 12 April 2011 to 19 April 2011 (DOY 102 to 109) and the second during winter, 17 June 2007 to 26 June 2007 (DOY 168 to 177). Locations M1, M2, B1 and B2 were measured during the April field campaign while M3 and B3 were measured during the June study. The results therefore span a range of substrate concentrations and climatic conditions.

Prior to starting the chamber measurements for the June study an ambient background measurement was made over several hours during daytime at the 3 m level. The average background recorded was  $1.22 \pm 0.03 \text{ ng m}^{-3}$  ( $p < 0.05$ ).

#### 3.1 Substrate characteristics

The mercuriferous substrates were granular sandy soils with grain size fractions of approximately 60 % granule, 20 % sand and 20 % silt plus clay. In contrast the background soils were 50 % silt plus clay, 30 % sand and 20 % granule. XRF analysis showed both mercuriferous and background soils to be approximately 50 % silicates with high Fe, Al, Ag, and Ti content. The mercuriferous soils were also high in Cu and As, while the background soils were below detection limit for these elements. Table 1 summarizes

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the mercuriferous and background THg content measured with the DMA-80, as well as soil organic matter and bulk density. Due to instrument malfunction soil moisture data are not available.

The THg substrate data consistently showed higher concentrations in the 5 to 10 cm layer than the 0 to 2 cm sample depth for all sites. The ratio of surface to depth THg concentrations is also consistent across all sites with an average ratio of 1.6.

Background sites B1 and B3 were bare soil sites whereas site B2 was a sparsely vegetated site with mostly  $O_e$  horizon leaf litter, twigs, and grass with some  $O_i$  horizon grass. Samples of these materials were collected and separated into two categories for analysis; (1) leaf litter and (2) grass and twigs. The samples were homogenized using a stainless steel coffee mill and analysed using a DMA-80 (EPA method 7473). Results of this analysis showed THg for leaf litter as  $0.041 \pm 0.001 \mu\text{g g}^{-1}$  and  $0.044 \pm 0.005 \mu\text{g g}^{-1}$  for grass & twigs. The litter results interestingly compare with  $O_e$  horizon litter THg reported recently by Obrist (2012) for 14 US forests. Lastly, it is noteworthy that Site B2 also had the highest SOM in the 0 to 2 cm soil sample, suggesting an  $O_a$  horizon. GEM flux and SOM will be discussed further in Sect. 3.3.

### 3.2 GEM flux measurements

The dynamic flux chamber measurements for the six sites were calculated half hourly and are reported here as average daytime and nighttime fluxes (defined by a net radiation threshold of  $20 \text{ W m}^{-2}$ ) and overall flux averages for the period measurements were made at that site. The data are presented in Table 2.

Mean day and night GEM fluxes showed both emission and deposition ranging from a maximum emission of  $174 \pm 13 \text{ ng m}^{-2} \text{ h}^{-1}$  for daytime flux over a mercuriferous substrate, to a deposition flux of  $-0.33 \pm 0.05 \text{ ng m}^{-2} \text{ h}^{-1}$  for a nighttime flux over a background substrate. Mean overall fluxes for all sites showed emission, with background sites displaying small net emissions and mercuriferous sites showing much higher net emissions. The mercuriferous site results are consistent with those observed by others (Edwards et al., 2001; Gustin et al., 1999a; Rasmussen et al., 1998).

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The average GEM flux from all background sites gave means of  $1.4 \pm 0.1 \text{ ng m}^{-2} \text{ h}^{-1}$  (daytime) and  $0.02 \pm 0.05 \text{ ng m}^{-2} \text{ h}^{-1}$  (nighttime) with an overall mean of  $0.36 \pm 0.06 \text{ ng m}^{-2} \text{ h}^{-1}$ . This small net emission from the background sites is congruent with data reported by others (Kuiken et al., 2008; Gustin et al., 2006) for low mercury containing substrates.

The naturally enriched substrates flux data (i.e. M1 to M3) were obtained over a range of substrate concentrations showing a clear relationship between flux and substrate concentration. This general relationship has been observed by others (Engle and Gustin, 2002; Edwards et al., 2001; Rasmussen et al., 1998) and will be explored further in Sect. 3.4.

### 3.3 Environmental correlates

To investigate environmental correlates, data from the June study were selected as these data offered longer time series for a mercuriferous and background measurement sites than the April data sets. Figure 3 presents diel composite graphs constructed using the half hour data obtained over four days at the mercuriferous site and four days at the background site.

Figure 3a, b shows the diel flux cycle for the mercuriferous and backgrounds sites respectively, along with the corresponding diel composite of total incoming solar radiation and UV-B ( $\times 10^3$ ) to illustrate the relationship with these components of radiation. The modality displayed in the composite diel flux graphs results from tree canopy shading variation with solar angle. The locations of the background and mercuriferous sites were different with respect to canopy architecture, thus the modality differences seen in the respective diel flux graphs.

It has been observed from laboratory based studies that UV-B radiation enhances GEM flux and it is suggested that this is due to UV-B directly reducing divalent forms of mercury to elemental in the soil compartment (Choi and Holsen, 2009; Carpi et al., 2007; Moore and Carpi, 2005). Figure 3a, b shows both the mercuriferous and

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background flux diel patterns better mirror total incoming solar than UV-B, especially later in the day where peaks in the flux seen at 16:00 h at both sites are followed by total solar but not by UV-B. Later in the day UV-B is seen to drop off. The relative role of UV-B and total solar is unclear. If UV-B plays a role in GEM evasion it may be a function of intensity. The stronger correlation with total solar suggests the fluxes are more temperature driven.

Figure 3c, d shows the same GEM diel flux data correlated with ambient air temperature and soil surface temperature, measured within the chamber and the chamber's soil surface footprint respectively. It is evident that the correlation with chamber air temperature is much stronger than with chamber footprint soil surface temperature.

During this study both soil surface temperature and chamber ambient air temperature were measured inside the chamber and just outside the chamber. This was done to investigate possible chamber influences on the natural temperature regime. Figure 4 shows the results of plotting these four temperatures with their corresponding half hourly GEM fluxes for emission only. It is clear from Fig. 4a, b compared to Fig. 4c, d that the GEM fluxes are significantly better correlated with air temperature than soil temperature. This further supports what was observed in the results presented in Fig. 3c, d.

Comparison of the emission flux versus temperature relationships for temperatures within and outside the chamber (i.e. Fig. 4a vs. Fig. 4b and Fig. 4d vs. Fig. 4d) suggests that the chamber is not significantly influencing the flux-temperature relationship. Figure 4a shows a strong exponential relationship between the flux and air temperature.

The data collected during the June study, as seen in Fig. 3, highlights that the background site displayed emission during the day and deposition at night. Xin and Gustin (2007) and Gustin et al. (2006) suggested that air GEM concentration is an important predictor of soil Hg flux direction and defined an associated compensation point concept for soils. The compensation point is the ambient air concentration at which the net GEM flux between the soil and the atmosphere go to zero. Following Xin and Gustin (2007), Fig. 5 was constructed from the June background flux data set.

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The dotted vertical line shows the ambient background Hg concentration measured during the June study (i.e.  $1.22 \pm 0.03 \text{ ng m}^{-3}$ ), delineating those emission fluxes that were removed from the linear correlation constructed in order to determine the compensation point. A strong linear relationship is seen, defining the compensation point at  $2.48 \text{ ng m}^{-3}$ . This corresponds well with the results of Xin and Gustin (2007).

Soil organic matter (SOM) has been suggested to influence the GEM flux from background soils (Moore and Castro, 2012; Yang et al., 2007; Grigal, 2003). While data from this study are not sufficient to fully explore this correlate, SOM does not appear to correlate with the mercuriferous site fluxes as would be expected (Mauclair et al., 2008). However, the data in Tables 1 and 2 do show that the GEM flux decreases with increasing SOM for the background sites.

### 3.4 Comparison with Northern Hemisphere data

In the absence of Southern Hemisphere air-surface exchange data, modelling efforts applied to SH domains have had to rely on NH data for emission factors, deposition rates and associated empiricism. As such, natural emissions inventories for Australia have been based on indirect estimates, rather than reliable data.

Over the past decade a series of field studies over naturally enriched and background terrestrial landscapes were carried out in Canada and USA, (Edwards et al., 2001, 2002, 2005; Schroeder et al., 2005; Gustin et al., 1999a, b; Lindberg et al., 1999). Data from these studies showed a strong relationship between emission flux and substrate concentration for naturally enriched sites and highlighted relationships with environmental factors such as temperature, solar radiation, precipitation, and soil moisture. For background substrates the relationship to substrate THg concentration is not as distinct, with air-surface exchange processes for these substrates being controlled by additional biological and chemical factors (Rasmussen et al., 2005). Figure 6 shows North American (NA) flux data plotted against substrate concentration from these studies.

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These data were collected over the NH summer season with an overall average temperature of 14.3°C. On the same plot we have displayed the data collected over the mercuriferous and background substrates presented herein where the overall average temperature for these measurements was 15.5°C. The Australian mercuriferous and background data behaved in a similar manner as the NA data, with the naturally enriched substrate data showing a strong relationship between emission flux and substrate concentration. The Australian background flux data behaved similar to the NA background flux data by not showing as strong a correlation with substrate THg.

Two separate trend lines were purposely fitted to the NA and Australian data from mercuriferous sites with a view to illustrating differences in the average behaviour of the naturally enriched data. The slopes for the two data sets are not significantly different but the intercept is displaced upwards for the Australian data. This displacement is due to an overall difference in environmental factors between the two data sets. Temperature is likely to be the principal reason for the shift upward of the Australian trend line as the average temperature of the Australian studies (i.e. autumn/winter season) and the NA data (i.e. summer) data were 1.2°C warmer for Australian data.

#### 4 Summary and conclusions

Prior to undertaking the GEM air-surface exchange measurements presented here there were no direct measurements of Hg exchange available for Australian landscapes. GEM fluxes were obtained over both naturally enriched and background soils and over a range of climatic conditions during Australian autumn and winter periods. Large GEM emissions were observed over the naturally enriched sites compared to small net emissions from background sites, with background sites displaying both emission and deposition.

Strong correlations were observed between the GEM flux and the climatic variables temperature and radiation. The relative role of UV-B and total solar radiation was not clear however pointed to the need for further full scale field experiments to address this

uncertainty. GEM emission from both naturally enriched and background substrates correlated strongly with temperature. The background soils with higher soil organic matter showed relatively lower fluxes to those with less soil organic matter content. Exploration of the compensation point for bare soils showed a good agreement with the concept.

Strong correlations were observed between substrate THg and fluxes for the mercuriferous sites but not for the background substrates. This general result is congruent with similar measurements in North America. Comparison of the Australian flux substrate relationship for mercuriferous substrates with the North American relationship showed good agreement with respect to slope however the intercept was offset. The intercept offset was explained by the average temperature difference between the NA and Australian data. This has interesting implications when it comes to using NA-based emission rates and empiricism to estimate Australian natural emission inventories.

Results from these two pilot studies strongly confirm the need for Australian specific data on air-surface exchange of atmospheric mercury. Characteristic and reliable data are needed that are representative of Australia's unique climatic conditions, vegetation types, land use patterns, soils, and spatial and temporal variability. With the ratio of natural to anthropogenic sources thought to be 9:1, Australia presents a unique opportunity to study and address uncertainties in the global biogeochemical cycle of mercury associated with land-atmosphere cycling.

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**Table 1.** Summary of average substrate THg concentration, soil organic matter, and bulk density.

	Depth (cm)	THg Concentration ( $\mu\text{g g}^{-1}$ )	Ratio of THg Conc. surface to depth	SOM (63 micron) (%)	Dry Bulk Density ( $\text{g cm}^{-3}$ )
Site M1	0–2	2.3	1.5	3.7	1.62
	5–10	3.4		3.0	
Site M2	0–2	3.3	1.5	5.8	
	5–10	5.0		3.7	
Site M3	0–2	0.250		4.2	
	5–10				
Site B1	0–2	0.044	1.6	6.2	
	5–10	0.07		3.9	
Site B2	0–2	0.065	1.6	11.7	
	5–10	0.102		5.2	
Site B3	0–2	0.050	1.7	9.4	1.46
	5–10	0.084		4.2	1.64

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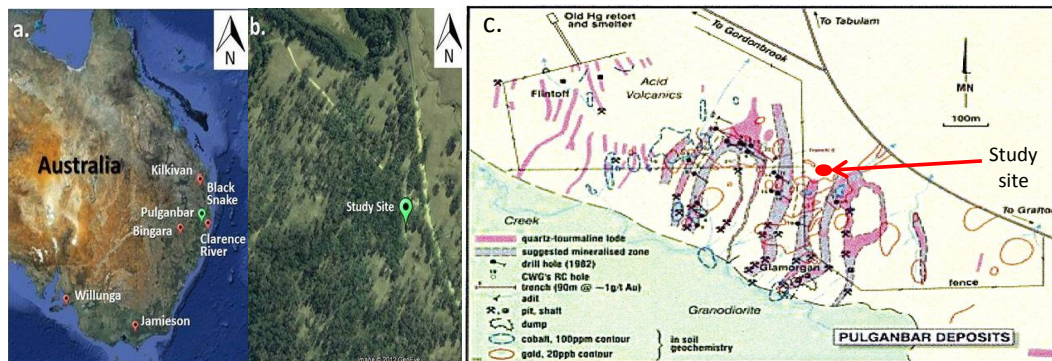


**Table 2.** Summary of GEM fluxes and environmental parameters measured over six locations at the Pulganbar site during the April and June field campaigns.

		Hg Flux ( $\text{ng m}^{-2} \text{h}^{-1}$ )	Air Temp. ( $^{\circ}\text{C}$ )	Soil Temp. ( $^{\circ}\text{C}$ )	Solar ( $\text{W m}^{-2}$ )	UV-B ( $\text{W m}^{-2}$ )
April						
Site M1	Day	$108 \pm 15$ ( $n = 12$ )	22	23	200	0.39
	Night	$76 \pm 6$ ( $n = 32$ )	15	18		
	Overall	$85 \pm 6$	17	19		
Site M2	Day	$174 \pm 13$ ( $n = 23$ )	21	20	148	0.21
	Night	$90 \pm 4$ ( $n = 60$ )	17	17		
	Overall	$113 \pm 6$	18	18		
Site B1	Day	$1.1 \pm 0.1$ ( $n = 21$ )	23	23	164	0.2
	Night	$0.73 \pm 0.05$ ( $n = 56$ )	18	19		
	Overall	$0.83 \pm 0.05$	19	20		
Site B2	Day	$0.37 \pm 0.06$ ( $n = 11$ )	26	22	156	0.22
	Night	$0.09 \pm 0.04$ ( $n = 27$ )	18	20		
	Overall	$0.17 \pm 0.04$	20	21		
June						
Site M3	Day	$33 \pm 3$ ( $n = 49$ )	18	17	118	0.13
	Night	$8 \pm 1$ ( $n = 164$ )	11	17		
	Overall	$14 \pm 1$	13	17		
Site B3	Day	$2.0 \pm 0.1$ ( $n = 32$ )	18	25	107	0.11
	Night	$-0.33 \pm 0.05$ ( $n = 120$ )	9	17		
	Overall	$0.17 \pm 0.09$	11	18		

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**Fig. 1.** Locations of pilot studies at Pulganbar, NSW. **(a)** Image from Google Earth maps 2010. **(b)** Aerial photograph of Pulganbar site (Google Earth image). **(c)** Pulganbar deposits (Central West Gold, 2008).

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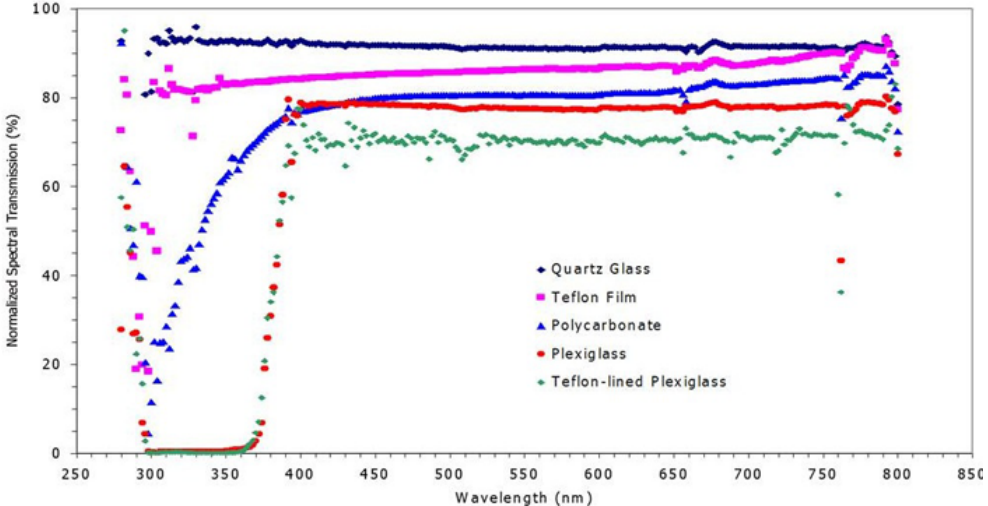


Fig. 2. Spectroradiograph measurements of materials commonly used for flux chamber construction (Kemp, 2001).

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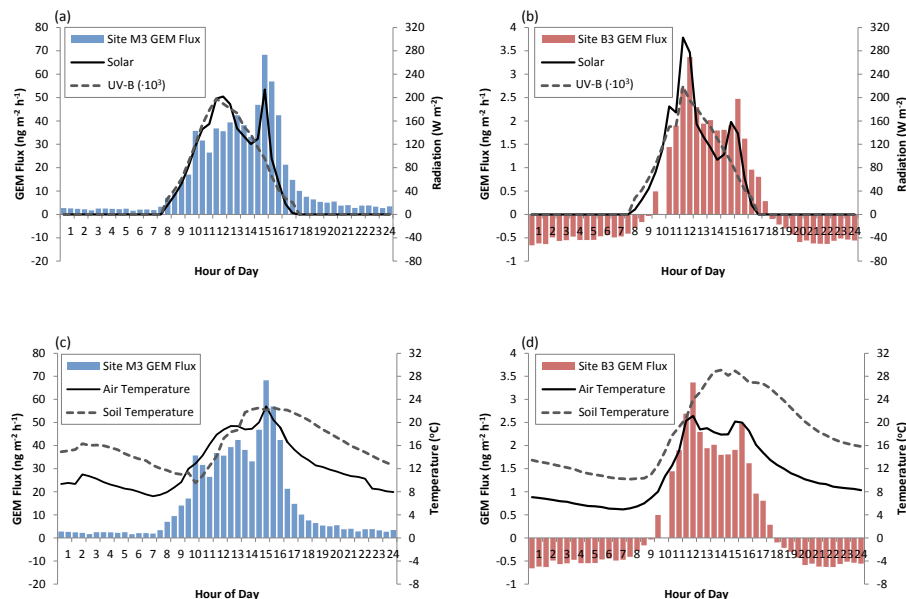
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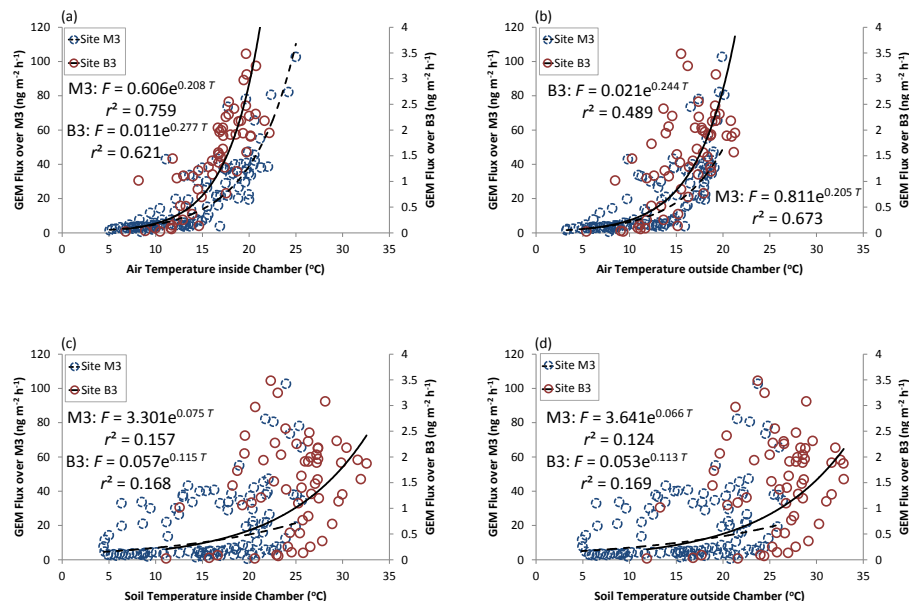


**Fig. 3.** Diel composite GEM flux, radiation and temperature data from June study. Top graphs compare GEM flux with radiation measurements taken over a mercuriferous **(a)** and background **(b)** site. Bottom graphs compare GEM flux with temperature measurements taken over a mercuriferous **(c)** and background **(d)** site.

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**Fig. 4.** GEM emissions fluxes compared to temperature measurements taken over mercuriferous site M3 (blue, dotted, left scale) and background site B3 (red, solid, right scale) during June study. Top graphs show air temperature data taken inside (a) and outside (b) the flux chamber. Bottom graphs show soil surface temperature data taken inside (c) and outside (d) the chamber footprint. Equations and  $r^2$  values are for least squares exponential curves, where  $F$  = GEM flux and  $T$  = temperature.

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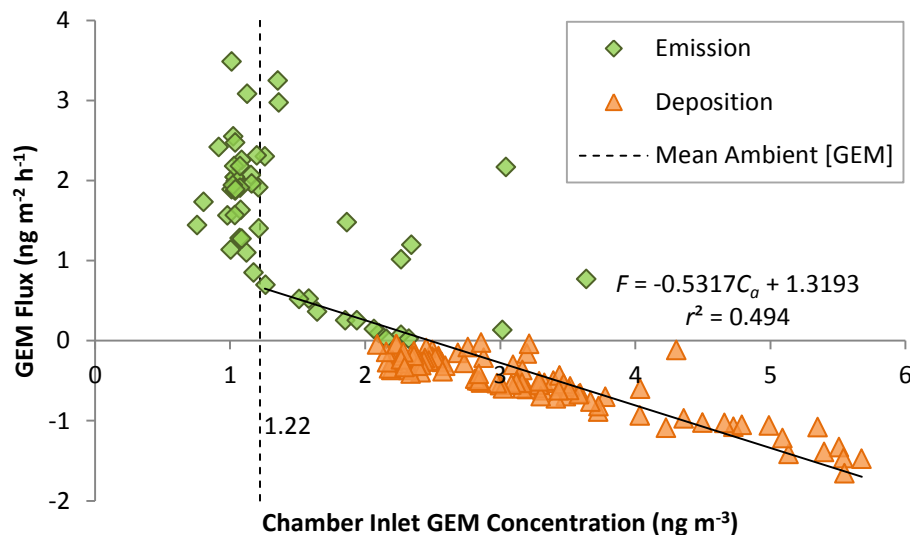
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**Fig. 5.** GEM fluxes, and GEM concentrations at the chamber inlet, taken over Site B3 during June study. Chamber inlet GEM concentrations greater than the mean ambient taken at the beginning of the study were used to construct the linear least squares trend, where  $F$  = GEM flux and  $C_a$  = GEM concentration.

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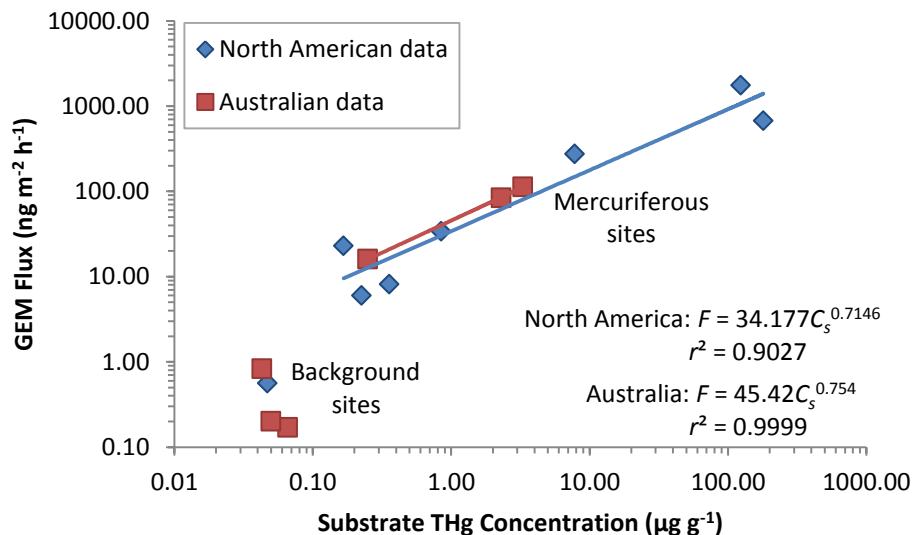
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**Fig. 6.** Mean GEM flux and substrate THg data taken over Australian and North American mercuriferous and background sites. Linear least squares lines are fitted to mercuriferous sites only, with  $F$  = GEM flux and  $C_s$  = substrate THg concentration.

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