Atmospheric dispersal of mercury from the Hindustan Lever Limited thermometer factory, Kodaikanal, Tamil Nadu, India, using lichen as a biomonitor.

Brigden, K. & Stringer, R.

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Greenpeace Research Laboratories, Department of Biological Sciences,
University of Exeter, Prince of Wales Road, Exeter EX4 6QX, UK.

1 EXECUTIVE SUMMARY

Kodaikanal is located within the Palni Hills in Tamil Nadu, south India. The Palni Hills are an offshoot of the Western Ghats, which are in part comprised of pristine forests and grasslands that are home to endangered flora and fauna. One of these areas is the Pambar Shola forest, a nature sanctuary protected by the Tamil Nadu State Government, which edges the town of Kodaikanal.

In 1983, a factory making mercury thermometers was relocated to Kodaikanal from the USA by Ponds India. The factory produced mercury thermometers for export, primarily using mercury imported from the United States. Activities at the site included the crushing of mercury contaminated glass and mercury distillation. In 1998, the factory was acquired by Hindustan Lever Limited (HLL), a subsidiary of the Anglo-Dutch multinational Unilever, who operated the factory until its closure in March 2001.

Mercury used in the production of thermometers is in the metallic (elemental) form. Metallic mercury slowly evaporates to form mercury vapour, a colourless and odourless gas. Mercury is extremely toxic in all its forms. Being an element, mercury is persistent and cannot be broken down in the environment but only changed from one form to another.

Inhalation of mercury vapour is often the most significant route of human exposure in industrial contexts. The central nervous system (CNS) and kidney are the main organs affected following exposure, though skin and stomach complaints have also been reported. In extreme cases, high levels of exposure to metallic mercury vapours can kill.

Ex-workers have expressed concerns over health and safety practices at the Kodaikanal thermometer factory during the time it was operating, as well as health effects from mercury exposure. These, and additional concerns over environmental impacts on the forest adjacent to the factory site, have continued subsequent to closure of the factory.

Ex-workers have also reported a wide range of health effects that occurred during and subsequent to their employment at HLL. Common symptoms reported by these workers match known effects that can result for exposure to metallic mercury through inhalation and dermal exposure.

An environmental site assessment of the HLL site, and immediate areas, has shown serious contamination of soil and sediment with mercury.

The current study was carried out to establish the extent of atmospherically transported mercury from the Hindustan Lever Ltd. Site, adressing a wider geographical coverage than previously studied.

In November 2002, samples of lichen (*Lobaria discolor* and *Parmotrema reticulatum*) were collected from locations around the factory and Kodaikanal. Concentrations of mercury in lichen are a good indicator of averaged atmospheric mercury concentrations.

The concentration of mercury in the lichen samples ranged from 0.08 to 1.99 mg/kg dry weight. It has also been found that the different lichen species accumulate different concentrations of mercury under the same conditions. Had it been possible to collect the more sensitive *P. reticulatum* in the forest environments, it is expected that even higher

concentrations of mercury would have been found in lichen from the most contaminated areas.

The most contaminated samples were those collected closest to the HLL site, either to the northwest or northeast - along the principal wind directions from the site. Two of these samples were collected in the Bombay Shola forest and another in Vattakanal Shola. These results extend the geographical area over which atmospheric mercury distribution has been demonstrated.

As part of the environmental assessment commissioned by HLL, samples of lichen from locations both within and outside the factory site were analysed. Elevated concentrations were found in samples collected outside and close to the factory site, generally between 2.2 and 9 mg/kg mercury, though significantly higher concentrations were found immediately north and south of the site.

Samples collected for the current study were collected at greater distances from the site, and, as would be expected, contained lower concentrations of mercury. These data, however, better show the distance to which the impact can be detected.

The observed mercury elevation in lichen samples gives an indication of elevated atmospheric mercury concentrations at their collection locations, which may result in impacts on ecosystems at these locations. Where elevated atmospheric mercury levels are present, other plant species, which also form part of the diet of fauna, will similarly accumulate mercury to higher concentrations, transferring it into the wider ecosystem.

To address the contamination caused by the factory, Hindustan Lever Ltd. has agreed to carry out limited remediation work. The HLL site has already been partially decontaminated through the removal of 289 tonnes of mercury contaminated material. It is currently proposed that future remediation will be carried out for soil that is contaminated with mercury above a concentration of 10 mg/kg, a limit defined under Dutch guidelines as an intervention value for seriously contaminated multifunctional land to ensure protection of both human health and terrestrial ecosystems.

The Dutch regulations, however, set two different concentration limits for mercury; an intervention value of 10 mg/kg and a target value of 0.3 mg/kg. The intervention value is defined as serious contamination, although the regulations recognise that in some cases concentrations below this value may also result in serious contamination. The target value indicates the level at which there is judged to be a fully recovered sustainable soil quality providing long term negligible risks to the ecosystem.

The Dutch target value is a level equivalent to background levels in the Netherlands, a highly industrialised and densely populated country. A very different situation applies at this site with the pristine sanctuary forest in close proximity, where background levels can be expected to be significantly lower. Furthermore, a large proportion of the contaminating mercury at the Kodaikanal site is likely to be present in metallic form, which has a greater mobility through atmospheric transport than many other forms of mercury. Therefore, clean-up to a level below that of the Dutch target value (0.3 mg/kg) may be needed to achieve long term negligible risks to the ecosystem, the goal of the Dutch target value.

Following cleanup, all contaminated material removed from the site must be dealt with appropriately to prevent future releases of mercury. Unless correctly stored, waste material containing mercury is a recognized source of ongoing releases of mercury to the biosphere. Furthermore, if the mercury is recovered from the wastes and returned to the marketplace, its future use in products and processes will result in ongoing releases of this mercury to the global biosphere.

Removal from future use is the only long-term solution for the mercury recovered from the HLL site that will prevent future releases. Adequate long-term waste management for contaminated material, or mercury recovered from it, can only be achieved through monitored and retrievable permanent storage. The generator of the hazardous waste must bear full legal and financial responsibilities for this.

On a global scale, atmospheric releases of mercury from the HLL site may be small, but virtually any local source contributes to the global pool, adding to the total mercury burden on the biosphere by anthropogenic emissions. As a consequence, mercury issues must be addressed at both local and global levels.

A cessation in the use of mercury in products and processes though substitution with non-mercury alternatives is required. Alternatives are commercially available for virtually all applications of mercury, including mercury thermometers. In many countries, national regulations and recommendations currently exist with the aim of eliminating or restricting uses of mercury in products and processes. Many of these include specific restrictions on mercury medical thermometers. Substitution must be accompanied with the retiring of mercury currently in use, or on the market place, through terminal storage.

2 INTRODUCTION

The Palni Hills in Tamil Nadu, south India, are an offshoot of the Western Ghats, which separate Tamil Nadu from the neighbouring state of Kerala. Although settlement, agriculture and forestry have encroached on the forests and grasslands, a significant part of them remain pristine and home to endangered flora and fauna. Among them is the Pambar Shola forest, a nature sanctuary protected by the Tamil Nadu State Government and edging the town of Kodaikanal.

With its elevation of 2100m above sea level, giving it a cool climate and spectacular views, tourism has for many years been a major source of income in Kodaikanal. However, this changed in the 1980s, when a factory making mercury thermometers commenced operations. The factory was originally located at Watertown, New York, USA and operated by Chesebrough Ponds. It was relocated from this site to Kodaikanal by Ponds India in 1983. The factory produced mercury thermometers primarily for export to Europe, USA, South America and Australia. The mercury used was primarily imported from the United States. Activities at the site included the crushing of mercury contaminated glass and mercury distillation. In 1998, the factory was acquired by Hindustan Lever Limited (HLL), a subsidiary of the Anglo-Dutch multinational Unilever, who operated the factory until its closure in the March 2001 (URS Dames & Moore 2002).

Mercury used in the production of thermometers is in the metallic (elemental) form. Elemental mercury is a silver-white metal. It is a liquid at room temperature, and is reasonably volatile at ordinary temperatures. If not enclosed, metallic mercury will slowly evaporate to form mercury vapour, a colourless and odourless gas. It can subsequently be distributed over long distances, even on a global scale.

All forms of mercury are extremely toxic. It is a non-essential trace metal, with no biochemical or nutritional function. Once absorbed, it can be excreted from the body, but slowly and with residues tending to remain in certain organs, notably the brain and kidneys (WHO 1989). Being an element, mercury is persistent. It cannot be broken down in the environment, only its form can change.

Metallic mercury is readily absorbed by the lungs, and approximately 80% of inhaled mercury vapour is absorbed into the bloodstream (Bernard 1997, WHO 1991). In the industrial context this is likely to be the most significant route of exposure, although ingestion and dermal absorption may also occur. Effects of exposure are mainly seen on the central nervous system (CNS) and kidney, though skin and stomach complaints have also been reported. CNS effects include tremors, spasms, loss of memory, increased excitability, severe depression, personality changes, even delirium and hallucination. Kidney damage has been reported in chronically exposed workers (Ratcliffe *et al.* 1996, Goyer 1996). In extreme cases, high levels of exposure to metallic mercury vapours can kill (ATSDR 2000).

Effects have previously been reported for workers exposed to elemental mercury vapour through occupational exposure, including at a thermometer factory. These included tremors, dysdiadochokinesia (impairment of the ability to perform rapidly alternating movements) and difficulty with heel-to-toe gait linked to chronic exposure (Ehrenberg *et al.* 1991).

Ex-workers from the Kodaikanal thermometer factory have reported concerns over health and safety practices employed, along with health effects from mercury exposure. These, and additional concerns over environmental impacts on the forest adjacent to the factory site have continued subsequent to closure of the factory (Bhargava 2002).

As a result of these concerns, a tribunal was formed to investigate possible impacts resulting from activities at the factory. As part of this tribunal, ex-workers reported a wide range of health effects that occurred during and subsequent to their employment at the factory (Bhargava 2002).

Common symptoms reported by these workers include fatigue, headaches, nausea and other stomach dysfunctions, giddiness, blurring of vision, skin complaints including burns and dermatitis, respiratory disorders, kidney dysfunction (supported by high urea/creatinine levels). CNS effects were also reported including loss of memory, tremors, depression and mood changes. A number of workers were reported to have suffered from epileptic seizures after commencing work at the factory (Bhargava 2002).

Many of the reported symptoms match known effects that can result for exposure to metallic mercury through inhalation and dermal absorption (ATSDR 2000, Goyer 1996, Ratcliffe *et al.* 1996, WHO 1991).

The public tribunal also collated statements from ex-workers describing inadequate health and safety practices at the factory. Many reported that they were never informed about the dangers of working with mercury, nor of the health problems related to long-term mercury exposure. They also reported that shop floor practices included the sweeping of mercury containing broken thermometer bulbs and glass stems into buckets or drains (Bhargava 2002). These practices would have mobilised metallic mercury into the atmosphere.

Ex-workers also reported the contamination of their clothing with mercury, leading to the transfer of mercury to their homes (Bhargava 2002). This would have put their families at risk of exposure. Research into other facilities using mercury, including thermometer factories, found the homes of workers to be contaminated with mercury which was thought to have been transferred in the workers' clothing and on their shoes (ATSDR 2000, Hudson *et al.* 1987, Zirschky 1990). The families were indeed found to have been exposed. At the time of exposure, the children of the workers had levels of mercury in their urine at levels about five times those of children in unaffected homes (Hudson *et al.* 1987).

Ongoing environmental and human health concerns subsequent to the closure of the factory prompted HLL to commission the consultancy firm URS Dames & Moore to conduct an environmental site assessment and preliminary risk assessment for the factory site (URS Dames & Moore 2002).

This report includes data for air concentrations measured within parts of the factory site while operations were ongoing (URS Dames & Moore 2002). These data include a number of measurements where concentrations exceeded 0.05 mg/m^3, a widely recognised limit for occupational exposure (NIOSH 1992)(see Appendix 2 for more detail). In a summary environmental site assessment and preliminary risk assessment report on the HLL site that was released in May 2001, a maximum spot reading for mercury in air of 0.48 mg/m^3 was reported (URS Dames & Moore 2001).

Very few data are available for off-site atmospheric mercury concentrations in Kodaikanal. Krishna *et al.* (2003) directly sampled the atmospheric mercury concentration near the thermometer factory site, reporting a concentration of 1.32 μg/m^3 (0.00132 mg/m^3). The International Programme on Chemical Safety (IPCS) has identified 0.2 μg/m^3 (0.0002 mg/m^3) as a guidance value for long-term inhalation exposure of the general public to metallic mercury vapour (WHO/IPCS, 2002). This value is seven times lower that the atmospheric concentration recorded outside the HLL site (Krishna *et al.* 2003). The recorded level was also much higher than typical concentrations for non-contaminated areas (0.5-10 ng/m^3 or 0.0005-0.010 μg/m3) (Horvat *et al.* 2000, USEPA 1997).

Unfortunately, the precise location and date for the collection of the air sample is not reported, so it is not clear if the factory was in operation at the time. Moreover, a one-off recording of this type does not give any indication of the average concentration, or variability, over time. Nevertheless, the measurement of even a single value many fold higher than the WHO/IPCS guidance value provides further cause for concern about the impacts from mercury emitted from the site.

The Hindustan Lever Ltd. thermometer factory site and surrounding areas are known to be contaminated with very high levels of mercury (URS Dames & Moore 2002). In addition, since 1992 many tonnes of mercury-bearing wastes from the HLL factory, including broken thermometers, have been sold to other businesses. The latest sale was to a local scrap yard where the material was inappropriately stored within a residential/commercial area of Kodaikanal (URS Dames & Moore 2002). In June 2001, HLL arranged the recovery of 7.4 tonnes of mercury-contaminated material from this scrap yard.

As part of the URS Dames & Moore study, samples of soil, sediment, water, bark and lichen were collected for analysis, from locations both within and outside the factory site. Data presented in the report show significant and wide-ranging mercury contamination of soil and sediment.

However, the geographical extent of the URS Dames & Moore lichen sampling program was limited. Since metallic mercury, which was vented to atmosphere from the factory over a number of years, is known to be carried quite efficiently in the air, it was considered likely that it had spread beyond the URS Dames & Moore study area. The purpose of the current survey, therefore, was to establish as far as possible the extent of atmospherically transported mercury contamination emanating from the HLL site.

3 SAMPLING PROGRAM

A number of techniques are available for the direct measurement of mercury concentrations in air. Atmospheric levels of mercury in air are, however, generally very low and therefore potentially subject to analytical errors. Direct air monitoring typically requires pre-concentration techniques even in areas with elevated concentrations.

Atmospheric levels at a location can fluctuate significantly over time. Direct measurements are usually averaged over short timeframes, typically from minutes to one day (Horvat *et al.* 2000). Without taking many measurements over long timeframes, direct measurements are not able to provide a long-term representation of atmospheric mercury concentrations.

Many of the limitations for studying long term atmospheric contamination using direct measurement can be overcome through the use of mercury-accumulating plants. Lichens have no roots or analogous structures and so obtain most of their nutrients directly from the atmosphere, largely through wet and dry fallout. This process results in the accumulation of atmospheric pollutants in the lichen tissues. Lichens are slow growing, do not shed parts during growth and are generally very tolerant of high metal concentrations.

These factors make lichen suitable long-term spatial biomonitors for atmospheric pollutants, including mercury, from both diffuse and point sources (Mulgrew & Williams 2000). Analysis of lichen enables integration over extended time periods, averaging over seasonal variations and accounting for short-term effects from changing temperature and wind direction. It has also been demonstrated that lichens do not accumulate mercury from the bark of the trees on which they grow (Sloof & Wolterbeek 1993).

Through comparison with direct air measurements, studies have demonstrated that mercury concentrations in lichen are a good indicator of atmospheric mercury concentrations (Burton 1986, Horvat *et al.* 2000, Wolterbeek *et al.* 1996). Where lichen has been transplanted into an area close to a mercury source, mercury has been shown to accumulate in the lichen to levels that correlated with the distance from the source within months of transplantation (Horvat *et al.* 2000, Makholm & Bennett 1998).

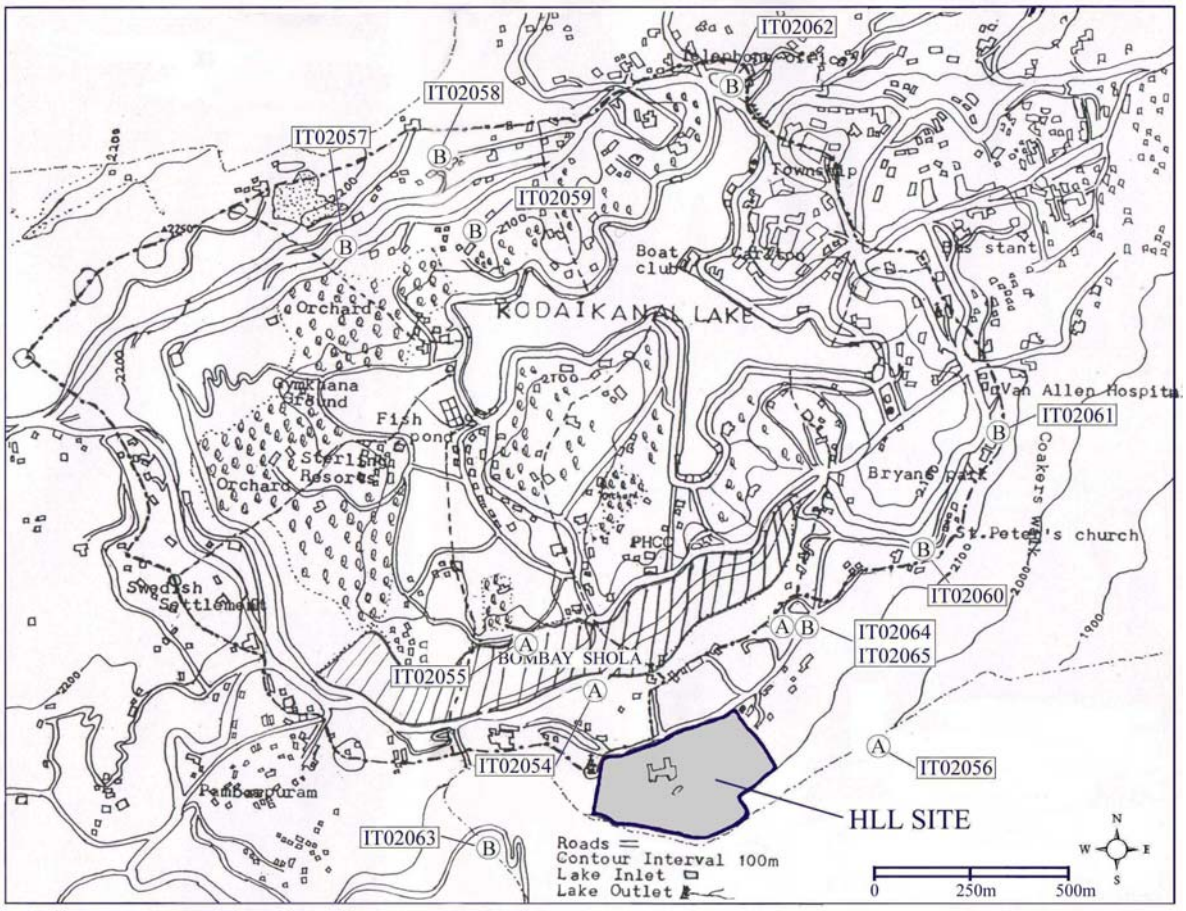

Figure 1. Map of lichen sampling locations, Kodaikanal, Tamil Nadu, India, 2002. Adapted from a map prepared by the Palni Hills Conservation Council (PHCC), based upon the Survey of India Map. The location of each sample is indicated by 'A' for samples of L. discolor and 'B' for P. reticulatum.

Lichen have been used in numerous studies in different geographical regions as biomonitors of atmospheric mercury concentrations, demonstrating significant correlations between the mercury concentrations found in lichen and their growing distance from mercury sources of various type. Mercury concentrations in lichen tend to decline exponentially with distance from the source, as atmospheric mercury concentrations decline (Bargagli *et al.* 1987, Bargagli *et al.* 1989, Makholm & Bennett 1998, Garty 1993, Ikingura & Akagi 2002, Riget *et al.* 2000).

In November 2002, twelve samples of epiphytic lichen (lichen living on the surface of another plant) were collected from the trunks and branches of trees in the forests around the HLL factory and in the grounds of residential and commercial premises in Kodaikanal. A map showing the locations of these samples in presented in Figure 1.

The samples were collected directly into 100ml glass bottles that had been rinsed thoroughly with Aristar grade nitric acid and analytical grade pentane in order to remove all heavy metals and organic residues.

The samples of lichen were collected from locations outside the factory perimeter, at varying distances and directions from the site. A single lichen species was not available for sampling at all of the locations. Hence, two different species of lichen, *Lobaria discolor* and *Parmotrema reticulatum* (Figure 2), were collected to give the desired geographical coverage.

Mercury accumulation by lichen is dependent on a number of factors including the rate of growth of the lichen. To determine whether the two lichen species used in this study accumulated mercury to differing degrees under the same conditions, samples of both types of lichen were collected from the final sampling location, a residential property on St Mary's Road (IT02064 and IT02065).

All samples were immediately sealed and cooled upon collection. The samples were returned to the Greenpeace Research Laboratories in the UK for analysis.

Type A, Lobaria discolor *Type B, Parmotrema reticulatum*

Figure 2. The two lichen species collected. Classification of species collected was kindly undertaken by Dr Michael Proctor of Exeter University, UK and Dr Brian Coppins, Royal Botanic Garden, UK.

4 MATERIALS AND METHODS

Prior to analysis, the lichen samples were manually cleaned in a similar method to that employed in other studies (Ikingura & Akagi 2002, Krishna *et al.* 2003). Foreign adhered material was manually removed and the lichen rinsed twice with deionised water to remove any remaining small particles and dust. The cleaned lichens were dried at 30°C and powdered prior to analysis.

The samples were analysed for total mercury using an acid digest extraction method employing concentrated nitric acid (69%) at 80°C followed by analysis using Cold Vapour Inductively Coupled Plasma Atomic Emission Spectrometry (CV-ICPAES). A certified reference material of known mercury content (CRM482, trace elements in lichen; epiphytic lichen species *Pseudevernia furfuracea*) was analysed in an identical manner to determine the efficiency of the method employed. Detailed descriptions of sample preparation and analytical procedures are presented in Appendix 1.

5 RESULTS AND DISCUSSION

The results of the total mercury concentrations in the lichen samples are presented below in Table 1.

The concentrations of total mercury in the lichen samples collected for this study (IT02054-IT02065) ranged from 0.08 to 1.99 mg/kg dry weight. Comparison of the two samples from the residential site closest to the factory (IT02064 and IT02065) indicates that *P. reticulatum* (lichen type B) accumulates mercury to approximately twice the concentration as *L. discolor* (lichen type A) under the same conditions.

Sample no.	Lichen Species	Approximate distance and direction from HLL	Location	Mercury concentration (mg/kg dry weight)
IT02054	A	200m, northwest	Bombay Shola	1.99
IT02055	A	400m, northwest	Bombay Shola, below Upper Lake Road	0.38
IT02056	A	300m, east-northeast	Vattakanal Shola	0.32
IT02057	B	1500m, northwest	Residential property Observatory Road	0.12
IT02058	B	1600m, northwest	North of Observatory Road	0.08
IT02059	B	1400m, northwest	Residential property below Observatory Road	0.08
IT02060	B	700m, northeast	Property by cliff edge at Coakers Point	0.15
IT02061	B	1000m, northeast	Property above Coakers Point	0.16
IT02062	B	1700m, north	Kodaikanal lake, northeast shore by bridge	0.17
IT02063	B	300m, west-southwest	Across gorge to the southwest of the factory	0.11
IT02064	B	300m, northeast	Residential property, St Mary's Road	0.38
IT02065	A	300m, northeast	Residential property, St Mary's Road	0.17

Table 1. Mercury concentrations for samples of lichen collected in the vicinity of the closed Hindistan Lever Limited mercury thermometer factory, Kodaikanal, India. Lichen identification; Type A - Lobaria discolor; Type B- Parmotrema reticulatum

The four samples with the highest concentrations of mercury were those collected closest to the Hindustan Lever Ltd site. Of these, the highest concentration was found in sample IT02054 (*L. discolor*), which contained 1.99 mg/kg total mercury, 25 times that in the least contaminated samples (0.08 mg/kg in samples IT02058 and IT02059, *P. reticulatum*).

This most contaminated sample was collected closest to the HLL site on the edge of Bombay Shola, approximately 200 metres northwest of the Hindustan Lever site, in the direction of Kodaikanal Lake. Two further samples of *L. discolor* from the ancient forests around the factory contained significantly elevated concentrations of mercury. The sample collected from Bombay Shola forest below Upper Lake Road, approximately 400 metres northwest of the site, (IT02055) contained 0.38 mg/kg total mercury, while the sample collected from Vattakanal Shola, approximately 300 metres east-northeast of the site (IT02056) contained 0.32 mg/kg total mercury. In addition, one sample of *P. reticulatum* (IT02064) contained a significantly elevated concentration of mercury: 0.38 mg/kg total mercury. This was collected at a residential property on St Mary's Road, approximately 300 metres northeast of the site.

Upon release, mercury vapour is dispersed in the atmosphere, predominantly in the principal wind directions. The terrain in the vicinity of the factory site is, however, highly varied and includes a 1300m precipitous slope immediately to the south of the site. This topography may complicate the dispersal of mercury vapour from the site and reduce correlation between offsite mercury contamination and principal wind directions.

Data compiled from the meteorological records of 1995 to 2000 by URS Dames & Moore demonstrates that the principal wind directions in Kodaikanal are northeast/north-northeast, northwest/north-northwest and southeast (URS Dames & Moore 2002). The most contaminated lichen samples are to the northeast and northwest, as would be expected. Immediately to the southest of the site is a precipitous drop of 1300m preventing collection of a sample to the southeast. However, sample IT02056 demonstrates atmospheric spread of mercury towards the east.

The remaining samples, which were less contaminated, containing total mercury concentrations in the range 0.08-0.17 mg/kg, were generally collected from further away from the site, and were mostly *P. reticulatum*. There were two exceptions to this.

Firstly, sample IT02063, which although it was *P. reticulatum* and collected only 300 metres from the site, was collected to the west-southwest of Hindustan Lever Ltd in a location less exposed to the prevailing air flows from the mercury contaminated areas.

The second exception was IT02065. This was a sample of *L. discolor* that was collected at the same location as one of the more highly contaminated samples (IT02064, *P. reticulatum*), on St Mary's Road. The *L. discolor* sample from St Mary's Road (IT02065) contained 0.17 mg/kg total mercury, while the concentration in the sample of *P. reticulatum* (IT02064) was 0.38 mg/kg. These data demonstrate that under the same conditions the lichen *P. reticulatum* can accumulate mercury to a greater concentration than *L. discolor*. The concentration in the sample of *P. reticulatum* was higher than in that of *L. discolor* by a factor of 2.2. Variation in accumulated mercury concentrations between different lichen species has been previously observed (Garty 1993).

The higher accumulation of mercury in *P. reticulatum* (IT02064) compared to *L. discolor* (IT02065) can be expected to be similarly reflected in the remaining lichen samples analysed. Had it been possible to collect the more sensitive *P.reticulatum* in the forest environments, it is likely that even higher concentrations of mercury would have been found in lichen from the most contaminated areas.

As discussed previously, the URS Dames & Moore assessment study commissioned by HLL included analysis of soil, sediment, water, bark and lichen from locations both within and outside the factory site (URS Dames & Moore 2002). Data presented in the report showed significant contamination of soil in and around the factory site with mercury. Further, 35 samples of lichen were analysed; 7 from within the factory site and 28 from offsite. The data presented for these samples show elevated levels of mercury in many of these samples, including some collected immediately outside the factory site. However, the species collected were not recorded, making it more difficult to compare these data with that from other studies.

In addition to the report published by URS Dames & Moore, a further study of mercury pollution in the vicinity of the HLL thermometer factory was published in August 2003 (Krishna *et al.* 2003). This study analysed samples of lichen and mosses to demonstrate elevated atmospheric mercury levels around the factory site. The lichen species investigated by Krishna *et al.* was the epiphytic lichen *Parmelia sulcata*. However, data are presented for only a few samples and neither precise locations nor sampling dates are given.

Data for background levels in these other two studies are consistent with data obtained in the current study. URS Dames & Moore reported concentrations of less than 0.2 mg/kg total mercury. The study by Krishna *et al.* (2003) similarly reported background mercury concentrations in lichen of 'about 0.2 mg/kg', at a site 20 kilometers from the HLL site.

Lichen samples collected within the HLL site for the URS Dames & Moore report were found to contain mercury up to 87 mg/kg dry weight (URS Dames & Moore 2002). Many soil samples collected within the site were also found to contain extremely elevated mercury concentrations, many in excess of 50 mg/kg and a few samples over 500 mg/kg.

Lichen samples collected off site, but in the immediate vicinity of the HLL site, were also found to contain elevated mercury concentrations, generally between 2.2 and 9 mg/kg. A few samples, however, contained significantly higher concentrations; up to 68 mg/kg immediately north of the site and up to 80 mg/kg immediately south of the site. The source of mercury in the highly elevated samples was attributed to mercury area exhaust fans as well as the mercury distillation area and the glass crusher room (URS Dames & Moore 2002). These data underline the seriousness of the contamination of the factory and its immediate environment, but most samples were collected too close to the site to reveal the full geographical extent of the contamination.

The highest concentrations of mercury in the lichen (*P. sulcata*) collected off site by Krishna *et al.* (2003) approached 8 mg/kg. The highest concentration was for a sample collected "near the thermometer factory", though precise collected locations were not given.

The data for lichen samples collected within and in the immediate vicinity of the HLL site reflect high atmospheric mercury concentrations that would have been expected close to the source of the emissions (Krishna *et al.* 2003, URS Dames & Moore 2002). Samples

analysed in the current study were collected from locations at greater distances from the HLL site. As would be expected, these samples did not contain mercury at such high concentrations but better show the distance to which the impact can be detected.

The many-fold increases in accumulated mercury in the elevated lichen samples (IT02054, IT02055, IT02056 and IT02064) gives an indication of potential impacts on ecosystems at these locations. Where elevated atmospheric mercury concentrations are present, many other media will accumulate mercury to far greater concentrations than they would in uncontaminated areas, including other plant species (Markert 1993). In the immediate vicinity of the HLL site, highly elevated mercury concentrations have already been found in tree bark (URS Dames & Moore 2002).

Plants that accumulate atmospheric mercury are likely to form part of the diet of fauna living in the vicinity. Through such processes mercury can be transferred into the wider ecosystem, possibly bioaccumulating in some species.

It should also be noted that the mobilisation of mercury is a continuous process. In addition to being absorbed by plant and other material, atmospheric mercury will over time be re-deposited to land and water. Much of the deposited mercury, however, will subsequently be re-mobilised to the atmosphere in an ongoing process (UNEP 2002).

6 CONCLUSIONS

The site assessment commissioned by Hindustan Lever Ltd. (HLL) has previously shown severe mercury contamination within the factory site. Off site research was restricted to the immediate vicinity of the site, other than for specifically defined areas such as along the course of streams flowing from the site. The current study has demonstrated the spread of mercury though the atmosphere to areas that the HLL commissioned assessment failed to investigate. These comprised Bombay Shola, Vattakanal Shola and a residential area.

Given the known atmospheric mobility of metallic mercury, which would have been exacerbated by the practice of venting contaminated air from the factory, the possibility of such widespread contamination should have been investigated before in order to facilitate a fully informed discussion of impacts resulting from the factory's operation.

To address the contamination in and around the site that has resulted from activities at the thermometer factory, Hindustan Lever Ltd. has agreed to carry out limited remediation work. They have stated that "the objective of remediation is to remediate to the extent required to ensure protection of human health and the environment and not to clean up to background" (HLL 2002).

To date, the cleanup of Kodaikanal has only proceeded so far as the removal of some of the most contaminated wastes. In early 2003 some 289 tonnes of mercury-contaminated material was transported to the Bethlehem Apparatus Inc. mercury recycling facility in the United States. The intention is for the mercury to be recovered from the waste material and to be returned to the marketplace for alternative future uses.

The removal of this highly contaminated material from the site was a first step in ensuring the protection the sensitive ecosystem around Kodaikanal. HLL next plan a partial

decontamination of the site through the removal of approximately 4100 m^3 of soil and stream sediment containing mercury above a concentration of 10 mg/kg. Contaminated soil and sediment from the cleanup operation will be temporarily stored on the HLL site in secure containers until an appropriate site for its disposal, or intermediate storage pending final disposal, has been identified (URS Dames & Moore 2002).

The site assessment commissioned by HLL concluded that remediation was only required for soil contaminated with mercury above a concentration of 10 mg/kg (URS Dames & Moore 2002). This value is taken from Dutch guidelines for multifunctional land for the protection of both human health and terrestrial ecosystems. It is an "intervention value" and is defined as the concentration above which there can be said to be a case of serious contamination. If the intervention value is exceeded the functional properties of the soil for humans, flora or fauna is deemed to have been seriously diminished or in danger of being seriously diminished (MHSPE 1994).

However, the Dutch regulations also state that, in specific cases, concentrations below the intervention value may nevertheless seriously diminish or potentially diminish the functional properties of the soil for humans, flora and fauna and place them at risk, resulting in a case of serious contamination (MHSPE 1994).

Moreover, the Dutch regulations do not only contain an intervention value, but also a target value of 0.3 mg/kg (MHSPE 1994). The target value indicates the level at which there is judged to be a sustainable soil quality; that is, the level that has to be achieved to fully recover the functional properties of the soil for humans and plant and animal life. Furthermore, the target value gives an indication of the benchmark for environmental quality in the long term on the assumption of negligible risks to the ecosystem (MHSPE 1994). This target value can therefore be regarded as the minimum level to which remediation must be carried if a fully recovered ecosystem is to be achieved with long term negligible risks to the ecosystem. This is obviously far more appropriate for an otherwise non-industrial site such as that in Kodaikanal, and should be regarded as the minimum requirement for cleanup.

The circumstances in Kodaikanal are such that remediation may be required to a level below that of the Dutch target value (0.3 mg/kg) to achieve long term protection of the ecosystem, the objective of the Dutch target value. Close to the factory site is a pristine sanctuary forest, where background levels can be expected to be significantly lower than those found in the highly industrial and densely populated country such as the Netherlands. In addition, contamination in and around the Kodaikanal site is likely to be dominated by metallic mercury, a form with a greater potential for mobility through atmospheric transport than many other forms of mercury.

It should be noted that while cleanup to more stringent levels than those proposed by HLL will partially address the legacy of mercury in Kodaikanal, it may not be possible to reverse all the damage to protected forest areas.

Waste material containing mercury is itself a recognized source of ongoing releases of mercury to the biosphere. Where the mercury is in metallic (elemental) form, as is expected for the vast majority of the wastes from and contaminated soil within the HLL site, such releases will primarily be in the form of gaseous elemental mercury emitted to the

atmosphere (UNEP 2002). This material must be dealt with appropriately to prevent future releases of this mercury to the biosphere.

There are as yet no agreed estimates for the total releases of mercury from the Kodaikanal thermometer factory site. Some estimates have been made (URS Dames & Moore 2002), but there remains an ongoing debate as to the accuracy of such estimates by environmental and local groups (Greenpeace 2003).

These releases are, of course, in addition to all the mercury that has been dispersed from HLL in the form of its finished product. Given that any mercury released will add to the total anthropogenic mercury burden on the biosphere (UNEP 2002), the releases cannot be regarded as insignificant.

Recent data from the United States has estimated that discards of mercury used in medical thermometers to municipal solid waste has amounted to over 15 tonnes annually (USEPA 1997). This is indicative of the large scale of the potential wastage from the accumulation of even such small events as the disposal of a single thermometer.

Impacts from mercury are of global as well as local concern, and mercury issues must be addressed at both local and global levels to be fully effective. The consequences of the relocation of the old thermometer factory from the USA to Kodaikanal can be regarded as an example of a sequence of events that has happened before and will, in all likelihood, be repeated elsewhere. The trend towards the phasing out of mercury-based products and technologies in the older industrialised countries, largely driven by legislation to prevent damage to human health and the environment, is being reflected in increasing mercury imports to the newly industrialised nations, including India.

Some mercury-contaminated wastes from Kodaikanal have already been exported to the USA with the intention that the mercury be recovered for sale. This could conceivably be resold to India where further releases and impacts will be inevitable. Ultimately, the only way to prevent this would be for governments and the corporations who trade in mercury- including Unilever, HLL's multinational parent company- to commit to removing it from commerce permanently and place it in terminal storage.

The United Nations Environment Program (UNEP) recently released a comprehensive document on mercury. This document highlights the global aspects of the environmental and human health impacts associated with this toxic metal (UNEP 2002). This document states that, globally, mercury is now present in many environmental media and at levels that adversely affect humans and wildlife (UNEP 2002).

UNEP concluded that a near-total phase-out of mercury is possible. In many countries, including Canada, Denmark, France, Norway and Sweden, national regulations and recommendations currently exist with the aim of eliminating or restricting uses of mercury in products and processes. Many of these include specific restrictions on mercury medical thermometers (UNEP 2002).

In Sweden the manufacture, import and sale of many mercury products, including thermometers, has been banned since 1993 (Government of Sweden 1991). In Denmark, mercury and mercury-containing products, including thermometers, are regulated. A general ban on the sale of such products has been in place since 1994 and their export has

been banned since 1998, though certain exceptions do exist (Government of Denmark 1998). In the USA several states have banned the use of mercury fever thermometers and most major retailers no longer sell them (UNEP 2002). In keeping with such concerns, the export of metallic mercury, as well as chemical compounds and preparations containing mercury, have been prohibited in Sweden (Government of Sweden 1998).

Alternatives are commercially available for virtually all applications of mercury, including thermometers. There are a variety of options for replacement of mercury thermometers, including electrical and electronic thermometers, which have become standard in many countries. Although initial costs for such alternatives may appear higher, these cost differences do not take into account additional costs resulting from impacts to human health and the environment in places such as Kodaikanal through the manufacture and disposal of mercury thermometers.

Substitution must be accompanied with the retiring of mercury currently in use, or on the market place, through terminal storage. The return of recycled mercury to the market will result in future releases of mercury to the biosphere; UNEP (2002) recognises the ongoing trade in mercury, noting that some is illegal, uncontrolled and/or unregulated. "Recycled mercury recovered from spent products and industrial wastes" is specified as one source of mercury for such trade. The UNEP report also notes that "even under current regulations and restrictions, many of the uses and movements of mercury and mercury containing products are likely to eventually result in the release of mercury to the global environment".

Removal from future use is the only long-term solution for the mercury recovered from the HLL site that will prevent its future release to the global biosphere. Such long-term waste management can only be achieved through monitored and retrievable terminal storage, with the repository maintained in a way that minimises emissions by all routes to the greatest extent possible, a method internationally recognized by the United Nations Environment Program (UNEP) (UNEP 2002). The generator of this hazardous waste must bear full legal and financial responsibility for its stewardship. The need for permanent storage is presently acknowledged in Sweden, where the Swedish Environmental Protection Agency has stated that final storage is the only acceptable disposal of mercury waste as "Mercury is a substance that remains a threat to human health and the environment in perpetuity, and for this reason it should not be recycled" (SEPA 1997).

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APPENDIX 1
ANALYTICAL METHODOLOGY

A1.1 Preparation of lichen samples for mercury analysis

All chemicals were of High Purity Aristar Grade. All glassware was cleaned in detergent, rinsed with tap water and deionised water, soaked in 10% nitric acid overnight, rinsed with deionised water and dried in an oven.

A1.1.1 Sample preparation

On arrival, the samples were stored in a freezer at -20°C. Prior to cleaning, the samples were moved to a fridge and allowed to defrost and stored at between 0°C and 5°C.

The lichen samples were cleaned by manual removal of foreign adhered material with stainless steel instruments, followed by two rinses with deionised water to remove any remaining small particles and dust. The method of cleaning employed is comparable to that used in similar studies (Ikingura & Akagi 2002, Krishna *et al.* 2003).

The cleaned samples were dried at 30°C until weighing readings became constant (approx. 2 days). They were then crumbled into a coarse powder and ground in an agate mortar to obtain a powder. Approximately 0.4 g of sample was weighed into a glass 100 ml boiling tube and to this 3ml of concentrated nitric acid (69%) was added. The samples were placed onto a Gerhardt Kjeldatherm digestion block (40 space) connected to a Gerhardt Turbosog scrubber unit (filled with 10% w/v sodium hydroxide) and digested at 80°C for four hours to obtain a near clear solution.

After cooling to ambient temperature, 5 ml of deionised water was slowly added to each sample and the solution filtered into 10 ml volumetric flasks. The digestion tubes and filter papers were washed twice with 1 ml deionised water, the solutions made up to a volume of 10 ml and mixed. A blank sample and a Standard Reference Material, CRM482 (Trace elements in lichen, certified by the Commission of the European Communities, Brussels) were analysed as part of the same batch as the Indian lichens. The lichen used as a standard reference material was the epiphytic lichen species *Pseudevernia furfuracea*.

A1.2.2 Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES)

Following preparation, samples were analysed for mercury content by Cold Vapour Generation Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES), using a Varian Liberty-100 Sequential Spectrometer.

For each solution, Hg (II) was reduced to Hg (0), i.e. a vapour, following reduction of the samples with sodium borohydride (0.6% w/v), sodium hydroxide (0.5% w/v) and hydrochloric acid (10 molar). The vapour was carried in a stream of argon into the spectrophotometer. Two calibration standards were prepared, at 10 μg/l and 100 μg/l,

matrix matched to the samples (i.e. in 30% v/v nitric acid). The calibration was validated using two quality control standards (10 μg/l and 80 μg/l), prepared internally from different reagent stocks. Any sample exceeding the calibration range was diluted accordingly, in duplicate, and re-analysed.

The concentration determined in the reference material was 0.41 mg/kg dry weight. The certified value of this material is 0.48 mg/kg dry weight, indicating a recovery efficiency of 86% for the method. The methods employed by Krishna *et al.* (2003) gave similar recovery efficiencies (87-89%).

APPENDIX 2
BACKGROUND INFORMATION FOR MERCURY

Mercury is the only metal that can exist as both a liquid and a vapour at ambient temperatures. Its environmental behaviour therefore differs from that of most other toxic elements (ATSDR 2000, WHO 1989). Being an element, mercury is persistent as it cannot be broken down in the environment, but can only change between different forms that exhibit greater or lesser toxicity. Mercury can exist in three valence states, Hg (0), Hg (I) and Hg (II). In the atmosphere, elemental mercury is by far the most common form, and, as a vapour, it is responsible for the long-range global cycling of mercury (UNEP 2002). In addition, though to a far lesser degree, mercury may be associated with particulates, which are removed from the atmosphere by dry or wet deposition. Atmospheric inputs may be more significant in areas where other sources, such as aquatic inputs, are absent (ATSDR 2000, WHO 1993).

Globally, anthropogenic emissions are thought to account for between 40-75% of the total annual input to the global atmosphere, and current atmospheric concentrations are thought to be 2 - 3 times those of pre-industrial levels (USEPA 1997).

A2.1 Toxicity

All forms of mercury are extremely toxic, with the precise toxicity being dependent on the route of exposure and the form of mercury; metallic (elemental), inorganic or organic. Mercury is a non-essential trace metal, with no biochemical or nutritional function; it can be excreted from the body, but slowly and with residues tending to remain in certain organs, notably the brain and kidneys (WHO 1989). In organic forms, mercury is the only metal known to biomagnify, i.e. progressively accumulate though the food chain (WHO 1989).

Elemental (metallic) mercury vapour is readily absorbed by the lungs; approximately 80% of the inhaled vapour is absorbed into the bloodstream (Bernard 1997, WHO 1991). In the industrial context, this is likely to be the most significant route of exposure, although ingestion and dermal absorption may also occur. Effects of exposure are mainly on the central nervous system (CNS) and the kidney, though damage to the skin and stomach have also been reported (ATSDR 2000). In extreme cases, high levels of exposure to metallic mercury vapours can kill.

Inhaled metallic mercury readily reaches the foetuses of pregnant women. Children and foetuses are particularly sensitive to the harmful effects of metallic mercury on the nervous system (ATSDR 2000).

Acute inhalation of high levels of mercury vapour may cause nausea, vomiting, diarrhoea, increase in blood pressure or heart rate, skin rashes, eye irritation, corrosive bronchitis and pneumonitis. In extreme cases, high levels of exposure to metallic mercury vapours can result in death through respiratory failure (ATSDR 2000). If not fatal, exposure may also be associated with central nervous system (CNS) effects such as tremor or increased excitability (ATSDR 2000, Goyer 1996). With chronic exposure, the major effects are on the CNS (tremor, spasms, loss of memory, increased excitability, severe depression, personality changes, even delirium and hallucination). Renal (kidney) damage has also

been shown for chronically exposed workers (Ratcliffe *et al.* 1996, Goyer 1996). Many of these effects are not specific to humans, having also been reported in animal studies (ATSDR 2000).

A2.2 Regulations and recommendations

Regulations and recommendations for concentrations of mercury in air are mainly set for occupational exposure. Notable examples include the US National Institute for Occupational Safety and Health (NIOSH) recommended airborne exposure limit of 0.05 mg/m^3 averaged over a 8-hour work shift (NIOSH 1992). The American Conference of Governmental Industrial Hygienists (ACGIH) recommends an airborne exposure limit of 0.025 mg/m^3 averaged over an 8-hour work shift (ATSDR 2000).

Various studies have lead to the setting of minimal risk level (MRL) and inhalation reference concentration (RfC) for mercury, at significantly lower concentrations than the above. A MRL is an estimate of daily human exposure below which a person is likely to be without an appreciable risk of adverse noncancer health effects. RfCs are used by the United States Environmental Protection Authority (USEPA) as estimates of continuous inhalation exposure to the human population (including sensitive subgroups) below which a person is likely to be without appreciable risk of deleterious noncancer effects during a lifetime. These values are not direct estimators of risk but rather reference points used to gauge potential effects. At exposures increasingly greater than the MRL or RfC values, the potential for adverse health effects increases. The MRL and RfC values are derived from Lowest-Observed-Adverse-Effect Levels (LOAELs), the lowest exposure level of mercury in a study, or group of studies, that produced statistically or biologically significant increases in frequency or severity of adverse effects between the exposed population and an appropriate control population. It should be noted that MRL and RfC values do incorporate uncertainty and modifying factors in their calculation from LOAELs (ATSDR 2000).

In one study, a minimal risk level (MRL) of 0.0002 mg/m^3 was derived for chronic-duration inhalation exposure (1 year or more) to metallic mercury vapour. The study concerned a group of 26 mercury-exposed workers from three industries who were exposed to low levels of mercury for an average of 15 years (Fawer *et al.* 1983). A separate study of occupationally exposed dentists attributed adverse neurological effects to even lower average level of exposure than that reported by Fawer *et al.* (1983). However, a MRL was not derived from these data due to some uncertainties in the study (Ngim *et al.* 1992).

The International Programme on Chemical Safety (IPCS) has identified 0.2 $\mu g/m^3$ (0.0002 mg/m^3) as a guidance value for long-term inhalation exposure of the general public to metallic mercury vapour (WHO/IPCS 2002). The USEPA quotes an inhalation reference concentration (RfC) for elemental mercury of 0.0003 mg/m^3 based on central nervous system (CNS) effects in humans from a number of separate studies (USEPA 1999).

A2.3 References

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