Exposure to polycyclic aromatic hydrocarbons in coal liquefaction workers: impact of a workwear policy on excretion of urinary 1-hydroxypyrene

R Quinlan, G Kowalczyk, K Gardiner, I Calvert

Abstract

Objective—This study was undertaken to assess whether contaminated personal clothing worn beneath a coverall (normal workwear) is a source of potentially significant dermal exposure to polycyclic aromatic hydrocarbons (PAHs) in coal liquefaction workers.

Methods—An intervention study was conducted over a two week period involving 10 workers that reflected the range of activities performed at the factory. A cross over design was used to examine the influence of normal workwear (personal clothing worn beneath a coverall) and intervention workwear (new coverall, shirt, trousers, underwear, socks, and boots) upon excretion of urinary 1-hydroxypyrene (1-OHP) and skin pad deposition of pyrene.

Results—The impact of intervention was noted in three ways:

(1) A notable reduction (55%) in the mass of 1-OHP excreted on the first day of the intervention phase was found. The median reduction in mass excreted (22.7 nmol) was significant from zero at the 5% level; (95% confidence interval (95% CI) 9.5–40.8 nmol).

(2) A notable reduction (82%) in skin pad deposition of pyrene on the first day of the intervention phase was found. The median reduction of 13.20 ng.cm⁻² was significant from zero at the 5% level; (95% CI 7.3–26.4 ng.cm⁻²).

(3) About a 50% reduction in 1-OHP concentration over the working week occurred during the intervention phase; an increase of 2.07 µmol/mol creatinine was found from the start to the end of the work period during the intervention phase compared with an increase of 4.06 µmol/mol creatinine during the normal phase. This reduction was not significant at the 5% level.

Conclusion—The results indicate that on the first day of the working week investigated, significant reductions in absorption (as measured by excretion of urinary 1-OHP) and deposition of PAHs (as measured by skin pad deposition of pyrene) can be effected by improvements in workwear policy. The impact of the improved workwear regimen was also detected by reduction in spot urinary 1-OHP concentrations, although this effect was less pronounced. One implication of the findings is that exposure to

PAHs may arise from workers' own contaminated personal clothing. As a consequence of this study an improved workwear policy has been implemented at the factory.

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Keywords: polycyclic aromatic hydrocarbons, 1-hydroxypyrene, workwear

Coal liquefaction involves the transformation of coal into hydrocarbon fractions that can be used as transport fuels. The process has been previously described¹ and only a brief summary is included here. In the extraction stage coal is digested in anthracene oil, which possesses hydrogen donation properties. About 85% by weight of coal passes into solution. The digest is filtered to remove undissolved coal and mineral matter, and clean coal solution is catalytically hydrocracked in a trickle bed reactor. The main products are naphtha (initial boiling point up to 170°C), mid-distillates (170°C–450°C), and a small amount of heavy products (>450°C). The naphtha and mid-distillates need to be chemically refined to make quality petrol and diesel fuels. The pilot plant converts 2.5 tonnes of coal a day into light distillates (35%–47% distillate/tonne of coal received).

The process stream consists of complex mixtures that are mainly polycyclic aromatic hydrocarbons (PAHs) of which some are known carcinogens.² Previous studies at the coal liquefaction factory have concluded that: (1) Measurement of 1-hydroxypyrene (1-OHP) in urine specimens taken at the end of the work shift is a valid measure of total exposure to and absorption of PAHs in coal liquefaction workers.¹

(2) Group median 1-OHP concentrations can be used as an effective indicator of hygiene performance at the factory.³

(3) The potential for exposure to the process stream materials occurs during both routine and non-routine tasks and is primarily dermal.³

Control of health risks at the factory is effected by a detailed occupational health programme that involves routine medical surveillance, annual biological and atmospheric monitoring, and adherence to good occupational hygiene practices. As well as showering facilities and skin luminoscope examinations, hygiene practices at the factory rely in part on provision of protective clothing to minimise any possibility of dermal exposure to coal

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liquefaction materials. Workers are issued with protective coveralls, protective gloves, and boots; beneath these items workers wear their own personal clothing and underwear.

Workwear permeability studies (Walton, personal communication) have shown that Arco Coroguard TP Coveralls (Proban-100% cotton drill; 305 g.m²) provide the best protection against penetration by process liquids. Each worker at the factory is provided with two coveralls for routine everyday use. Although these items provided by the factory are laundered regularly, responsibility for laundering personal clothing worn beneath coveralls is left to each worker.

Previous studies of occupational hygiene practices at the factory have highlighted the potential for dermal exposure and absorption of PAHs, and measures have been taken through improved working practices to increase worker awareness of skin contamination. These activities have resulted in successive annual reductions in excretion of 1-OHP at the factory. This provides a quantitative measure of improvements in occupational hygiene. Nevertheless, despite these improvements, relatively high concentrations of 1-OHP are still being detected in workers and further action to reduce these exposures is therefore still warranted.

Van Rooij and co-workers have shown that a substantial amount of dermal contamination by PAHs can arise on areas of skin covered by workwear, which suggests that when contaminated this may be an important source of dermal exposure.^{4,5} Our observations of working practices have indicated that contaminated workwear may also be a potential source of exposure to PAHs in coal liquefaction workers.

To determine whether the workwear practices (personal clothing worn beneath a coverall) in the liquefaction factory could contribute to total exposure of PAHs, the impact of an improved workwear regimen (new coverall, shirt, trousers, underwear, socks, and boots) was assessed principally by monitoring changes in excretion of 1-OHP. Should the normal workwear be a source of exposure to PAHs, then reductions in 1-OHP would be anticipated after improvements in the workwear regimen. Also skin pads were used in this study to obtain evidence of direct deposition on the skin at sites covered by workwear.

Several methods are available for measuring dermal deposition; garment samplers, surrogate skin samples, fluorescent tracer techniques, skin washing, tape stripping, and surface wipes. The appropriateness and limitations of these techniques have been recently reviewed by Fenske.⁶ We chose to use skin pads as a surrogate skin sampler, as these had been successfully applied in previous studies of dermal exposure to PAHs.^{4,5,7,8} Although skin pads have been used as measures of total dermal deposition of substances and possibly also of the quantity of dermally absorbed material, our application of the technique was of a more limited nature and was aimed at finding whether a difference in pad deposition

was obvious between the two workwear phases. The skin pads were located on sites covered by protective clothing and were designed to detect any transfer of PAHs that may arise either through the protective clothing or from PAHs already deposited on the clothing. Two skin pads per person were used on the first day back at work when the maximum benefit of the intervention strategy was likely.

Materials and methods

DESIGN AND SAMPLE SIZE	
An intervention study ⁹ with a crossover design ¹⁰ was used in which the differences in excretion of 1-OHP and skin pad deposition of pyrene in a group of exposed workers were compared during a normal workwear phase and an intervention workwear phase (table 1). The advantages of the crossover design were threefold; firstly the design allowed more observations for the given sample size, secondly every worker acted as his own control thus eliminating variations between workers, and finally the design aimed to reduce the potential bias from variability in exposure over two consecutive work periods (weeks).	
At the liquefaction factory workers who may be exposed to the process material are engineers, responsible for monitoring plant equipment and sampling process streams, and maintenance workers who are required to attend routine maintenance procedures and unexpected occurrences, for example process stream blockages. The study took place during two consecutive working weeks. For the engineers each working week consisted of four 12 hour shifts and consecutive weeks were separated by four days off. For the maintenance staff each working week consisted of five eight hour shifts and consecutive weeks were separated by two days off.	
There was a total population of 35 workers available for investigation. Ten workers (five engineers and five technicians) were selected as follows; of the 20 engineers on site (four shift teams, each of five workers) one shift team (E2, E9, E10, E11, and E12) was randomly selected and consisted entirely of non-smoking Caucasian men aged 36 to 53	
Table 1 Study design	
Weeks:	
One	
Normal workwear	M2, M6, E2, E10, E11
Intervention workwear	M1, M4, M8, E9, E12
Two	
Normal workwear	M1, M4, M8, E9, E12
Intervention workwear	M2, M6, E2, E10, E11
Workwear:	
Normal	Coverall, gloves, boots; workers' own clothing and undergarments.
Intervention	New coveralls, new boots, new gloves, new shirts, new trousers, and new undergarments.
Measurements:	
1-OHP:	Urine collection 24 h on day 1 of each study week plus five spot samples (four taken on days 1 and 2 and the fifth at the end of the working week).
Skin pads	Groin and ankle sites on day 1 of each study week.

years (mean 43.6). Their job experience at the factory was between three and five years (mean 3.8) and all were clinically in good health. Of the 15 technicians on site five were selected to give a stratified sample that reflected the range of activities performed by maintenance workers and consisted of three mechanical technicians (M4, M6, and M8), one instrument technician (M2), and one manual technician (M1). All were Caucasian men aged 31 to 52 years (mean 44.6 years) with between one and five years (mean 2.4) job experience at the factory. All were clinically in good health. Three of them were smokers, but smoking was strictly prohibited at the workplace. The 10 workers were randomly assigned to one of two study groups; intervention workwear phase followed by normal workwear phase for workers M1, M4, M8, E9, and E12, and normal workwear phase followed by intervention workwear phase for workers M2, M6, E2, E10, and E11. An information sheet was distributed to the participants to brief them of all the relevant facets of the study. A self administered diary card was used for each week and included information on age, smoking habits, diet, medication, and nature of workwear. Informed consent was obtained and each worker had the right to withdraw from the study at any time. After analysis and interpretation of the data, workers were informed of their own individual results and the employer was provided with group data.

STRATEGY OF URINARY 1-OHP SAMPLING

Six urine samples were taken from each person during each of the two consecutive working weeks. These were one 24 hour urine collection on the first day of each working week, four spot samples during the first 24 hours after the start of each working week (start of shift on day 1, end of shift on day 1, midway between shifts on days 1 and 2, and start of shift on day 2) and a further spot sample at the end of each working week. A total of 120 samples were taken for analysis.

The impact of intervention workwear was assessed by changes in: (a) the mass of 1-OHP excreted in the first 24 hours of each sampling period—that is, on return to work after a break—and by (b) the changes between concentrations of 1-OHP at the start and the end of the working week. Taking additional spot samples—that is, at the end of the first shift, midway between shifts, and at the start of the second shift—allowed for a comparison to be made between the mass of excreted 1-OHP derived from a 24 hour urine collection, with the value obtained by calculating area under the excretion rate/time curves.

STRATEGY FOR SKIN PAD SAMPLING

As the impact of intervention workwear was likely to be most significant on the first day of each working week (the potential benefits of new clothing would be likely to be diminished by contamination with process material on subsequent days) skin pads were worn only on these days. Exposure pads consisted of a soft

polypropylene adsorbing material surrounded by self adhesive tape for fixation to the skin.⁷ Each worker had two exposure pads to monitor dermal contamination during the entire work shift at body sites most likely to be contaminated by penetration and permeation through clothing. The groin (junctional region between thigh and abdomen) and the forefoot on the dominant side were used.

ANALYSES

Urine samples

After collection, urine samples were stored at -20°C . All samples were coded and the person who undertook the analysis was blind to the sample sources. Urinary 1-OHP concentrations were measured by high performance liquid chromatography (HPLC). The method was based on those described by Jongeneelen *et al*,¹¹ Tolos *et al*,¹² and Gardiner *et al*¹³ but incorporated modifications as previously reported in detail.¹

Exposure pads

After removal exposure pads were packed in aluminium foil and kept in the dark at 4°C until subsequent analysis for pyrene and benz(a)pyrene. The method involved extraction by dichloromethane followed by high performance liquid chromatographic separation and fluorescence detection.⁷

Treatment of data

Differences between the effects of normal workwear and intervention workwear on excretion of 1-OHP and skin pad deposition of pyrene were examined. Non-parametric statistical methods (Wilcoxon signed rank test), with Minitab software, were used as the number of subjects was small and the observations were not expected to be normally distributed.

Results

The study investigated two workwear phases, (normal workwear *v* intervention workwear), two consecutive work periods (each period containing a mixture of normal and intervention workwear) and two exposure groups (one in the first instance wearing normal workwear followed by intervention workwear, and the other wearing intervention workwear followed by normal workwear).

Only the first comparison, normal workwear phase *v* intervention workwear phase, showed a significant difference in excretion of 1-OHP and skin pad deposition of pyrene. No significant difference in excretion of 1-OHP or skin pad deposition of pyrene was found when the two consecutive working weeks or the two exposure groups were compared.

Tables 2 and 3 relate to information obtained on day 1 of each working week. Table 2 shows the difference between the effects of normal workwear and intervention workwear on mass of 1-OHP excreted over one day as determined from a complete 24 hour urine collection. A notable reduction (55%) was achieved during the intervention

Table 2 Mass of 1-OHP excreted (nmol; 24 h urine collection) on day 1 of each working week (normal *v* intervention workwear) for engineers and maintenance workers (n = 10)

Worker number	Normal workwear day 1	Intervention workwear day 1	Change in mass excreted
E2	37.92	12.35	-25.57
E9	125.78	60.48	-65.30
E10	42.41	22.57	-19.84
E11	43.64	14.98	-28.66
E12	8.43	15.76	+ 7.33
M1	69.12	34.84	-34.28
M2	65.70	24.78	-40.92
M4	69.31	52.73	-16.58
M6	67.77	51.57	-16.20
M8	67.55	65.14	- 2.41
Median (n = 10)	66.62	29.81	-22.70

phase; normal workwear phase median 66.62 (range 8.43–125.78) nmol *v* intervention workwear phase median 29.81 (range 12.35–65.14) nmol. The median reduction in mass excreted (22.7 nmol) was significantly different from zero at the 5% level (95% CI 9.5–40.8 nmol). Only one person (E12) did not show a decrease in mass of 1-OHP excreted during the intervention strategy, but this may be due to an abnormally low result (median 8.43 *v* 66.2 nmol) recorded during the normal workwear period. Review of the diary cards of work activities undertaken did not provide an explanation for this finding: the low urine collection over 24 hours (0.68 l) may have been a factor, but the total creatinine output of 7.8 mmol over 24 hours, although low was not dissimilar from findings during the intervention phase (10 mmol excreted in 2.22 l collected). This 24 hour excretion data for this person is also inconsistent with the skin pad deposition data (see next paragraph and table 3) and remains unexplained, although a discrepancy in the 24 hour urine collections is a possible explanation as is excretion of 1-OHP arising from dietary and environmental sources during the intervention period that were not adequately recorded in the diary card.

The effects of the intervention phase upon skin pad deposition (table 3) supports the findings already discussed. A notable reduction in deposition (82%) was achieved with intervention; normal workwear median 16 (range 6.90–44.50) ng.cm⁻² *v* intervention

workwear median 2.80 (range 2.80–7.05) ng.cm⁻². The median reduction, 13.20 ng.cm⁻² was significant at the 5% level (95% CI 7.3–26.4 ng.cm⁻²). The skin pads were also analysed for benzo(a)pyrene but all results were below the limit of detection (0.05 ng). Considering that the pyrene to benzo(a)-pyrene ratio is greater than 450 in both airborne and liquid phases³ a much higher (about 10-fold) pyrene deposition, in the order of 150 ng.cm⁻² per pad would have been required for benzo(a)pyrene to reach the level of detection.

The impact of intervention over the whole working week was also assessed (table 4). A reduction in the concentration of 1-OHP at the end of the working week was found; intervention *v* normal workwear median 5.73 *v* 7.46 µmol/mol creatinine. Also, the increase in concentration of 1-OHP was on average 50% lower during intervention 2.07 *v* 4.06 µmol/mol creatinine. Neither result, however, achieved significance at the 5% level.

The results were also analysed after excluding values for workers M4 and M6 as they worked through the weekend before the normal workwear phase and therefore their samples from the start of the work period could not be regarded as true baselines. The value at the end of the work period for E2 during the normal workwear phase was also excluded on the basis of a low creatinine value (<4mmol/l),⁸ which might possibly have resulted in a high 1-OHP reading. With these exclusions there was still a substantial impact of intervention upon excretion of 1-OHP although significance was not reached.

Discussion

A large and significant effect was seen on the first day of intervention, as measured by 24 hour mass of 1-OHP and skin pad deposition of pyrene (tables 2 and 3). The magnitude of this effect, however, was not maintained at the end of the study period where the impact upon the concentrations of 1-OHP at the end of the working week was smaller and not significant (table 4). A possible explanation might be that as intervention clothing becomes contaminated during the working

Table 3 Deposition of pyrene (ng.cm⁻²) on skin pads on day 1 of each working week (normal *v* intervention workwear) for engineers and maintenance workers (n = 10)

	Normal workwear day 1			Intervention workwear day 1			Changes in pad deposition
	Skin pad sites		Mean value	Skin pad sites		Mean value	
Worker number	Foot	Groin		Foot	Groin		
E2	24:00	14:10	17.50	ND	ND	2.80	- 14.70
E9	41:60	47:70	44.50	11:30	ND	7.05	- 37.00
E10	14:10	14:90	14.50	ND	ND	2.80	- 11.70
E11	15:10	21:00	18.05	ND	ND	2.80	- 15.25
E12	ND	22:40	12.60	ND	ND	2.80	- 9.80
M1	ND	16:80	9.80	ND	ND	2.80	- 7.00
M2	7:50	29:80	18.65	ND	ND	2.80	- 15.85
M4	10:40	78:30	44.35	ND	ND	2.80	- 41.55
M6	ND	11:30	6.90	ND	ND	2.80	- 4.10
M8	11:30	16:80	9.05	ND	9:30	6.05	- 3.00
Median (n = 10)			16.00			2.80	- 13.20

ND = not detectable (<5.6 ng.cm⁻²), to show average pad concentrations, 50% of the detection limit = 2.8 ng.cm⁻² was used in calculations.

Table 4 1-OHP results ($\mu\text{mol/mol}$ creatinine) for normal v intervention workwear for engineers over a four day working week (96 h) and maintenance workers over a five day working week (104 h) ($n = 10$)

Worker number	Normal workwear			Intervention workwear		
	Start of week	End of week	Change over working week	Start of week	End of week	Change over working week
E2	0.74	48.47	+ 47.73	0.38	8.33	+ 7.95
E9	5.08	36.33	+ 31.25	3.62	8.32	+ 4.70
E10	1.40	3.83	+ 2.43	1.15	7.33	+ 6.18
E11	0.83	7.96	+ 7.13	0.37	4.48	+ 4.11
E12	0.73	2.79	+ 2.06	1.04	4.41	+ 3.10
M1	2.19	4.33	+ 2.14	1.94	3.22	+ 1.28
M2	3.30	8.85	+ 5.55	2.78	3.88	+ 1.11
M4	9.59	5.48	- 4.11	5.99	4.81	- 1.18
M6	15.43	8.07	- 7.36	3.79	6.65	+ 2.86
M8	3.25	7.04	+ 3.79	4.57	7.65	+ 3.08
Median ($n = 10$)	2.72	7.46	+ 4.62	2.36	5.73	+ 2.07

week, the benefit of the new clothing may be diminished over time; maximum effect of intervention would therefore be anticipated to arise on the first day.

On review of the data for the first day of each study phase, there was evidence of some association between mass of 1-OHP excreted and the deposition of pyrene on the skin pads during both the intervention and normal workwear phases (figure). The lack of a correlation is not unexpected, for although mass of 1-OHP is a reflection of total dose of pyrene absorbed, the deposition of pyrene on two selectively placed skin pads is not of itself a direct index of deposition of pyrene over the whole body, nor more importantly of whole body uptake of pyrene. The gradient, however, was steeper for the normal workwear phase, which may suggest that deposition on the skin contributes more to total exposure during the normal workwear phase than the intervention workwear phase. This is further reinforced by the finding that the highest pad contamination was found during the normal workwear phase and corresponded with the highest mass of 1-OHP excreted and the concentration at the end of the working week (engineer E9; normal working period, tables

2, 3, and 4). This implies that intervention prevented deposition on the skin and that the excretion of 1-OHP for the intervention group may be largely attributable to inhalation.

The mass of 1-OHP excreted during the normal workwear period was consistent with previously estimated values at the factory³ and was similar in magnitude to results in other studies.^{4 5 7 8 14 15}

A comparison was made between the mass of 1-OHP excreted from 24 hour urine collections and calculation from area under excretion rate/time curves derived from data of 1-OHP concentration as previously reported.³ In summary this involved the calculation of excretion rates of 1-OHP of individual people ($\mu\text{mol/h}$) and subsequent determination of the area under excretion rate/time curves to estimate total mass excreted over a given time—that is, in 24 hours. With only four data points; start of the working week, end of the first shift, midway between shifts, and at the start of the second shift, area under the curve calculations tended to overestimate the mass excreted by about threefold. This finding shows the limitation inherent in attempting to use a few spot urine results to estimate mass excreted. From our findings many more than four data points would be needed to convert spot sample data into a reasonable estimate of total mass excreted.

There has been much criticism of the use of deposition pads as quantitative measures of dermal contamination,⁶ but the use in this study was of a much more limited nature, because the magnitude of deposition was used as a comparative indicator of deposition over the phases of the study under investigation. No prediction of the total amount of dermally deposited material, nor of the material absorbed by the skin, was attempted from our deposition data. We consider that the use of skin deposition pads in this limited way is entirely justified, and can provide an important source of information.

The study further shows the usefulness of urinary 1-OHP in securing improvements in workplace control and protection for groups of workers who may be exposed to PAHs. Although this study investigated the effect of changes in workwear policy upon the excretion profile of 1-OHP, the biological

Relation between mass of 1-OHP excreted and skin pad deposition of pyrene for normal and intervention workwear.

monitoring technique lends itself to the assessment of other controls that may influence exposure in the workplace—for example, improved ventilation, washing facilities, or changes to working practices, such as shift patterns. Also, a crossover design allows workers to act as their own controls and removes many of the confounding factors in such investigations.

Successive studies^{1,3} at the factory have shown small reductions in median urinary concentrations of 1-OHP at the end of the work period as working practices and hygiene awareness have improved. The effect of the intervention workwear policy upon this value has so far provided the highest impact, and has reduced the previously lowest median value of 7 $\mu\text{mol/mol}$ creatinine to 5.46. This is a clear indication that a further improvement in hygiene at the factory can be achieved by changes in workwear policy. Ideally the workwear policy should be daily provision of clean clothing and frequent chemical cleaning of this clothing would be desirable; however, this ideal was precluded on cost grounds. As a result, the following recommendations were made to management; no personal clothing should be worn and workers should be provided with two sets of workwear, including underclothes. During normal working activities, workwear should be changed at least weekly. Outer clothing should preferably be laundered by a dry cleaning method, as normal washing procedures are not effective in removing contamination of PAHs (Van Rooij, personal communication). Additional workwear should be available for workers accidentally contaminated by process material.

It is anticipated that the impact of these changes will further improve the occupational hygiene practices at the factory and will reduce even further potential exposure to and absorption of process material.

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