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Determinants of Isocyanate Exposures in Auto Body Repair and Refinishing Shops

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As part of the Survey of Painters and Repairers of Auto bodies by Yale (SPRAY), the determinants of isocyanate exposure in auto body repair shops were evaluated. Measurements (*n* = 380) of hexamethylene diisocyanate-based monomer and polyisocyanate and isophorone diisocyanate-based polyisocyanate were collected from 33 auto body shops. The median total reactive isocyanate concentrations expressed as mass concentration of the NCO functional group were: 206 μg NCO/m³ for *spray* operations; 0.93 μg NCO/m³ for samples collected in the vicinity of spray operations done on the shop floor (*near spray*); 0.05 μg NCO/m³ for office or other shop areas adjacent to spray areas (*workplace background*); 0.17 μg NCO/m³ for paint mixing and gun cleaning operations (*mixing*); 0.27 μg NCO/m³ for *sanding* operations. Exposure determinants for the sample NCO mass load were identified using linear regression, tobit regression and logistic regression models. For *spray* samples in a spray booth the significant determinants were the number of milliliters of NCO applied, the gallons of clear coat used by the shop each month and the type of spray booth used (custom built crossdraft, prefabricated crossdraft or downdraft/semi-downdraft). For *near spray* (bystander) samples, outdoor temperature >65°F (18°C) and shop size >5000 feet² (465 m²) were significant determinants of exposure levels. For *workplace background* samples the shop annual income was the most important determinant. For *sanding* samples, the shop annual income and outdoor temperature >65°F (18°C) were the most significant determinants. Identification of these key exposure determinants will be useful in targeting exposure evaluation and control efforts to reduce isocyanate exposures.

Keywords: auto body; exposure determinants; isocyanate; statistical modeling

INTRODUCTION

This exposure assessment was developed as part of the Survey of Painters and Repairers of Auto bodies by Yale (SPRAY). In 1997, Yale and the University of Massachusetts–Lowell initiated a series of epidemiological studies of the respiratory, immunological and physiological responses to isocyanate exposures among auto body shop workers (Redlich *et al.*, 2001, 2002). Polyurethane paints containing isocyanate hardeners were introduced in the US car refinishing market in the late 1960s and began to be widely used in the 1980s. In 1999 there were ~35 000 automotive refinishing facilities in the USA, employing ~207 000 people. In 1999 Connecticut had almost 500 auto body shops employing ~2700 workers (United States Census Bureau, 1999).

Isocyanates have been identified as a major cause of occupational asthma in industrialized countries (Chan-Yeung and Malo, 1995). A reasonable estimate of the prevalence rate of exposed workers is believed to be 5–10% (Torling *et al.*, 1990; Deschamps *et al.*, 1998), although figures as high as 20–30% have been reported for high risk occupations (Vandenplas *et al.*, 1993). Isocyanate asthma is produced by sensitization after exposure to isocyanates and may persist even after removal from exposure. Numerous studies have described cases of asthma in auto body painters (Cockcroft and Mink, 1979; Belin *et al.*, 1981; Clarke and Aldons, 1981; Malo *et al.*, 1983). Therefore, implementation of effective control strategies is very important in

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reducing the incidence and prevalence of isocyanate asthma.

The objective of this paper is to evaluate the determinants of isocyanate exposure for all workers in an auto body repair shop. Identification of the important factors influencing exposure levels between shops and between jobs and locations within a shop will enable the development of models to estimate the exposures of unsampled workers, as well as to provide a basis to prioritize future exposure evaluations and intervention efforts.

METHODS

Air sampling and analysis

Air sampling was performed according to US National Institute of Occupational Safety and Health (NIOSH) method 5525 for total isocyanate group in air (NIOSH, 2002). Air samples were collected at 2 l/min using an IOM inhalable sampler (SKC, Eighty Four, PA) and Gilian (Wayne, NJ) personal sampling pumps calibrated before and after sampling with a DryCal DC-Lite primary flowmeter (Bios International Co., Pompton Plains, NJ). IOM samplers with stainless steel cassettes were loaded with 25 mm quartz fiber filters impregnated with 500 μg of 1-(9-anthracenylmethyl)piperazine (MAP) (NIOSH, Cincinnati, OH). Filters still inside their stainless steel cassettes were transferred immediately after sampling to a jar containing 10 ml of a solution of 1×10^{-4} M MAP in acetonitrile. All supplies and samples were stored and shipped in cooled containers. Comparison of the IOM MAP-treated filter sampler with MAP impinger samples found that impingers and treated filter IOMs perform equally well with respect to collection efficiency for the monomer and total oligomeric hexamethylene diisocyanate (HDI) (Bello *et al.*, 2002a).

Samples were prepared and analyzed according to NIOSH method 5525 using an Agilent HP1100 HPLC system and the Agilent 'Chemstation' software. Slight modifications were made in the gradient to improve resolution for oligomeric aliphatic isocyanates. Modifications of the method, including the pH gradient, and a detailed evaluation of its performance are provided in a companion paper (Bello *et al.*, 2002b).

All exposure units in this paper are expressed as total reactive isocyanate group mass (TRIG), also called the NCO mass (μg). The isocyanate (N=C=O) functional group metric offers many advantages which have been thoroughly discussed in a companion paper by Bello, Woskie, Streicher *et al.* (submitted for publication). They include the ease and consistency of calculation across all types of isocyanates, the relationship of the NCO functional group to health effects, the insensitivity of the metric to market changes in product formulations and the ability to quantify exposures to materials comprised of a mixture of pure isocyanate products using this metric. The limit of detection (LOD) for the method was 4 ng for HDI monomer and 5 ng for HDI and isophorone diisocyanate (IPDI) polyisocyanates (Bello *et al.*, 2002b). For samples below the LOD, a value of LOD/2 was used in the statistical analysis (Hornung and Reed, 1990).

Sampling strategy

The exposure assessment strategy was developed as part of a series of epidemiological studies of the respiratory, immunological and physiological responses to isocyanate exposures among auto body shop workers. The approach developed was to estimate daily exposure level by collecting limited task-based air samples and combining them with worker-specific estimates of time in the task collected from a daily task checklist. This approach was developed as a cost-effective way to estimate the daily exposure for each member of the cohort during their cross-week evaluation of respiratory effects.

The task-based sampling strategy involved collecting samples to represent several common exposure situations in the auto body work environment: samples during spray operations using isocyanate coatings both inside and outside spray booths (*spray*), samples representing the bystander exposure of workers near a spraying operation done outside a spray booth (*near spray*), samples representing bystander exposures in rooms or offices adjacent to where isocyanate operations occurred (*workplace background*), mixing of coatings and spray gun cleaning operations (*mixing*) and sanding or compounding operations done on isocyanate-coated surfaces (*sanding*).

Shops were visited for 4 days, which included 2 days during which medical testing was done and 2 days during which air sampling was done. In each shop an attempt was made to collect one or more samples of each type. Sample collection times varied and were influenced by concerns about reaching the LOD for the isocyanate analysis method. In most cases, personal breathing zone *spray* samples were collected for the time it took to complete the application of all isocyanate-containing coats for a single layer (primer, sealer or clear coat). In some cases, samples spanned several of these layers. *Near spray* samples were collected as personal breathing zone or area samples adjacent to spray operations done outside the booth in the shop. Samples were begun at the start of the spray application and were run for ~2 h to ensure collection of any residual aerosol or vapor in the shop. *Workplace background* samples were set up as area samples in offices or rooms without activities that could generate airborne isocyanates. These samples were generally collected for the whole day to ensure collection of any residual aerosol or vapor from the shop. *Mixing* operation samples were turned on and off during multiple mixing and spray gun cleaning operations done during the day. *Sanding* operations were sampled for the duration of the task.

In conjunction with sample collection, data on a wide range of potential exposure determinants were collected. Some of the information collected for each shop concerned the shop size, business activity, materials or equipment used, engineering controls, work practices used and personal protective equipment available and used. This information is summarized in a companion paper (Sparer *et al.*, 2004). Additional information on a set of potential exposure determinants specific to the sample was also collected. This information included weather conditions, the quantity, mix ratio and type of coating sprayed, whether sampling was done in a spray booth, what the air exchange rate of the booth was, whether bay doors were open during sampling and the sampled workers' distance from a nearby spraying operation. For spray samples, bulk sample analysis allowed estimation of the milliliters of NCO applied based on the volume of coating applied, the percentage of NCO in the hardener and the mix ratio of the hardener, reducer and coating. For sanding, information was collected on whether a wet or dry technique was used, a mechanical or hand method applied, primer/sealer or clear coat was the coating being sanded and how long ago the coating was applied.

Statistical analysis

Sampling results were skewed to the right and truncated on the left due to the presence of LOD values. Hence, exposure levels were described by the median and the 25th and 75th percentiles. Exposure determinant modeling was done using several approaches depending on the degree of truncation of the sample distribution and the normality of the distribution of logged exposures. All modeling was done using SAS Version 8.2 (SAS Inc., Cary, NC). For each type of sample (*spray*, *near spray*, *workplace background*, *mixing* and *sanding*) single predictor models were run to evaluate the significance of a variety of potential exposure determinants. Using a significance level of $P < 0.15$ for entry, multiple regression models were built in a forward selection process. Before building the multiple regression models, determinants were evaluated for potential co-linearity by examining their correlation. Only determinants with, at most, moderate correlations (Pearson $r < 0.7$) were placed in a multiple regression model together.

Spray samples were modeled using linear regression on the natural log of the exposure level (Proc GLM). For *near spray* samples, where 16% of the samples were below the LOD for HDI polyisocyanate, tobit regression on the natural log of the exposure level was used to adjust for the truncation of the distribution. The technique is related to parametric survival analysis, the difference being that left censoring (LOD values) rather than right censoring occurs in this application. Tobit regression uses the data above the LOD and the proportion of data below the reporting limit to compute the slope coefficient by maximum likelihood estimation (Helsel, 1990). The SAS procedure Proc Lifereg was used for the tobit regression. For *workplace background*, *mixing* and *sanding* samples, where over 50% of the samples were below the LOD for HDI polyisocyanate, logistic regression was used (Proc Logistic). The cut-off point for the logistic regression was the 80th percentile of the sample distribution in order to try to find those determinants that were predictive of 'higher' exposures within these tasks. Use of the 80th percentile for the logistic models meant that the occurrence of higher levels was not a common one, ensuring that the odds ratios from the models would closely approximate the rate ratios for that predictor.

RESULTS

Air concentrations

During the study, 380 samples for airborne isocyanate were collected from 33 auto body shops. Sample concentrations can be expressed as the NCO group mass concentration from HDI monomer exposures (Table 1), HDI polyisocyanate exposures (Table 2) or IPDI polyisocyanate exposures (Table 3).

Table 1. HDI monomer (μg/m^3 NCO)

Sample type	*n*	Concentration (μg/m^3)		LOD (%)[a]	>70 μg/m^3 (%)[b]
		Max	Median		
Spray	166	56.14	1.69	10	0
Near spray	37	1.17	0.03	46	0
Workplace background	107	0.24	0.01	60	0
Mixing	45	2.38	0.04	62	0
Sanding	25	0.71	0.05	72	0

[a]LOD (%), percentage of samples below the LOD.
[b]70 μg NCO/m^3 is equivalent to the NIOSH monomer ceiling standard for HDI and the UK-HSE 10 min STEL for any isocyanate.

Table 2. HDI polyisocyanate (biuret and isocyanurate) (μg/m³ NCO)

Sample type	*n*	Concentration (μg/m³)		LOD (%)[a]	>220 μg/m³ (%)[b]	>70 μg/m³ (%)[b]
		Max	Median			
Spray	166	3119.63	190.52	0	45	76
Near spray	37	108.06	0.89	16	0	5
Workplace background	107	12.55	0.02	50	0	0
Mixing	45	117.48	0.10	56	0	4
Sanding	25	35.43	0.12	48	0	0

[a]LOD (%), percentage of samples below the LOD.
[b]70 μg NCO/m³ is equivalent to the NIOSH monomer ceiling standard for HDI and the UK-HSE 10 min STEL for any isocyanate. 220 μg NCO/m³ is equivalent to the Oregon and Bayer Corp. STEL for HDI-based polyisocyanates.

Table 3. IPDI polyisocyanate (μg/m³ NCO)

Sample type	*n*	Concentration (μg/m³)		LOD (%)[a]	>220 μg/m³ (%)[b]	>70 μg/m³ (%)[b]
		Max	Median			
Spray	103	2375.6	38.75	37	18	41
Near spray	16	5.03	0.03	67	0	0
Workplace background	63	6.08	0.01	51	0	0
Mixing	28	23.33	0.05	66	0	0
Sanding	11	1.96	0.06	73	0	0

[a]LOD (%), percentage of samples below the LOD.
[b]70 μg NCO/m³ is equivalent to the NIOSH monomer ceiling standard for HDI and the UK-HSE 10 min STEL for any isocyanate. 220 μg NCO/m³ is equivalent to the Oregon and Bayer Corp. STEL for HDI-based polyisocyanates.

The NIOSH ceiling (10 min) occupational exposure limit (OEL) for HDI monomer is equivalent to 70 μg/m³ NCO (Streicher *et al.*, 2000). The Bayer Corp. (Myer *et al.*, 1993) and Oregon (Janko *et al.*, 1992) have promulgated a short-term exposure limit (STEL) (15 min) equivalent to 220 μg/m³ NCO for the HDI-based polyisocyanates biuret and isocyanurate (Bello, Woskie, Streicher *et al.*, submitted for publication). The UK Health and Safety Executive (HSE) total reactive isocyanate group (TRIG) OEL is 70 μg/m³ NCO for a 10 min STEL exposure to any isocyanate (Silk and Hardy, 1983).

None of the HDI monomer samples were above the NIOSH OEL of 70 μg/m³ (Table 1). For *workplace background*, *mixing* and *sanding* over 60% of samples were less than the LOD, while for *near spray* about half (46%) were below the LOD. Ten percent of the spray samples were below the LOD.

For the HDI polyisocyanate exposures (Table 2), only *spray* samples were above the 220 μg/m³ NCO Bayer/Oregon STEL (45%). For the more stringent UK limit of 70 μg/m³ NCO, 76% of the HDI polyisocyanate spray samples and a small number (two each) of the *near spray* (5%) and *mixing* (4%) samples exceeded this STEL. For *workplace background*, *mixin*g and *sanding* close to 50% of the samples were less than the LOD. Sixteen percent of the *near spray* samples were below the LOD. None of the *spray* samples were below the LOD.

For the IPDI polyisocyanate exposures (Table 3), only *spray* samples were above the 220 μg/m³ NCO Bayer/Oregon STEL (18%) or the more stringent UK limit of 70 μg/m³ NCO (41%). For *workplace background*, *near spray*, *mixing* and *sanding* over 50% (51–73%) of the samples were less than the LOD. For *spray* 37% of the samples were below the LOD.

The relative toxicity of HDI monomer compared with that of the homopolymers biuret and isocyanurate is still under debate. Nevertheless, the percentage of the total NCO contributed by the monomer is very small. In *spray* operations the median monomer fraction of the total NCO was 0.9% (upper quartile 1.4%). Although there is no toxicity data on the polymeric form of IPDI, toxicity data comparing other polyisocyanates suggest little difference in toxicity (Bello, Woskie, Streicher *et al.*, submitted for publication) On this basis the concentrations (μg/m³ NCO) for all the isocyanate exposures were combined into a total NCO concentration metric (Table 4). This exposure metric is comparable to the UK TRIG standard. Using this exposure metric, 49% of the *spray* samples exceeded the recommended Bayer/Oregon exposure limit translated to total μg/m³ NCO, and 79% exceeded the UK TRIG OEL of 70 μg/m³ NCO. A small number (two each) of the *near spray* (5%) and *mixing* (4%) samples also exceeded the UK OEL.

Exposure determinant modeling

Despite efforts to standardize the collection of air samples, the reality of the complex and changing patterns of work in the auto body industry resulted in

Table 4. Total NCO concentration (µg/m^3)

Sample type	n	Concentration (µg/m^3)			>220 µg/m^3 (%)[a]	>70 µg/m^3 (%)[a]
		Median	25–75 percentile	Max		
Spray	166	205.53	81.77–519.46	5519.02	49	79
Near spray	37	0.93	0.33–6.49	108.70	0	5
Workplace background	107	0.05	0.01–0.23	12.61	0	0
Mixing	45	0.17	0.07–1.15	118.30	0	4
Sanding	25	0.27	0.11–1.11	36.14	0	0

[a]70 µg NCO/m^3 is equivalent to the NIOSH monomer ceiling standard for HDI and the UK-HSE 10 min STEL for any isocyanate. 220 µg NCO/m^3 is equivalent to the Oregon and Bayer Corp. STEL for HDI-based polyisocyanates.

Table 5. Sampling time (min) and total NCO filter load (µg)

Sample type	n	Sampling time (min)			Filter load (µg NCO)		
		Min	Mean	Max	Median	25–75 percentile	Max
Spray	166	1	15	67	4.19	1.44–16.85	46.05
Near spray	37	30	83	267	0.14	0.05–1.00	10.13
Workplace background	107	10	294	492	0.02	0.01–0.10	9.12
Mixing	45	1	74	360	0.008	0.006–0.070	5.50
Sanding	25	7	38	177	0.02	0.01–0.40	0.51

a wide range of sampling times for a given task (Table 5). Some samples were collected for only a short time while the immediate task was completed; for example, some *spray* samples were collected only during the short time when one thin coat was applied or *near spray* samples that had to be turned off shortly after the nearby spray job was finished because sanding was to begin immediately adjacent to the sampler. Other samples were allowed to run for longer periods, potentially diluting the air concentration; for example, some *spray* samples were collected over the application of multiple coats with breaks in between to fill the spray gun and for drying or *near spray* samples were collected over the full 4 h of the protocol, even though the nearby spray job was of short duration. Many samples were below the LOD. If half the LOD mass was used for these samples and the sampling time varied, the estimated LOD concentration could vary quite widely, biasing the exposure distribution. Therefore, as an alternative to air concentration, NCO mass collected on the filter for each task type was also calculated (Table 5).

An OEL for the simple NCO mass metric could be estimated if a sampling rate of 2 l/min is assumed for the Bayer/Oregon 15 min STEL of 220 µg/m^3 or the UK 10 min STEL of 70 µg/m^3 (6.6 and 1.4 µg NCO, respectively). When these NCO mass OEL estimates are compared with the sampling data, slightly fewer *spray* samples and slightly more *near spray*, *workplace background* and *mixing* samples exceed the limits than when using the air concentration data.

Due to these concerns about the effect of sampling time on the concentration estimates, exposure determinant modeling was attempted using the total NCO mass on the filter (µg) rather than the NCO mass concentration (µg/m^3) (Table 5). For the exposure determinant modeling, *spray* samples were stratified into two data sets, those taken inside spray booths and those taken on the shop floor, on the assumption that the exposure determinants for the two locations would be different. For example, the interpretation of shop characteristics for booth samples would be different than that for shop floor samples. Characteristics associated with a 'large shop' might include a high volume of painting and multiple jobs per day in the booth, while for the shop floor samples the category of 'large shop' might suggest a larger less cramped work area with greater general dilution ventilation. Thus, the impact of a determinant like 'large shop' could depend on the type of spraying being done (inside or outside the booth). In addition, preliminary analysis suggested that the quantity and type of coatings sprayed were significant determinants of exposure. To evaluate the importance of these factors in combination with location (spraying inside the booth or outside) the data were stratified into in booth and outside booth jobs, since coating applications inside the booth use more isocyanate (median 12 ml NCO, range 1–133 ml) than coating applications on the shop floor (median 5 ml NCO, range <1–34 ml) and spraying outside the booth is most often a primer or sealer coat, while the top clear coat is almost exclusively applied inside a booth.

Spray in booth. The simple linear regression models (Table 6) for *spray* operations in the booth found that milliliters of NCO applied was the most significant predictor of the natural log of the total

Table 6. Exposure determinants

Determinant (variable name)	Regression model point estimate (*P* value)		Tobit model point estimate (*P* value)	Logistic model odds ratio (*P* value)		
	Spray in booth ln(total NCO μg)	Spray no booth ln(total NCO μg)	Near spray ln(total NCO μg)	Workplace background total NCO μg >80 percentile	Mixing total NCO μg >80 percentile	Sanding total NCO μg >80 percentile
Annual Income (US $100 000)	–0.02 (0.29)	–0.02 (0.71)	**–0.20 (0.003)**	**0.90 (0.08)**	**1.09 (.15)**	**1.26 (0.12)**
Cars painted per month	–0.001 (0.74)	0.001 (0.89)	**–0.04 (<0.001)**	0.99 (0.18)	1.00 (0.89)	1.00 (0.86)
Shop size (feet^2)	**–0.005 (0.06)**	–0.002 (0.58)	**–0.03 (<0.001)**	0.99 (0.35)	1.01 (0.43)	1.00 (0.85)
Shop workers (full–time equivalent)	–0.05 (0.17)	–0.07 (0.25)	**–0.25 (<0.001)**	0.98 (0.75)	1.06 (0.49)	**1.37 (0.10)**
Number of painters	–0.04 (0.85)	**–0.57 (0.13)**	–0.34 (0.43)	1.31 (0.41)	0.70 (0.57)	1.78 (0.40)
Clear coat gallons applied/month	**–0.07 (0.05)**	–0.08 (0.22)	**–0.21 (<0.001)**	0.99 (0.92)	0.99 (0.91)	1.67 (0.21)
Hustle (cars painted per month/shop worker)	**0.06 (0.14)**	0.08 (0.20)	**0.21 (0.13)**	**0.87 (0.11)**	0.86 (0.19)	0.88 (0.49)
Crowding (shop workers/shop size)	**–0.002 (0.09)**	**0.003 (0.10)**	0.001 (0.74)	**0.99 (0.05)**	1.00 (0.44)	1.00 (0.52)
Shop size/cars painted per month	**–0.003 (0.06)**	–0.006 (0.24)	**–0.01 (0.13)**	1.00 (0.17)	1.00 (0.77)	1.00 (0.67)
Cars painted per month/painter	–0.001 (0.90)	0.006 (0.67)	**–0.06 (0.003)**	**0.96 (0.12)**	0.99 (0.50)	0.99 (0.50)
Cars painted per month/shop size	**0.659 (0.02)**	**0.81 (0.10)**	1.28 (0.20)	1.02 (0.96)	0.67 (0.56)	1.56 (0.63)
Outdoor temperature (°F)	–0.003 (0.76)	0.02 (0.16)	**–0.04(0.13)**	1.02 (0.30)	1.02 (0.37)	0.96 (0.19)
Indoor temperature (°F)	0.01 (0.42)	0.018 (0.57)	**–0.19(<0.001)**	1.04 (0.16)	1.02 (0.60)	**0.88 (0.11)**
Outdoor relative humidity (%)	0.004 (0.72)	–0.03 (0.09)	**–0.07(<0.001)**	1.00 (0.78)	1.00 (0.87)	**0.94 (0.12)**
Indoor relative humidity (%)	0.003 (0.82)	–0.01 (0.72)	**–0.05 (0.12)**	1.02 (0.40)	1.03 (0.32)	**0.84 (0.03)**
Shop bay doors open	–0.67 (0.46)	–0.49 (0.52)	1.55 (0.17)	1.28 (0.80)	1.83 (0.66)	0.67 (0.76)
NCO applied (ml)	**0.03 (<0.001)**	**0.10 (0.02)**				
Number of coats applied	0.23 (0.31)	–0.30 (0.39)				
Spray in custom built crossdraft booth versus downdraft/semi-downdraft	**0.85 (0.03)**					
Spray in prefabricated crossdraft booth versus downdraft/semi-downdraft	**0.87 (0.05)**					
Distance from nearby spraying operation (feet)			**–0.09 (0.09)**			
Mixing room versus other location for mixing					0.76 (0.76)	
Dry sanding versus wet sanding						1.50 (0.69)
Mechanical sanding versus hand sanding						3.2 (0.34)
Sanding primer/sealer versus sanding clear coat						2.15 (0.53)
Sanding paint <24 h old versus > 24 h old						2.50 (0.39)

Bold text indicates statistically significant findings, $P < 0.15$.

NCO mass collected. In the stepwise model development, the kind of booth used was the next most important predictor of exposures. Custom built crossdraft booth exposures were higher than downdraft/semi-downdraft booth exposures and prefabricated crossdraft booth exposures higher than downdraft/semi-downdraft booth exposures. In addition, several shopwide characteristics were significant. These included shop size, gallons of clear coat used per month, crowding (number of shop workers/square foot shop size), hustle (cars painted per month/shop worker), cramped work space (cars painted per month/square foot shop space). In each case, as the indicator of a larger or busier shop increased, exposures decreased. In the case of cramped work, a high value for the predictor indicated both a busier shop and a smaller size, increasing exposures. Since the significant shop characteristics were correlated (Pearson $r \geq 0.7$), only one could be used in the final model. After testing each of the shop characteristics in the multiple regression model, the gallons of clear coat used by the shop each month was chosen because it produced the best fitting model and because it is relatively easy to acquire from the shops. The final multiple regression model for *spray* operations inside spray booths ($R^2 = 0.39$) was: ln(NCO mass) = 0.95 ($P < 0.001$) + 0.04 ml NCO applied ($P < 0.001$) – 0.05 gallons clear coat used per month ($P = 0.002$) + 1.09 if custom built crossdraft booth ($P = 0.002$) + 0.72 if prefabricated crossdraft booth ($P = 0.09$).

Spray outside booth. The simple linear regression models (Table 6) for *spray* operations outside the booth found that milliliters of NCO applied was the most significant predictor of the natural log of the total NCO mass collected. As with samples in spray booths, many of the shop characteristics had a negative coefficient, indicating that larger or busier shops had lower exposures. The three determinants that were significant were the number of painters in the shop, crowding (number shop workers/square foot shop size) and cramped work (cars painted per month/square foot shop space). Shop size (square feet), as a continuous variable, was not significant. However, a dichotomous variable for shop size (≤5000 feet^2 or >5000 feet^2) showed that larger shops had lower exposures and was significant ($n = 29$, $F = 7.01$, $P = 0.01$). The final multiple regression model for *spray* operations outside spray booths ($R^2 = 0.34$) was: ln(NCO mass) = 0.99 ($P = 0.04$) + 0.09 ml NCO applied ($P = 0.03$) – 1.33 if shop >5000 feet^2 (465 m^2) ($P = 0.01$).

Near spray. The single predictor tobit regression models (Table 6) for *near spray* operations found that several shop characteristics were significant. The most significant were cars painted per month, shop size, number of shop workers and gallons of clear coat applied per month. In each case, as indicators of a larger or busier shop increased, exposures decreased. The models also showed that when seasonal factors increased (indoor temperature and humidity and outdoor temperature and humidity), the exposures significantly decreased.

Since the number of shop workers was highly correlated (Pearson $r \geq 0.7$) with clear coat gallons used per month, cars painted per month and shop size, it was not included in the final multiple tobit regression model for *near spray* operations. Shop size was chosen as the first variable to enter the model because it was available for most shops and was intuitively related to the *near spray* concentration. Among the weather variables, the outdoor temperature was chosen because it is relatively easy to acquire from local weather records. Both shop size and temperature were used as dichotomous variables in the final model. Shop size was dichotomized at 5000 feet^2 (465 m^2), outdoor temperature at 65°F (18°C), the US National Oceanic and Atmospheric Administration (NOAA) cut-off point used to differentiate heating days from non-heating days. The final multiple tobit regression model for *near spray* operations (log likelihood = –50.7) was: ln(NCO mass) = –0.06 ($P = 0.93$) – 2.39 if outdoor temperature >65°F (18°C) ($P < 0.01$) – 1.48 if shop size >5000 feet^2 (465 m^2) ($P = 0.12$). A regular multiple regression model had similar parameter estimates and P values and an R^2 of 0.34.

Since so many *workplace background*, *mixing* and *sanding* samples were below the LOD, logistic models were used to predict the probability of a sample being in the top 20% of the distribution of the sample levels. Significant odds ratios >1 in a single predictor model suggest that the particular determinant being modeled increases the probability of being in the top 20% of the sample distribution. Alternatively, odds ratios <1 suggest that the determinant decreases exposures.

Workplace background. As indicators of a larger or busier shop increased, the likelihood of a *workplace background* area being more highly exposed (in the top 20% of the distribution) decreased. However, only annual income, hustle (cars painted/month/shop worker) and crowding (shop workers/square foot shop size) were statistically significant. An increase in indoor temperature increased the likelihood of being more highly exposed, however, these findings were marginally significant, and outdoor temperature was not a significant determinant. Efforts to dichotomize the indoor and outdoor temperature using the NOAA heating day cut-off point of 65°F (18°C) did not produce significant results. The final model for *workplace background* was a single predictor logistic model using shop annual income (odds ratio 0.90, $P = 0.08$ –2 log likelihood 8081.98), suggesting that larger volume shops have lower exposures.

Mixing. Logistic models showed no consistent or significant exposure determinants for the *mixing*/gun cleaning samples in the top 20% of the distribution.

Sanding. Logistic models found that as many of the indicators of a larger or busier shop increased, the likelihood of *sanding* exposures being higher (in the top 20% of the distribution) increased. However, only annual income and the number of shop workers were statistically significant. In addition, indoor temperature and relative humidity were significant, with increasing indoor temperature and humidity (indoor and outdoor) associated with a decrease in the likelihood of *sanding* exposures being higher. Although not statistically significant, large odds ratios were found for several determinants related to the nature of the process. Dry sanding, mechanical sanding, sanding primer/sealer and sanding freshly painted surfaces (<24 h old) all increased the likelihood of higher exposures (being in the top 20% of the exposure distribution). The final logistic model for *sanding* included only shop annual income (odds ratio 1.5, $P = 0.07$) and outdoor temperature >65°F (18°C) (odds ratio 0.92, $P = 0.13$). Outdoor temperature was used because it can be ascertained for any day using public records and hence it has greater utility in a predictive model for unsampled workdays and for understanding the impact of season on exposure levels. It should be noted that none of the *sanding* samples exceeded any current OEL and the maximum NCO filter load for *sanding* was only 0.5 μg, less than 40% of the UK TRIG standard equivalent.

DISCUSSION

Aliphatic monomer and polyisocyanate concentrations during spray painting in auto body shops have been reported in several studies during the past 15 years. A direct comparison of the results of the SPRAY study results with these previous studies is difficult for a number of reasons. Often the sampling times are quite different, the results were described with a variety of descriptive statistics (arithmetic means, geometric means, etc.), the aims of the studies varied greatly (sampler comparisons, method evaluations, surveillance and epidemiology) and the studies were conducted in different countries and covered different time spans (years to a single day/shop). In addition, in previous studies many different materials were used as calibration standards and different units were used to express the exposure concentrations (mg/m^3 bulk product mass, total reactive isocyanate group mass, diisocyanate monomer mass, etc.).

As part of our work on isocyanates, we have proposed that conversion to a metric based on the total reactive isocyanate group (NCO mass) concentration would help simplify both exposure comparisons and the development of occupational exposure limits in the future (Bello, Woskie, Streicher *et al.*, submitted for publication). A thorough summary of previous isocyanate auto body sampling studies converted to the μg/m^3 NCO metric is reported in a companion paper (Sparer *et al.*, 2004).

The spray operations measured as part of the SPRAY study had a median HDI polyisocyanate concentration of 0.191 mg/m^3 NCO (maximum 3.120 mg/m^3). A study of southern Australian auto body shops reported a geometric mean concentration of 0.07 mg/m^3 NCO (range <0.01–3.5 mg/m^3 NCO) (Pisaniello and Muriale, 1989). Swedish auto body shop exposures during spray painting were between 0.26 and 1.1 mg/m^3 NCO (Torling *et al.*, 1990). The results from this and other studies listed here have been converted to mg/m^3 NCO. A survey of Oregon auto body shops measured a geometric mean concentration of 0.35 mg/m^3 NCO with a maximum of 4 mg/m^3 NCO (Janko *et al.*, 1992). Samples collected during spray operations in a study sponsored by isocyanate manufacturers found a geometric mean level of 0.59 mg/m^3 NCO (range 0.09–2.1 mg/m^3 NCO). A study of French auto body spray operations reported an arithmetic mean level of 0.33 mg/m^3 NCO (range 0.05–0.65 mg/m^3 NCO) (Maître *et al.*, 1996). These findings suggest that the levels measured during auto body operations during our SPRAY study are in the range of most previously reported polyisocyanate exposures.

HDI monomer exposures in each of these studies were all quite low, as was reported in this study. This is the first study to report IPDI polyisocyanate exposure levels in auto body spray painting operations. It would appear that this aliphatic isocyanate is increasingly used in auto body coatings (Sparer *et al.*, 2004).

Little work has been done to date to evaluate the determinants of isocyanate exposure levels in the auto body repair industry. Exposure control evaluations have focused on the effect of specific controls on exposure levels during spray painting, rather than more generic questions of what worker activities or what process and environmental conditions increase the risk of isocyanate exposure. The total NCO mass collected was the metric used as the outcome measure for the exposure determinant modeling. Although one might presume that sample time would be correlated with NCO load, it was not for most types of samples due to the many LOD samples and because of problems faced by the field staff in standardizing the task sampling time.

In the case of spray painting, the final multiple regression models suggest that the amount of material containing isocyanate (ml NCO) is the most important predictor of the total amount of NCO exposure a worker receives during a spraying task. ml NCO applied also acts as an imperfect surrogate for task sample time (Pearson $r = 0.59$) since not all spray samples were restricted to application time only. Another significant predictor was a shop characteristic signifying larger versus smaller shops (gallons of clear coat applied per month). In this case, larger shops with higher clear coat application volumes tended to have lower exposures. This may be because the larger shops are more spacious, have more modern equipment or have better maintenance. The model also predicts that the use of a custom built crossdraft booth during spraying produces significantly higher exposure, with a lesser increase in exposure resulting from the use of a prefabricated crossdraft booth, compared to the use of a downdraft or semi-downdraft booth. A χ^2 evaluation of booth type versus shop size [≤5000 feet^2 and >5000 feet^2 (465 m^2)] showed that smaller shops were significantly more likely to have crossdraft booths and big shops were significantly more likely to have downdraft or semi-downdraft booths (χ^2 32.08, n = 125, df = 2, $P < 0.001$).

Studies to evaluate spray painting controls under experimental conditions have shown that downdraft booths produce lower particulate exposures for workers than do crossdraft or semi-downdraft (Heitbrink *et al.*, 1995). Although our SPRAY study found that exposures in crossdraft booths were higher than other booths, we did not find a statistically significant difference between downdraft and semi-downdraft booths, so they were combined. Lesage *et al.* (1992) also found that crossdraft booths had higher exposures than downdraft booths. This may be related to booth flow rate, since in our study crossdraft booths had lower flow rates (median 0.5 air changes/min) than either semi-downdraft (median 2.0 air changes/min) or downdraft (median 2.7 air changes/min) (Sparer *et al.*, 2004). Other authors have shown that increasing booth flow rate decreases exposure levels (Torling *et al.*, 1990; Goyer, 1995). Although the use of high volume–low pressure (HVLP) spray painting guns has been shown to decrease worker solvent exposures (Heitbrink *et al.*, 1996), this study could not evaluate the impact of HVLP guns because all the shops had adopted their use as the result of a Connecticut State Department of Environmental Protection permit requirement (Sparer *et al.*, 2004). It should also be noted that 30% of the shops used supplied air respirators during spray painting and 86% provided either a reusable or a disposable half-face respirator with organic vapor cartridges and N or P 95 prefilters for use by the painters (Sparer *et al.*, 2004).

Less sampling has been reported for non-spray painting activities or locations within the auto body shop. Exposures during paint mixing and spray gun washing were reported to be between 1 and 27 μg/m^3 NCO HDI polyisocyanate (Torling *et al.*, 1990). These levels are higher than the 0.2 μg/m^3 NCO reported in this study (maximum 118 μg/m^3 NCO). This may be in part because the sampling times for *mixing* ranged from 1 to 360 min, even though the actual procedure of mixing or gun cleaning often takes only 5 min. In addition, during the study the Connecticut State Department of Environmental Protection implemented a permit requirement for the use of enclosed gun cleaners that may have resulted in lower exposures during cleaning tasks. No significant exposure determinants were identified for the *mixing* samples. One possible determinant that was not collected or evaluated was the type of gun cleaning procedure used. If gun cleaning was done outside an enclosure, it can include aerosolizing the cleaning fluid, which may produce higher exposures.

Sanding exposures of 6.3 and 19 μg/m^3 NCO were reported by Pisaniello and Muriale (1989). These levels are higher than the median 0.27 μg/m^3 NCO reported for this study (maximum 36 μg/m^3 NCO). Although most isocyanate exposures were quite low in sanding operations, the logistic model for sanding indicated that larger volume shops (higher annual income) had higher exposures and that outdoor temperature >65°F (18°C) decreased exposures. Higher volume shops could do larger jobs requiring more sanding or might have a higher density of sanding operations within the shop area. Lower exposures with increasing temperature suggests that open doors and windows might reduce sanding dust levels within the shop. Other predictors specifically related to the sanding operation were not statistically significant, which may be partially a function of the small sample size (n = 25). However, the results suggest that some operations may be more likely to produce sanding aerosols that still contain active NCO functional groups. These include dry sanding, mechanical sanding, sanding primer/sealer and sanding freshly painted surfaces (<24 h old).

Samples taken in areas adjoining the spray painting operations were reported to be 2.5 μg/m^3 NCO HDI polyisocyanate by Pisaniello and Muriale (1989), while 100% of samples sampled by Myer *et al.* (1993) were below the limit of quantification (6.5 μg/m^3 NCO HDI). Janko *et al.* (1992) sampled areas adjacent to spraying operations in two surveys and found that all nine were below the LOD (<10.9 μg/m^3 NCO) in one shop and in the other shop fell in the range non-detectable to 39.1 μg/m^3 NCO (mean 8.7 μg/m^3 NCO). In our SPRAY study, non-spray exposures were characterized as *near spray* (area samples in the shop when isocyanate paints were applied on the shop floor) with a median of 0. 93 μg/m^3 NCO (maximum 109 μg/m^3 NCO) or *workplace background* (in areas adjacent to spraying operations) with a median of 0.05 μg/m^3 NCO (maximum 13 μg/m^3 NCO). Thus, our measurements appear to be at the low end of previous sampling results. Nevertheless, between 5 and 14% of the *near spray* samples, 0 and 5% of the *workplace background* samples and 4 and 9% of the *mixing* samples are over the UK OEL of 70 μg/m^3 NCO, depending on whether the raw data or concentration estimated from mass load is used.

To develop information on how to reduce the exposures of non-painter auto body shop personnel, the circumstances producing higher exposures were examined using tobit and logisitic models. For *near spray* workers in the vicinity of an isocyanate application (usually primer or sealer sprayed on the shop floor) exposures were significantly higher in small shops [≤5000 feet^2 (465 m^2)] and when temperatures were <65°F (18°C). Other non-isocyanate studies have also found that warm weather decreases exposure (Woskie *et al.*, 1994; Preller *et al.*, 1995). The final model for *workplace background* was a single predictor logistic model suggesting that larger volume (higher annual income) shops have lower exposures. Thus, it would appear that in the case of workers not engaged in spray painting themselves, the highest risk of isocyanate exposures are found in shops of small size and painting volume, especially during the colder months when doors and windows may not be open.

In the case of *workplace background*, *mixing* and *sanding* samples, the high number of samples below the LOD presented a statistical modeling challenge. Although several studies have used logistic models to evaluate exposure determinants (Nieuwenhuijsen *et al.*, 1996; Gomez, 1997), their use in the occupational hygiene field to examine exposure distributions truncated by many LOD samples has been minimal (Teshke *et al.*, 1995, 2002). Likewise, the use of tobit models to examine mildly truncated exposure distributions has not been previously reported in the occupational literature. The tobit model reported here produced similar results to a linear regression model, which helps in the interpretation of these models. Unfortunately, the interpretation and evaluation of overall model fit for the logistic models is not as clear cut as linear regression approaches. The issue of modeling the determinants of occupational and environmental exposures when air sampling results are truncated and skewed is increasingly a challenge as exposure levels are reduced and exposure assessments are not targeted only at the high end exposures. This area is one prime for the development of new approaches.

CONCLUSION

Isocyanates are well known asthmagens and respiratory irritants, yet exposures during spray painting continue to exceed occupational exposure limits in many auto body shops. In addition, bystanders to spraying operations (*near spray* and *workplace background*) and those performing *mixing*/gun cleaning operations can receive significant isocyanate exposures. Evaluation of exposure determinants for isocyanate exposures points to small low volume shops as a consistent predictor of higher exposures, especially during the colder months when buildings are closed up and general ventilation reduced. In addition, the use of custom built crossdraft spray booths is also predictive of higher isocyanate exposures. Identification of these key determinants is helpful in targeting exposure evaluation and control efforts to reduce isocyanate exposures. Based on work done to date, the SPRAY project team has begun a new initiative to introduce and evaluate exposure control methods in auto body shops. In addition to their utility in targeting exposure controls, these exposure determinant models will be used as part of the retrospective and prospective exposure assessment algorithms developed as part of the SPRAY epidemiological studies.

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