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Historical occupational isocyanate exposure levels in two Canadian provinces

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ABSTRACT

Isocyanates such as toluene 2, 4-diisocyanate (TDI), methylene bisphenyl isocyanate (MDI), and hexamethylene diisocyanate (HDI) are known sensitizers and exposure to these chemicals can result in isocyanate-induced asthma—the leading cause of occupational asthma. A newly created exposure database was available containing occupational isocyanate measurements spanning 1981–1996 from Ontario and British Columbia (BC)—two of the largest provinces in Canada. The aim was to describe the historical measurements relative to exposure thresholds, ascertain differences in the data between provinces, and identify time trends. Descriptive statistics of the observations were summarized and stratified by isocyanate species and province. Chi-square tests and Student's *t*-test were performed to determine differences between provinces. To investigate time trends in the odds of a measurement exceeding the limit of detection (LOD) and time-weighted average (TWA), mixed effects logistic regression models were constructed. In total, 6,984 isocyanate measurements were analyzed, the majority of which were below the LOD (79%). Overall, 8.3% of samples were in excess of the 2014 TLV-TWA of 0.005 ppm. Comparing the two provinces, the proportion of samples exceeding the LOD and TLV-TWA was greater in BC for all isocyanate species. Differences in time trends were also observed between provinces—the odds of a sample exceeding the TLV-TWA decreased over time in the case of MDI (Ontario only), TDI (both Ontario and BC), and other isocyanates (BC only). Our finding that a majority of the exposure measurements was below the LOD is similar to that reported by others. Differences between provinces may be due the fact that isocyanates are classified as a designated substance in Ontario and must adhere to specific exposure control regulations. Limitations of the database, such as finite number of variables and measurements available until 1996 only, presents challenges for more in-depth analysis and generalization of results. An argument is made that a Canadian occupational exposure database be maintained to facilitate risk assessments as well as for occupational epidemiology research.

KEYWORDS

Exposure database; isocyanates; historical exposure; occupational exposure; time trends

Introduction

Isocyanates are low molecular weight substances characterized by the presence of an N=C=O group.^[1] On an annual basis, nearly nine million tonnes of isocyanates are produced globally and demand for these agents is expected to continue to increase over time.^[2,3] Isocyanates remain the most common cause of occupational asthma worldwide and exposure can also lead to contact allergy and irritation of both the skin and respiratory system.^[4–7] The three most prevalent isocyanate species are toluene 2,4-diisocyanate (TDI), methylene

bisphenyl isocyanate (MDI), and hexamethylene diisocyanate (HDI).^[8] Isocyanates are found in a variety of materials including automotive paints, polyurethane foams, coatings, and adhesives.^[8–11]

Jobs at risk of exposure to isocyanates via the skin and respiratory tract include automotive painting, foam-blowing, and the manufacture of various polyurethane products such as furniture, adhesives, and insulation.^[7,12] Despite the broad potential for exposure, very little information exists on the exposure levels (measured or estimated) in Canadian workplaces, either currently or in the past. Using data for 2006, CAREX Canada estimated

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approximately 24,000 Canadians were occupationally exposed to TDI alone.^[13]

It is often assumed that exposure levels in workplaces are generally decreasing over time due to better controls and workplace health and safety requirements,^[14,15] but little quantitative and longitudinal data exists for isocyanate exposure. In the United Kingdom, Stocks et al.^[16] analyzed biomonitoring data from 2006–2014 and found that the odds of detectable isocyanate metabolites in urine seemed to be decreasing over this period among occupations known to be at risk of exposure to isocyanates (motor vehicle, bus, boat, trailer, or aircraft repair). In Canada, specifically the province of Ontario, previous research suggest that there has been a decrease in the number of compensated cases of isocyanate-induced occupational asthma over time^[17] but there has been no investigation of exposure levels over time. Moreover, given that occupational health and safety is provincially governed in Canada, there is a dearth of literature comparing levels of exposure between provinces in the country.

Occupational exposure measurements for a wide variety of exposures (including isocyanates) were recently combined to form a new database, the Canadian Workplace Exposure Database (CWED)—80% of which are from two of Canada's largest provinces, Ontario and British Columbia (BC).^[18] The CWED contains mostly compliance-sampling data from provincial government agencies in Canada and is primarily historical in nature (most provinces no longer have centralized data collection as part of their prevention and/or enforcement programs). The uses of such a database have been outlined previously. Briefly, they include the examination of changes in occupational regulations over time, quantitative risk assessment, identification of high-risk groups, priority setting, and comparison between provinces or with other countries.^[19]

From the CWED, isocyanate exposure measurements collected in BC and Ontario were extracted. The aim was to describe the historical isocyanate exposure levels, to explore provincial differences in the isocyanate measurements, and to estimate trends over time in relation to select thresholds of exposure. To our knowledge, historical isocyanate exposure data from Canadian workplaces has not been explored before. Additionally, as the source of these historical measurements is from a newly-formed database, commentary regarding the utility of this database with regards to research applications is offered.

Methods

A historical occupational exposure database for isocyanates from the Canadian provinces of Ontario and BC was extracted from a larger database—the CWED. The

variables available included sampling date and company name, type of sample (area or personal), job title (though this was not complete for BC), sample duration, air volume, sample mass, and name of the company where sampling took place.

The data were collected for a variety of purposes, including compliance, complaints, routine sampling, and/or research. Based on the name of the company where sampling took place, workplaces were coded according to the 2002 North American Industry Classification System (NAICS).^[20] Observations in which the sampling duration was less than 15 min were excluded from analyses as the goal was to compare measurements to the time-weighted average (TWA) threshold level, and shorter samples were assumed to be less relevant to full-shift exposure levels. Observations with missing or incomplete data were also excluded from analyses.

Since limit of detection (LOD) values were not available in the Ontario dataset, LOD values by sampling year and by species from the BC database were used to populate corresponding Ontario observations. To confirm the validity of the LOD values from BC, the values were cross-referenced with historical LODs in the National Institute of Occupational Safety and Health (NIOSH) Manual of Analytical Methods (NMAM) (methods 5521, 2535, and 5522). The LODs used were: MDI 0.4 µg/sample (all years), TDI 0.5 µg/sample (all years), HDI 0.35 µg/sample (all years) and “other” 0.4 µg/sample (1984–1986) as well as 0.2 µg/sample (1987–1994). The reported exposure measurements in mass were compared to these aforementioned LODs and whether a value was above or below the corresponding LODs was noted. The resulting isocyanate concentration was subsequently calculated based on the mass (numerator) and the associated sampling volume (denominator) and reported in units of parts per million (ppm). Since the majority of samples were collected for compliance purposes by government inspectors/officers, an assumption was made that the calculated concentration was equivalent to the 8-hr time-weighted average.

Descriptive statistics (frequencies and percentiles) were used to summarize the measurement data available. These descriptive statistics were stratified by isocyanate species, type of sample (area or personal), and province where the measurements were collected. Where isocyanate mass isocyanates exceeded the AL and TLV-TWA relative to MDI, HDI, and TDI. The percentiles are shown in values were greater than the LOD, the sample concentrations were compared to two threshold levels: (i) the 2014 ACGIH TLV-TWA of 0.005 ppm (which is applicable to all isocyanate species) and (ii) the corresponding Action Level (AL) of 0.0025 ppm (or 50% of the TLV-TWA). As a best practice, actions such as additional exposure monitoring or the implementation of control

Table 1. Description of occupational isocyanate measurement data.

	N	n (%) > LOD	75 th %ile	95 th %ile	Max. (ppm)	n (%) > AL	n (%) > TLV-TWA
<i>Overall</i>	6984	1464 (21%)	< LOD	0.014	35.37	860 (12.3%)	578 (8.3%)
BC	1164	401 (34.4%)	0.0041	0.034	2.15	341 (29.3%)	258 (22.2%)
Ontario	5820	1063 (18.3%)	< LOD	0.0076	35.37	519 (8.9%)	320 (5.5%)
Area	6650	1307 (19.6%)	< LOD	0.011	5.37	742 (11.1%)	492 (7.4%)
Personal	334	157 (47%)	0.0062	0.053	35.37	118 (35.3%)	86 (25.8%)
MDI	3444	729 (21.2%)	< LOD	0.014	35.37	407 (11.8%)	290 (8.4%)
HDI	500	43 (8.6%)	< LOD	0.0051	4.04	32 (6.4%)	17 (3.4%)
TDI	2613	569 (21.8%)	< LOD	0.0088	1.17	311 (11.9%)	189 (7.2%)
Other	427	123 (28.8%)	0.0030	0.074	2.15	110 (45.8%)	82 (19.2%)

measures should be considered for any exposure concentration above the AL.

Differences between provinces were investigated using Student's *t*-test (sample duration) and Chi-square tests (> LOD, > AL, >TLV-TWA). The number of samples collected by year was described, including the annual breakdown by sample type and isocyanate species. In order to investigate time trends where a significant portion of data was below the LOD, the exposure data was simplified into binary outcomes: yes/no – above LOD; yes/no – above TLV-TWA. Mixed effects logistic regression models (PROC GLIMMIX) were constructed to investigate time trends in the odds of a measurement exceeding the LOD and the TLV-TWA separately.^[9] A random intercept for industry was included in the models to account for correlation between measurements collected in the same industry. The predictor variable of primary interest in these models was year of measurement (continuous variable). Models were run using area samples only due to the much smaller proportion of personal samples (4.8% of database), and constructed separately by isocyanate species, adjusting for province and sampling duration. Subsequently, models stratified by province were constructed by isocyanate species, and adjusted only for sampling duration. The level of statistical significance was set at $p < 0.05$. Statistical analyses were performed using Statistical Analytical System (SAS) 9.2 software (Cary, NC).

Results

Overall

In total, 7,463 isocyanate measurements were collected in Ontario and BC between 1981 and 1996. There were 479 samples excluded from the final dataset because they had mass units that could not be verified ($n = 225$), the sample air volumes were unavailable ($n = 26$), the sampling durations were less than 15 min ($n = 119$), or the industry descriptions were unavailable ($n = 109$). This resulted in a total of 6,984 observations available for the current analysis. A large majority of measurements were area samples

($n = 6,650$), compared to just 334 personal exposure samples. Most of the samples were collected in Ontario ($n = 5,820$, 83%) with the remainder from BC ($n = 1,164$, 17%).

Overall, only 21.0% ($n = 1,464$) of the reported mass values exceeded the LOD. Compared to exposure thresholds, 12.3% of the measurements exceeded the AL of 0.0025 ppm and 8.3% of samples were in excess of the TLV-TWA of 0.005 ppm. A larger proportion of the personal samples exceeded the exposure thresholds compared with the area samples (% > AL 35.3 vs. 11.1% and % > TLV-TWA 25.8 vs. 7.4%, respectively). When stratified by species, the dataset consisted of 3,444 MDI samples, 2,613 TDI samples, 500 HDI samples, and the remainder ($n = 427$) were categorized into a group referred to as “other” which included bitolyene diisocyanate ($n = 6$), isophorone diisocyanate ($n = 178$), methylene bis-(4-cyclohexylisocyanate) ($n = 9$), and isocyanates not otherwise specified ($n = 234$). A larger proportion of the “other” isocyanates exceeded the AL and TLV-TWA relative to MDI, HDI, and TDI. The percentiles are shown in Table 1.

When stratified by industry, the sectors (and associated NAICS code) with the highest proportion of observations in excess of the TLV-TWA for each isocyanate species were: 811 - Repair and Maintenance (HDI), 339 - Miscellaneous Manufacturing (MDI), 333 - Machinery Manufacturing (TDI), and 332 - Fabricated Metal Product Manufacturing (Other) (not shown). A summary of all measurements by North American Industry Classification System (NAICS) at both the 2-digit and 3-digit level is provided in a Supplementary Table (online), including the n (%) above each of the LOD, the action level (AL), and the time weighted average value (TLV-TWA).

Comparison between provinces

Table 2 shows a comparison of the isocyanate exposure data between the two provinces. Although there were a larger number of samples collected from Ontario, the proportion of samples exceeding the LOD, AL and TLV-TWA was greater in the BC data for all isocyanate species. The overall mean sampling duration was longer in Ontario

Table 2. Comparison of measurement data from British Columbia (BC) and Ontario (ON). All results shown as n (column %) unless otherwise noted.

		BC	ON ^a
Overall	Number of samples, n	1164	5820
	> LOD	401 (34%)	1063 (18%)
	> AL	341 (29%)	519 (8.9%)
	> TLV-TWA	258 (22%)	320 (5.5%)
	Duration in minutes, mean (sd; range)	59.0 (58.6; 15–435)	93.7 (53.1; 15–375)
MDI	Number of samples, n	707	2737
	> LOD	174 (25%)	555 (20%)
	> AL	142 (20%)	265 (10%)
	> TLV-TWA	118 (17%)	172 (6.3%)
	Duration in minutes, mean (sd; range)	63.0 (63.8; 15–384)	87.3 (49.2; 15–360)
HDI	Number of samples, n	100	400
	> LOD	27 (27%)	16 (4.0%)
	> AL	20 (20%)	12 (3.0%)
	> TLV-TWA	10 (10%)	7 (1.8%)
	Duration in minutes, mean (sd; range)	43.9 (19.0; 15–95)	99.7 (64.2; 15–375)
TDI	Number of samples, n	164	2449
	> LOD	83 (51%)	486 (20%)
	> AL	74 (45%)	237 (10%)
	> TLV-TWA	51 (31%)	138 (5.6%)
	Duration in minutes, mean (sd; range)	56.6 (46.0; 15–274)	98.9 (53.0; 15–360)
Other	Number of samples, n	193	234
	> LOD	117 (61%)	6 (2.6%)
	> AL	105 (54%)	5 (2.1%)
	> TLV-TWA	79 (41%)	3 (1.3%)
	Duration in minutes, mean (sd; range)	53.6 (60.3; 15–435)	96.0 (68.6; 15–340)

^aIn all instances, the differences between the two provinces was statistically significant ($p < 0.05$).

(93.7 min) than in BC (59.0 min) and this increased sampling duration in Ontario was found for all isocyanate species (Table 2).

The relative number of area and personal samples (Figure 1) was similar between Ontario and British Columbia; 91.8% of samples were area samples in BC vs. 95.9% of samples in Ontario. MDI was the predominant species sampled in British Columbia while in Ontario measurements of MDI and TDI were most common. All comparisons between provinces were statistically significant ($p < 0.05$).

Trends in exposure over time

Given the small proportion of personal samples (<5% overall), only area samples were examined in the time

trend analysis. Time trends for having a detectable (> LOD) sample and a sample exceeding the TLV-TWA are shown in Table 3, stratified by isocyanate species for both provinces combined and individually.

The time trend for having a detectable sample (>LOD) varied between provinces and isocyanate species. In BC, significant decreasing trends of having a sample > LOD over time were observed for HDI (OR 0.18, 95% CI 0.04–0.70) and other isocyanates (OR 0.42, 95% CI 0.26–0.68). In Ontario, the only significant result was increasing odds of a detectable TDI sample over time (OR 1.05, 95% CI 1.01–1.10). Due to a limited number of samples in excess of the LOD, time trends for other isocyanates species from Ontario could not be explored.

The odds of a sample exceeding the TLV-TWA decreased over time in the case of MDI (Ontario only), TDI (both Ontario and BC), and other isocyanates (BC only). No significant time trends were observed for the odds of an HDI sample exceeding the TLV-TWA in either province, perhaps due in part to a smaller sample size for this isocyanate species.

In models that combined data from both BC and Ontario, the time trend models were adjusted for province as well as sample duration. In the combined models for HDI and TDI, samples from British Columbia were consistently more likely to exceed both the LOD and the TLV-TWA. This was not the case for MDI where there was no statistically significant difference between the provinces. Inter-provincial differences were not investigated in the combined model for other isocyanates as there were so few measurements from Ontario that exceeded either threshold.

In the models investigating the odds of a sample exceeding the TLV-TWA, an increase in sample duration of 10 minutes was associated with a lower odds of a sample exceeding the TLV-TWA for MDI (OR 0.89, 95% CI 0.85–0.93) and other isocyanates (OR 0.82, 95% CI 0.72–0.94) (Table 3).

In the models investigating the odds of a sample exceeding the LOD, the results were less consistent. In BC, among HDI samples, an increase of 10 min in sample duration was associated with an increase in the odds of having a sample above the LOD (OR 1.51, 95% CI 1.10–2.07). Among TDI samples from Ontario, an increase of 10 min in sample duration was associated with small increase in the odds of a sample being above the LOD (OR 1.04, 95% CI 1.01–1.06).

Discussion

There were nearly 7,000 isocyanate measurements in the database consisting of data from BC and Ontario. An overwhelming majority of the data (79%) was below the

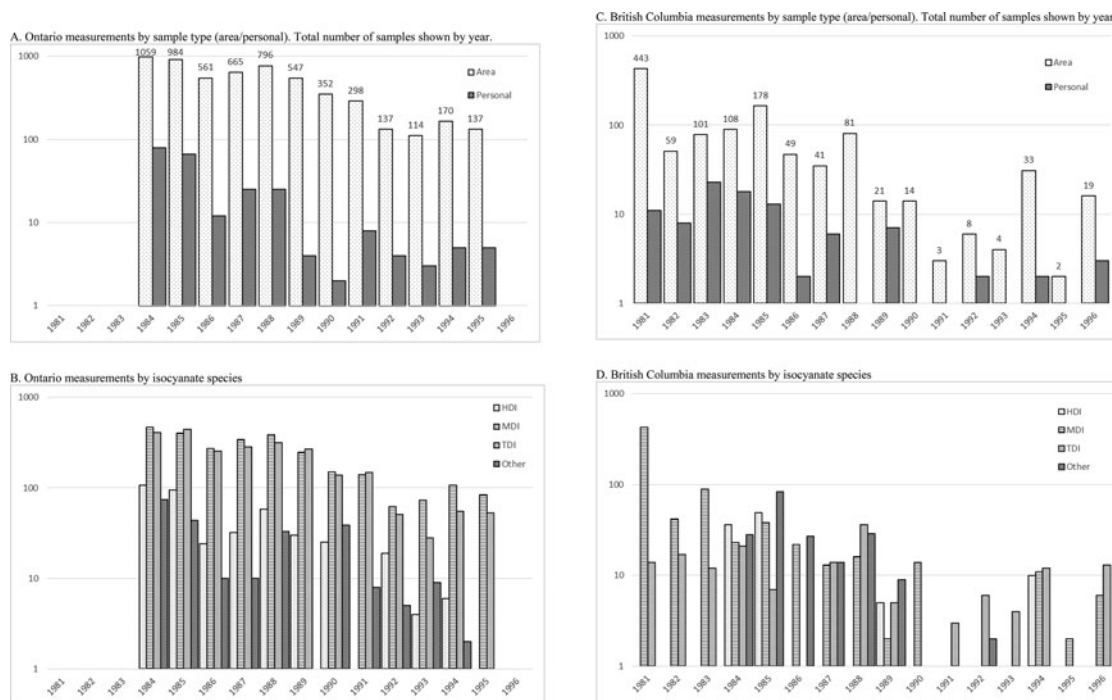


Figure 1. A-D: Number of measurements by year and by sample type (A, C) and species (B, D), shown for Ontario (A, B) and British Columbia (C, D) separately. A. Ontario measurements by sample type (area/personal). Total number of samples shown by year. B. Ontario measurements by isocyanate species. C. British Columbia measurements by sample type (area/personal). Total number of samples shown by year. D. British Columbia measurements by isocyanate species.

LOD which posed significant challenges for analysis of the exposure data. The use of a binary outcome provides an alternative and this approach has been used in previous studies of isocyanate exposure.^[9,16] The general trend observed was that samples were less likely to exceed the TLV-TWA over time and this finding is consistent with other airborne contaminants.^[21-24] Similar time trends were observed in models constructed to examine the odds of a sample that exceeded the AL over time (results not shown). The trends observed for detectable ($> \text{LOD}$) samples over time was less consistent and varied by isocyanate species and province. Despite these limitations, our findings of very low exposure levels in a reasonably large exposure database for isocyanates were also observed by Booth and colleagues in their study of MDI exposures in which 75% of the data, collected from various manufacturing processes from workplaces in the United States, was below the LOD.^[25]

The database consisted of exposure measurements from two of the largest provinces in Canada. Although a majority of these samples originated from Ontario, BC had a larger proportion of samples that were $> \text{LOD}$, $> \text{AL}$ and $> \text{TLV-TWA}$ for all isocyanate species. Although the mean sampling duration in British Columbia was significantly shorter, there was no clear association between sample duration and proportion exceeding LOD, AL, or TLV-TWA. While the database did not allow for an

investigation of the reasons for these provincial differences, it is possible that differences in the regulatory environments could have led to differences in risk, such that exposure was truly higher in BC. Ontario's Occupational Health and Safety Act has a *Designated Substances* regulation for select chemical hazards that includes isocyanates (<http://www.ontario.ca/laws/regulation/090490>). This regulation, developed in the 1980's, requires that special considerations are given to isocyanate exposure in Ontario workplaces, including a "duty to limit airborne exposure" with specific requirements surrounding exposure controls at multiple levels within the hierarchy of controls. These exposure control programs also included a duty to monitor and post the results of any monitoring. No such regulation exists in BC, and this could at least partly explain the higher number of exposure measurements collected in Ontario as well as the higher levels of exposure observed in BC. In addition, a review of Statistics Canada labor force survey data from 1981-1996 indicates that Ontario had more manufacturing than BC which might explain the differences in the number and type of isocyanates sampled.^[26]

The number of occupational exposure samples collected in Canada (as elsewhere) has decreased substantially over time^[18] and this presents a challenge in interpretation of results. In such environments, it is conceivable that sample selection would be limited to

Table 3. Linear time trends for the odds of a detectable sample (A) and a sample that exceeded the threshold limit value time-weighted average (TLV-TWA) (B) for each isocyanate species. Models included area samples only. Each line represents a separate model.

		OR (95% CI)			
A) Outcome: ≥ LOD	Total n	n > LOD or TLV-TWA	Sample Year	Sample Duration ^a	British Columbia (BC) vs. Ontario (ON)
All Data					
HDI ^b	493	40	0.68 (0.48–0.96)	1.05 (0.96–1.15)	6.22 (1.97–19.7)
MDI ^b	3265	650	1.00 (0.96–1.03)	0.98 (0.96–1.00)	0.96 (0.71–1.30)
TDI ^b	2479	504	1.02 (0.98–1.06)	1.03 (1.01–1.05)	3.64 (2.43–5.48)
Other ^c	413	113	0.40 (0.25–0.65)	0.90 (0.82–0.98)	—
British Columbia Only					
HDI ^c	97	25	0.18 (0.04–0.70)	1.51 (1.10–2.07)	—
MDI ^c	670	170	1.05 (0.96–1.15)	0.96 (0.93–1.01)	—
TDI ^c	122	61	0.93 (0.85–1.01)	0.96 (0.89–1.05)	—
Other ^c	180	108	0.42 (0.26–0.68)	1.01 (0.93–1.09)	—
Ontario Only					
HDI ^c	396	15	0.67 (0.43–1.03)	0.94 (0.82–1.08)	—
MDI ^c	2595	480	1.00 (0.96–1.04)	0.99 (0.96–1.01)	—
TDI ^c	2357	443	1.05 (1.01–1.10)	1.04 (1.01–1.06)	—
Other ^{c,d}	233	5	^d	^d	—
B) Outcome: ≥ TLV-TWA					
All Data					
HDI ^b	493	16	0.77 (0.52–1.12)	0.99 (0.87–1.13)	2.48 (0.64–9.66)
MDI ^b	3265	245	0.89 (0.83–0.95)	0.89 (0.85–0.93)	1.34 (0.86–2.08)
TDI ^b	2479	157	0.88 (0.83–0.94)	0.98 (0.95–1.02)	4.84 (2.95–7.93)
Other ^c	413	74	0.25 (0.14–0.47)	0.82 (0.72–0.94)	—
British Columbia Only					
HDI ^c	97	10	0.32 (0.06–1.80)	1.10 (0.85–1.42)	—
MDI ^c	670	116	1.02 (0.92–1.14)	0.92 (0.86–0.98)	—
TDI ^c	122	36	0.86 (0.78–0.95)	0.93 (0.81–1.06)	—
Other ^c	180	72	0.26 (0.14–0.48)	0.89 (0.74–1.07)	—
Ontario Only					
HDI ^c	396	6	0.84 (0.55–1.30)	0.90 (0.75–1.09)	—
MDI ^c	2595	129	0.87 (0.79–0.94)	0.89 (0.84–0.93)	—
TDI ^c	2357	121	0.88 (0.82–0.96)	0.99 (0.95–1.03)	—
Other ^{c,d}	233	2	^d	^d	—

^aSample duration shows the change in OR per 10-min increase in sample duration. ^bAdjusted for sample duration and province. ^cAdjusted for sample duration only. ^dDid not converge. - Not included in the model.

high-risk or “worst-case” workplaces or exposed workers in more recent years, as an attempt by hygienists to capture the higher-risk workers with less resources.^[27] If worst-case scenarios were sampled preferentially, the odds of having a detectable sample may increase over time. But, as noted earlier, exposure levels are often assumed to be decreasing over time due to better controls and workplace health and safety requirements,^[14,15] which would lead to a decrease in the odds of a measurement exceeding the TLV-TWA over time; this interpretation is supported by our findings for MDI and TDI, particularly in Ontario.

A large majority (95%) of the observations in the dataset were area samples. This is likely due to the fact that, at the time of sample collection, compliance monitoring for isocyanate was measured using impingers^[28] and, given the fragile nature of these devices, they are less desirable for use in personal sampling.^[6] Due to the small number of personal samples, only area sample were analyzed in the time trend models and, as such, our results cannot speak directly to any trends in the detectable samples or samples exceeding the TLV-TWA in the breathing

zone of workers. This is a problem with isocyanates exposure assessment in general, and indeed for other exposures that are difficult to measure for various reasons.

Additional limitations should be discussed regarding the data. The “other” isocyanate species category (which included bitolylene diisocyanate, isophorone diisocyanate, methylene bis-(4-cyclohexylisocyanate), and isocyanates not otherwise specified) had a higher proportion of samples exceed the TLV-TWA compared to TDI, HDI, and MDI. Although in some cases these “other” samples were identified as specific isocyanate species in the databases, it is likely that many samples included in the “other” category and listed as “isocyanates not otherwise specified” were actually TDI, MDI, or HDI. If these samples had been identified appropriately, a higher proportion of samples may have exceeded the selected thresholds, but this would not appreciably change our interpretation of the analyses.

With regards to data quality, no LODs were reported from Ontario; these values were extrapolated from the corresponding sampling year from the BC database. However, given the standard laboratory methods in use

at the time in Canada, it is likely that the LOD substitutions made were reasonable, and no obvious anomalies in the data were observed by using these substitutions.

As with many other exposure databases, there is a lack of detailed occupational hygiene information in the CWED including the rationale for sample collection and whether any control measures (such as personal protective equipment or ventilation) were in place. Although some of the exposure measurements exceeded current exposure limits, no information on the frequency or the duration of isocyanate exposures for workers in the sampled workplaces was available. It is also not possible to know to what degree the difficulty of sampling mechanisms for isocyanates impacted the hygienists/technicians' ability to accurately measure exposure to isocyanates.

Analysis of the data, which originated from a newly formed exposure database has revealed some interesting findings; however, it should be noted that it is limited in its ability to draw generalizable conclusions. This speaks to the need for continual maintenance of occupational exposure databases, as has been called for previously by Hall and colleagues in Canada.^[18] In the database under investigation, analysis of time trends was limited to the years 1981–1996. No measurements beyond 1996 were available for Ontario due to a government decision to close the laboratory responsible for analyses; limited data was available beyond 1996 for BC, but none met the requirements for inclusion. These two provinces have since ceased the centralized collection and maintenance of occupational exposure data, as is the case in most other Canadian jurisdictions with the exception of Quebec. This shifting of responsibility for exposure monitoring from regulatory bodies to employers that occurred in the 1990s has led to decreased access to up-to-date exposure data, with broad-reaching consequences for exposure assessment science and beyond.^[18]

Further, the exposure databases that do exist (including CWED) are not always complete (e.g. missing reason for sampling, no LOD reported, etc.). This fact has been discussed and attempts have been made to address this shortcoming—most specifically via an expert-based workshop about appropriate exposure database design in 1999 in London.^[29] In order to have continued value for research, policy and practice in occupational health and disease prevention, current and complete (as much as possible) data is required. The benefits of such a system of cataloging include, but are not limited to, more timely surveillance of exposure levels over time, the impact to assess changes or differences in regulations, the ability to associate trends in occupational illness and injury claims with measured exposure levels, and identification of determinants of exposure as targets for exposure

prevention efforts. More broadly speaking, it allows for accurate determination of exposure for use in risk assessment and for occupational epidemiology.

On a positive note, the work to improve exposure databases in Canada continues on several fronts, particularly with the ongoing development of the CWED, which seeks to include recent data shared by private organizations and provinces, and in Québec through the development of new databases using exposure data available in the peer reviewed literature.^[30] These initiatives and others will serve to provide a much improved source of information on exposure levels across the country, which in turn can support the development of exposure assessment tools for large epidemiological studies of occupational disease as well as lend evidence-based information when developing policies to protect the health and well-being of workers.

Conclusion

Our findings suggest that isocyanate exposure in BC and Ontario decreased over the time period examined and that there were differences in exposure between the two provinces. This conclusion is not without its caveats as this study also demonstrated a general lack of reliable exposure monitoring data in Canada with a marked decrease in sampling frequency over time. In conjunction with changes over time in legislation and/or practices on the shop floor, this should add to the general call for better exposure assessment of hazardous exposures in occupational settings.

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References

- [1] Nakashima, K., T. Takeshita, and K. Morimoto: Review of the occupational exposure to isocyanates: Mechanisms of action. *Environ. Health Prev. Med.* 7(1):1–6 (2002).
- [2] Ceresana: “Market Study: Polyurethanes (PUR) and Isocyanates (MDI & TDI).” Available at <http://www.ceresana.com/en/market-studies/plastics/pur-isocyanates> (accessed January 1, 2015).
- [3] Fisseler-Eckhoff, A., H. Bartsch, R. Zinsky, and J. Schirren: Environmental isocyanate-induced asthma: Morphologic and pathogenetic aspects of an increasing occupational disease. *Int. J. Environ. Res. Public Health* 8:3672–3687 (2011).

- [4] Meredith, S.K., J. Bugler, and R.L. Clark: Isocyanate exposure and occupational asthma: a case-referent study. *Occup. Environ. Med.* 57(Mdi):830–836 (2000).
- [5] Roberge, B., S. Aubin, C. Ostiguy, and J. Lesage: "Guide for Safe Use of Isocyanates - An Industrial Hygiene Approach." Available at <http://www.irsst.qc.ca/media/documents/PubIRSST/RG-773.pdf> (accessed January 1, 2015).
- [6] Reh, B.D.: "A Summary of Health Hazard Evaluations: Isocyanates, 1989 to 2002." Available at <http://www.cdc.gov/niosh/docs/2004-116/pdfs/2004-116.pdf> (accessed January 1, 2015).
- [7] Reeb-Whitaker, C., N.J. Anderson, and D.K. Bonauto: Prevention guidance for isocyanate-induced asthma using occupational surveillance data. *J. Occup. Environ. Hyg.* 10:597–608 (2013).
- [8] LeBreton, E.: "Isocyanates." Available at <http://www.tc.gc.ca/eng/canutec/articles-isocyanate-461.htm> (accessed June 4, 2015).
- [9] Pronk, A., F. Yu, J. Vlaanderen, et al.: Dermal, inhalation, and internal exposure to 1, 6-HDI and its oligomers in car body repair shop workers and industrial spray painters. *Occup. Environ. Med.* 63(9):624–631 (2006).
- [10] Bello, D., C. Redlich, M.H. Stowe, et al.: Skin exposure to aliphatic polyisocyanates in the auto body repair and refinishing industry: II. A quantitative assessment. *Ann. Occup. Hyg.* 52:1171–24 (2008).
- [11] Vangronsveld, E., S. Berckmans, K. Verbinnen, C. Van Leeuw, and C. Bormans: Isocyanate and total inhalable particulate air measurements in the European wood panel industry. *Int. J. Hyg. Environ. Health* 213(6):475–488 (2010).
- [12] Occupational Safety & Health Administration: "Isocyanates." Available at <https://www.osha.gov/SLTC/isocyanates/index.html> (accessed January 1, 2015).
- [13] CAREX: "Toluene Diisocyanates." Available at http://www.carexcanada.ca/en/toluene_diisocyanates/occupational_estimate/ (accessed January 1, 2015).
- [14] Creely, K.S., H. Cowie, M. Van Tongeren, H. Kromhout, J. Tickner, and J.W. Cherrie: Trends in inhalation exposure - A review of the data in the published scientific literature. *Ann. Occup. Hyg.* 51(8):665–78 (2007).
- [15] Symanski, E., L.L. Kupper, and S.M. Rappaport: Comprehensive evaluation of long-term trends in occupational exposure: Part 1. Description of the database. *Occup. Environ. Med.* 55(5):300–309 (1998).
- [16] Stocks, S.J., K. Jones, M. Piney, and R.M. Agius: Isocyanate exposure and asthma in the UK vehicle repair industry. *Occup. Med.* 65(9):713–718 (2015).
- [17] Ribeiro, M., S.M. Tarlo, A. Czyrka, et al.: Diisocyanate and non-diisocyanate sensitizer-induced occupational asthma frequency during 2003 to 2007 in Ontario, Canada. *J. Occup. Environ. Med.* 56(9):1001–1007 (2014).
- [18] Hall, A.L., C.E. Peters, P.A. Demers, and H.W. Davies: Exposed! Or not? The diminishing record of workplace exposure in Canada. *Can. J. Public Heal.* 105(3):e214–7 (2014).
- [19] Peters, C.E., C.B. Ge, A.L. Hall, H.W. Davies, and P.A. Demers: CAREX Canada: an enhanced model for assessing occupational carcinogen exposure. *Occup. Environ. Med.* 64–71 (2014).
- [20] Statistics Canada: North American Industry Classification System (NAICS) 2002 – Canada. Available at <http://www.statcan.gc.ca/subjects-sujets/standard-norme/naics-scian/2002/naics-scian02l-eng.htm> (accessed October 4, 2013).
- [21] Okun, A., G. Cooper, A.J. Bailer, J. Bena, and L. Stayner: Trends in occupational lead exposure since the 1978 OSHA lead standard. *Am. J. Ind. Med.* 45:558–572 (2004).
- [22] Vermeulen, R., J. de Hartog, P. Swuste, and H. Kromhout: Trends in exposure to inhalable particulate and dermal contamination in the rubber manufacturing industry: Effectiveness of control measures implemented over a nine-year period. *Ann. Occup. Hyg.* 44(5):343–354 (2000).
- [23] Symanski, E., C.C. Chang, and W. Chan: Long-term trends in exposures to nickel aerosols. *AIHA J.* 61(May 2015):324–333 (2010).
- [24] Coble, J.B., P.S. Lees, and G. Matanoski: Time trends in exposure measurements from OSHA compliance inspections of the pulp and paper industry. *Appl. Occup. Environ. Hyg.* 16:263–270 (2001).
- [25] Booth, K., B. Cummings, W.J. Karoly, et al.: Measurements of airborne methylene diphenyl diisocyanate (MDI) concentration in the U.S. workplace. *J. Occup. Environ. Hyg.* 6:228–238 (2009).
- [26] Statistics Canada: "Table 282–0008- Labour Force Survey Estimates (LFS), by North American Industry Classification System (NAICS), Sex and Age Group." Available at <http://www5.statcan.gc.ca/cansim/a26?lang=eng&id=2820008> (accessed May 18, 2016).
- [27] Lavoue, J., M.C. Friesen, and I. Burstyn: Workplace measurements by the US Occupational Safety and Health Administration since 1979: descriptive analysis and potential uses for exposure assessment. *Ann. Occup. Hyg.* 57(1):77–97 (2013).
- [28] Levine, S., K. Hillig, V. Dharmarajan, M. Spence, and M. Baker: Critical review of methods of sampling, analysis, and monitoring for TDI and MDI. *Am. Ind. Hyg. Assoc. J.* 56(6):581–589 (1995).
- [29] Tickner, J., T.W. Armstrong, and T.F. Bloom: Workshop on harmonization of serving future needs with occupational exposure databases? Inhalation modeling, Session IIIA. *Appl. Occup. Environ. Hyg.* 16(2):309–314 (2001).
- [30] Beaudry, C., J. Lavoué, J-F. Sauvé, et al.: Occupational exposure to silica in construction workers: a literature-based exposure database. *J. Occup. Environ. Hyg.* 10:71–77 (2013).

SUPPLEMENTAL TABLE (ONLINE ONLY)

Historical occupational isocyanate exposure levels in two Canadian provinces

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Supplementary Table 1 Summary of Measurements Collected, by industry (NAICS) and species. Data shown as:

n % > LOD / % > AL / % > TLV-TWA

NAICS		HDI	MDI	Other	TDI	Total
11	Agriculture, forestry, fishing and hunting	0 0/0/0	30 13.3/0/0	30 0/0/0	0 0/0/0	60
113	Forestry and logging	0 0/0/0	30 13.3/0/0	30 0/0/0	0 0/0/0	60
21	Mining and oil and gas extraction	0 0/0/0	21 28.6/14.3/4.8	8 0/0/0	9 0/0/0	38
212	Mining (except oil and gas)	0 0/0/0	13 30.8/7.7/7.7	0 0/0/0	9 0/0/0	22
213	Support activities for mining and oil and gas extraction	0 0/0/0	8 25/25/0	8 0/0/0	0 0/0/0	16
23	Construction	14 0/0/0	67 4.5/3.0/3.0	4 0/0/0	45 28.9/0/0	130
236	Construction of buildings	4 0/0/0	21 0/0/0	4 0/0/0	27 37.0/0/0	56
237	Heavy and civil engineering construction	2 0/0/0	8 0/0/0	0 0/0/0	18 16.7/0/0	28
238	Specialty trade contractors	8 0/0/0	38 7.9/5.3/5.3	0 0/0/0	0 0/0/0	46
31	Manufacturing	328 4.3/2.7/1.2	3092 21.4/12.0/8.5	233 26.6/21.9/12.0	2501 22.1/12.3/7.5	6154
313	Textile mills	0 0/0/0	0 0/0/0	0 0/0/0	24 25.0/16.7/8.3	24
314	Textile product mills	0 0/0/0	43 6.98/0/0	0 0/0/0	26 76.9/38.5/26.9	69
315	Clothing manufacturing	0 0/0/0	5 0/0/0	0 0/0/0	0 0/0/0	5

316	Leather and allied product manufacturing	0 0/0/0	35 0/0/0	0 0/0/0	21 0/0/0	56
321	Wood product manufacturing	12 0/0/0	131 28.2/19.8/11.4	16 0/0/0	0 0/0/0	159
322	Paper manufacturing	0 0/0/0	3 0/0/0	0 0/0/0	29 17.2/17.2/6.9	32
323	Printing and related support activities	0 0/0/0	12 0/0/0	0 0/0/0	86 1.2/0/0	98
325	Chemical manufacturing	57 12.3/7.0/0	164 11.6/4.9/1.2	16 0/0/0	132 5.3/0/0	369
326	Plastics and rubber products manufacturing	21 14.3/4.8/0	443 15.8/5.2/2.3	40 60.0/50.0/10.0	1316 27.0/17.4/10.2	1820
327	Non-metallic mineral product manufacturing	3 0/0/0	28 0/0/0	9 22.2/22.2/11.1	28 0/0/0	68
331	Primary metal manufacturing	3 0/0/0	635 29.0/19.8/14.8	11 18.2/18.2/18.2	36 0/0/0	685
332	Fabricated metal product manufacturing	25 0/0/0	339 28.9/21.5/18.6	21 76.2/71.4/71.4	66 16.7/6.1/6.1	451
333	Machinery manufacturing	10 0/0/0	244 11.9/5.3/2.9	3 0/0/0	46 47.8/39.1/17.4	303
334	Computer and electronic product manufacturing	12 0/0/0	59 3.4/1.7/0	0 0/0/0	31 19.4/16.1/9.7	102
335	Electrical equipment, appliance and component manufacturing	18 0/0/0	39 15.4/7.7/7.7	0 0/0/0	123 8.9/4.1/3.2	180
336	Transportation equipment manufacturing	140 2.9/2.9/2.9	757 23.9/11.5/8.3	105 17.1/11.4/5.7	326 24.2/5.2/4.3	1328
337	Furniture and related product manufacturing	3 0/0/0	99 2.02/1.01/1.01	0 0/0/0	95 4.2/2.1/2.1	197
339	Miscellaneous manufacturing	24 0/0/0	56 55.4/16.1/10.7	12 0/0/0	116 22.4/7.8/6.0	208

41	Wholesale trade	0 0/0/0	26 11.5/7.7/3.8	6 0/0/0	9 0/0/0	41
413	Food, beverage and tobacco wholesaler-distributors	0 0/0/0	0 0/0/0	3 0/0/0	0 0/0/0	3
414	Personal and household goods wholesaler-distributors	0 0/0/0	0 0/0/0	0 0/0/0	6 0/0/0	6
416	Building material and supplies wholesaler-distributors	0 0/0/0	6 50.0/33.3/16.7	0 0/0/0	0 0/0/0	6
417	Machinery, equipment and supplies wholesaler-distributors	0 0/0/0	8 0/0/0	3 0/0/0	3 0/0/0	14
418	Miscellaneous wholesaler- distributors	0 0/0/0	12 0/0/0	0 0/0/0	0 0/0/0	12
44	Retail	54 5.6/3.7/1.8	31 25.8/0/0	20 5.0/5.0/5.0	19 10.5/10.5/5.3	114
441	Motor vehicle and parts dealers	54 5.6/3.7/1.8	21 28.6/0/0	20 5.0/5.0/5.0	19 10.5/10.5/5.3	124
444	Building material and garden equipment and supplies dealers	0 0/0/0	8 25/0/0	0 0/0/0	0 0/0/0	8
445	Food and beverage stores	0 0/0/0	2 0/0/0	0 0/0/0	0 0/0/0	2
48	Transportation and warehousing	10 0/0/0	18 0/0/0	0 0/0/0	0 0/0/0	28
485	Transit and ground passenger transportation	10 0/0/0	10 0/0/0	0 0/0/0	0 0/0/0	20
488	Support activities for transportation	0 0/0/0	8 0/0/0	0 0/0/0	0 0/0/0	8
51	Information and cultural industries	8 0/0/0	2 0/0/0	0 0/0/0	0 0/0/0	10
511	Publishing industries (except Internet)	0 0/0/0	2 0/0/0	0 0/0/0	0 0/0/0	2

515	Broadcasting (except Internet)	8 0/0/0	0 0/0/0	0 0/0/0	0 0/0/0	8
52	Finance and insurance	0 0/0/0	0 0/0/0	2 0/0/0	0 0/0/0	2
524	Insurance carriers and related activities	0 0/0/0	0 0/0/0	2 0/0/0	0 0/0/0	2
53	Real estate and rental and leasing	0 0/0/0	3 66.7/0/0	0 0/0/0	0 0/0/0	3
532	Rental and leasing services	0 0/0/0	3 66.7/0/0	0 0/0/0	0 0/0/0	3
54	Professional, scientific and technical services	0 0/0/0	3 100/0/0	13 0/0/0	0 0/0/0	16
541	Professional, scientific and technical services	0 0/0/0	3 100/0/0	13 0/0/0	0 0/0/0	16
56	Administrative and support, waste	0 0/0/0	16 31.2/6.2/6.2	0 0/0/0	0 0/0/0	16
561	Administrative and support services	0 0/0/0	16 31.2/6.2/6.2	0 0/0/0	0 0/0/0	16
61	Educational services	7 0/0/0	4 0/0/0	7 0/0/0	0 0/0/0	18
611	Educational services	7 0/0/0	4 0/0/0	7 0/0/0	0 0/0/0	18
62	Health care and social assistance	0 0/0/0	98 25.5/21.4/13.3	0 0/0/0	6 0/0/0	104
621	Ambulatory health care services	0 0/0/0	56 10.7/10.7/10.7	0 0/0/0	0 0/0/0	56
622	Hospitals	0 0/0/0	42 45.2/35.7/16.7	0 0/0/0	6 0/0/0	48
81	Other services (except public administration)	79 32.9/26.6/15.2	33 24.2/24.2/24.2	104 57.7/55.8/51.0	24 4.2/4.2/4.2	240
811	Repair and maintenance	79 32.9/26.6/15.2	33 24.2/24.2/24.2	104 57.7/55.8/51.0	19 5.3/5.3/5.3	235

813	Religious, grant-making, civic, and professional and similar organizations	0 0/0/0	0 0/0/0	0 0/0/0	5 0/0/0	5
Total		500	3444	427	2613	

TLV-TWA = 2014 Threshold Limit Value – Time-Weighted Average of 0.005 ppm by ACGIH

AL = Action Level of 0.0025 ppm (50% of TLV-TWA)