University of Massachusetts Lowell &
Center for *Construction Research and Training (CPWR)*

Occupational Exposures to Epoxy Resins Among Construction Painters: Methods to Monitor Exposures and Urinary Biomarkers

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Overview

- Health concerns of exposures to epoxy compounds
- Methods, applications and main findings
  - Air and skin
  - Urinary biomonitoring
- Observations on PPEs and work practices
- Permeation and penetration testing on select garments
- Opportunities for reducing exposures
- Q&A

Health concerns of epoxy exposures

- **Irritant and allergic contact dermatitis** (hands, forearms, face)
  - *Construction workers have high rates of contact dermatitis*
  - *25-47% self-reported;[1] 20% ACD [2]*

- **Other potential health effects** [3]
  - Occupational asthma
  - Acute decline in lung function
  - Hypersensitivity pneumonitis (epoxy-resin lung)
  - ↑ Plasma Follicle Stimulating Hormone (FSH) in workers
  - Other effects in animal studies and cells (lipid metabolism & endocrine)
  - *Recommendation to monitor human exposure to BADGE and its byproducts at least as intensely as BPA…"* [4]

Contact dermitits on hand

Figure 1 credit : CPWR report on epoxy resins in construction

Figure 2 credit DermNet NZ

[1] Timmerman et al 2016 (Dutch study; [2] Aalto-Korte et al., 2015 (Finland); [3] Bello et al 2021; [4] Marqueno et al.2019

Cancers

- Occupational exposure as a painter has been classified as a Group 1 carcinogen by IARC, based on an increased risk of lung and urinary bladder cancers
  - Limited evidence for mesothelioma and childhood leukemia
  - Originally in 1989; reconfirmed in 2010; and again in 2021
  - Job title, not epoxies
  - Exact chemical/s causing cancer are not known

- Inhalation, skin and Ingestion - all potentially important exposure pathways

Study motivation and objectives

Needs

- No data on epoxy exposures in occupational settings
- Lack of suitable sampling and analytical methods
  - Herrick and Smith, 1987, 1988 – important contributions, but not replicated

Objectives

- To develop methods for quantitation of epoxies – individual species and total epoxide group
- To apply both methods for characterization of inhalation and potential skin exposures in bridge painters
- Urinary biomonitoring in these workers
- Work practices and PPEs

Methods details can be found in these recent pubs

Annals of Work Exposures and Health, 2020, 1–15
doi: 10.1093/annweh/wxaa138
Original Article
BOHS The Chartered Society for Worker Health Protection
OXFORD

Original Article

Characterization and Quantitation of Personal Exposures to Epoxy Paints in Construction Using a Combination of Novel Personal Samplers and Analytical Techniques: CIP-10MI, Liquid Chromatography–Tandem Mass Spectrometry and Ion Chromatography

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Urinary biomonitoring of occupational exposures to Bisphenol A Diglycidyl Ether (BADGE) – based epoxy resins among construction painters in metal structure coating

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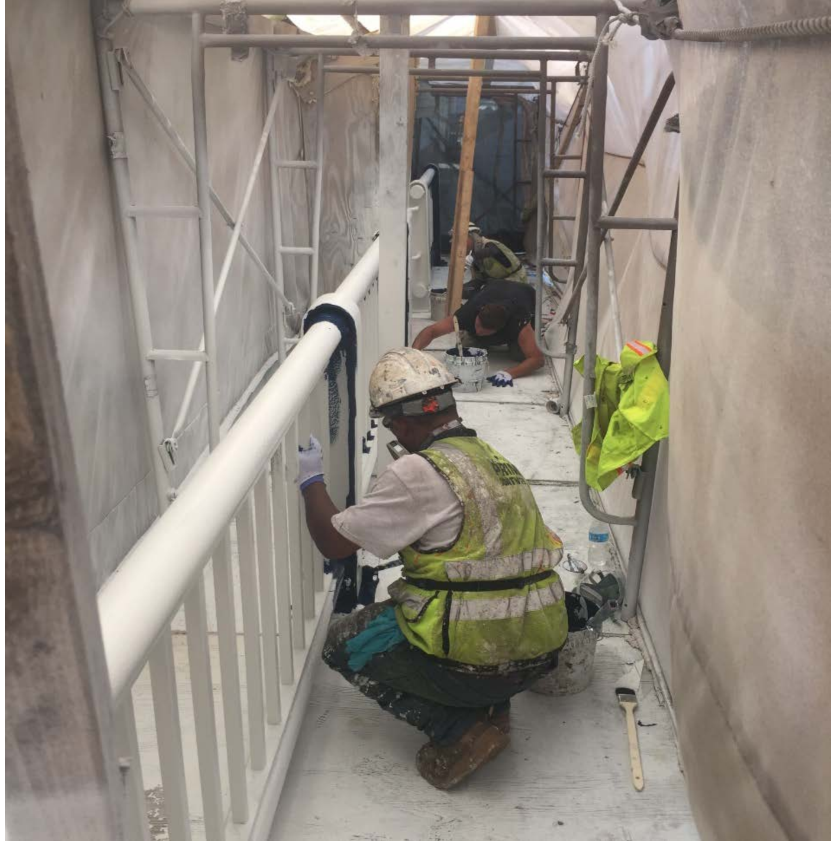



Protective Steel Structure Coatings

- 2015 Annual global cost of corrosion in steel structures = $2.5 trillion, 3.4% of GDP; (Am. Soc. Prot. Coatings)
- Better anti-corrosion coatings can produce annual savings of $375B - $875B
- New $2.3 trillion infrastructure plan calls to spend $115B for roads & bridges
- EU has invested $42.8B in new windfarms (S: WindEurope)
- Thousands of workers exposed to epoxies daily

Protective Anti-corrosion Exterior Metal Structure Coatings

Coating systems

Sequential coating layers

Surface cleaning/sand blasting

Primer

-High solids, Zn-rich, epoxy rich - polyamide formulation

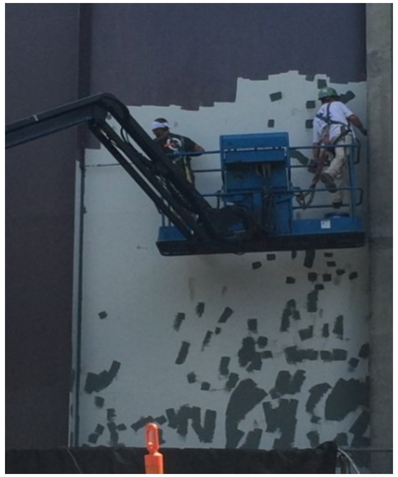
Mid-coat

-Epoxy-based reactive systems

Top-coat

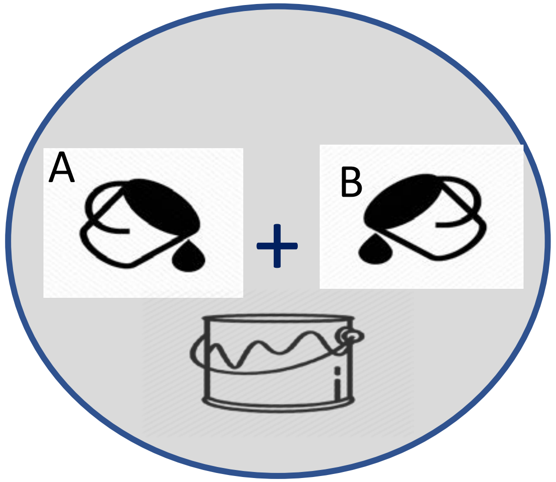
-Aliphatic isocyanate-based reactive systems

Reactive Systems: Steel structure coatings example



Part A

EPOXY
or
Isocyanate



ROLLER/BRUSH or/and SPRAY

Part B

HARDENERS: AMINES, etc.

SOLVENTS

Nano FILLERS
Zn, TiO_2,Fe_2O_3, nano/Quartz, etc.

CATALYSTS

Other additives

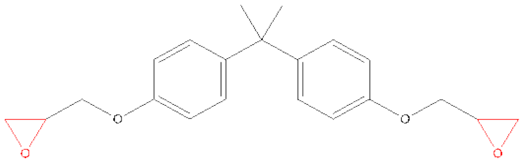
<u>Epoxy resins are</u> pre-polymers contain two or more epoxide groups in their molecule

1) Bisphenol A diglycidyl ether (BADGE)

2) Bisphenol F diglycidyl ether (BFDGE)

3) BADGE dimer

4) Generic structure of a BADGE-based polymer with ‘n’ monomeric units



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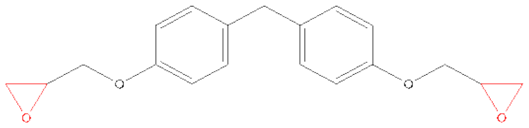

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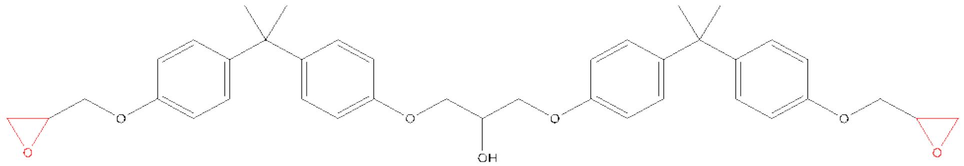

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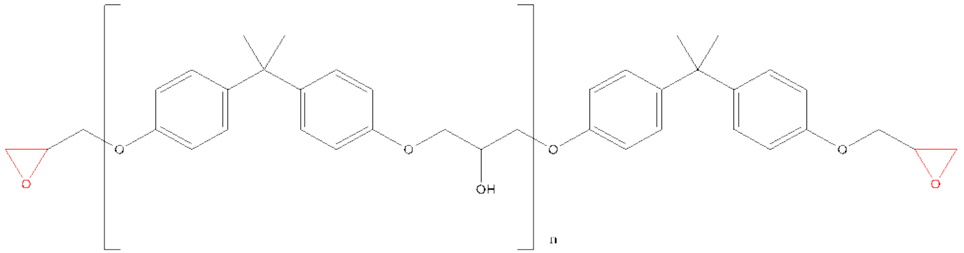


(4)

Considerations in developing sampling and analysis methods for epoxies

- Reactive chemical systems consisting of mixtures
  - Share similar features with isocyanate systems
  - New chemical species formed as a function of curing rate, env. temp, surface temp, RH, pot-life, application method
  - Measure individual epoxy species
  - Measure TOTAL EPOXY group
  - No prior data or experiences
- Measure inhalation exposure
- Measure skin exposure
- Provide context for urinary biomonitoring results

Overall approach

Workplace observations

- Site layout, job size,
- Worker activities, task duration
- Product & composition (SDS)
- Environmental conditions (T, RH, climate)

Inhalation exposures

- Personal inhalable sampler: CIP10-MI

Skin exposures

- Cotton gloves, workers gloves

Urinary biomonitoring

- Spot urine collection pre- and post-work shift

Inhalation exposures

Personal inhalable sampler - CIP10-MI

Used successfully for sampling of pMDI (fast cure) in SPF applications and aliphatic isocyanates on bridges (slow cure)

- Filled with 2mL Dimethylformamide (DMF)
- 7600 RPM, 10L/min
- Integrated system (no tubing) – [ideal impinger (?)]
- Relatively robust
- Well-tolerated by workers and preferred over traditional active sampling pumps
- Suitable for spray applications – large aerosols (with MMAD ~10um)

CIP-10MI unit collects >95% of aerosol particles with aerodynamic diameter >2.8 μm and >50% for particles >1.8 μm (Gorner et al., 2006).

Potential skin exposure

- Cotton gloves or workers gloves
  - Anatomical site with one of the highest likelihood for skin exposure
  - In SPF applicators, it was second only to head and neck

- Transferred in jars with DMF
- Median sampling duration 90 min (range 20 - 230 min)
- Sample prep: Mixing, aliquoting, filtration, dilution, analysis

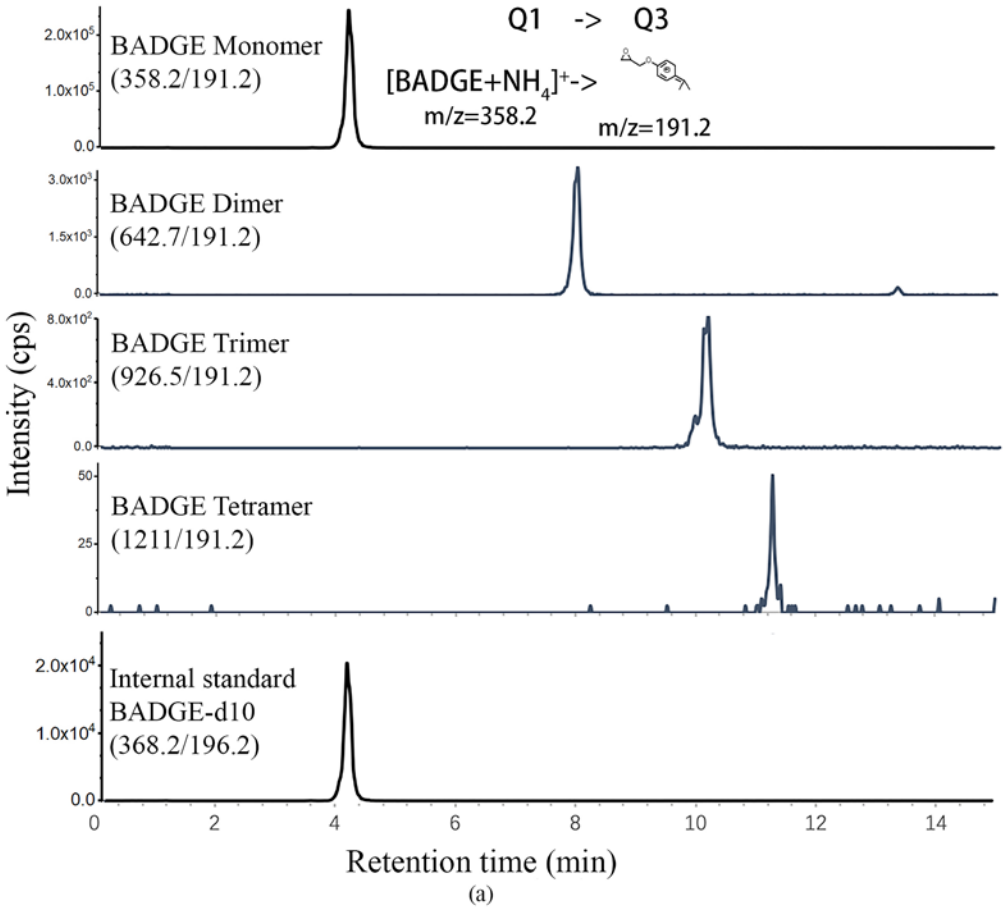
Sites and Activities

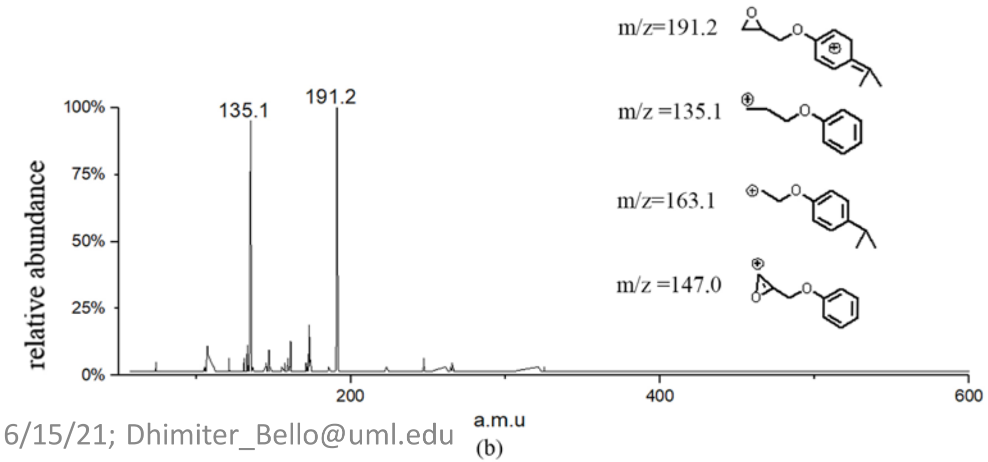
Table 1

Characteristics of metal structure coating sites investigated in this work, site activities, products and tasks, number of workers and urine samples collected.

Sites	Nr. of sampling sites	Activity	Tasks performed by workers at the day of sampling (n)	Product used onsite at the day of urine sampling	Urine samples, N	
					Pre-shift	Post-shift
Mid-coat application	4	Bridge and indoor shop painting	Spraying (7) Rolling and brushing (4) Helping (3) Bystanders (3) *Total = 17*	Zinc CLAD ® 4100 and Macropoxy® 646	14	17
Top-coat application	8	Bridges, reactor dome, wind turbine, water tank and indoor shop painting	Spraying (7) Rolling and brushing (14) Helping (3) Bystanders (3) *Total = 27*	ACROLON™ 218 HS Hi-solids Polyurethane Semi-Gloss (Part S)	24	26
Total	12 coating sites		44 participants		38	43

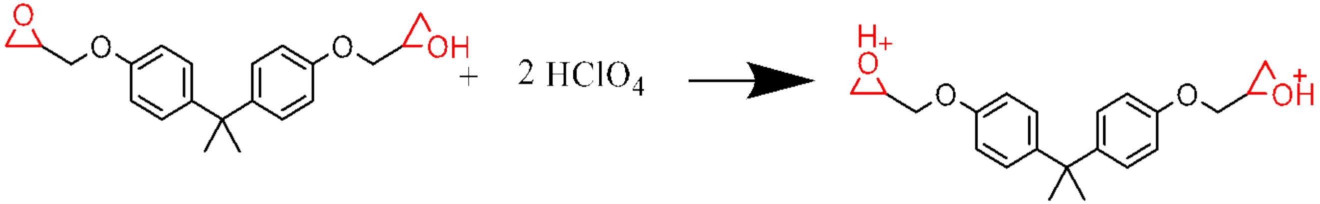
Analytical method for air and gloves: LC-ESI-MS/MS





Analyte	Nominal concentrations (ng/mL)	Accuracy (%)	RSD (%)
BADGE	500	99.8	2.3
	100	105	0.8
	10	96.0	1.2
	Average	100.2	1.4
Dimer	1000	100	0.3
	500	97.0	1.2
	50	88.2	1.5
	Average	95.1	1.0
Trimer	1200	96.5	5.3
	600	94.1	4.6
	60	103	5.3
	Average	97.8	5.1

Basis of TEG by Ion Chromatography



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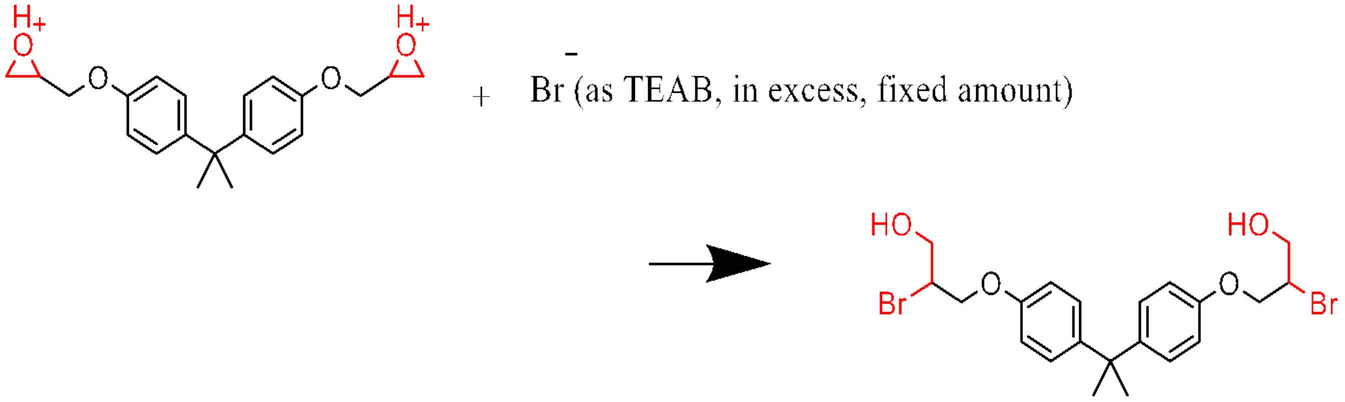
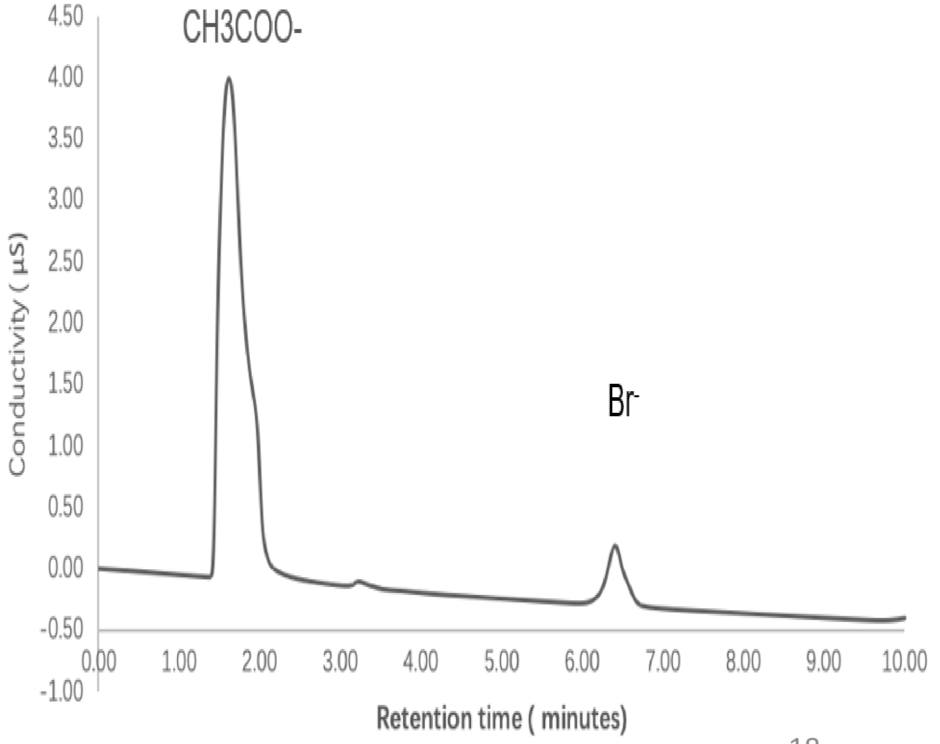


(2)

Ion chromatogram of a typical sample



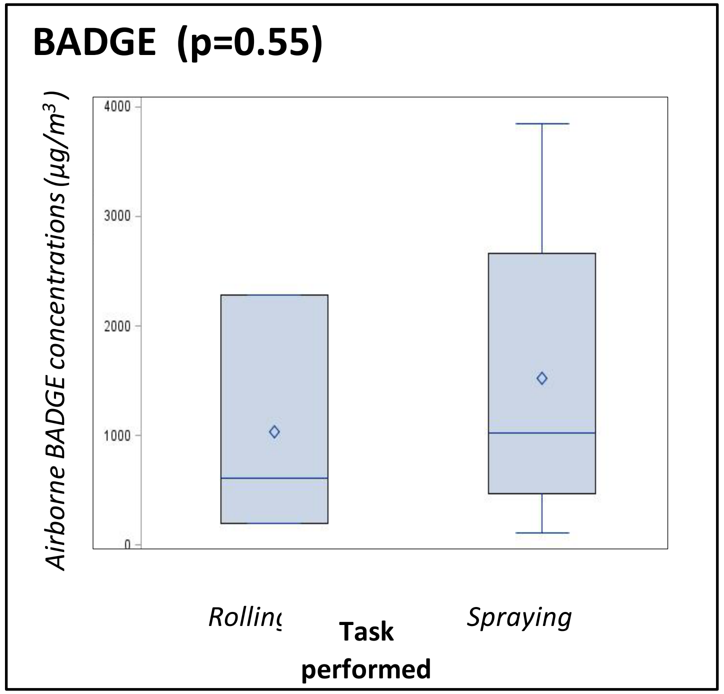
The total epoxy group (TEG) content of the sample was determined as the difference between the total Br⁻ added in the reaction medium and residual Br⁻ measured by IC. TEAB, tetraethylammonium bromide.

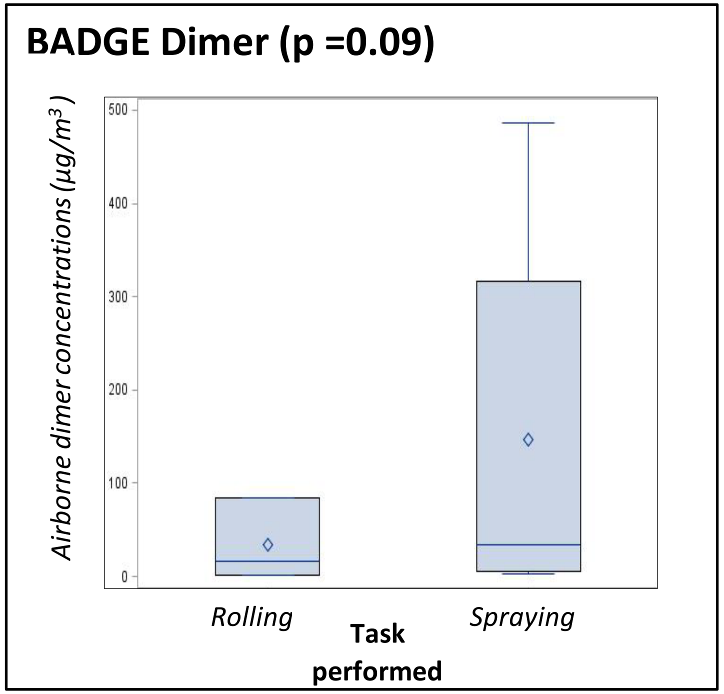
Composition of products used at sampled sites

Table 2. Compositional analysis of two typical bulk products used at sampling sites and the reference material [Poly (Bisphenol A-co-epichlorohydrin), glycidyl end-capped]. Linear regression fit: TEG (by IC) = 1.18 × TEG (by LC–MS/MS), R^2 = 0.999.

Materials	% w/w				% TEG eq. by LC–MS/MS (w/w)	% TEG by IC (w/w)	TEG ratio IC:MS/MS
	Monomer	Dimer	Trimer	Tetramer			
Reference: material	71.9	26.2	1.9	0.2	23.9	28.3	1.18
Product I: Mid-Coat	19.3	5.9	1.4	0.6	4.7	5.2	1.10
Product II: Primer	18.9	3.5	9.3	10.2	7.0	8.7	1.24

Results: Inhalation Exposures





Epoxy species	ND N (%)	GM (GSD)* µg/m³	Range µg/m³
BADGE	**0 (0)**	**803 (3.2)**	**111-3,850**
BADGE Dimer	0 (0)	26 (7.2)	2-478
BADGE Trimer	1 (11)	13.1 (10.2)	ND-325

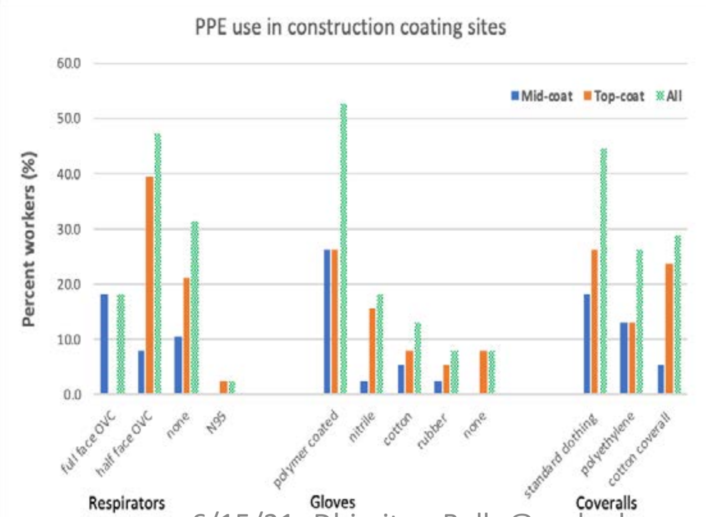
* Task duration, median 90 min (20-230 min)

There are no occupational standards for epoxies!

- GMs are comparable with *Herrick et al 1988* area samples
- We report much higher max values
- Highest conc. in tarp-enclosed areas, consistent with other observations on isocyanate exposures

High potential for skin exposures !

Fig. 2. a. Personal protective equipment observed in use at sampling sites (n workers = 30, n sites = 10). Observations report on the use of PPE during active tasks involving handling, preparation, and application of isocyanate formulations – spraying, roller/brush, mixing, cleanup.



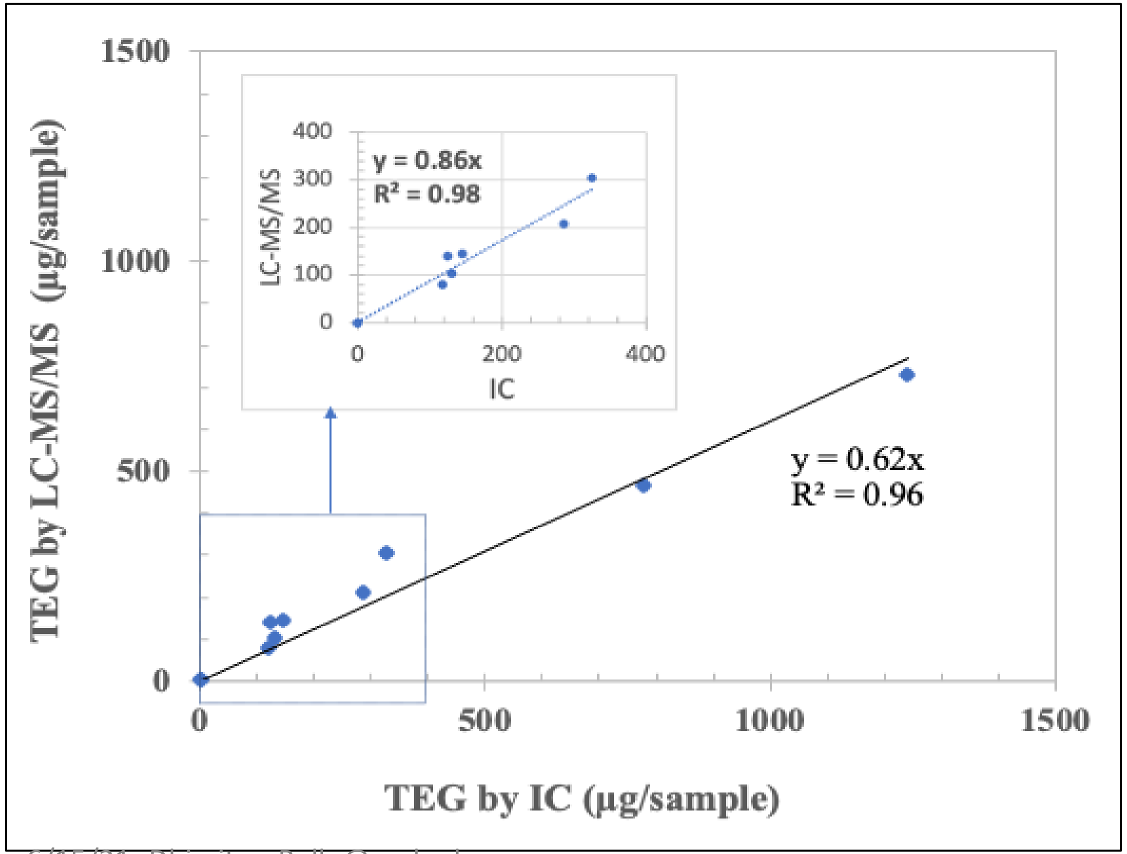
Potential Skin Exposure (n=11)

Epoxy Species	N<LOD n (%)	Glove loading (mg/pair)		
		GM (GSD)		Range
BADGE MW=340.42	0 (0)	547.2	(2.9)	55-1,963
Dimer MW=624.77	0 (0)	10.7	(4.5)	0.5-70.7
Trimer MW=909.13	1 (9)	8.3	(3.0)	0.6-23.0
TEG, IC	0 (0)	173.1	(3.0)	18.4-752
Calc. TEG by LC-MS/MS	-	141.1	(2.9)	14.0-506

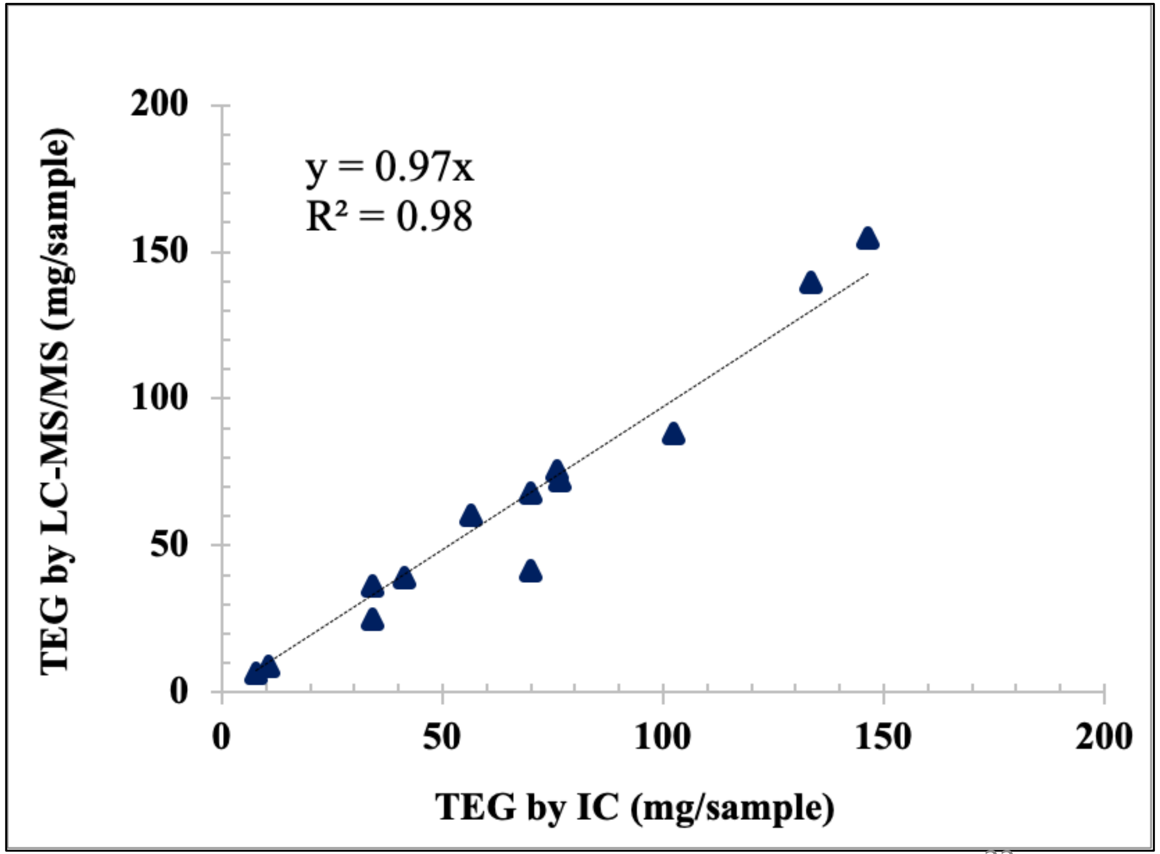
TEG, mg epoxy group/ pair; Epoxy EW, 43 g/mol

Comparison of TEG by LC-ESI-MS/MS & IC

Air samples



Glove samples



Urinary biomonitoring: Objectives

- To determine urinary concentrations of epoxy biomarkers among construction painters in metal structure coating sites

- To quantify cross-shift changes of epoxy biomarkers in urine to gain insights into body uptake during the shift and assess the adequacy of exposure controls

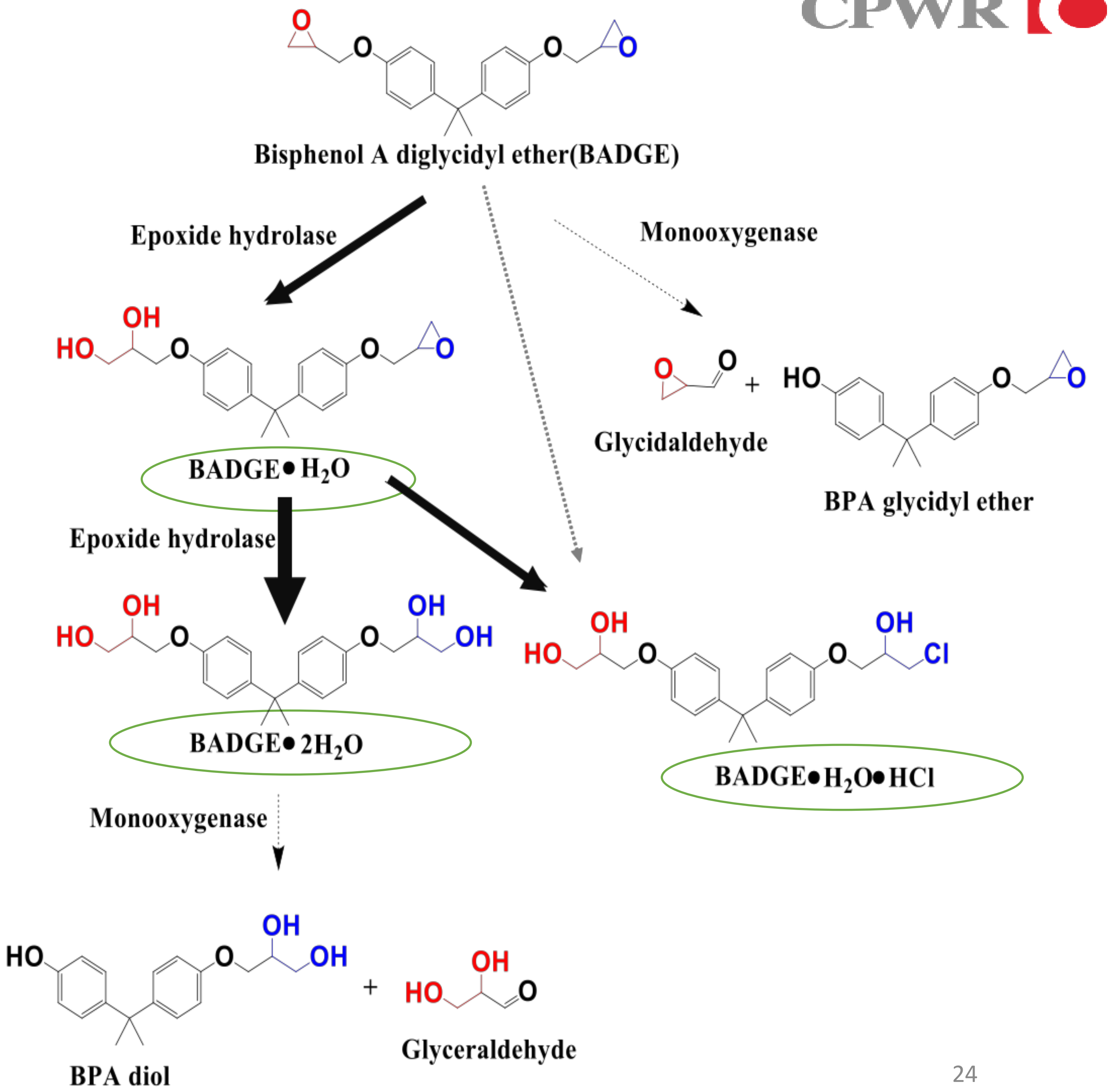
- To compare three distinct exposure groups
  - Epoxy-exposed group in mid-coats – applied epoxies on the day of sampling
  - Isocyanate exposed group in top-coats (possible exposure to epoxies 1-7 days earlier)
  - SPF workers, non-occupational epoxy exposure

Metabolic biotransformation pathways of bisphenol A diglycidyl ether (BADGE) in humans

No ADME data in humans
Few studies in animals (*Climie et al 1981; Boogaard et al 2000*)

BADGE biomarkers in urine*

- **BADGE·H_2O (**mono-diol epoxide of BADGE)
- **BADGE·$2H_2O$ (**bis-diol epoxide of BADGE)
- **BADGE·HCl·H_2O** (chlorohydroxy derivatives of BADGE)



ADME of BADGE

INGESTION (Climie et al. 1981)

- Oral administration of 14C-BADGE in mice (Climie el al. 1981a) resulted in rapid uptake by the ingestion route
- And fast elimination – 88% in three days
    - mostly via feces (80%)
    - urine (11%)

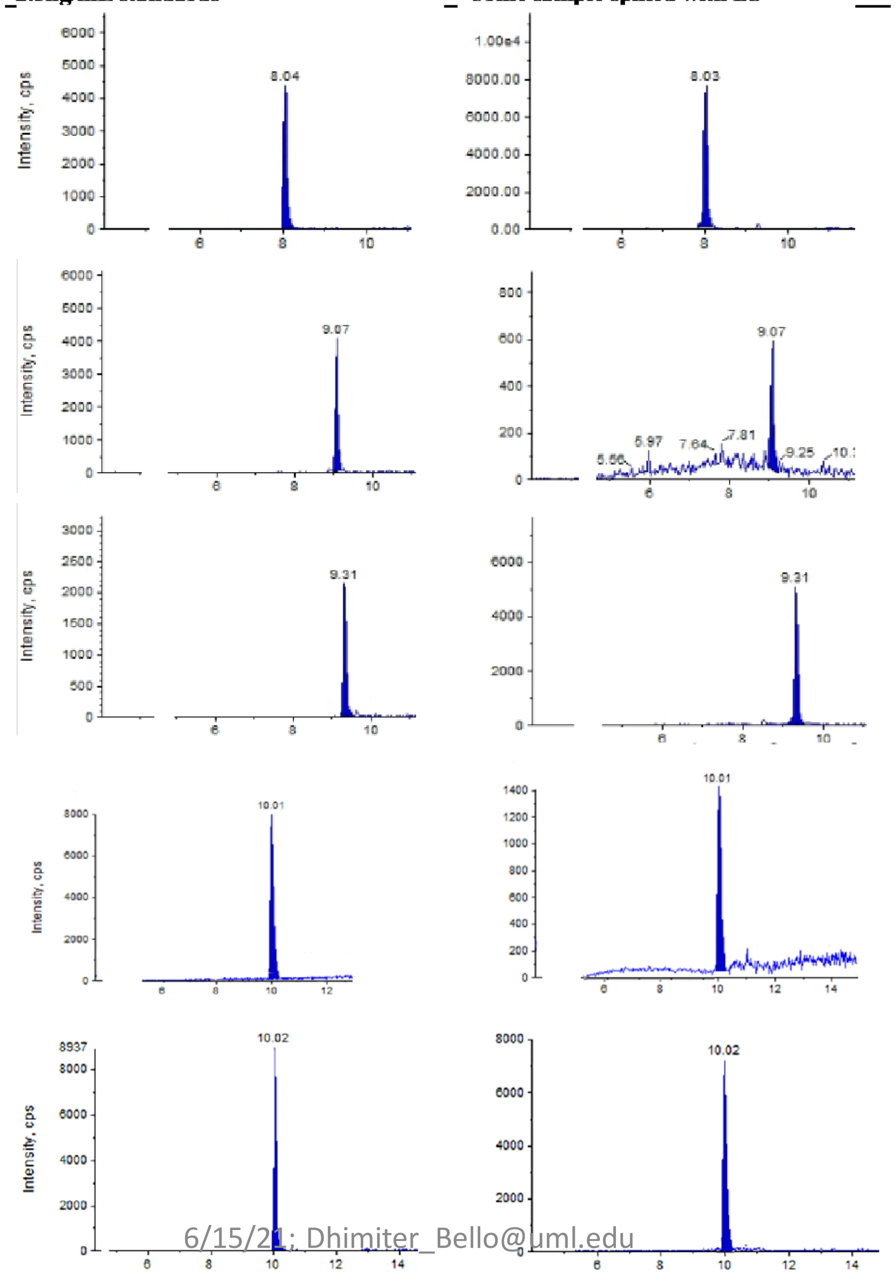
SKIN EXPOSURE

BADGE is absorbed very slowly from the mouse skin, with ~90% of the administered dose (as 14C-BADGE) remaining in the skin at 24 hrs post-application and 40% after 8 days (Climie el al. 1981a).

INHALATION

No human or animal inhalation data

Quantitation by LC-ESI-MS/MS



BADGE-2H2O
394.3>209.1

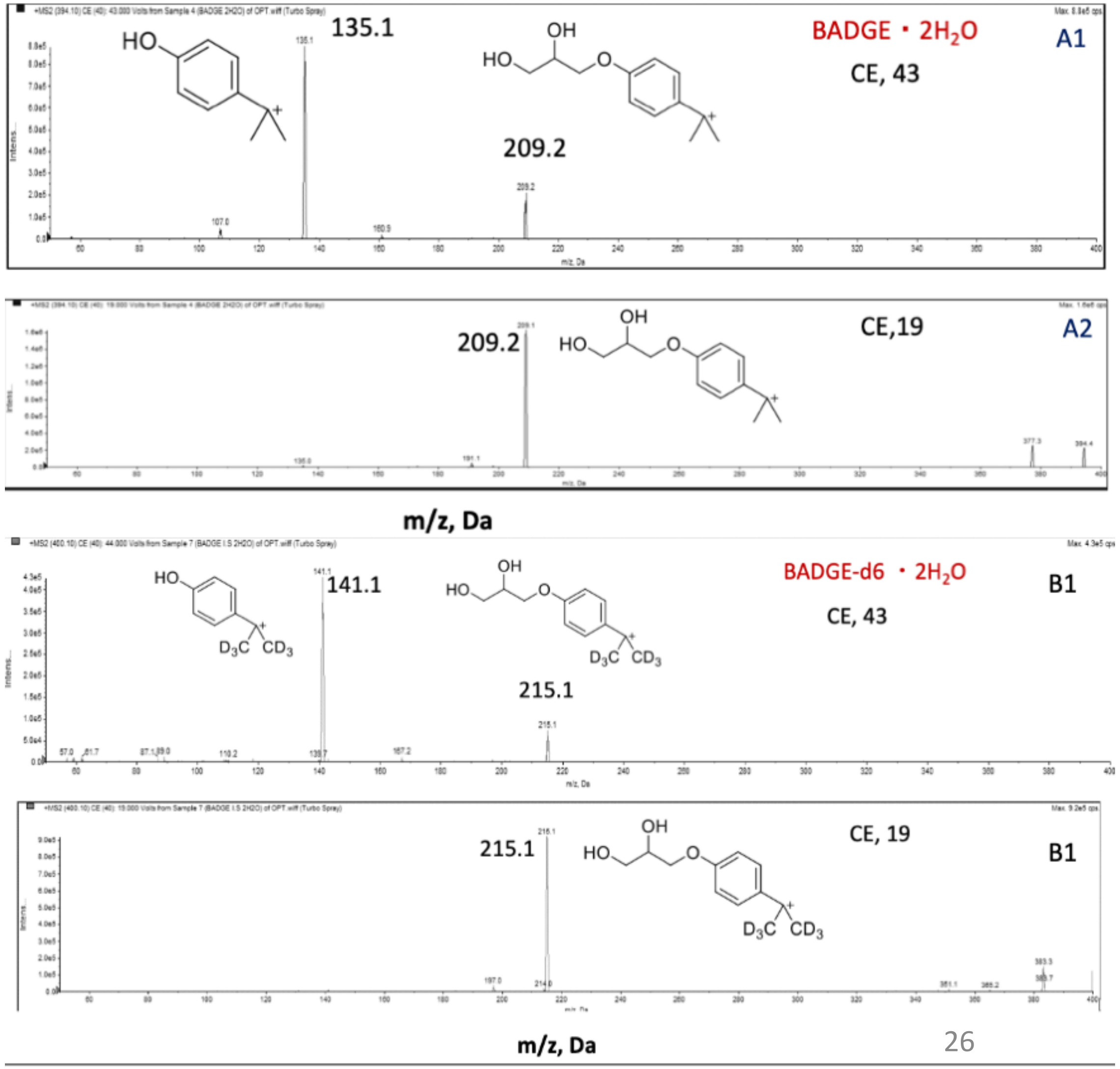
BADGE-H2O
376.2>209.1

BADGE-H2O-HCl
412.1>227.1

BADGE
358.2>191.2

BADGE-D6 (IS)
364.2>197.2

2nd transition



Urinary epoxy biomarkers among study participants

Activity / Urinary Biomarkers [a]	GM (GSD) (ng/mL), SG normalized			
	Free BADGE [b]	BADGE • $2H_2O$	BADGE • HCl • H_2O	Total BADGE
Mid-coat				
Pre-Shift, n=14	0.04 (3.0)	0.50 (2.0)	0.17 (3.1)	0.69 (2.2)
Post-Shift, n=17	0.04 (2.5)	1.46 (3.6)	0.17 (2.3)	1.66 (3.2)
Range	nd - 0.16	0.20 - 18.7	nd – 0.59	0.24 -17.2

- BADGE *H_2O was quantified only in 10% of samples; Omitted from table
- BADGE*$2H_2O$ – Major BIOMARKER; ~9:1 ratio relative to BADGE*H2O*HCl
- Post-/pre-shift GM ratios of BADGE*H_2O =2.9 fold (increase)
- Maximum of BADGE*H_2O = 18.7 ng/mL
- No cross-shift changes for BADGE*H2O*HCl and free BADGE

Urinary biomonitoring results, cont'd

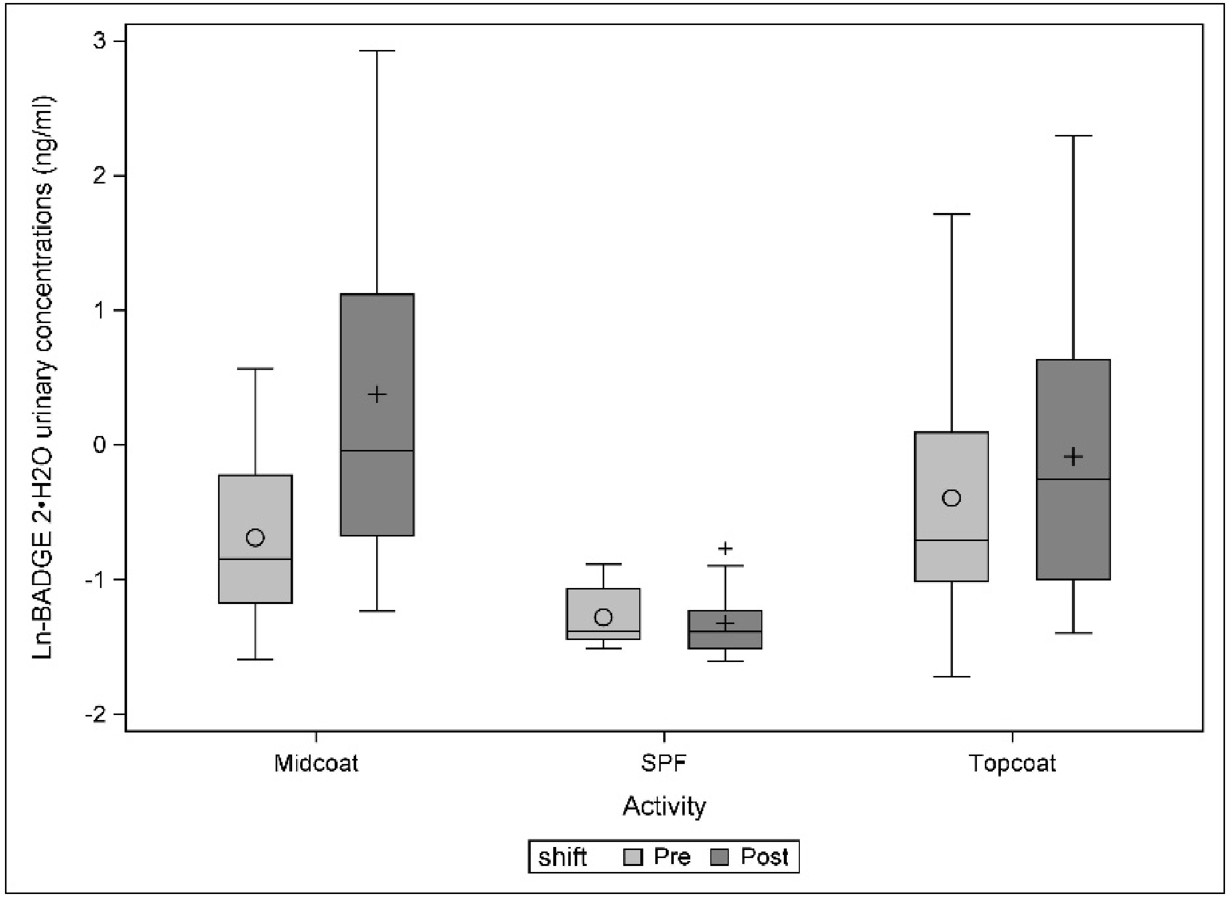
Activity	BAGDE*$2H_2O$ GM(GSD) ng/mL SGN	Cross-shift changes (ratio of GMs)
Mid coat (epoxy)		
Pre-shift, n=14	0.50 (2.0)	
Post-shift, n=17	1.46 (3.6)	**2.9x increase**
Range	0.20 - 18.7	
Top coat (previous epoxy)		
Pre-shift, n=24	0.67 (2.4)	**1.34x higher than in mid coats**
Post-shift, n=26	0.91 (3.0)	**1.35x increase from pre-s; 3.7x higher than SPF**
Range	0.18 - 9.97	**High maximum values**
SPF (no epoxy)		
Pre-shift, n=14	0.28 (1.3)	
Post-shift, n=14	0.27 (1.3)	**No increase**
Range	0.20 - 0.46	**Max < 0.5 ng/mL**

Urinary biomonitoring results, cont’d

Activity	GM(GSD) ng/mL SGN BAGDE*H_2O *HCl	Cross-shift changes
Mid coat (epoxy)		
Pre-shift, n=14	0.17 (3.1)	
Post-shift, n=17	0.17 (2.3)	No change
Range	nd – 0.59	
Top coat (previous epoxy)		
Pre-shift, n=24	0.21 (2.4)	
Post-shift, n=26	0.29 (3.0)	1.38 fold increase
Range	0.06 - 5.18	High max value
SPF (no epoxy)		
Pre-shift, n=14	0.10 (1.5)	
Post-shift, n=14	0.09 (1.7)	No change
Range	0.05 - 0.22	

BADGE·$2H_2O$ - A biomarker of occupat. exposures to epoxies

Significant uptake of BADGE during work shift 2.9x; Uptake continues days later

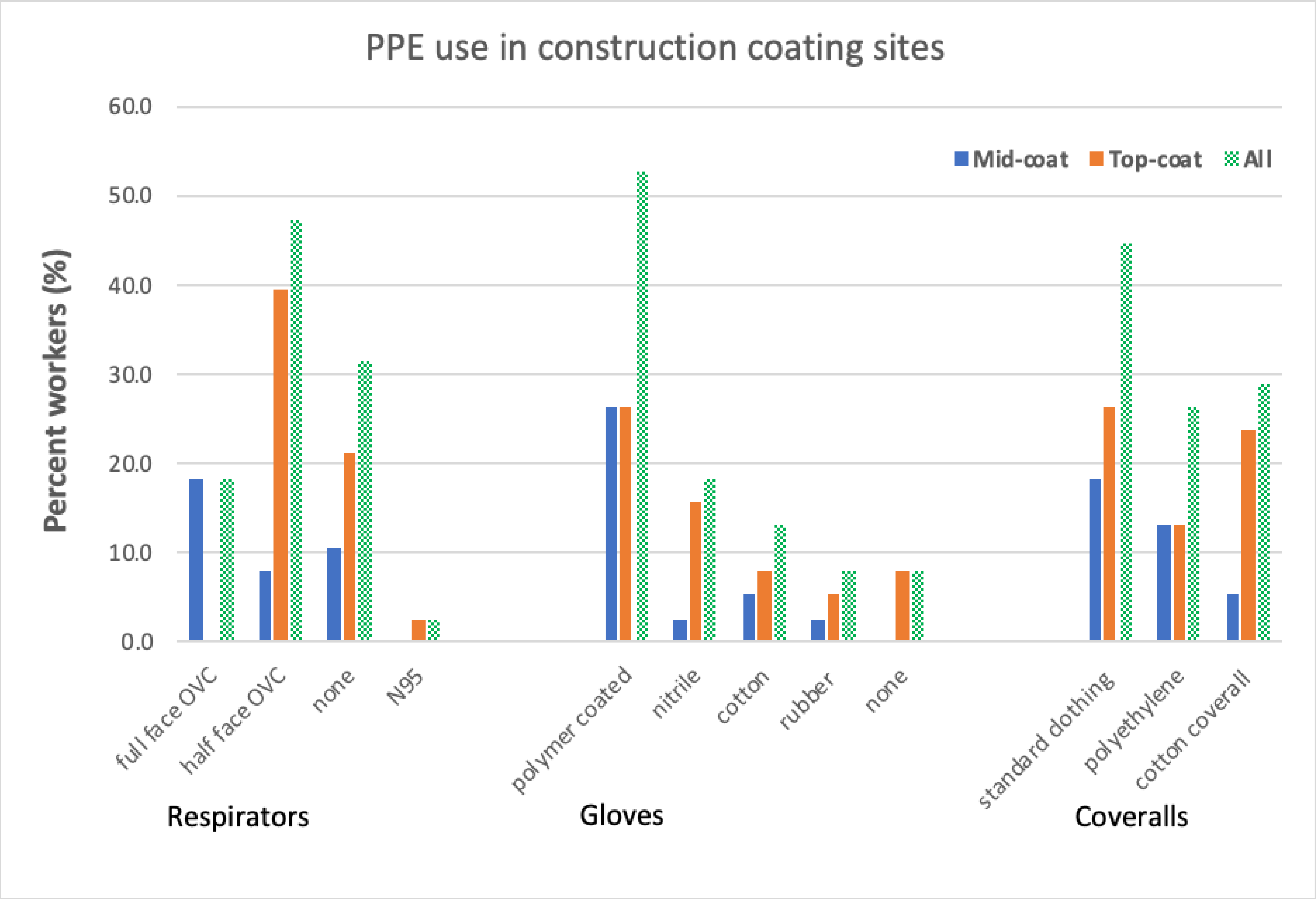


- BADGE*$2H_2O$ is a sensitive and suitable urinary biomarker of occupational exposures to BADGE/epoxies

To discriminate occupational from non-occupational exposures:

- We propose an initial guidance value for BADGE·$2H_2O$ of 0.5 ng/mL as the threshold for non-occupational exposures
- Results > 0.5 ng/mL warrant better occupational hygiene and more effective PPE
- 75% of urine samples were > 0.5 ng/mL
- Other biomarkers have limited utility
- BADGE*H_2O*HCl – ingestion ?

Fig. 2. a. Personal protective equipment observed in use at sampling sites (n workers = 30, n sites = 10). Observations report on the use of PPE during active tasks involving handling, preparation, and application of isocyanate formulations – spraying, roller/brush, mixing, cleanup.


PPE use in construction coating sites
Mid-coat
Top-coat
All
Percent workers (%)
60.0
50.0
40.0
30.0
20.0
10.0
0.0
full face OVC
half face OVC
none
N95
polymer coated
nitrile
cotton
rubber
none
standard clothing
polyethylene
cotton coverall
Respirators
Gloves
Coveralls

Panel study: Permeation and penetration of 2-part epoxy formulations

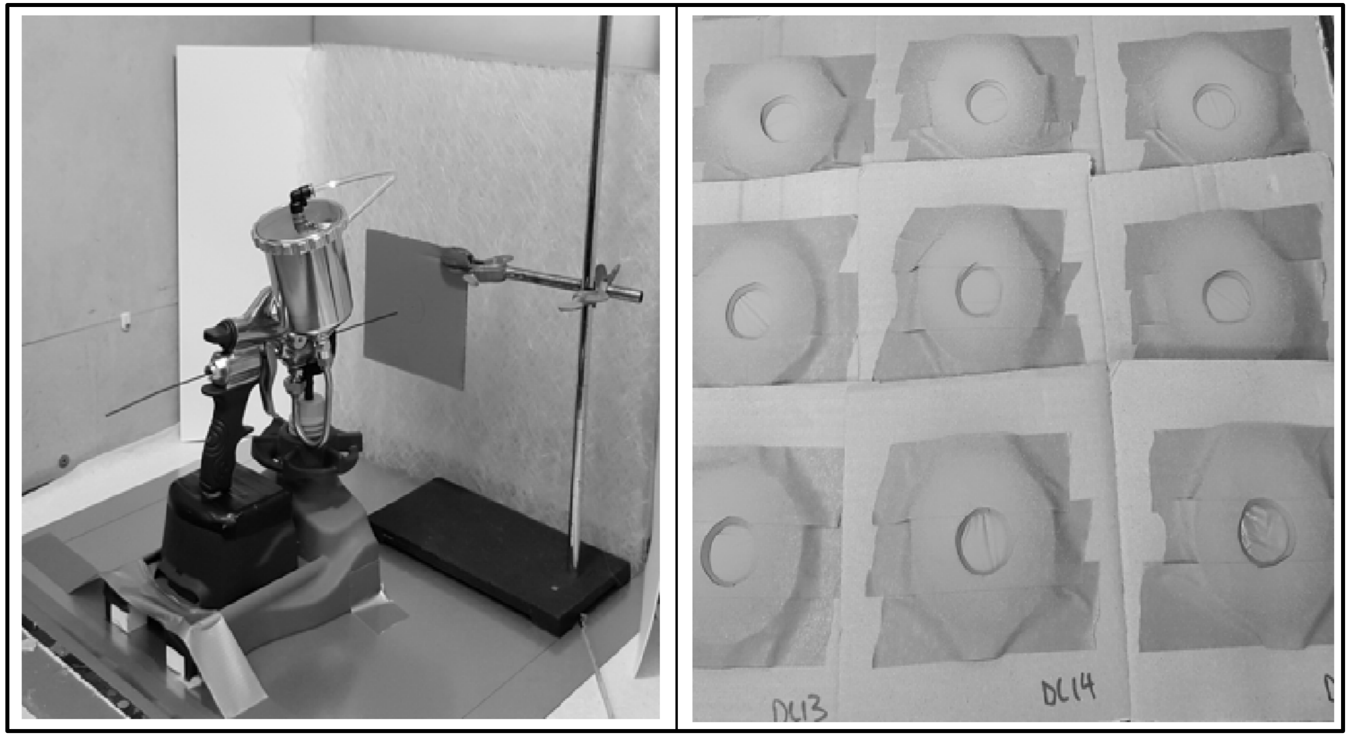


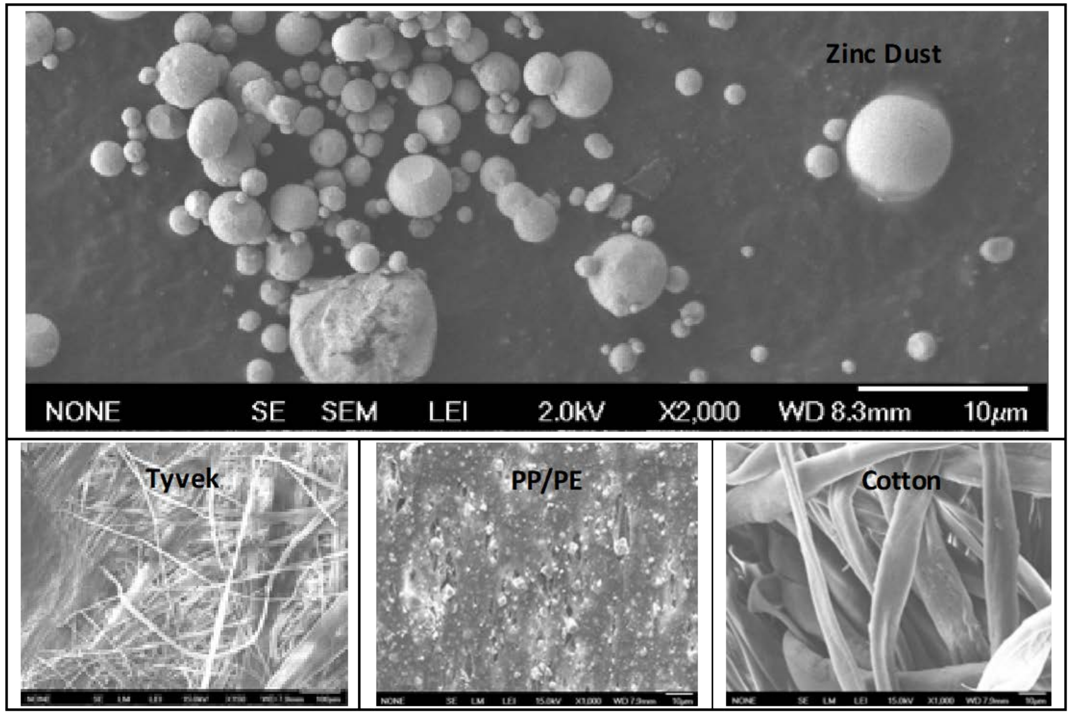
Table 1. Tests garment materials and measured thicknesses.

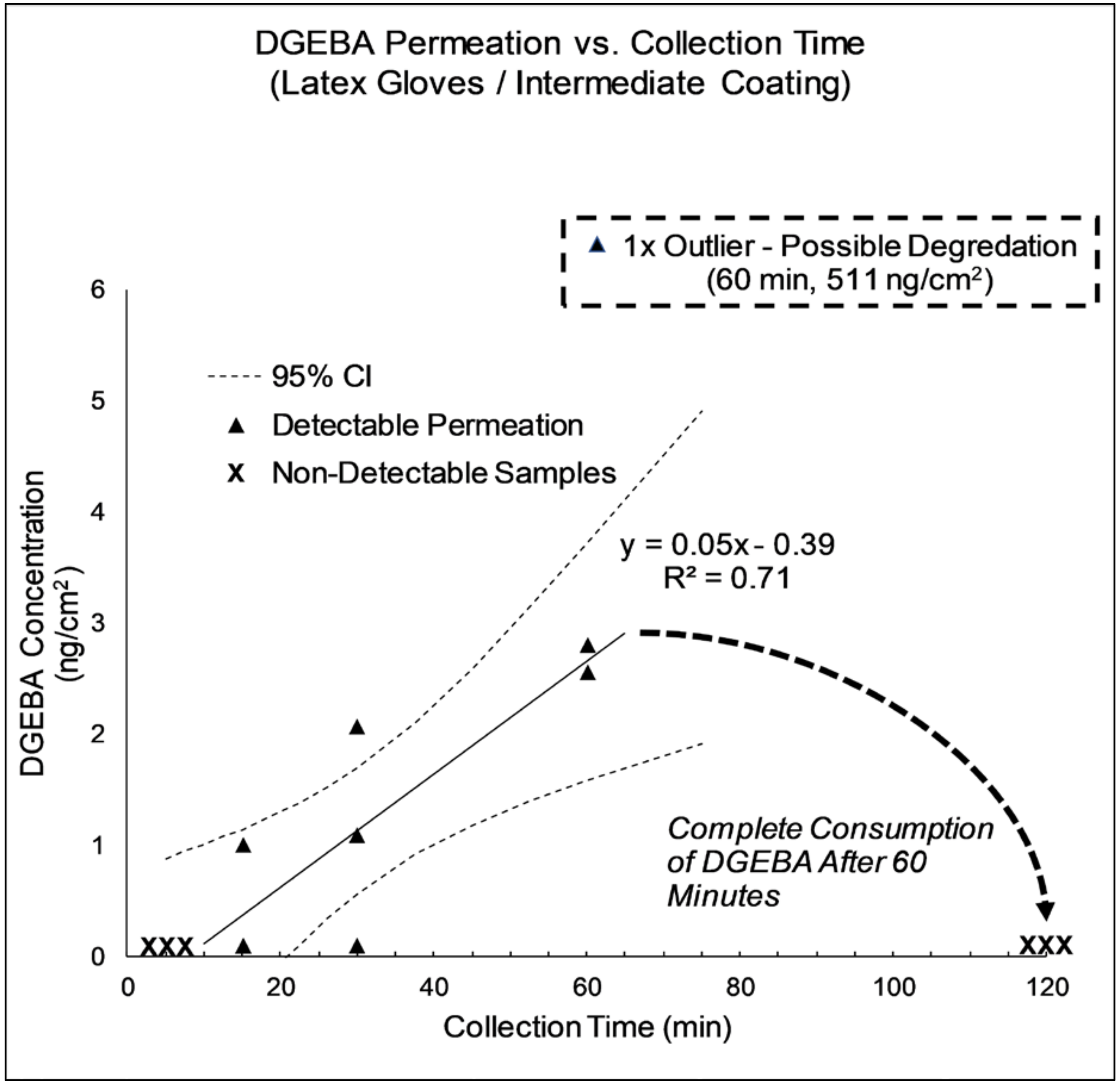
	Manufacturer	Product #	Thickness (mm [mils])[a,b]
Disposable gloves			
Latex	HDX	432202	0.068 [2.7]
Nitrile	HDX	953849	0.073 [2.9]
Clothing			
Tyvek Coverall	Trimaco	14113	0.105 [4.1]
PE/PP[c] Coverall	3M	4540+	0.105 [4.1]
Cotton Shirt	Champion	T425	0.317 [12.5]

[a] Average thickness of all specimens measured with a dial caliper.
[b] 1 mil = 0.001 in.
[c] Polypropylene and polyethylene (PE/PP) laminate film

- **NEPCOAT approved Zn-rich primer and mid coat**
  - Primer: 3 component system: A (11.5% polyamide & solvents) , B (epoxy polymer & solvents), F (Zn powder)
  - Mid coat: two-component: A (polyamide +TiO_2 &solvent) + B (10-25% epoxy + SiO_2 & solvents)

Permeation for epoxy mid coat





Unpublished data: Mike Mellette's ScD thesis

Table 4.3. Maximum penetration of DGEBA (ng/cm^2), % of cumulative permeation threshold, and protection factors for each coating and garment combination.

Garment Type	Zinc-Rich Primer					Intermediate Coating				
	Maximum DGEBA penetration (ng/cm^2)	Collection time (min)[a]	Detectable samples (#)[b]	Penetration threshold (%)[c]	Protection Factor[d]	Maximum DGEBA penetration (ng/cm^2)	Collection time (min)[a]	Detectable samples (#)[b]	Penetration threshold (%)[c]	Protection factor
Disposable gloves										
Latex	21.7	120	2	1.1	12615	511.1	60	6	25.5	308
Nitrile	<0.1	--[e]	--[e]	--[e]	--[e]	<0.1	--[e]	--[e]	--[e]	--[e]
Clothing										
Tyvek Coverall	<0.1	--[e]	--[e]	--[e]	--[e]	6.6	15	1	0.1	23847
PE/PP Coverall	5.0	15	1	0.1	54750	599.4	30	2	6.0	263
Cotton Shirt	127.5	5	15	1.3	2147	28.0	120	10	0.3	5621

[a] Time-period when the sample with maximum cumulative permeation was collected.
[b] Number of detectible samples in each batch (n=15) for a garment/coating combination.
[c] Percent of recommended permeation threshold for garment type: 2 $\mu g/cm^2$ gloves; 10 $\mu g/cm^2$ clothing (Henricks et al., 2015). Percent was calculated as the (ratio of maximum DGEBA penetration / recommended permeation threshold) x 100.
[d] The protection factor is the ratio of the average direct loading concentration for each coating divided by the maximum DGEBA penetration for each coating and garment combination.
[e] All results below limit of detection (LOD).

Unpublished data, please do not distribute

CONCLUSIONS

- First set of biomonitoring data of epoxy exposures in occupational settings, including construction
- BADGE·$2H_2O$ biomarker, the most predominant urinary biomarker and suitable for routine biomonitoring
- Considerable work shift exposures confirmed with urinary biomonitoring results
- Better exposure controls are needed at these construction sites
- Sampling and analysis methods developed for exposures and urinary biomonitoring of epoxies can be used for larger scale investigations

Coating systems

Sequential coating layers

Surface cleaning/sand blasting

Primer

-High solids, Zn-rich, epoxy rich - polyamide formulation

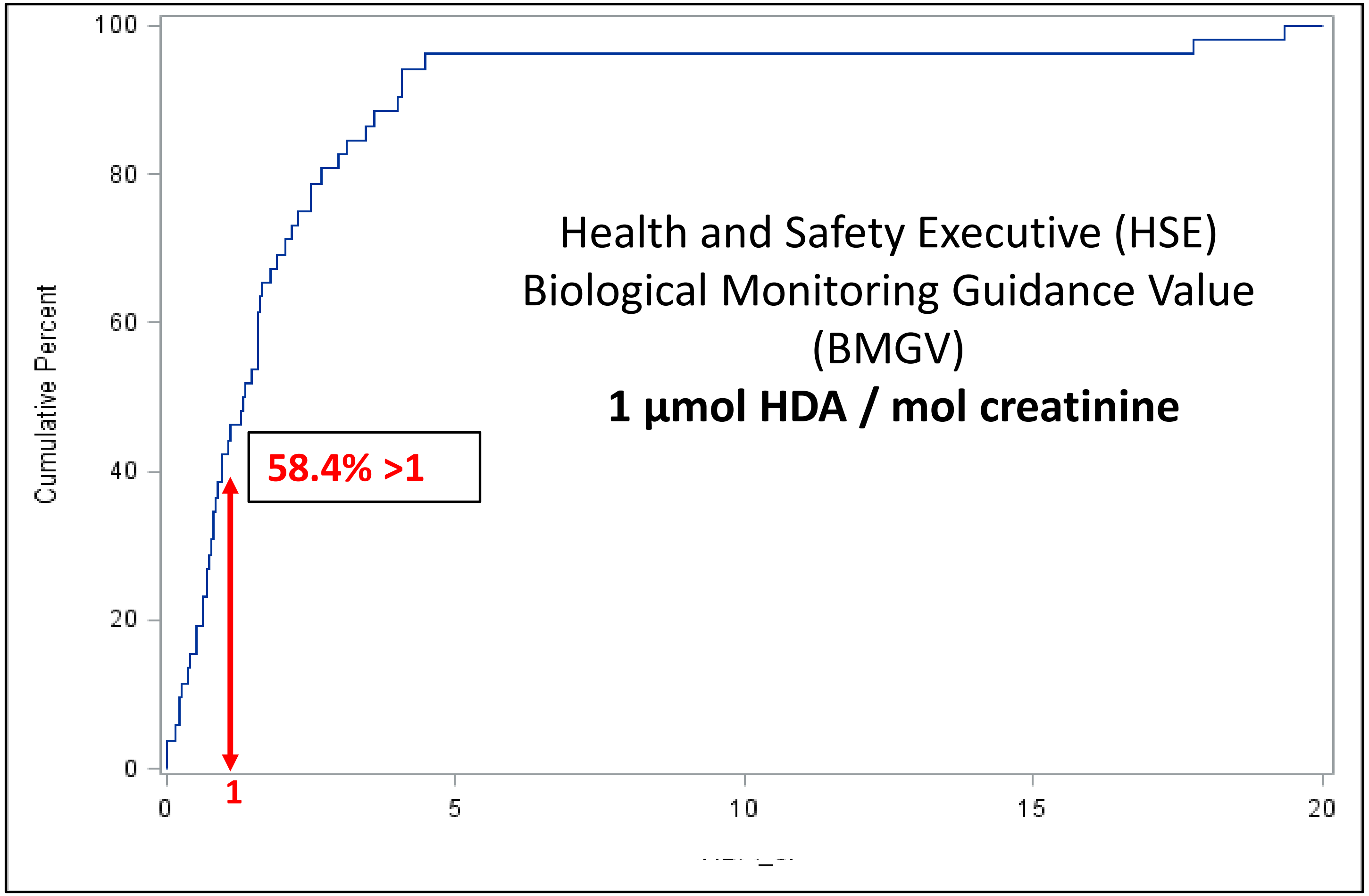
Mid-coat

-Epoxy-based reactive systems

Top-coat

-Aliphatic isocyanate-based reactive systems

Industrial coatings: Urinary HDA exceeds Biological Monitoring Guidance Values (BMGV) in ~60% of samples



HDA adjusted to creatinine **(µmol HDA / mol creatinine)**

- 2.5 x cross-shift increase in urinary HDA-
- 20-35% of air samples exceeded an OEL/REL
- Similarly high potential; for skin exposure (mg isocyanate load)

Opportunities for interventions

1. Currently working with the CPWR team to develop a package of interventions that integrate across various domains - engineering controls and PPEs, heat stress/dehydration, improvements in work practices, and awareness and training
    - Inhalation exposures
    - Skin exposures

2. Ongoing work on Part B – amines, solvents, ENMs/fillers

3. National scale biomonitoring study of industrial painters

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- Rebecca Gore for assistance with statistical analysis

Thank you for your attention!

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Relevant Publications

Annals of Work Exposures and Health, 2020, 1–15
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Original Article
BOHS The Chartered Society for Worker Health Protection
OXFORD

Original Article

Characterization and Quantitation of Personal Exposures to Epoxy Paints in Construction Using a Combination of Novel Personal Samplers and Analytical Techniques: CIP-10MI, Liquid Chromatography–Tandem Mass Spectrometry and Ion Chromatography

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Urinary biomonitoring of occupational exposures to Bisphenol A Diglycidyl Ether (BADGE) – based epoxy resins among construction painters in metal structure coating

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Exposures and urinary biomonitoring of aliphatic isocyanates in construction metal structure coating

Anila Bello[a,*], Yalong Xue[b], Rebecca Gore[c], Susan Woskie[a], Dhimiter Bello[d]

Annals of Work Exposures and Health, 2019, 1–12
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Original Article
BOHS The Chartered Society for Worker Health Protection
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Original Article

Evaluation of Disposable Protective Garments against Isocyanate Permeation and Penetration from Polyurethane Anticorrosion Coatings

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