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Historical Occupational Trichloroethylene Air Concentrations Based on Inspection Measurements From Shanghai, China

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ABSTRACT

Purpose: Trichloroethylene (TCE) is a carcinogen that has been linked to kidney cancer and possibly other cancer sites including non-Hodgkin lymphoma. Its use in China has increased since the early 1990s with China's growing metal, electronic, and telecommunications industries. We examined historical occupational TCE air concentration patterns in a database of TCE inspection measurements collected in Shanghai, China to identify temporal trends and broad contrasts among occupations and industries.
Methods: Using a database of 932 short-term, area TCE air inspection measurements collected in Shanghai worksites from 1968 through 2000 (median year 1986), we developed mixed-effects models to evaluate job-, industry-, and time-specific TCE air concentrations.
Results: Models of TCE air concentrations from Shanghai work sites predicted that exposures decreased 5–10% per year between 1968 and 2000. Measurements collected near launderers and dry cleaners had the highest predicted geometric means (GM for 1986 = 150–190 mg m^{-3}). The majority (53%) of the measurements were collected in metal treatment jobs. In a model restricted to measurements in metal treatment jobs, predicted GMs for 1986 varied 35-fold across industries, from 11 mg m^{-3} in 'other metal products/repair' industries to 390 mg m^{-3} in 'ships/aircrafts' industries.
Conclusions: TCE workplace air concentrations appeared to have dropped over time in Shanghai, China between 1968 and 2000. Understanding differences in TCE concentrations across time, occupations, and industries may assist future epidemiologic studies in China.

KEYWORDS: China; occupational exposures; population-based studies; statistical model; trichloroethylene

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INTRODUCTION

Trichloroethylene (TCE), a chlorinated solvent predominantly used to clean and degrease metal parts, was reclassified by the International Agency for Research on Cancer (IARC) from a probable human carcinogen based on a 1995 review (Kennedy *et al.*, 2000) to a definite human carcinogen (Group 1) in 2012 because of sufficient evidence from epidemiologic studies linking TCE exposure to kidney cancer (Guha *et al.*, 2012). Increased risks at other sites are also suspected, including non-Hodgkin lymphoma and cancers of the liver, prostate, bladder, and esophagus (Scott and Chiu, 2006). TCE exposure has also been linked to generalized skin disorders (Kamijima *et al.*, 2008), the autoimmune disease scleroderma (Cooper *et al.*, 2009), and other acute toxicities involving the central nervous system, liver, kidney, and immune system (EPA, 2011).

Although TCE use began to decline in the late 1970s in the USA (Bakke *et al.*, 2007) and in Europe (Heathcote, 1993) because of environmental and health concerns, TCE use in China began increasing in the 1990s as its electronic, microelectronics, and telecommunications industries rapidly expanded and required efficient chemical degreasers that did not leave residues (Boulton, 1997; Luthje, 2003). Reported TCE use in China rapidly increased from ~4–5 kilotons between 1980 and 1990 (China Chemical Reporter, 1996), to 40 kilotons in 2000 (Xie, 2005), and 165 kilotons in 2010 (China Chlor-Alkali Industry Association, 2013). In the early 2000s, an estimated 60–75% of the TCE consumed in China was used for degreasing, 20–25% for chemical feedstock, 10% for agricultural uses, and 5% for pharmaceutical manufacturing and other uses (Zhang, 2004; Youzhong, 2005).

In light of the continuing use of TCE in China and its suggested link to several cancer sites, it remains important to examine TCE's health effects. In this paper, we describe job-, industry-, and time-specific patterns of short-term area TCE measurements (i.e. ≤20 min) recorded in a database of measurements collected during health and safety inspections in Shanghai, China.

METHODS

Shanghai database of inspection measurements

To identify patterns in TCE air concentrations in Shanghai, China, TCE measurements were extracted from a measurement database that recorded the results of short-term, area air measurements collected between 1968 and 2000 as part of health and safety inspections by the Shanghai Center for Disease Control and Prevention (formerly the Shanghai Health and Anti-epidemic Station). This database was obtained by NCI researchers in the early 2000s to aid exposure assessment efforts for the Shanghai Women's Health Study, a prospective cohort of ~75 000 Shanghai women who were enumerated into the study in the early 2000s (Zheng *et al.*, 2005). Monitoring records for 1990 and 1991 were missing for TCE and other agents (Friesen *et al.*, 2012; Koh *et al.*, 2014). The database included the sampling date, free-text factory name (70 unique factory names for TCE), free-text location of the sampling device, and air concentration in mg m^{-3} (1 ppm = 5.37 mg m^{-3} at 25°C and 1 atmosphere pressure). It also included the industry and occupation represented by each measurement, coded using the 1982 3-digit coding scheme of the Standard Chinese Classification of Industries (SIC) and Occupations (SOC) for the third national census, respectively.

The database did not record the sampling and analytical methods or the quality assurance methods used to evaluate TCE and limited information on analytical methods was available from the Shanghai CDC. Our communication with Shanghai CDC researchers and discussion with industrial hygienists in China suggested that through the 1990s, the industrial hygienists predominantly used colourimetric samplers that collected 1.5 l of air using a pump with a flow rate of 0.5 l min^{-1} (3 min per sample) that passed the air through a porous glass tube containing 10 ml of pyridine solution (1970s: estimated 90%; 1980s: estimated 75%). The color change of the pyridine solution was then compared to prepared concentration standards. Potential interferences included perchloroethylene, carbon tetrachloride, and chloroform. In the 1990s, the use of activated charcoal sorbent tubes to collect 10-min air samples using a pump at a flow rate of 0.1 l min^{-1} became more common (1990s: estimated 50%). These air samples were likely desorbed with carbon disulfide and analyzed using gas chromatography using a flame ionizing detector. In 1998, the previous charcoal tube method was modified to use a pump flow rate of 0.5 l min^{-1} for 20 min to collect a 10 l air sample.

Statistical analyses

Descriptive statistics and graphical representations of the TCE measurements were conducted using Stata/SE version 11.2 (StataCorp LP, College Station, TX, USA).

The inspection measurements were lognormally distributed and thus the natural log-transformed values were used in all analyses. The inspection data set did not include information on the limit of detection (LOD) and did not directly identify samples that were less than the LOD. It appears that undetected samples were mostly reported as '0.1', the most common and lowest TCE value reported in the database; however, this value is much lower than the potential detection limits of the TCE methods. It is also likely that some nondetected samples were entered with the value of the detection limit for that sample. Thus, we used an empirical approach to account for samples less than the LOD. We examined q-q plots that compared the natural log-transformed measurements to a normal distribution for three time periods with expected similar sampling and analytical methods based on input from local industrial hygiene experts: <1990 (colourimetric), 1990–1997 (transition from colourimetric to charcoal tubes), and ≥1998 (charcoal tubes with larger air volume). For each period, we visually identified where the measurements deviated from a normal distribution at the low-end of the scale (the inflection point) to estimate the LOD. This approach may characterize some truly detectable values as less than the LOD, but this conservative approach does not bias the means and variance, whereas setting the LOD too low will bias these parameters (Lubin *et al.*, 2004). Based on this review, we assumed a LOD of 4 mg m^{-3} for measurements collected up to and including 1989, 1 mg m^{-3} for 1990 through 1997, and 0.2 mg m^{-3} for 1998 onwards. Multiple values for each measurement below the LOD were imputed using a previously developed SAS procedure that assumed that the data were lognormally distributed with distribution parameters defined based on the measurements above the LOD (Lubin *et al.*, 2004). A value for each LOD sample was randomly drawn from the exposure distribution of the measurements by implementing the macro in SAS version 9.3 (SAS Institute Inc., Cary, NC, USA). The draw was repeated 10 times, to obtain 10 data sets. The concentration values for measurements above the assumed LOD were unchanged in all 10 data sets.

Predictors of TCE air concentrations were evaluated in SAS using PROC MIXED based on a restricted maximum likelihood estimation method. The model structure is shown in equation 1.

$$\text{Ln}(Y)_{ij} = \beta_0 + \beta(\text{T-1986}) + \sum\beta\text{Occ} + b\text{OccurrenceID}_i + \varepsilon_{ij} \quad (1)$$

$\text{Ln}(Y)_{ij}$ represented the natural log-transformed TCE concentration of the *i*th measurement on the *j*th measurement day. β_0 represented the model intercept. *T* represented the measurement's calendar year and was incorporated as a linear function that was centered on the median measurement year of 1986. Nonlinear time trends were evaluated in preliminary analyses based on both b-splines and penalized b-splines, but a linear function provided the best fit based on AIC (Akaike Information Criteria), thus only models incorporating a linear time trend are shown. 'Occ' represented occupation groups based on the SOC, using the most frequently monitored occupation (e.g. metal treatment jobs, 2-digit SOC: 72) as the reference group. Occupation groups were predominantly kept at the 3-digit SOC level. However, we grouped all metal treatment jobs into one occupation group because TCE use was expected to be similar across these occupations (all occupations within the 2-digit SOC: 72), although likely different within this occupation by industry. Similarly, we also grouped all textile jobs into one occupation group due to their similar uses of TCE (all occupations within the 2-digit SOC: 75). Additionally, other occupation groups at the 3-digit level that had <10 measurements were combined into a 'miscellaneous' group. OccurrenceID represented the measurement occurrence number (unique sampling date/ factory name combination, $n = 134$) and was incorporated as a random effect term to account for correlation between repeated measurements collected within the same factory on the same sampling date. On average, seven measurements were collected per occurrence (median: 4, maximum: 64). 'ε' represented the residual within-occurrence error. The between- and within-occurrence variance components were assumed independent and normally distributed, with means of 0.

Each model was estimated separately on each of the 10 imputed data sets. The resulting variable and variance parameters from the 10 sets were combined using PROC MIANALYZE (Rubin and Schenker, 1991; Lubin *et al.*, 2004). A 'null model', with only the occurrence variable included as a potential explanatory variable, was also estimated using PROC MIXED to estimate how much of the variance was explained by occupation, year, and industry [(variance in null model – variance in multivariable model)/variance in null model].

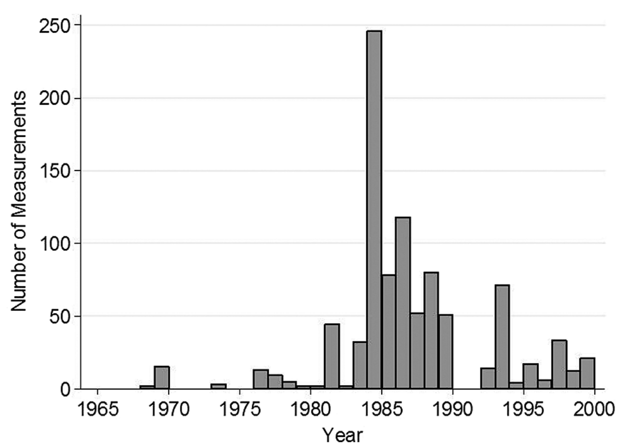
We examined the influence of industry-specific differences in air concentrations for measurements collected in metal treatment jobs, the most commonly monitored group. We restricted the analyses to measurements collected for 2-digit SOC = 72 and used the model structure in equation 1, but substituted an industry variable based on 2-digit SIC groups for the occupation variable.

Sensitivity analyses

Several sensitivity analyses were conducted. Because the use of TCE pre-1980 was expected to be very limited in China, pre-1980 measurements may reflect coding errors that assigned the wrong agent to a measurement record or were collected under unusual circumstances. To examine the influence of pre-1980 measurements, a model was also constructed restricting the data to measurements collected ≥1980. To evaluate the potential influence of change in the sampling and analytical methods, the model was further restricted to measurements collected ≥1990. Because 53% of the measurements were collected in metal treatment jobs, we also examined the time trend for nonmetal treatment occupations separately. To evaluate the influence of high concentrations and possible outliers, the main models were also rerun restricting the analyses to measurements <2000 mg m^{-3}, a concentration above which we observed visual deviations from normality in the q-q plots. We also evaluated whether the time trend varied by industry by including interaction terms for all industry groups (based on 2-digit SIC) with at least 20 measurements (reference = electronic components industry, SIC2: 45) in the respective models.

RESULTS

The inspection database contained 932 TCE measurements collected between 1968 and 2000, after removing duplicate and nonroutine (i.e. measurements not within a worker's proximity) measurements. The majority of the measurements (95%) were collected in the mid- to late 1980s, with a median collection year of 1986 and interquartile range from 1984 to 1988 indicating that the measurements predominantly represented a narrow time window (Fig. 1). Only 49 (5%) measurements were collected pre-1980. In 1969, 12 measurements were collected in the equipment generation/transmission/distribution of electricity industry (SIC 441) and 3 in the machinery for the everyday use industry (SIC 431). In the mid- to late 1970s, 20 measurements were collected in the laundry and dyeing industry (SIC 666), 8 in the clocks, watches, and timing instruments industry (SIC 483), 2 in the manufacturing/repair of precision instruments industry (SIC 499), and 2 in the general industrial machinery manufacturing industry (SIC 428).

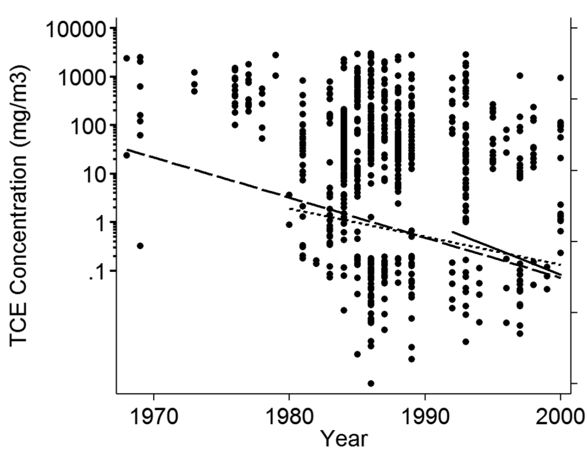


1 Distribution of TCE measurements by year in the Shanghai exposure database of inspection measurements.

Overall TCE air concentrations in the database appeared to slightly decline from the 1980s through 2000 (Fig. 2); formal evaluations of the time trends were examined in statistical models. The proportion of samples below the assumed LOD was 20% for pre-1990, 21% for 1990–1998, and 15% for 1998–2000; these measurements are included in Fig. 2 using their imputed values from the first imputation data set. Measurements >2000 mg m^{-3} accounted for 2% of the pre-1990 measurements and 0.6% of the ≥1990 measurements.

Descriptive statistics of TCE concentrations for occupations monitored by the Shanghai CDC are listed in Table 1. Area measurements were collected in occupations representing 27 unique 3-digit SOC codes, but only 16 occupations had at least 10 measurements. The arithmetic means (AMs) of 17 occupations and the geometric means (GMs) of 11 occupations exceeded the Chinese occupational ceiling limit of 30 mg m^{-3} set in 1990; however, most measurements predated the ceiling limit. The area measurements for two occupations were all <3 mg m^{-3} (SOC: 742, 823). Area measurements in metal treatment jobs (SOC: 724, 726, 729) had a short-term AM of 230 mg m^{-3}, a GM of 30 mg m^{-3}, and a geometric SD (GSD) of 16. Area measurement-based AMs were >200 mg m^{-3} for eleven 3-digit SOC groups, GMs were >30 mg m^{-3} for 12 groups, and maximum concentrations were >1000 mg m^{-3} for 7 groups. We expect that most occupation groups likely used TCE for cleaning and degreasing metal parts or were in areas near degreasing operations (e.g. warehouse jobs). However, the highest AMs were observed for textile dyeing and printing jobs (AM 770 mg m^{-3}; SOC: 756) and laundry jobs (which includes dry cleaners) (AM 710 mg m^{-3}; SOC: 520) where we expect that TCE was likely used as a textile scouring agent (textile jobs) or a cleaner on textiles (both).

In the null model (not shown), the between-occurrence variance (7.7) and within-occurrence variance (3.2) accounted for 71 and 29% of the total variability in the TCE measurement database, respectively. Table 2 lists the model parameters, variance components, and predicted area measurement-based GMs for the year 1986 for occupations with at least ten measurements. Compared to the null model, occupation and calendar year explained 25% of the between-occurrence variability and 1% of the within-occurrence variability. Once adjusted for the time trend, the highest predicted GMs were observed for laundry/dry clean jobs (1986 GM = 190 mg m^{-3}), occupations involved in the production of cultural, education, and sports articles (1986 GM = 130 mg m^{-3}), and other electric/electronic equipment installers/maintainers (1986 GM = 89 mg m^{-3}). Overall, the monitored occupations had predicted area measurement-based GMs for the year 1986 between <3 and 190 mg m^{-3}, with metal treatment jobs predicted to have a 1986 GM of 34 mg m^{-3}. Predicted area measurement-based TCE concentrations decreased 10.2%



2 TCE concentrations across all years (long dashed line) in the Shanghai exposure database of inspection measurements, with time trends from linear regression shown from 1980 onwards (short dashed line) and from 1990s onwards (solid line).

Table 1. Short-term area exposure concentrations by associated 2- and 3-digit standardized occupation codes (SOC) in the Shanghai database of inspection measurements

3-digit SOC and description		TCE concentration, in mg m^{-3}					Median (IQR) measurement year
		N	AM	GM	GSD	Maximum	
034	Mechanical Engineering Personnel	62	45	25	4.3	190	1984 (1984–1984)
520	Laundering (Dry Clean), Dyeing, Repair Garments	23	710	570	2.0	2200	1977 (1976–1977)
522	Cleaning/Waste Disposal Public Surroundings	10	11	<3	16	80	1986 (1986–1986)
523	Odds-and-Ends Workers	18	18	<3	22	78	1986 (1985–1988)
	All SOC2: 52	51	330	15	55	2200	1985 (1977–1986)
724	Workers in Heat Treatment of Metals	4	240	19	74	840	1986 (1985–1986)
726	Workers in Surface Treatment of Metals	14	430	140	6.0	1200	1993 (1993–1993)
729	Other Ore Smelting/Metal Processing	475	220	28	16	3000	1986 (1984–1989)
	All SOC2: 72	493	230	30	16	3000	1986 (1984–1989)
739	Other Chemicals Workers	11	70	11	36	240	1988 (1988–1999)
742	Plastic Processing Machine Operators	12	<3	<3	—	<3	1986 (1986–1986)
753	Install/Maintain Machinery Used for Weaving/Knitting	17	17	<3	26	150	1997 (1997–1998)
754	Cloth-Weaving Workers	3	60	50	2.2	100	2000 (2000)
755	Knitting Workers	3	98	95	1.4	120	1989 (1989–1989)
756	Textile Printing & Dyeing Workers	5	770	680	1.8	1400	1986 (1983–1986)
	All SOC2: 75	28	170	9	38	1400	1997 (1989–1998)
822	Plate Makers	20	510	74	20	2200	1984 (1984–1985)
823	Printing Workers	8	<3	<3	—	<3	1984 (1983–1986)
	All SOC2: 82	28	370	13	50	2200	1984 (1983–1985)
842	Fitters	4	13	<3	41	25	1988 (1988–1989)
849	Other Forge, Tool Makers, Machine Tool Installers	11	<3	<3	—	7	1983 (1983–1992)
	All SOC2: 84	15	5	<3	2.7	25	1988 (1983–1992)
851	Machinery Installation & Assembly	21	25	<3	24	300	1988 (1988–1994)
853	Clocks, Watches, Precision Instrument Production	5	230	9	120	800	1983 (1978–1989)
	All SOC2: 85	26	64	<3	35	800	1988 (1987–1994)
863	Install/Assembly Electric/Electronic Equipment	52	78	6	12	2600	1988 (1982–1988)

Table 1. *Continued*

3-digit SOC and description		TCE concentration, in mg m^{-3}					Median (IQR) measurement year
		N	AM	GM	GSD	Maximum	
869	Other Electric/Electronic Equipment Install/Maintenance	20	180	34	15	840	1983 (1981–1986)
	All SOC2: 86	72	110	10	14	2600	1986 (1982–1988)
901	Painters	2	180	160	1.9	250	1976 (1976–1976)
916	Production of Cultural, Education, Sports Articles (includes sport shoe makers)	12	630	47	65	2900	1996 (1994–1998)
917	Jewelers	4	250	220	2.0	380	1985 (1985–1985)
	All SOC2: 91	16	540	68	39	2900	1995 (1989–1997)
941	Loading/Unloading Workers & Movers	107	41	19	7.1	140	1984 (1984–1984)
961	Examiners/Inspectors	8	27	4	29	80	1996 (1992–1996)
994	Warehouse Workers	1	<3	<3	—	4	1980 (1980–1980)

GSD, unitless.
IQR, interquartile range: 25th to 75th percentile; *N*, number of measurements.

per year [(1 − exp(−0.108))*100%] in the model using all data (Table 2).

In measurements collected for metal treatment jobs, predicted GMs decreased 5.5% per year overall (Table 3). The predicted GMs for the year 1986 near metal treatment jobs varied by industry, ranging from 11 mg m^{-3} in 'other metal products/repair metal' to 390 mg m^{-3} in 'ships/aircrafts'. GMs >100 mg m^{-3} were also predicted for the industries 'metal products' (130 mg m^{-3}), 'machinery for everyday use' (110 mg m^{-3}), and 'clocks, watches, and timing instruments' (350 mg m^{-3}). Electric/electronic component manufacturing, which represented 37% of the measurements for metal treatment jobs, had a predicted GM of 43 mg m^{-3}.

Sensitivity analyses

In a model restricted to measurements collected in 1980 or later, the predicted time trend decreased to 8.1% per year [(1 – exp(−0.0846))*100%] compared to 10.2% per year based on all data (Table 2); however, the confidence intervals overlap. In this restricted model, predicted GMs were generally 5–25% lower for most occupation groups than in the model that used all data. For instance, the predicted GM decreased from 34 to 29 mg m^{-3} for measurements collected in metal treatment jobs. However, the predicted GM increased from 89 to 120 mg m^{-3} for measurements collected in other electric/electronic equipment installers/maintainers, from 20 to 35 mg m^{-3} for other chemical occupations. In a model restricting measurements to those collected from 1990 onwards, exposures decreased 6.2% per year (not shown). All industry × year interaction terms were not statistically significant and thus we found no significant deviation from the overall time trend in any model.

Removing measurements >2000 mg m^{-3}, but including all years, reduced the predicted GMs and time trend to approximately the same estimates as those estimated in the model restricted to measurements from 1980 onwards (data not shown). The two exceptions were other electric/electronic equipment installers/maintainers and laundry/dry clean occupations, which remained the same as in the model that used all data (data not shown).

In the model restricted to measurements in occupations other than metal treatment jobs, the time trend decreased 13.9% per year [(1 − exp(−0.15))*100%] (Table 2). Predicted GMs were generally similar to the model of the overall data and data ≥1980; however, lower predicted GMs were observed for mechanical engineering personnel, install/assembly of electric/

Table 2. Model parameter estimates and predicted GMs based on short-term area TCE measurements reported in the Shanghai exposure database of inspection measurements, by occupation group

	All TCE data			Data restricted to year ≥1980			Excluding metal treatment occupations[b]		
Model parameter	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})
Intercept	3.53	0.29	—	3.38	0.34	—	2.23	0.73	
Year: 1986	−0.108	0.039	—	−0.085	0.058	—	−0.15	0.07	
2- or 3-digit standardized occupation group									
Mechanical Engineering Personnel (SOC3: 034)	−0.21	0.30	28	−0.20	0.30	24	−0.05	0.27	9
Laundering (Dry Clean), Dyeing, Repair Garments (SOC3: 520)	1.74	0.99	190	1.64	1.80	150	2.99	1.36	180
Cleaning/ Waste Disposal Public Surroundings (SOC3: 522)	−3.97	2.48	<3	−3.82	2.62	<3	−2.67	2.74	<3
Odds-and-Ends Workers (SOC3: 523)	−3.05	0.96	<3	−2.94	1.00	<2	−1.82	1.02	<3
Metal Treatment Workers (SOC2: 72)	Reference		34	Reference		29	Excluded		—

Table 2. *Continued*

Model parameter	All TCE data			Data restricted to year ≥1980			Excluding metal treatment occupations[b]		
	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})
Other Chemicals Workers (SOC3: 739)	−0.54	0.73	20	−0.47	0.74	35	−0.55	1.18	5
Textile Workers (SOC2: 75)	0.19	1.01	41	0.17	1.07	35	1.82	1.33	57
Plate Makers (SOC3: 822)	0.52	1.21	57	0.71	1.28	60	1.78	1.43	55
Machinery Installation & Assembly (SOC3: 851)	−3.10	0.79	<3	−3.07	0.81	<3	−2.61	1.40	<3
Install/ Assembly Electric/ Electronic Equipment (SOC3: 863)	−1.27	0.56	10	−1.14	0.58	9	−1.05	1.03	<3
Other Electric/ Electronic Equipment Install/ Maintenance (SOC3: 869)	0.96	1.00	89	1.45	1.11	120	2.11	1.20	77
Production of Cultural, Education, Sports Articles (SOC3: 916))	1.36	1.39	130	1.29	1.47	110	1.33	2.95	35

Table 2. *Continued*

Model parameter	All TCE data			Data restricted to year ≥1980			Excluding metal treatment occupations[b]		
	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})	β	SE	Predicted GM, 1986[a] (in mg m^{-3})
Loading/ Unloading Workers & Movers (SOC3: 941)	−0.38	0.25	23	−0.37	0.25	20	Reference		9
Miscellaneous Jobs	−1.25	0.79	10	−1.32	0.84	8	−0.11	0.89	8
Variance components									
Between-occurrence	5.70	0.20		6.37	1.11		6.67	3.84	
Within-occurrence	3.15	0.20		3.22	0.21		2.19	0.21	

SOC2 and SOC3, 2- and 3-digit standardized occupation code, respectively.
[a]Predicted GM for the median measurement year, 1986. Calculation: GM = exp($\beta_{intercept} - \beta_{year}$*(Year-1986) + Occupation Group Estimate). For the reference group, the 'occupation group estimate' = 0.
[b]Model for metal treatment workers is shown in Table 4.

Table 3. Model parameter estimates and predicted GMs based on short-term area TCE air measurements collected in metal treatment occupations reported in the Shanghai database of inspection measurements

Model parameter	*N*	β	SE	Predicted GM, 1986[a] (in mg m^{-3})
Intercept		3.76	0.51	
Year: 1986		−0.057	0.052	
2-digit standardized industry group				
Metal Products (SIC2: 40)	17	1.12	1.32	130
Other Metal Products/Repair Metal (SIC2: 41)	158	−1.38	0.82	11
Industrial Machinery (SIC2: 42)	36	0.03	1.24	44
Machinery for Everyday Use (SIC2: 43)	25	0.94	1.35	110
Electric/Electronic (SIC2: 44)	183	Reference		43
Electronic Components (SIC2: 45)	13	0.74	1.18	90
Ships/Aircrafts (SIC2: 47)	20	2.21	1.10	390
Clocks, Watches & Timing Instruments (SIC2: 48)	15	2.10	1.14	350
Miscellaneous (all other SIC)	157	−0.73	1.14	21
Variance components				
Between-occurrence		4.38	0.96	
Within-occurrence		3.13	0.28	

SOC2 and SOC3, 2- and 3-digit standardized occupation code, respectively.
[a]Predicted GM for the median measurement year, 1986. Calculation: GM = $\exp(\beta_{intercept} - \beta_{year}$*(Year-1986) + Industry Group Estimate). For the reference group, the 'industry group estimate' = 0.

electronic equipment, production of cultural, education and sports article jobs, and loading/unloading and moving jobs.

DISCUSSION

This paper describes and evaluates the TCE air concentrations reported in a database of TCE airborne air measurements collected during routine health and safety inspections of workplaces in Shanghai, China. TCE air concentrations reported in the inspection database decreased approximately linearly on a log-scale over the 1980s and 1990s and were consistent with exposure time trends reported for other volatile organic compounds (Symanski *et al.*, 1998; Kromhout and Vermeulen, 2000; Creely *et al.*, 2007; de Vocht *et al.*, 2008; Friesen *et al.*, 2012). For instance, Creely *et al.* (2007) observed declines in TCE concentrations in Denmark of 4% per year between 1947 and 1964 and 15% per year between 1964 and 1989; this data represented a variety of industries, including iron and metal, painting, dry cleaning, chemical, electronic, printing, and shoe manufacturing. Similarly, Symanski *et al.* (1998) found an average decline of 7% per year based on vapor measurements from a variety of industries. We found that the magnitude of the time trend differed somewhat depending on whether the model was based on all measurements (10% per year), measurements collected ≥1980 (8% per year), measurements collected pre-1990 (6% per year), measurements collected on metal treatment jobs (5%), and measurements collected on nonmetal treatment jobs (14%). Analyses of industry-specific time trends found no statistically significant deviations from the overall time trend, but differences in trends may have been missed due to sparse data.

The occupational exposure limit for TCE in China was designated 30 mg m^{-3} as a maximum (ceiling) allowable concentration in 1990. In a 2002 update, the occupational exposure limit was changed from a ceiling limit to an 8-h occupational exposure limit set at 30 mg m^{-3} (occupational exposure limit for hazardous agents in the workplace, GBZ2-2002, China). These occupational exposure limits were exceeded by the predicted short-term area measurement-based GMs for 1986 for 7 of the 14 occupation groups examined in the models (Table 2, all data); however, this is a qualitative, not legal, comparison because these measurements were predominantly collected before this ceiling limit was set. The highest predicted TCE air concentrations for Shanghai work sites (1986 GM = 150–190 mg m^{-3}) were observed for the textile, laundering (dry cleaning), and dyeing jobs (SOC2: 75) that may have used TCE as a spot cleaner or as a dry cleaning fluid. Air concentrations in metal treatment jobs (SOC2: 72) that likely used TCE as a degreaser had an overall predicted GM of 29–34 mg m^{-3}; however, concentrations varied over 35-fold by industry with the highest air concentrations observed in the ship and aircraft industries. We expect that this wide heterogeneity across industries may relate to the scale and quantity of the pieces to be degreased, which vary from very small parts in the electronics industry to very large parts in the machinery and aircraft industries, as well as the methods used to degrease (e.g. from small squeeze bottles, large vats with manual dipping, to automated degreasing lines), engineering controls, layout of the operation, economic resources, and other variables (e.g. von Grote *et al.*, 2003; Hellweg *et al.*, 2005; Kikuchi *et al.*, 2012). However, because no information on the tasks performed, types of parts, or degreasing methods was available in either the database of inspection measurements, we were unable to examine these potential determinants, limiting our interpretation to only broad differences in TCE concentrations between occupations and industries across time.

Predicted short-term GM concentrations for metal treatment jobs in Shanghai [1986 GM = 29–34 mg m^{-3} (Table 2)] approximately corresponded to AMs [based on a typical GSD of 2.5 for organic vapors (Kromhout *et al.*, 1993)] of ~44–52 mg m^{-3}. We observed some industry-specific differences, with higher predicted concentrations in the metal products industry, machinery for everyday use industry, ships and aircraft industry, and clocks, watches, and timing instruments industries that were generally similar to the AMs for Chinese work sites that were reported in several English-language studies published in the year 2000 (Table 4). These published AMs ranged from 10 to 1428 mg m^{-3} but were predominantly <200 mg m^{-3} and were mostly based on small numbers of measurements and were collected in work sites with known TCE-induced skin disorder cases. The two studies with the most measurements were conducted by Inoue *et al.* (1989) and Lan *et al.* (2010). Inoue *et al.* (1989) collected 140 airborne measurements using personal diffusive samplers in two factories in northeast China: in one factory the workers produced TCE and in the other factory workers used TCE to degrease metal parts prior to metal plating and found similar TCE concentrations for both types of workers (AMs 62–78 mg m^{-3}) that was similar in magnitude to the predicted concentrations observed here. Lan *et al.* (2010) collected ~160 airborne personal measurements using 3M organic vapor monitoring badges in six factories with TCE metal cleaning operations (AM = 119 mg m^{-3}), which was higher than the overall predicted AM for measurements collected in metal treatment jobs but lower than we observed in metal treatment jobs in the mostly highly exposed industries.

Limitations

Inspection data can be a useful source of identifying exposure patterns and in particular broad contrasts between groups (e.g. Friesen *et al.*, 2012; Koh *et al.*, 2014). However, these patterns must be interpreted in conjunction with the data's limitations, which have been previously mentioned in analyses of benzene measurements (Armstrong *et al.*, 2011; Friesen *et al.*, 2012) and lead measurements (Koh *et al.*, 2014) from this database. First, these samples represent short-term, stationary sampling (hence the high variance components for within-occurrence), which can either over- or underestimate the average personal exposure depending on the placement of the monitoring device relative to the workers' activities. Second, no information was available to evaluate whether the location of

Table 4. Occupational TCE concentrations in air (mg m^{-3}) in China and Singapore reported in published English-language scientific articles

Reference	Study purpose/subjects monitored	Products made	Reported job title/task/area	Year (s) measured	Sample type[a]	Sample duration[b]	*N*	AM (SD)	Range
China									
Inoue *et al.*, 1989[c]	Exposure study of TCE exposure and urinary metabolite levels	TCE	TCE production	NR	P	F	78	NR	11–505
		Metal parts	Degreasing metal parts	NR	P	F	62	NR	5–338
	Subjects: likely representative exposed workers								
Li *et al.*, 2007[c]	Case-control study of a TCE-induced skin disorder indentifying biomarkers for genetic susceptibility	Electronic elements, metal plating	Cleaning and degreasing metals	NR	NR	NR	NR	NR (median = 483)	371–4242
	Subjects: cases and exposed workers performing the same work								
Kamijima *et al.*, 2008	Exposure study in factories with reported skin disorder cases and factories with no cases	Computer keyboards	Soaking and wiping parts	2003	P	F	5	155 (127)	80.7–382
			General worker (bystander)	2002	P	F	2	66.4	65.1,67.7
		Metal parts for copy machines	Degreasing machine operator	2002	P	F	3	128 (40.5)	94.8–175
	Subjects: bystanders and exposed workers performing the same work as cases		Carry products (bystander)	2002	P	F	2	60.3	54.8, 65.8

Table 4. *Continued*

Reference	Study purpose/subjects monitored	Products made	Reported job title/task/area	Year (s) measured	Sample type[a]	Sample duration[b]	*N*	AM (SD)	Range
		Metal parts for electronic devices	Degreasing machine operator	2003	P	F	8	30.8 (29.4)	2.1–74.9
			Near shielded degreasing machines	2003	A	S	NR	NR	11–27
		Watch parts	Metal stamping	2002	P	F	6	93.9 (40.7)	61.5–164
			Metal polishing (bystander)	2002	P	F	2	12.2	11.9, 12.4
			Near metal stamping work	2003	A	S	NR	NR	43–163
		Metal watchbands	Soaking and wiping parts	2003	P	F	9	102 (68)	22.4–248
			Soaking parts	2003	P	F	5	1428 (553)	452–1803
		Printed circuit boards	Degreasing parts with brush	2002	P	F	6	131 (90.2)	52.7–287
			Placing parts (bystander)	2002	P	F	2	9.7	9.5, 9.9
			Near degreasing with brush	2003	A	S	NR	NR	14–243
		Metal parts for electronic devices	Soaking parts	2003	P	F	9	677 (763)	113–2330
			Outside workshop while immersing parts in degreasing tank	2003	A	S	NR	NR	>1620

Table 4. *Continued*

Reference	Study purpose/subjects monitored	Products made	Reported job title/task/area	Year (s) measured	Sample type[a]	Sample duration[b]	*N*	AM (SD)	Range
		Watchbands and electronic parts	NR	2003	A	NR	NR	NR	11.9–80.2
		Keyboards, watchbands, electronic parts	Wiping and packaging area	2003	A	S	NR	NR	<27
		Keyboards, watchbands, electronic parts	Above degreasing tank after immersing parts	2003	A	S	NR	NR	324–432
Xu *et al.*, 2009	Health and exposure study of skin disorder cases	Electronics, hardware products, electroplating	Degreasing metal or electronic products	2003–2005	A	F	NR	NR	18–683
	Subjects: cases	Factories below OEL[d]		2003–2005	A	F	NR	17.6 (7.4)	NR
		Factories above OEL		2003–2005	A	F	NR	45.7 (13.5)	NR
Lan *et al.*, 2010[c]	Cross-sectional molecular epidemiology study	Metal, optical lenses, circuit boards	Cleaning and degreasing	2006	P	F	≥160[e]	119 (193)	NR
	Subjects: representative exposed and unexposed workers								

N, number of measurements; NR, not reported.
[a]A = area sample, P = personal sample.
[b]F = full shift or 8 h, S = short-term using detector tubes.
[c]Results converted from ppm to mg m^{-3} assuming 25°C and 1 atmosphere pressure.
[d]Occupational exposure limit of 30 mg m^{-3}, as recommended by the Ministry of Public Health of China (Xu *et al.*, 2009).
[e]Estimated, based on two to three measurements per subject for 80 exposed subjects.

the area measurements matched well with the assigned occupation code. Third, we were unable to account for temporal correlation in the measurements within worksites because factory name was not recorded in a consistent manner. Fourth, the measurements represent only factories and exposure scenarios that were monitored under the Shanghai CDC health and safety inspection program, which focused on government-owned factories and likely had limited coverage of joint venture and private companies. These inspections may have also focused on reported cases of TCE poisoning. Fourth, information on sampling and analytical methods was lacking at a measurement level. Sensitivity analyses that showed that the change in time trend from 10.2% for all data to 8.1% per year for measurements collected from 1980 onwards to 6.2% per year for 1990 onwards may, in part, reflect a systematic change due to analytical methods. However, it may also reflect changes in the types of occupations, industries, and factories targeted by inspection activities. In addition, models that included interaction terms to examine industry-specific time trends found no deviation from the overall time trend; however, this may reflect the limited amount of measurements available with which to detect differences. Fifth, the LOD and the samples below the LOD were not identified and thus we assumed time-dependent LOD values that were based on deviations from a normal distribution plot. In addition, there is potentially underreporting of measurements below the LOD into the inspection database, as was previously reported by Friesen *et al.* (2012) and Armstrong *et al.* (2011) for benzene measurements. Finally, there was little descriptive information as to workplace factors (e.g. the task, presence of ventilation) to help interpret the measurements.

Despite these limitations, the magnitudes of the time trends and the exposure concentrations were consistent with trends and measurements reported in the published literature and our sensitivity analyses were robust. The predicted concentrations observed here should be interpreted and used cautiously and supplemented with additional information, including expert judgment from industrial hygienists familiar with TCE exposure in Chinese workplaces. For instance, our previous exposure assessments for benzene and lead (Friesen *et al.*, 2012; Koh *et al.*, 2014) used measurements from the Shanghai inspection exposure database to systematically calibrate expert-based estimates from job-exposure matrices to identify broad differences across time, occupation, and industry for the Shanghai Women's Health Study. To assess benzene exposure in another Shanghai epidemiologic study, Armstrong *et al.* (2011) combined the expert judgment of industrial hygienists with exposure information from multiple sources, including the Shanghai inspection database, factory records, onsight investigations, and measurements reported in the literature. Although limited in scope and number of measurements, the TCE models may be similarly useful in helping industrial hygienists characterize TCE exposure for epidemiologic studies.

CONCLUSIONS

TCE air concentrations appeared to have declined over time in Shanghai, China between 1968 and 2000. Understanding differences in TCE air concentrations across time, occupations, and industries may assist future epidemiologic studies in China.

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