Occup Environ Med 1998;**55**:300–309

Comprehensive evaluation of long term trends in occupational exposure: part 1. Description of the database

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Accepted 31 October 1997

Abstract

***Objectives*—To conduct a comprehensive evaluation of long term changes in occupational exposure among a broad cross section of industries worldwide.**

***Methods*—A review of the scientific literature identified studies that reported historical changes in exposure. About 700 sets of data from 119 published and several unpublished sources were compiled. Data were published over a 30 year period in 25 journals that spanned a range of disciplines. For each data set, the average exposure level was compiled for each period and details on the contaminant, the industry and location, changes in the threshold limit value (TLV), as well as the type of sampling method were recorded. Spearman rank correlation coefficients were used to identify monotonic changes in exposure over time and simple linear regression analyses were used to characterise trends in exposure.**

***Results*—About 78% of the natural log transformed data showed linear trends towards lower exposure levels whereas 22% indicated increasing trends. (The Spearman rank correlation analyses produced a similar breakdown between exposures monotonically increasing or decreasing over time.) Although the rates of reduction for the data showing downward trends ranged from −1% to −62% per year, most exposures declined at rates between −4% and −14% per year (the interquartile range), with a median value of −8% per year. Exposures seemed to increase at rates that were slightly lower than those of exposures which have declined over time. Data sets that showed downward (versus upward) trends were influenced by several factors including type and carcinogenicity of the contaminant, type of monitoring, historical changes in the threshold limit values (TLVs), and period of sampling.**

***Conclusions*—This review supports the notion that occupational exposures are generally lower today than they were years or decades ago. However, such trends seem to have been affected by factors related to the contaminant, as well as to the period and type of sampling.**

(*Occup Environ Med* 1998;**55**:300–309)

Keywords: occupational exposure; exposure assessment; long term trends

It is generally recognised that levels of airborne contaminants in occupational settings change over time due to a host of factors.[1–6] As these factors tend to systematically influence the process giving rise to exposure, the underlying distribution of exposures may no longer be characterised by a mean value independent of time. In this instance, the distribution is said to be non-stationary.[7] As many conventional statistical methods are based on the assumption that the parameters of the underlying exposure distribution do not change over time, the failure to recognise non-stationary behaviour may compromise our ability to assess exposures accurately.[6]

Several different workplace conditions can give rise to non-stationary exposure distributions. For example, concentrations in air may steadily rise during start up operations or during periods of increased rates of production, or, conversely, may fall as output diminishes due to such economic factors as downsizing or reduced demand. Changes to the process, the installation of new control equipment, or the introduction of improved work practices should also lower exposures. Thus altogether, it would be anticipated that long term trends would be towards lower exposures, except for periods of heightened activity due to economic growth or start up operations. Moreover, the continuous lowering of exposure limits for many air contaminants provides additional, albeit indirect, evidence that exposures have generally been declining over time.[8]

Although occupational exposure levels are likely to have changed over time, relatively few studies have been conducted to investigate the long term behaviour of exposure.[9–14] More often, evidence of systematic changes in exposure is found in studies designed for other purposes—such as for reconstructing historical exposures as part of epidemiological investigations or for examining the relation between air and biological monitoring. As these investigations are reported in publications covering a wide range of disciplines, it becomes problematic to identify those that characterise long term exposures. As a result, it is difficult to support general statements about temporal trends in concentrations of airborne contaminants.

In the light of a lack of understanding of the historical behaviour of occupational exposures, we undertook a comprehensive evaluation of long term changes in exposure among a broad cross section of industries. Our primary aim was to review the scientific literature, identify

Table 1 Breakdown (number of data sets) of industries, by type of contaminant, for aerosols represented in the database

- Aluminum fluoride:
  - Aluminum fluoride production (1)
- Asbestos:
  - Asbestos cement production (9)
  - Asbestos milling (2)
  - Asbestos mining (4)
  - Asbestos textile production (16)
  - Vermiculite mining and milling (2)
- Arsenic:
  - Copper smelter (25)
  - Decorative glassware manufacturing (2)
- Benzene-soluble fraction of total particulates:
  - Steel plant (1)
- Beryllium:
  - Beryllium extraction and manufacturing (16)
- Cadmium:
  - Battery manufacturing (4)
  - Cadmium production (9)
  - Cd pigment production (1)
  - Cd alloy manufacturing (4)
  - Zinc processing plants(2)
- Chromium:
  - Chomeplating industry (1)
- Coal dust:
  - Coal mining (35)
- Cobalt:
  - Porcelain factories (1)
- Dusts:
  - Aluminum production (5)
  - Cement production (8)
  - Flax mills (15)
  - Grain elevators (1)
  - Granite sheds (9)
  - Haematite iron ore mining (1)
  - Nitrate-fertiliser manufacturing (2)
  - Sulphur, cobalt, zinc plants(4)
  - Talc milling (12)
  - Talc mining (6)
  - Vermiculite milling (2)
- Fluorides (mixed exposure, predominantly particulate):
  - Aluminum smelter (1)
  - Aluminum production (11)
- Hydroquinone:
  - Hydroquinone manufacturing (1)
- Lead:
  - Ammunition manufacture (2)
  - Automotive assembly (3)
  - Battery manufacturing (21)
  - Chemicals manufacturing (1)
  - Crystal glass industry (1)
- Lead (continued):
  - Foundries (8)
  - Lead smelting (4)
  - Metal scrap industry (1)
  - Metal producing plants (2)
  - Motor car repair shops (2)
  - Polyvinyl chloride plastics industry (1)
  - Railroad machine shops (1)
  - Sonar equipment manufacturing (1)
- Machining fluid:
  - Automotive plants (3)
- Manganese:
  - Manganese ore milling (1)
- MBOCA:
  - Polyurethane plastics (1)
- Nickel:
  - Copper refining (9)
  - Nickel alloy manufacturing (3)
  - Nickel mining (3)
  - Nickel milling (16)
  - Nickel smelting (34)
  - Nickel refining (80)
- Nickel oxides:
  - Nickel smelting (5)
- Nickel sulphides:
  - Nickel refining (3)
- Nitrate esters:
  - Dynamite manufacturing (4)
  - Nitrate fertiliser plant (4)
- Polynuclear aromatic hydrocarbons:
  - Needle coke manufacturing (2)
- Silica:
  - Coal mining (7)
  - Copper mining (1)
  - Pottery making (15)
  - Mining of metal ores (65)
- Sulphuric acid:
  - Battery factories (2)
  - Galvanising factories (2)
- Thallium:
  - Alloy anode plate manufacturing (1)
- Trichlorfon:
  - Pesticide production (1)
- Trimellitic anhydride:
  - Plastics, epoxy resins, and paint manufacturing (3)
- Trinitrotoluene (mixed exposure, predominantly particulate):
  - Munitions plant (2)
- Uranium:
  - Uranium milling (1)

studies that reported historical changes in exposure, and compile the exposure data reported therein. We then wanted to measure changes in exposure, determine whether consistent trends have occurred industry wide, and explain why some exposures have changed more than others. This paper describes our review of the literature and measurement of temporal effects; an accompanying article[15] focuses on our assessment of factors which influenced changes in exposure over time.

Methods

COMPILATION OF THE DATABASE

The scientific literature, primarily in the fields of occupational hygiene and occupational epidemiology, was reviewed to identify studies that reported historical occupational exposures. Two databases available from other investigations, one of chemical exposures from various industries worldwide,[16] and the other of nickel exposures derived exclusively from industrial sources, were also accessed to identify data suitable for analysis. At least three average exposure levels (estimated from area, personal, or biological monitoring data) had to be reported over a period of three years or longer. Data presented either in tabular or graphical form were used after being abstracted as they were reported (including any transformation performed by the original authors—for example, conversion from particle number concentrations to mass units). The midpoint of the sampling interval was computed when mean exposure levels had been averaged over periods extending for two or more years. In studies presenting both arithmetic and geometric mean (median) exposures, arithmetic means were used. If an investigator had published more than one study with the same set of data, only one was included in the database.

As well as compiling average exposure levels by period for each data set, details on the source and type of contaminant, along with features of the sampling regimen, were recorded. The United Nations international standard industrial classification (ISIC) of all economic activities was used to assign industrial codes.[17] The countries in which the data had been collected were identified. The International Agency for Research on Cancer (IARC) system of classification was used to group contaminants according to their carcinogenicity.[18–24] Also, IARC classification during the period over which the data were collected was compiled.[25–30] Information about the type of monitoring (biological, area, or

*Table 2 Breakdown (number of data sets) of industries, by type of contaminant, for gases and vapours represented in the database**

- Acetone:
  - Automotive assembly (1)
  - Cellulose acetate yarn manufacturing (1)
- Allyl chloride:
  - Organochlorine production (1)
- 1,3-dichloropropene:
  - Organochlorine production
- Benzene:
  - Petroleum refining (16)
  - Rubber hydrochloride manufacturing (17)
- p-Benzoquinone:
  - Hydroquinone manufacturing (1)
- Bis-chloromethyl ether:
  - Anion exchange manufacturing plant (1)
- 1,3-Dichloropropene:
  - Organochlorine production (1)
- N,N-dimethlyformamide:
  - Urethane resin manufacturing (1)
- Carbon disulphide:
  - Rayon filament and CS_2 production (6)
  - Viscose rayon industry (7)
- Carbon disulphide and hydrogen sulphide:
  - Carbon disulphfide factory (1)
  - Viscose film factories (3)
  - Viscose rayon factories (8)
- Epichlorohydrin:
  - Organochlorine plant (1)
- Ethylene oxide:
  - Plant producing medical equipment (2)
- Formaldehyde:
  - Furniture manufacturing (1)
  - Urea formaldehyde resin manufacturing (1)
- Hexachlorobenzene:
  - Chlorinated solvents manufacturing (1)
- Hexachloropentadiene:
  - Organochlorine plant (1)
- Hydrogen chloride:
  - Chemical manufacturing plant (2)
- Isocyanates (toluene diisocyanates or diphenyl methane isocyanates):
  - Automobile parts manufacturing (4)
- Isopropyl alcohol:
  - Automotive assembly (1)
- Isopropyl biphenyls:
  - Capacitor manufacturing (3)
- Mercury vapour:
  - Chloralkali industry (2)
  - Fluorescent lamp factory (2)
- Methyl methacrylate:
  - Acrylic fibre manufacturing (1)
  - Chemical manufacturing (1)
- Methylene chloride:
  - Film manufacturing plant (1)
- Mineral spirits:
  - Painting shops (4)
- Nitrogen dioxide:
  - Fertiliser manufacturing (1)
- Organic vapour:
  - Pesticide manufacturing (3)
- Petroleum distillates:
  - Automotive assembly (1)
- Piperazine:
  - Chemical manufacturing (2)
- Solvents:
  - Furniture manufacturing (1)
- Styrene:
  - Automotive assembly (1)
  - Chemical manufacturing (1)
  - Reinforced boat manufacturing (1)
  - Wholesale and commission trade (1)
- Sulphur dioxide:
  - Copper smelter (6)
  - Pulp and paper mill (2)
  - Sulphur, cobalt, zinc plants (4)
- Tetrachlorethylene:
  - Automotive assembly (1)
- Toluene:
  - Automotive industry (1)
  - Painting shops (5)
  - Photogravure printing (2)
- 1,1,1,-Trichlorethane:
  - Automotive assembly (1)
- Trichloroethylene:
  - Automotive industry (1)
- Vinyl chloride:
  - Polyvinyl chloride plants (5)
- VM and P Naptha:
  - Automotive assembly (1)
- Xylenes:
  - Painting shops (5)

*Data sets averaging exposure levels across industries are not tabulated.

personal sampling) was recorded. Short term personal samples collected in the breathing zone were distinguished from shift long personal samples. Data on the aggregation of exposure levels (by job group or work area, by factory or plant, across factories, or across industries) were compiled as well. If provided, any information about changes in sampling or analytical methods during the period monitored was noted. To assess time dependent effects, data were stratified into three periods based on the period of sampling—that is, up to and including 1972, both before and after 1972, or from 1972 onwards. To evaluate the possibility of a relation between trends toward lower air contaminant levels and the reduction of exposure limits over time, historical changes in the threshold limit values (TLVs) recommended by the American Conference of Governmental Industrial Hygienists (ACGIH) were recorded over the period during which each set of data had been collected.[31 32]

Finally, as some studies summarised exposures with both biological and airborne monitoring data, reported both personal and area monitoring results, or averaged the data in more than one way—for example, by job group and across job groups—a variable was coded to identify duplicated data. Occasionally, different investigators used the same set of data in their studies. These duplicates were identified whenever possible and were omitted in all analyses.

Table 3 Breakdown of the database by selected characteristics related to the contaminant or to the sampling regimen (n=696 data sets)

Factor	*n*	*%**
Region:		
Western Europe	251	36
North America	289	42
Japan	23	3
Eastern Europe	10	1
Other	123	18
Industry:		
Manufacturing	523	75
Mining and quarrying	163	23
Other	10	1
Type of contaminant:		
Vapours	170	24
Non-metallic aerosols	256	37
Metals and metalloids	270	39
Carcinogenicity†:		
1	320	46
2A	100	14
2B	79	11
3	30	4
Not classified	167	24
Type of monitoring:		
Personal	309	44
Stationary	253	36
Personal and stationary	41	6
Biological	93	13
Mean exposure levels:		
By job group or work area	436	63
By plant or factory	105	15
Across factories	147	21
Across industries	8	1
Period of sampling:		
Before 1972	83	12
Before and after 1972	269	39
From 1972 onwards	344	49
Observations (n):		
3–4	294	42
5–10	256	37
11–51	146	21
Duration of sampling (y):		
⩽5	125	18
6–10	163	23
11–20	200	29
⩾21	208	30
TLV‡ reductions (n):		
0	361	52
1	166	24
2	162	23
3	7	1

*Percentages may not add up to 100 due to rounding.
†Based on the most recent IARC classification of contaminants.
‡TLV=threshold limit value of the American Conference of Governmental Industrial Hygienists (ACGIH).

When selecting data sets to include, preference was given to airborne monitoring data, personal sampling results, and to data averaged over the smallest possible group of workers.

ANALYSIS OF TEMPORAL CHANGES IN EXPOSURE

Scatter plots of exposure levels and their natural logarithms over time were generated. For each data set, the non-parametric Spearman rank correlation coefficient was calculated to assess whether there was any evidence of a monotonic change in exposure over time. As linear trends in the log transformed values of exposure levels over time have been found in several previous investigations,[33–35] a simple regression of the logged exposure data on time was carried out. The model can be expressed as follows:

$$E(Y_{it_{ij}})=E(\ln X_{it_{ij}})=\alpha_i+\beta_i t_{ij} \qquad (1)$$

for i=1, 2, ..., n data sets and where t_{ij} is the j-th timepoint for the i-th data set, j=1,2, ..,n_i . Note that the time points are not necessarily evenly spaced and are indexed by i indicating that each data set may vary in terms of the

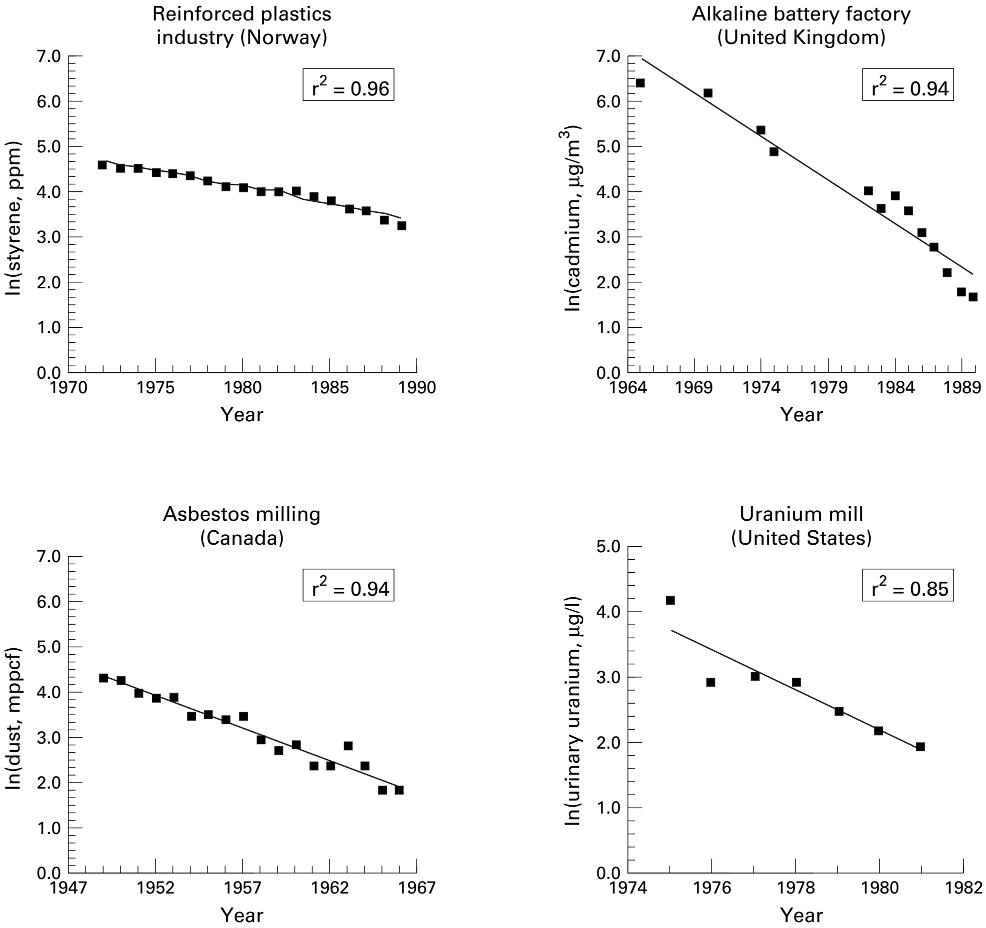

Figure 1 Examples of declining exposures to styrene in the reinforced plastics industry,[84] dust in asbestos milling,[37] cadmium in a battery factory,[34] and uranium as measured in the urine of workers at a uranium mill.[55]

number of occasions at which exposure levels had been evaluated. Here, $X_{it_{ij}}$ represents the mean exposure level for data set i at time point t_{ij}, $Y_{it_{ij}}$ is the natural logarithm of the exposure level for data set i at time point t_{ij}, α_i is the intercept term, and β_i is the regression coefficient associated with time for the i-th data set.

The unweighted least squares slope estimate ($\hat{\beta}_i$) was then used to compute the quantity $100(\hat{\theta}_i)$, the estimated median percentage change in exposure per year, where $\hat{\theta}_i = (e^{\beta_i}-1)$. The theory supporting the use of $\hat{\theta}_i$ is presented in the appendix. (In cases in which monthly data were recorded over periods of several years, the slope estimate was first multiplied by a factor of 12 to obtain the corresponding annual value.) Note that the regression model assumes a fixed percentage increase ($\beta_i > 0$) or decrease ($\beta_i < 0$) in the exposure level from one year to the next. Correlation and regression analyses were performed with SAS procedures (SAS Institute, Cary, NC). Although we largely report our findings on the basis of whether the trend estimates were positive or negative, results evaluated at a significance level of 0.05 are summarised as well.

EVALUATION OF MULTIPLE COMPARISON ISSUES

The issue of multiple testing was considered in two ways. Firstly, we examined our overall findings by using a χ^2 test to evaluate the likelihood of obtaining the observed proportion of data with negative trends assuming that exposure levels had not changed over time—that is, under the null hypothesis that there was an equally likely chance of obtaining either a negative or positive estimated trend result for each of the n data sets. Secondly, given that n independent regression analyses were examined for significance ($p<0.05$) under the null hypothesis that exposure levels remained the same over time, the expected number of significant results if all n of the individual null hypotheses were true (namely, 0.05n) was compared with the observed number of data sets with significant trends.

Results

CHARACTERISTICS OF THE DATABASE

In total, 119 studies that reported exposure levels over time were identified, along with several sources of unpublished data. In many cases, more than one set of data was available within an individual study, so that a total number of 696 data sets were compiled. Data were found in 25 journals that spanned a broad range of disciplines, including occupational hygiene, epidemiology, medicine, environmental health, and toxicology. Although most studies (71%) were published in epidemiological or medical journals, relatively few (11%) came from the classic occupational hygiene literature. Taken together, the studies were published over almost a 30 year period, with the earliest paper appearing in 1967 and the latest in 1996.

Tables 1 and 2 show that the database represents a broad range of exposures to both aerosols and vapours arising in various different industries. Although most studies focused on investigations of workplaces in North America,[33,35–70] the United Kingdom,[34,71–84] and in Scandinavia,[2,9,10,12–14,84–112] other countries were also represented—namely, Czechoslovakia,[113,114] France,[115] Germany,[22,116] Italy,[84,117–122] the Netherlands,[123–126] Poland,[127] Romania,[128] Japan,[129–133] China,[134–140] Singapore,[141] Greenland,[142] Israel,[143] South Africa,[144] and Australia (unpublished data). Table 3 summarises characteristics related to the source and type of contaminant represented by the data (n=696 data sets).

ANALYSIS OF TEMPORAL CHANGES IN EXPOSURE

Based on results from the Spearman rank correlation analyses, 76% of the data showed evidence of a decreasing monotonic change in exposure over time ($r_S < 0$), 23% indicated increasing exposures ($r_S > 0$), and few (1%) showed no change in either direction ($r_S=0$). The linear regression analyses produced similar results with 78% of the logged data indicating trends towards lower exposure levels (Pearson's $r < 0$). For the group of declining exposures, strong correlations were found with a median Spearman correlation coefficient of −0.80 (median Pearson's $r=-0.80$). On the other hand, increasing exposures showed, on average, much lower correlations with a median Spearman correlation coefficient of 0.37 (median Pearson's $r=0.34$).

Linear regressions of the logged exposures over time generally showed evidence of temporal effects on exposure levels, sometimes with clear decreasing linear trends of the logged data over time. Figure 1 shows this for styrene in the reinforced plastics industry,[84] dust in asbestos milling,[37] cadmium in a battery factory,[34] and uranium as measured in the urine among workers at a uranium mill.[55] Overall, the median r^2 value for data sets with declining trends was 0.65; the corresponding value for increasing exposures was 0.11.

Table 4 compares the results from the linear regression analyses stratified by various factors related to the contaminant or to the type of sampling performed. Our results suggest that the breakdown by geographical region, industrial sector, and type of contaminant was relatively similar for both downward and increasing exposures. On the other hand, 31% of the exposures evaluated by personal monitoring had increased over time compared with 14% of the biological monitoring data. Although the differences were not large, a greater percentage of carcinogens (designated as class 1, 2A, or 2B by IARC during the sampling period) (26%) increased over time compared with non-carcinogens (17%). When comparing the group of data collected before 1972 with those exposures evaluated after 1972, a larger proportion of the later data was characterised by increasing exposures (8% versus 31%). Table 4 also provides a breakdown by duration of sampling, sample size, and the number of reductions in the TLV that took place during the sampling period.

Table 4 Breakdown (%) of the trend results stratified by characteristics related to the contaminant or to the sampling regimen

	Downward trend (543 data sets)	*Upward trend (153 data sets)*
Region:		
Western Europe	202 (80)	49 (20)
North America	225 (78)	64 (22)
Japan	17 (74)	6 (26)
Eastern Europe	10 (100)	—
Other	89 (72)	34 (28)
Industry:		
Manufacturing	402 (77)	121 (23)
Mining and quarrying	133 (82)	30 (18)
Other	8 (80)	2 (20)
Type of contaminant:		
Vapour	135 (79)	35 (21)
Non-metallic aerosol	211 (82)	45 (18)
Metal or metalloid	197 (73)	73 (27)
Carcinogenicity*:		
Carcinogen	269 (74)	95 (26)
Non-carcinogen	274 (83)	58 (17)
Type of monitoring:		
Biological	80 (86)	13 (14)
Personal	213 (69)	96 (31)
Area	210 (83)	43 (17)
Area and personal	40 (98)	1 (2)
Exposure levels:		
By job group or work area	334 (77)	102 (23)
By factory	84 (80)	21 (20)
Across factories	119 (81)	28 (19)
Across industries	6 (75)	2 (25)
Period of sampling:		
Up to and including 1972	76 (92)	7 (8)
Before and after 1972	229 (85)	40 (15)
From 1972 onwards	238 (69)	106 (31)
Observations (n):		
3–4	248 (84)	46 (16)
5–10	192 (75)	64 (25)
11–51	103 (71)	43 (29)
Duration of sampling (y):		
⩽5	85 (68)	40 (32)
6–10	122 (75)	41 (25)
11–20	147 (75)	53 (25)
⩾21	189 (91)	19 (9)
TLV† reductions (n):		
Zero	253 (70)	108 (30)
One	134 (81)	32 (19)
Two or three	156 (92)	13 (8)

*Classified as a carcinogen if IARC designated the contaminant as a class 1, 2A, or 2B carcinogen by the end of the sampling period.

†TLV=threshold limit value of the American Conference of Governmental Industrial Hygienists (ACGIH).

Figure 2 A shows a cumulative frequency distribution of the rates of change in exposure for the entire set of data (n=696). The rates ranged from −62% to +121%, with values of −1%, −6%, and −11% per year for the 25th, 50th, and 75th percentiles, respectively. Figure 2 B shows the cumulative frequency distribution of the data showing downward trends in exposure (n=543). The rates of reduction ranged from −1% to −62% per year. However, most exposures declined at rates between −4% and −14% per year (the interquartile range), with a median value of −8% per year. The 25th, 50th, and 75th percentiles for those data with significantly negative trends ($p<0.05$) were −6%, −9%, and −17% per year (n=202, data not shown). Finally, for the data sets showing upward trends (n=153), the 25th, 50th, and 75th percentiles were +2%, +6%, and +12% per year (fig 2 C). (Of these data, only 13 data sets produced significantly positive trends ($p < 0.05$)).

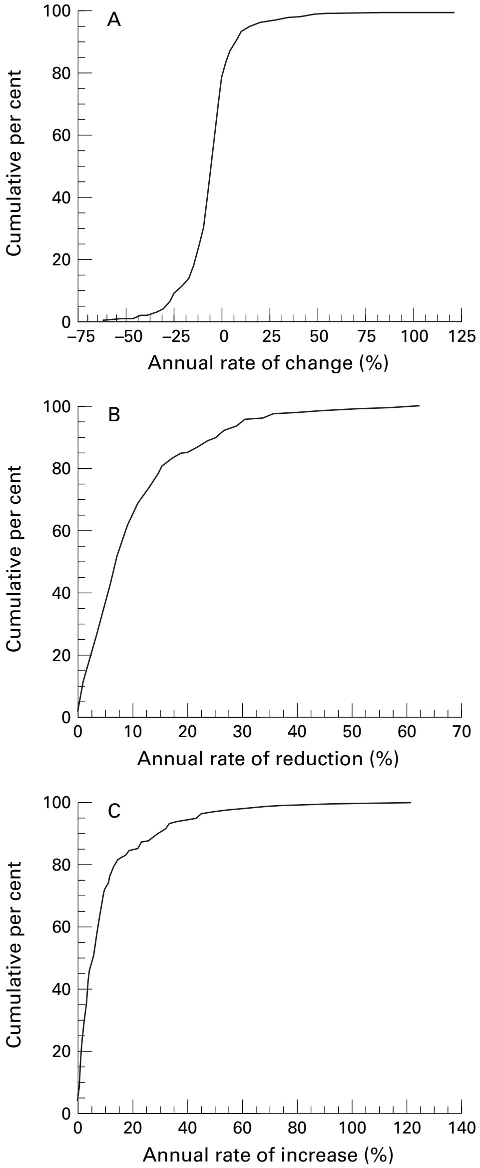

Figure 2 Cumulative frequency distributions of the rates of change for (A) decreasing and increasing trends (n=696), (B) decreasing trends (n=543), and (C) increasing trends (n=153).

EVALUATION OF MULTIPLE COMPARISON ISSUES

The χ^2 test compared the observed proportion of the 696 data sets with negative trends (namely, 0.78=543/696) with the null value of 0.50, and was highly significant. Secondly, under the null hypothesis that exposures had not changed, we would expect, on average, about 35 of the 696 regression analyses to be significant when each regression is evaluated at a significance level of 0.05. We found that 215 (many more than 35) data sets had either significantly negative (n=202) or significantly positive (n=13) trends. These results make it clear that our findings are not attributable to chance and that issues related to multiple comparisons have not contaminated our conclusions.

Discussion

Despite the widespread belief that exposures are generally lower today than they were years or decades ago, studies that offer general support for this notion are scarce. In this study, we investigated long term exposures to a wide range of airborne contaminants among a broad cross section of workplaces world wide. In total, 696 sets of data were identified spanning 69 years of measurements. Our results provide clear evidence of global downward trends in exposure (far more exposures declined than increased over time), at rates which generally ranged from −4% to −14% per year (fig 2 B). Exposures seem to have risen at lower rates than those which have declined over time. Despite the many statistical tests that were performed (which increased, for example, the probability of detecting significant trends due to chance alone), multiple testing does not seem to have invalidated our results.

Our findings also suggest that the simple log linear model described decreasing trends reasonably well (median r^2=0.65), but performed worse when exposures increased over time (median r^2=0.11). As the log-linear model seems to represent a reasonable approximation for declining exposures, model misspecification is unlikely to have presented significant problems in most cases. On the other hand, more complex models might better describe the pattern of increasing exposures and will be explored in a separate investigation.

Although only small differences were detected when the data were stratified by region or industrial sector, a greater proportion of data sets with upward trends was found for carcinogens than for non-carcinogens, and for metals than other types of contaminants (non-metallic aerosols or vapours). Similarly, a larger proportion of exposures assessed by personal rather than biological monitoring increased over time. Effects related to calendar period and number of reductions in TLVs were found as well. Interestingly, there was almost a fourfold difference in the percentage of increasing exposures for data collected after 1972 (31%) relative to data collected before 1972 (8%). As expected, a smaller percentage of the data was characterised by upward trends as the number of TLV reductions increased.

Changes in air sampling and analytical methods can make it difficult to compare results obtained from different periods. Such changes were reported in about 25% of the studies, but probably underrepresent the actual figure because information about the measurement protocol was often not provided. However, differences in the precision of the estimated exposure levels throughout the sampling period (which may also have resulted from unequal numbers of measurements contributing to each mean value) should not bias our slope estimates because such unweighted least squares estimators are unbiased (assuming that the model is correct) even when the error variances are unequal.[145] This lack of bias does not extend to assessments which relied on both area and personal sampling. In this instance, because measurements made with personal sampling are typically higher than those made with area samplers,[9 48 64 142 146–148] the estimated rates of reduction are likely to be smaller—that is, they are conservative—when earlier exposures were assessed by area sampling and later exposures by personal sampling. However, as relatively few data sets (6%) fell into this category, our overall results are unlikely to have been significantly affected.

Although area sampling data will generally underestimate workers' personal exposures, the average concentration at a particular location should reflect the general work environment at a particular time and should thereby bear some relation to workers' exposures received over the same period. Thus, such data were judged to be adequate to evaluate long term trends. Although there were limited data available to compare area and personal sampling data, consistent results were obtained when such comparisons were made.

As well as errors introduced by changes in air sampling and analytical methods, the quality of the data also depends upon the sampling strategy used. As sampling strategies are often targeted toward anticipated worst case exposures, assessments of time trends may not be representative of temporal changes in average exposure levels. If assessment strategies changed during the course of a study from monitoring worst case exposures to sampling workers randomly (where the biased results are likely to have yielded higher levels than what would have been obtained had random sampling been conducted),[2 149 150] then it is also possible that exposures could have seemed to decline even when they remained unchanged. Unfortunately, little information about the sampling strategies was provided in most cases; so it was not possible to evaluate the part that such changes in assessment practices may have played.

It is also clear from this review that few high quality studies have been conducted to assess long term trends in exposure. Notable among our findings was the high percentage of data sets (42%) comprised of few (three or four) observations. Although such small sample sizes may underrepresent data actually available from industrial sources, the sparseness of the published historical record can compromise the statistical power to detect trends—for example, only 31% of the data evaluated in this study showed evidence of a significant trend ($p<0.05$). In the future, it is incumbent on occupational hygienists, who have primary responsibilities for assessing exposures, to assume a greater role in reporting such assessments in the peer reviewed literature. Changes in the work environment and the process, as well as in the methods of data collection and analysis, should also be documented.

Furthermore, the overall poor quality of the historical record, documented herein, underscores the importance of implementing more rigorous assessment practices. The arguments in favour of statistically based sampling strategies have been clearly made[4] and rest largely with their use in considering a full range of questions related to overexposure, the selection of controls, the grouping of workers, and the assessment of exposure-response relations. Although such strategies take into account important random sources of variation in exposure experienced over time and between workers, the influence of temporal effects on such strategies has only recently been considered[6] and warrants further investigation.

Finally, inadequate historical data severely restrict our ability to evaluate health risks associated with occupational exposures. Although a wide variety of retrospective exposure assessment methods have been developed,[151] limited options are available when few or no measurement data exist. Thus, if it were possible to measure the rates of reduction in exposure for particular contaminants in specific industries, or to identify factors that affected these rates in a consistent fashion, then statistical models could be constructed to reconstruct past exposures. Such models would greatly assist in assessing exposure-response relations in situations where little or no historical exposure data are available.

We thank Sharon Cooper and Irva Hertz-Picciotto who reviewed the manuscript and provided helpful comments. This work was supported in part by the National Institute of Environmental Health Sciences (NIEHS) through training grant T32ES07018.

Appendix: Derivation of the estimator of the median proportionate change in exposure per year

Suppressing the subscripts i and j for notational convenience, the simple linear regression model to evaluate changes in exposure levels over time (t) can be specified as follows:

$$Y_t = \ln X_t = \alpha + \beta t + \varepsilon_t$$

Under the model, X_t is the mean exposure level at time t and Y_t is the log transformed value of the mean exposure level. It is assumed that ε_t is normally distributed with zero mean and variance σ_t^2 for all t, and that the $\{\varepsilon_t\}$ are mutually independent. It then follows that $Y_t \sim N(\alpha + \beta t, \sigma_t^2)$ and that the $\{Y_t\}$ are mutually independent. Thus, X_t has a lognormal density with $E(X_t) = e^{\alpha + \beta t + 1/2\sigma^2 t}$, and the $\{X_t\}$ are mutually independent.

Now, the relative change in exposure levels from year t to year t+1 is $\dfrac{X_{t+1} - X_t}{X_t} = \dfrac{X_{t+1}}{X_t} - 1$

and this is the random variable of interest. The distribution of this random variable can be determined as follows:

Since $\ln\left(\dfrac{X_{t+1}}{X_t}\right) = \ln X_{t+1} - \ln X_t = Y_{t+1} - Y_t$,

where $(Y_{t+1} - Y_t) \hat{\sim} N(\beta, \sigma_t^2 + \sigma_{t+1}^2)$, it follows

that $\dfrac{X_{t+1}}{X_t}$ is lognormal with

$$E\left(\frac{X_{t+1}}{X_t}\right) = e^{\beta + 1/2\left(\sigma_t^2 + \sigma_{t+1}^2\right)}$$

and that $\frac{X_{t+1}}{X_t} - 1$ is also lognormal with

$$E\left(\frac{X_{t+1}}{X_t} - 1\right) = e^{\beta + 1/2(\sigma_t^2 + \sigma_{t+1}^2)} - 1.$$

In our application, the median θ of the lognormal density of $\frac{X_{t+1} - X_t}{X_t}$ is the parameter of interest, and it satisfies the following relationship:

$$\frac{1}{2} = \text{pr}\left[\frac{X_{t+1} - X_t}{X_t} < \theta\right]$$

$$= \text{pr}\left[X_{t+1} < (1 + \theta)X_t\right]$$

$$= \text{pr}\left[\ln X_{t+1} < \ln(1 + \theta) + \ln X_t\right]$$

$$= \text{pr}\left[Y_{t+1} - Y_t < \ln(1 + \theta)\right]$$

$$= \text{pr}\left[\frac{(Y_{t+1} - Y_t) - \beta}{\sqrt{\sigma_t^2 + \sigma_{t+1}^2}} < \frac{\ln(1 + \theta) - \beta}{\sqrt{\sigma_t^2 + \sigma_{t+1}^2}}\right]$$

Since $\frac{(Y_{t+1} - Y_t) - \beta}{\sqrt{\sigma_t^2 + \sigma_{t+1}^2}}$ has a standard normal distribution, we must have:

$$\frac{\ln(1 + \theta) - \beta}{\sqrt{\sigma_t^2 + \sigma_{t+1}^2}} = 0, \text{ or } \ln(1 + \theta) = \beta, \text{ or } \theta = e^{\beta} - 1.$$

The median proportionate change in exposure per year can be estimated by $\hat{\theta} = e^{\hat{\beta}} - 1$. However, since the unweighted least squares estimator $\hat{\beta} \sim N(\beta, \sigma_{\hat{\beta}}^2)$ when the assumed straight line model is valid, $E(\hat{\theta}) = e^{\beta + 1/2\,\sigma^2 \hat{\beta}} - 1 > \theta$. So, $\hat{\theta}$ generally tends to be less negative than desired when $\beta < 0$, and it tends to be more positive than desired when $\beta > 0$. In general, the bias in $\hat{\theta}$ will be small when $\sigma_{\hat{\beta}}^2$ is small—that is, when the number of observations is large or when σ_t^2 is small for all t).

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