LBL-13195

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

ENERGY & ENVIRONMENT DIVISION

Presented at the New York Academy of Medicine Symposium on Health Aspects of Indoor Air Pollution, New York, NY, May 28-29, 1981

SOURCES AND CONCENTRATIONS OF ORGANIC COMPOUNDS IN INDOOR ENVIRONMENTS

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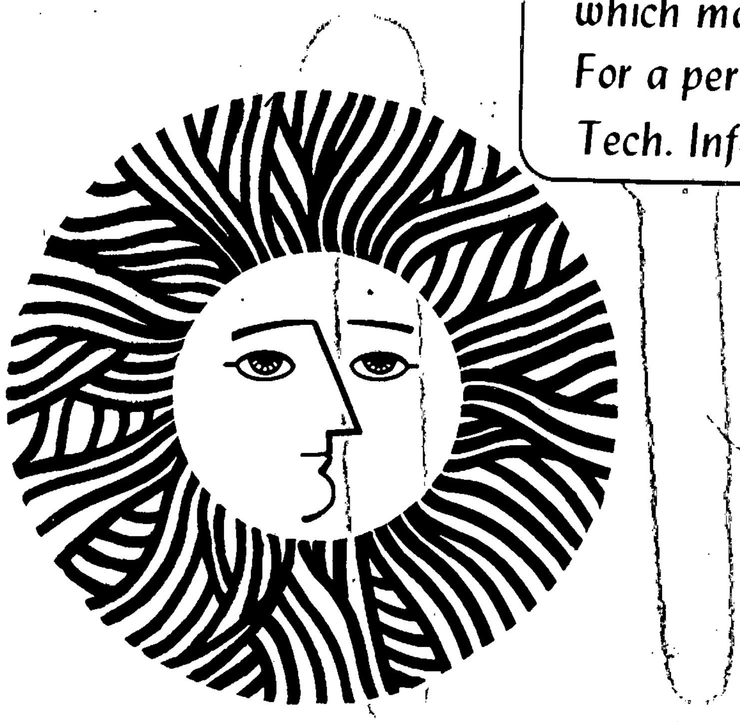

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

Presented at the
New York Academy of Medicine
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LBL-13195
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SOURCES AND CONCENTRATIONS OF ORGANIC COMPOUNDS IN INDOOR ENVIRONMENTS

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This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

All occupied buildings have various sources of indoor air pollution. Humans (and their household pets) generate carbon dioxide, moisture, odors, and microbes simply through normal living processes. Other more important sources of indoor air pollution are combustion appliances (gas stoves, unvented space heaters), building materials (used in construction, furnishings, and insulation), and soil under and around houses. These sources release carbon monoxide (CO), nitrogen dioxide (NO_2), formaldehyde (HCHO) and other organics, particulates, and radon. Table I summarizes the sources and types of air pollutants commonly found indoors.

This paper will discuss the sources and concentrations of organic compounds in indoor environments. Formaldehyde, as an indoor pollutant, has been extensively investigated; however, recent work at Lawrence Berkeley Laboratory (LBL) and elsewhere is now focussed on a broad range of organic compounds, in addition to formaldehyde.

Formaldehyde (HCHO) is an inexpensive, high-volume chemical used throughout the world in a variety of products, mainly in urea, phenolic, melamine, and acetal resins. These resins are present in insulation materials, particleboard, plywood, textiles, adhesives, etc. that are used in large quantities by the building trades. Although particleboard and urea-formaldehyde foam insulation have received the most attention, some of the combustion processes mentioned above also release formaldehyde. The pungent and characteristic odor of formaldehyde can be detected by most humans at levels below 100 $\mu g/m^3$. Several studies reported in the literature indicate that concentrations in the range of 100 to 200 $\mu g/m^3$ may be sufficient to cause swelling of the mucous membranes, depending on individual sensitivity and environmental conditions (temperature, humidity, etc.). Burning of the eyes, weeping, and irritation of the upper respiratory passages can also result from exposure to relatively low concentrations. High concentrations (>> 1000 $\mu g/m^3$) may produce coughing, constriction in the chest, and a sense of pressure in the head. There is concern that formaldehyde may have serious long-term health effects. Several foreign countries and various states in the United States are moving rapidly to establish standards for formaldehyde concentrations in indoor air. The range of these proposed standards is 120 to 600 $\mu g/m^3$. A summary of formaldehyde measurements in various indoor environments is given in Table II.[1]

Formaldehyde and total aliphatic aldehydes (formaldehyde plus other aliphatic aldehydes) have been measured by LBL at several energy-efficient research houses at various geographic locations in the U.S. Figure 1 shows a histogram of frequency of occurrence of concentrations of formaldehyde and total aliphatic aldehydes measured at an energy-efficient house with an air exchange rate of 0.2 ach. Data taken at an energy-efficient house in Mission Viejo, California, are shown in Table III. As shown, when the house did not contain furniture, formaldehyde levels were below 120 $\mu g/m^3$; when furniture was added, formaldehyde levels rose to almost twice the 120 $\mu g/m^3$ level. A further increase was noted when the house was occupied, very likely because of such activities as cooking with gas. When occupants opened windows to increase ventilation, the formaldehyde levels dropped substantially.

In the past few years, office workers throughout the country have registered numerous complaints of "bad air". These complaints come most frequently from workers occupying new office buildings with hermetically sealed windows. Although various government agencies have investigated these problems, the etiological agent(s) has frequently remained unidentified.

One of the primary contributors to poor indoor air quality in office buildings may be organic contaminants, which have numerous indoor sources: building materials, cleaning products, tobacco smoking, furnishings, common consumer products, and building occupants themselves. To date, however, there has been relatively little research on this topic. In 1980, LBL began a comprehensive DOE-sponsored research program in collaboration with the Center for Disease Control (CDC), and the National Institute of Occupational Safety and Health (NIOSH) to characterize indoor air pollution in "complaint" office buildings.

The results of work in one of the office buildings[2] is summarized in Table IV. Only total hydrocarbons exceeded air quality standards; no other indoor pollutants, including formaldehyde, exceeded air quality standards. The average total hydrocarbon concentration was 1627 ± 26 $\mu g/m^3$ (2.5 ppm expressed as methane). The average indoor concentration can be compared to the average outdoor concentration of 210 ± 60 $\mu g/m^3$ (0.32 ppm). These hydrocarbon concentrations, especially the indoor values, are well in excess of the National Ambient Air Quality Standard of 160 $\mu g/m^3$ (0.24 ppm). It must be emphasized, however, that this standard was established on the basis of hydrocarbons acting as precursors for photochemical smog, and does not necessarily imply that hydrocarbons themselves are harmful.

The observation of high total hydrocarbon concentrations led us to investigate in depth the organic compounds in several office buildings. Figure 2 shows typical comparative gas chromatograms of equal size air samples taken simultaneously inside and outside an office building where complaints had been registered[3]. Organic contaminants are greater in number and concentration indoors than outdoors as indicated by the sizes and number of peaks. For a few samples, comparison of the peak areas with those of external standards indicated that the largest peaks corresponded to air concentrations of a few parts per billion.

Samples were analyzed by gas chromatography-mass spectrometry (GC-MS) to establish identities. Generally, the largest peaks fell into one of three classes of compounds, the largest being aliphatic hydrocarbons including straight-chain and derivatives of cyclohexane. These hydrocarbons are derived petroleum distillate-type solvents. The second largest class was alkylated aromatic hydrocarbons, predominately toluene but also including xylenes, trimethyl- and other substituted benzenes, and even methyl- and dimethylnapthalenes. These compounds are either solvents themselves or constituents of naphenic-type petroleum solvent mixtures. The third class observed was chlorinated hydrocarbons, predominately tetrachloroethylene, 1,1,1,-trichloroethane and trichloroethylene. Miscellaneous other compounds observed were ketones, aldehydes, and benzene. Table V lists those organic compounds found to be at least five times as great inside offices as outdoors, and notes, where applicable, the standards of exposure promulgated by the Occupational Safety and Health Administration (OSHA) for the workplace environment. In research now in progress, we are quantitatively determining the concentrations of these organic compounds by current state-of-the-art analytic procedures, which, although they provide only rough estimates, are indicating concentrations ranging from 1 to 100 ppb. These levels are well below existing limits established by OSHA for occupational exposure but may be excessive for the general public for whom limits are typically ten times lower.

While no single compound was present in high enough concentration to be singled out as a health hazard by existing OSHA criteria, the potential health hazard from the combined effects of the organic compounds found in these samples cannot be assessed at this time. The existing OSHA health criteria may be inadequate given that: (1) additive or synergistic effects are not adequately addressed; (2) the criteria are generally based on acute exposure studies whereas here the exposure is chronic; (3) the population at risk is more diverse including women and elderly workers; (4) annoyance from odorant effects is not considered.

A summary of organic compounds identified in indoor environments is given in Table VI. Several of the sources of organic contaminants in closed office spaces can be categorized as: (1) new building materials; (2) aged building materials; (3) wet-process photocopiers; (4) tobacco smoke; and (5) building maintenance products. Table VII summarizes the source characteristics and generation pattern for organic contaminants in a typical office space.[4] New building materials are a source of organic contaminants because they contain residual solvents and other compounds remaining after the process of manufacture. Qualitative GC-MS analysis of the headspace vapor standing over a variety of new building materials in an LBL study has revealed a great number of compounds -- predominately toluene and aliphatic hydrocarbons. Ketonic solvents were observed as well as speciality compounds such as butylated hydroxytoluene (BHT).

Implementation of control strategies for organic contaminants in indoor environments must consider the nature and generation pattern of each source. For new building materials further research may define an acceptable waiting period prior to occupancy or a period of high ventilation rates while they "dry out". Workday pollution from photocopiers and tobacco smoke may be reduced by increased ventilation, but not as efficiently as source removal itself. Episodic contaminant generation from building maintenance products can be reduced by increased ventilation; but a better strategy may be to offset product use from the workday period.

ACKNOWLEDGEMENT

This work was supported by the Assistant Secretary for Conservation and Renewable Energy, Office of Building Energy Research and Development, Building Systems Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48.

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Table I

SUMMARY OF SOURCES AND TYPES OF INDOOR AIR POLLUTANTS

SOURCES	POLLUTANT TYPES
OUTDOOR	
Stationary Sources	SO_2, CO, NO, NO_2, O_3, Hydrocarbons, Particulates
Motor Vehicles	CO, NO, NO_2, Lead, Particulates
Soil	Radon
INDOOR	
Building Construction Materials	
Concrete, stone	Radon and other radioactive elements
Particleboard	Formaldehyde
Insulation	Formaldehyde, Fiberglass
Fire Reatrdant	Asbestos
Adhesives	Organics
Paint	Organics, Lead, Mercury
Building Contents	
Heating and cooking combustion appliances	CO, NO, NO_2, Formaldehyde, Particulates
Copy machines	Organics
Water service; natural gas	Radon
Human Occupants	
Metabolic activity	H_2O, CO_2, NH_3, Organics, Odors
Biological activity	Microorganisms
Human Activities	
Tobacco smoke	CO, NO_2, HCN. Organics, Odors Particulates
Aerosol spray devices	Flurocarbons, Vinyl Chloride, CO_2, Odors
Cleaning and cooking products	Organics, Odors
Hobbies and crafts	Organics, Odors

Table II

Summary of Formaldehyde Measurements in Various Indoor Environments

Sampling Site	Concentration[a] (ppm)[b] Range	Mean
Two mobile homes in Pittsburg, Pa.	0.1-0.8[c]	0.36
Mobile homes registering complaints in state of Washington	0-1.77	0.1-0.44
Mobile homes registering complaints in Minnesota	0-3.0	0.4
Mobile homes registering complaints in Wisconsin	0.02-4.2	0.88
Public buildings and energy-efficient homes, occupied and unoccupied	0-0.021 [0-0.23][c]	------ ------

[a]Formaldehyde, unless otherwise indicated.

[b]1 ppm = 1200 $\mu g/m^3$.

[c]Total aliphatic aldehydes

Source: National Research Council, National Academy of Sciences.[1]

Table III

Indoor Formaldehyde Concentrations

in a New

Residential Building

Condition	Formaldyhyde ($\mu g/m^3$)
Unoccupied, without furniture[a]	80 ± 9%
Unoccupied, with furniture[a]	223 ± 7%
Occupied, day[a]	261 ± 10%
Occupied, night[b]	140 ± 31%
Outdoor Air	<20

[a]Air exchange rate ≃ 0.4

[b]Windows open part of time; air exchange rate significantly greater than 0.4 and variable.

Table IV

Summary of Average Indoor Air Quality Measurements in an Office Building and Air Quality Standards

	Office Building Air Quality		Air Quality Standards	
Contaminant	Concentration	Averaging Time	Concentration	Averaging Time
Carbon monoxide	4.6 mg/m^3 (4 ppm)	1 hr	40 mg/m^3 (35 ppm)[a]	1 hr
Carbon dioxide	1800 mg/m^3 (1000 ppm)	8-10 hrs	9,000 mg/m^3[b] (5,000)	8 hrs
Nitrogen dioxide	60 μg/m^3 (30 ppb)	1 week	100 μg/m^3 (50 ppb)[a]	1 yr
Hydrocarbons (non-methane)	1627 μg/m^3 (2.5 ppm)	30 minutes	160 μg/m^3[a] (0.24 ppm)	3 hours (6-9 am)
Formaldehyde	49 μg/m^3 (41 ppb)	6 hours	120-840 μg/m^3[c] (100-700 ppb)	maximum
Aliphatic aldehydes	108 μg/m^3 (90 ppb)	6 hours	No standard	
Particulates	31 μg/m^3	12 hours	75 μg/m^3[a] 260 μg/m^3[a]	1 yr 24 hrs
Lead	0.2 μg/m^3	12 hours	1.5 μg/m^3[d]	3 months
Sulfur (as SO_4)	2.5 μg/m^3	12 hours	25 μg/m^3[d]	24 hrs
Ariborne Microbes	179 CFP/m^3	20 minutes	No standard	

[a] U.S. EPA Ambient Air Quality Standard for outdoor air.

[b] State of California Air Quality Standard.

[c] range of recommended standards

[d] U.S. Occupational Safety and Health Administration (OSHA).

Table V

Organic Compounds Detected in Office Buildings

Organic Compound	OSHA Permissible Exposure Limit (ppm)
HYDROCARBONS	
n-hexane	500
n-heptane	500
n-octane	500
n-nonane	
n-undecane	
2-methylpentane	
3-methylpentane	
2,5-dimethylheptane	
methylcyclopentane	
ethylcyclohexane	
methylcyclohexane	500
pentamethylheptane	
AROMATICS	
benzene	1
xylenes	100
toluene	200
HALOGENATED HYDROCARBONS	
tricloroethane	350
tricloroethylene	100
tetrachloroethylene	100
MISCELLANEOUS	
hexanal	
methylethylketone	200

Table VI

Organic Compounds in Indoor Environments

Compounds	Health Effects	Sources and/or Uses
Formaldehyde and other Aldehydes	Eye and respiratory irritation; may have more serious long term health effects	Out-gassing from building materials -- particle board, plywood and urea-formaldehyde insulation foam; also generated by cooking and smoking
C_n Alkanes N = 5 ∿ 16	Narcotic at high concentrations; moderately irritating	Gasoline, mineral spirits, solvents, etc.
C_n Alkenes N = 5 ∿ 16	Similar to that of alkanes	Similar to that of alkanes
Benzene	Respiratory irritation; recognized carcinogen	Plastic and rubber solvents; from cigarette smoking; used in paints and varnishes, including putty, filler, stains and finishes
Xylene	Narcotic; irritating; high concentrations may cause injury to heart, liver, kidney, and nervous system	Used as solvent for resins, enamels, etc.; also used in non-lead automobile fuels and in manufacture of pesticides, dyes, pharmaceuticals
Toluene	Narcotic; may cause anemia	Solvents; by-product of organic compounds used in several household products
Styrene	Narcotic; can cause headache, fatigue, stupor, depression, incoordination and possible eye injury	Widely used in manufacture of plastics, synthetic rubber and resins
1,1,1-Trichloroethane	Subject of OSHA carcinogenesis inquiry	Aerosol propellant, pesticide, cleaning solvents
Trichloroethylene	Animal carcinogen; subject of OSHA carcinogenesis inquiry	Oil and wax solvents, cleaning compounds, vapour degreasing products, dry cleaning operations; also used as an anaesthetic
Ethyl Benzene	Highly irritating to eyes, etc.	Solvents; used in Styrene related products
Chloro Benzenes	Strong narcotic; possible lung, liver, and kidney damage	Used in production of paint, varnish, pesticides, and various organic solvents
Polychlorinated Biphenyls (PCB's)	Sustected carcinogens	Used in various electrical components; may appear in waste oil supplies and in plastic and paper products in which PCB's are used as plasticizers
Pesticides	Suspected carcinogens	Used for insect control

Table VII

Characteristics of Sources of Organic Contaminants in a Typical Office Space[a]

Source	Nominal Emission Rate (gms/hr-office)	Generation Pattern	Major Known Types of Organic Contaminants Emitted
new building materials	10[b]	continuous	aliphatic hydrocarbons aromatic hydrocarbons ketones esters (formaldehyde) many miscellaneous
aged building materials	low	continuous	formaldehyde
wet-process photocopiers	25[c]	workday	aliphatic hydrocarbons
smokers	1[d] 0.5[d] 0.25[d] (4)[d]	workday	formaldehyde acrolein nicotine (total particulates) many miscellaneous
building maintenance products	100[e]	episodic	aliphatic hydrocarbons aromatic hydrocarbons formaldehyde amines chlorinated hydrocarbons many miscellaneous

[a]The typical office being considered has dimensions of 100 x 100 x 10 ft^3 with an occupancy of 40 workers.

[b]Emission rate calculated assuming 1.0 mg/ft^2 for wall-to-wall carpeting after several months.

[c]Emission rate calculated assuming one wet-process photocopier using 1000 gms of fluid per week.

[d]Calculated from data in Weber et al. (1977)

[e]Emission rate calculated assuming 100 gms of product (floor detergent or dusting fluid) applied in one hour.

Figure Captions

Figure 1 Histogram of indoor and outdoor formaldehyde concentrations at an energy efficient house

Figure 2 Comparative gas chromatograms of indoor and outdoor air at an office site

INDOOR/OUTDOOR FORMALDEHYDE AND ALDEHYDE CONCENTRATIONS

Energy Research House
Carrol County, Maryland

March/April, 1979

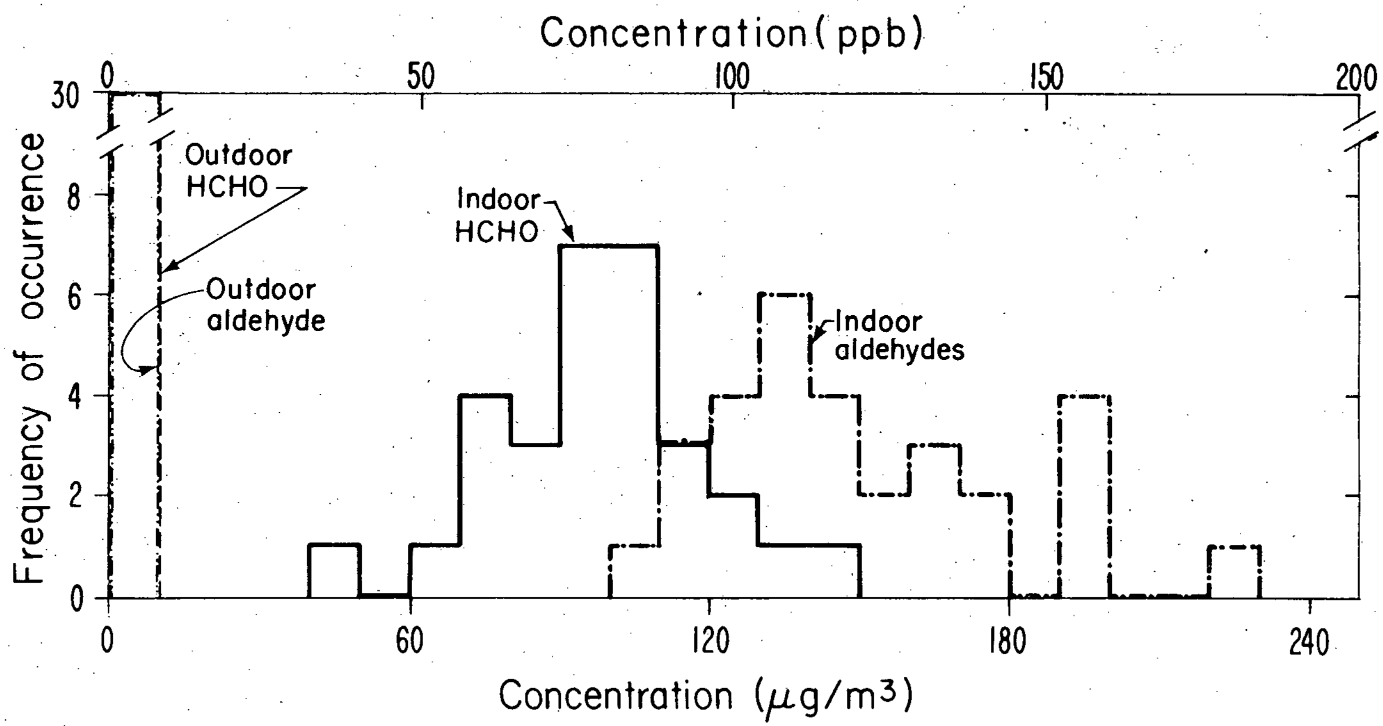

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Figure 1

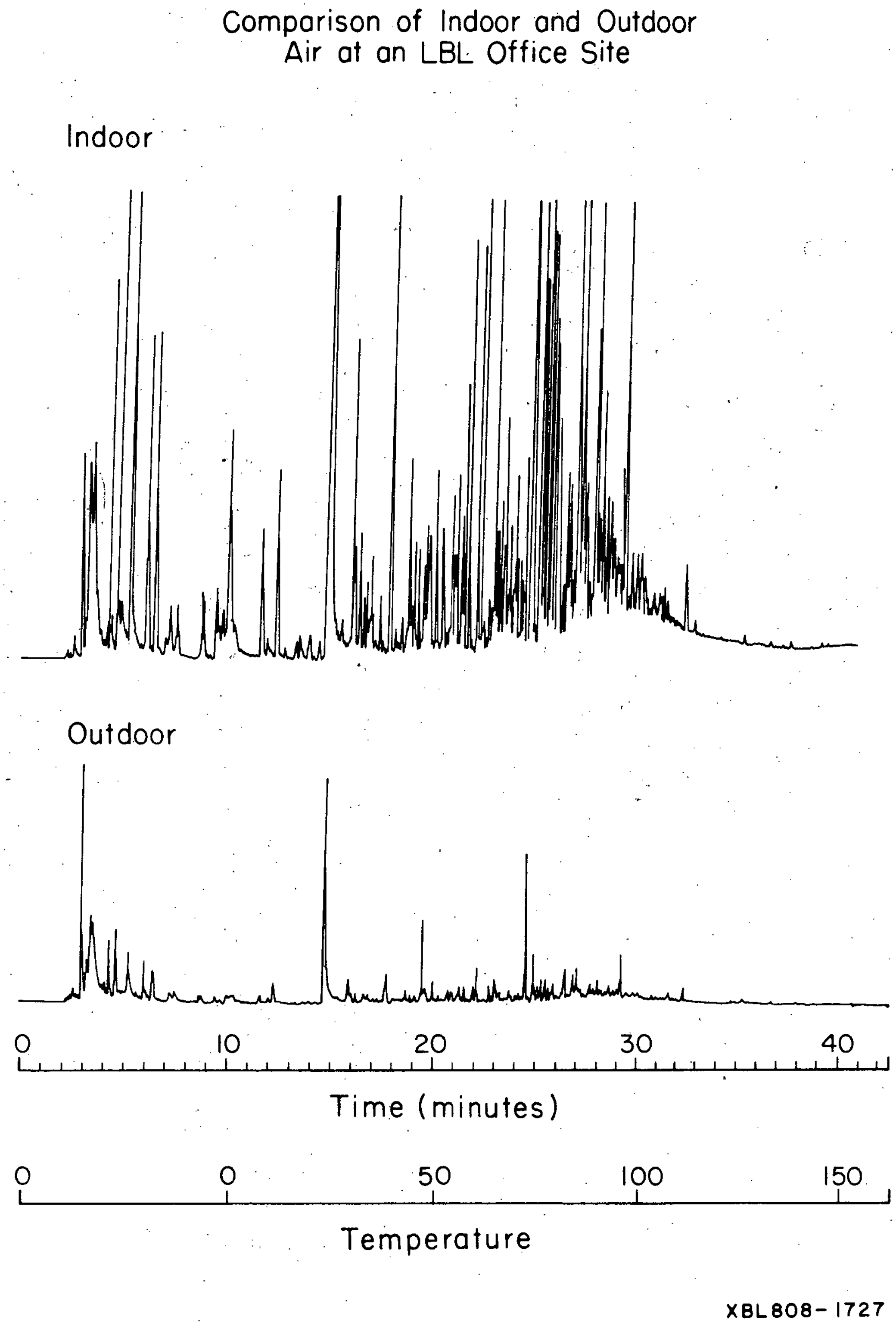
Comparison of Indoor and Outdoor
Air at an LBL Office Site
Indoor
Outdoor
0
10
20
30
40
Time (minutes)
0
0
50
100
150
Temperature
XBL808-1727

Figure 2

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