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EARTHWATCH

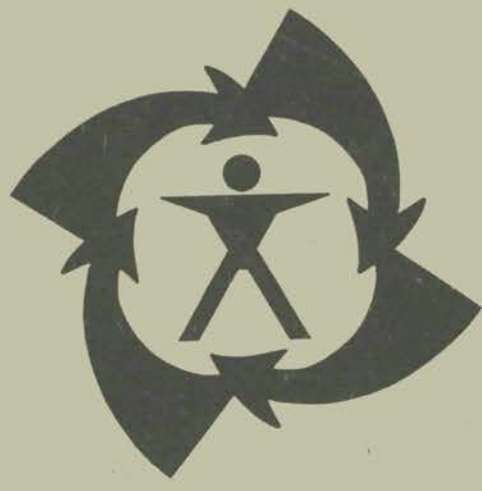
GLOBAL ENVIRONMENT MONITORING SYSTEM

EXPOSURE MONITORING OF NITROGEN DIOXIDE

An international pilot study within the WHO/UNEP Human Exposure Assessment Location (HEAL) Programme

Technical report prepared by

Hidetsuru Matsushita and Kiyoshi Tanabe
National Institute for Public Health
Tokyo, Japan



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A. PREFACE

Human exposure to a pollutant occurs when a person comes in contact with that pollutant through the air they breathe, the water and food they consume or through skin absorption. Knowing the degree of exposure is necessary to study the effects on health and to devise appropriate control strategies. In recent years new techniques have been developed directly to monitor such exposures, assess them accurately and evaluate contributions from the individual sources. An increasing number of countries are using this technology in a multidisciplinary approach to multi-media human exposure and assessment.

To assist the further development and application of the technique currently being applied in several countries, the WHO and UNEP through the GEMS are co-operating in a co-ordinated effort - Human Exposure Assessment Locations Studies (HEALs). This programme aims to improve exposure monitoring and assessment internationally leading to better protection of human health. Direct measurements of human exposure using established methodology, coupled with human activity patterns and social and cultural conditions provide the basic data enabling exposures in a given population to be determined. Accurate risk assessment studies can then be undertaken and cost effective strategies developed if require to further protect human health. of the technique currently being applied in several countries, the WHO and UNEP through the GEMS are co-operating in a co-ordinated effort - Human Exposure Assessment Locations Studies (HEALs). This programme aims to improve exposure monitoring and assessment internationally leading to better protection of human health. Direct measurements of human exposure using established methodology, coupled with human activity patterns and social and cultural conditions provide the basic data enabling exposures in a given population to be determined. Accurate risk assessment studies can then be undertaken and cost effective strategies developed if require to further protect human health.

For each study in the HEAL programme, one of the participating institutions in a particular country is designated as a Technical Co-ordinating Centre (TCC). The Centre is responsible for monitoring protocols, design and implementation of quality control programmes and other technical support to help participants implement the project. Each country has, however, a major responsibility for conducting their own study.

In the pilot phase seven countries were involved: Brazil, China, India, Japan, Sweden, U.S.A. and Yugoslavia, and three groups of chemicals were monitored by some of them, namely the heavy metals Pb and Cd, the organic chemicals hexachlorobenzene (HCB) and dichlorodiphenyltrichloroethane (DDT) and the gaseous substance nitrogen dioxide (NO_2). The substances were chosen for they may pose health risks and the participants were interested in determining exposure to such substances.

Reports of the three pilot phase studies are published separately. This report presents the results from the monitoring of nitrogen dioxide co-ordinated by a TCC in Japan as described in the section Institutions and Investigators. Laboratories in Japan, China, Sweden, Yugoslavia, India and the U.S.A. participated in the project. The results are presented as three sections in the report as some differences in experimental protocols and study design occurred.

Briefly, 10-15 non-smoking women were chosen from institute and staff in Japan, China, Sweden and Yugoslavia (Section I) while 17 non-smoking men and women were selected at random but living in the vicinity of the monitoring site in India (Section II) and the pilot phase undertaken approximately one year later. Nine non-smoking women were chosen by a random selection scheme in the USA study (Section III) as part of a larger survey. Other study design differences and protocol procedures are described in the relevant sections. The pilot project has successfully shown that human exposure monitoring can be undertaken internationally and draws attention to the limitations encountered in conventional ambient monitoring approaches being conducted at locations around the world.

B. SUMMARY

A pilot project has been undertaken to establish and validate a methodology for measuring in a standardized way personal exposures of volunteer subjects to NO_2 using badge type monitors in China, India, Japan, Sweden, Yugoslavia and the USA. Simultaneous measurements of indoor and outdoor NO_2 at the subject's home were obtained for comparison purposes.

The WHO has established for NO_2 a guideline value for 24-hour average exposure not to exceed 150 ug/m3 which is equivalent to 80 ppb. Mean personal exposures of the volunteer subjects ranged from 11 to 41 ppb with a range of 24-hour individual values of 8 to 88 ppb. These exposures were primarily influenced by fuel source, length of cooking time and time spent in the kitchen. Based on the limited pilot phase data, exposures were highest in Yokohama and lowest in Stockholm.

At all sites a significant relationship was found between the monitored personal exposures to NO_2 and the estimated exposures based on recorded times spent in various micro-environments indoors and outdoors.

In general, concentrations of NO_2 indoors were lower than outdoors when no internal sources of NO_2 were present and higher than outdoors when internal sources of NO_2 were present.

Despite the differences in geography, economic conditions and cultural norms, the pilot phase results have shown that the HEAL NO_2 methodology is a valid tool for carrying out comparable exposure studies internationally.

C. INSTITUTIONS AND INVESTIGATORS

The HEAL Pilot Project on Exposure Monitoring of NO_2 was co-ordinated by a Technical Co-ordinating Centre (TCC) in Japan in close collaboration with national institutions participating in the HEAL project on NO_2 monitoring. The TCC was organized as a committee with representatives of the National Institute of Public Health (Dr Yasutaka Osada, Chairman; Dr Masahiko Fujita, Secretary; Dr Hidetsuru Matsushita, Dr Yasumoto Magara, Dr Kiyoshi Tanabe, Dr Hiroyuki Nakazawa, Dr Shoichi Kunikane) and the National Institute of Hygienic Sciences (Dr Yukio Saito).

Following national institutions participated in both an analytical training component and in an exposure monitoring study.

CHINA

Institute of Environmental Health Monitoring, Chinese Academy of Preventive Medicine, Beijing; Beijing Sanitary and Anti-epidemic Station, Beijing
Principal investigator: Dr Chen Changjie

INDIA

National Institute of Occupational Health, Ahmedabad; Air Quality Monitoring and Research Laboratory, Bombay
Principal investigator: Dr S. K. Kashyap
Co-investigators: Prof. S. R. Kamat,
Mrs J. M. Deshpande, Dr D. J. Parikh
Responsible analysts:

AQM and RL, Bombay	- Mrs Sunita Naik
	Miss Devyani Parekh
	Mr Rajendra Karnik
NIOH Ahmedabad	- Mr C. V. Raiyani
	Dr S. P. Upadhyaya

JAPAN

National Institute of Public Health, Tokyo
Principal investigators: Dr Hidetsuru Matsushita,
Dr Kiyoshi Tanabe
Responsible analysts: Dr Taroh Kawamura,
Mr Hiroshi Kikawa, Yokohama City Institute of Health, Yokohama

SWEDEN

Karolinska Institutet, Institute of Environmental Medicine
Principal investigators: Dr Marie Vahter,
Dr Gunnar Bylin
Responsible analyst: Ms Marika Bergland

U.S.A.

U.S. Environmental Protection Agency, Washington D.C.;
Integrated Environmental Services, Irvine, California;
Environmental Research Centre, University of Nevada at
Las Vegas, Las Vegas, Nevada
Principal investigators: Steve Colome,
Michael J. Dellarco
Responsible analyst: Steve Colome, Dave McNelis

YUGOSLAVIA

Institute for Medical Research and Occupational Health,
University of Zagreb, Zagreb
Principal investigator: Dr Mirka Fugás
Responsible analyst: Ranka Paukovic

UNEP/WHO SECRETARIAT

Mr Guntis Ozolins, WHO, Geneva
Dr Henk W. de Koning, WHO, Geneva
Dr David Mage, WHO, Geneva
Dr Michael Gwynne, UNEP, Nairobi
Dr Veerle Vandeweerd, UNEP, Nairobi
Dr. Peter J. Peterson, UNEP/MARC, London

D. INTRODUCTION

Background and objectives

The UNEP/WHO Human Exposure Assessment Locations (HEAL) project was developed as part of the WHO Health-Related Monitoring Programme, which was started in 1973 and originally comprised urban air monitoring, water quality monitoring and food contamination monitoring. These three monitoring projects form part of the Global Environment Monitoring System (GEMS).

Between 1970 and 1980 several reports showing that data from outdoor air monitoring stations do not reflect the total human exposure appeared (e.g. Yocom et. al., 1971; Ott and Mage, 1975; Fugas, 1976; Cortese and Spengler, 1976). In 1977 a Government Designated Expert Group recommended that more attention should be given to human exposure assessment (WHO, 1977). Two pilot projects were developed, one dealing with health-related air pollution monitoring (WHO, 1982 a, b, c; WHO, 1985) and the other with biological monitoring of lead, cadmium and certain organochlorine compounds (Vahter, 1982; Friberg and Vahter, 1983; Slorach and Vaz, 1983; Bruaux and Svartengren, 1985). In 1981 a consultation on health-related monitoring (WHO, 1981) reviewed the ongoing activities and concluded that source-oriented monitoring was not providing adequate information to estimate human exposure and that there was a need to develop and agree on acceptable methodology to determine exposure to environmental pollutants. Therefore, ongoing exposure-oriented monitoring activities within the UNEP/WHO Human Exposure Assessment Locations (HEAL) project were initiated (WHO, 1982d; WHO 1983).

The main objectives of the HEAL project are (UNEP/WHO, 1985):

- to improve, field test, harmonize and demonstrate methods for the integrated monitoring and assessment of human exposure to environmental pollutants;
- to promote the assessment of human exposures to pollutants as a basis for the development of environmental control strategies for the protection of public health;
- to provide an overview of existing exposures of selected populations to pollutants and, if possible, to observe trends in this regard;
- to improve national capabilities for environmental monitoring and human exposure assessment, particularly in the developing countries.

Initially, exposure to Pb, Cd, DDT, HCB and NO_2 has been studied. Technical Co-ordinating Centres (TCCs) have been appointed for various types of pollutants (UNEP/WHO, 1986a). During this phase, seven countries are participating in the HEAL project: Brazil, China, India, Japan, Sweden, USA and Yugoslavia.

Environmental and exposure monitoring

For many commonly occurring environmental pollutants there is only a small difference between "normal" exposure levels and the exposure level at which the first signs of adverse health effects occur. Therefore, it is important to be able to detect small differences in exposure between different populations or groups of subjects, or small changes in exposure within a population over time. This requires sensitive methods for the detection of systematic errors in the analytical results. An analytical quality assurance programme was therefore developed to ensure the reliability and comparability of the NO_2 monitoring data. The reliability of analysis of each participating institution was confirmed through the analysis of quality control samples during a training phase. This analytical quality assurance was carried out in China, India, Japan, Sweden and Yugoslavia. The USA also participated in part of the analytical training.

The main purpose of the pilot phase NO_2 study was to develop and evaluate NO_2 exposure monitoring methodology and also to obtain preliminary information about NO_2 exposure levels at different HEAL sites. The pilot study was carried out with a small group of selected individuals at each HEAL sites except for the USA where individuals were chosen by a random selection procedure. The design of the pilot study was decided on at a meeting with representatives of the participating institutions in Tokyo in March, 1987 (GEMS 1987). It involved measurements of NO_2 personal exposure via air and NO_2 concentrations indoors and outdoors at the home and workplace for each subject during seven consecutive days. It also involved environmental monitoring at fixed environmental monitoring stations in high and low exposure areas in each HEAL site during the period of exposure monitoring.

Pilot NO_2 studies have been carried out in China, India, Japan, Sweden, U.S.A. and Yugoslavia, where the HEAL sites were Beijing, Bombay, Yokohama, Stockholm, Los Angeles and Zagreb respectively. The monitoring data in China, Japan, Sweden and Yugoslavia have been sent to the TCC, evaluated and appear in one report. The data for U.S.A. and the data for India, also evaluated by the TCC, appear as separate reports.

EXPOSURE MONITORING OF NITROGEN DIOXIDE AT LOCATIONS IN CHINA, JAPAN, SWEDEN AND YUGOSLAVIA

An international pilot study within the
WHO/UNEP Human Exposure Assessment Locations (HEAL) Programme

SECTION I

Technical report edited by

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2. MATERIALS AND METHODS

2.1. Exposure monitoring

It is important to know the level of human exposure to NO_2 in order to study the health effects dose-effect relationships and to devise appropriate pollution control strategies.

High temperature combustion is the main emission source of NO/NO_2 in air and NO is converted to NO_2 by photo-chemical oxidation in the environment. Unvented gas/kerosene cooling appliances and space heaters are major indoor emission sources while automobiles as well as installations burning fossil fuels are major outdoor emission sources. Emmissions from the outdoor sources are generally increasing throughout the world.

2.1.1. Selection of subjects

Since the main purposes of the pilot study were to establish the NO_2 exposure monitoring methodology and to obtain preliminary information on NO_2 exposure levels at HEAL sites, selection of subjects has been carried out according to the following guidelines agreed in the Tokyo meeting:

- Number of subjects should be at least 10-15;
- Subjects should be selected from healthy non-smoking women living in a high exposure area and in a low exposure area where environmental monitoring will be performed. It is preferable to select subjects of 25-55 years of age, some of whom live in houses with indoor NO_2 emission sources and some without the sources in well ventilated premises.
- Subjects should be reliable and able to perform the sampling and to complete questionnaires correctly. It was preferable to select the subjects from women employed at and/or related to the institutions participating in the HEAL project.

A description of subjects selected at the HEAL sites is given below:

Beijing: 15 non-smoking women, 31-51 years of age, working in Beijing Sanitary and Anti-epidemic Station (urban area). All of them were living in the urban area. Sampling period 1988/3/22-29.

Stockholm: 15 non-smoking women, 27-46 years of age, working in Karolinska Institute area (urban area). Three of them were living in the urban area, nine in suburban and three in rural area. Sampling period 1988/2/29 - 3/17.

Yokohama: 10 non-smoking women, 35-54 years of age. Six of them were housewives (two of them had part-time jobs), three were working in Yokohama City Institute of Health (urban area), one was working in a school (urban area). All subjects were living in the urban area. Sampling period 1988/3/11-18.

Zagreb: 16 non-smoking women, 22-65 years of age, working in Institute for Medical Research and Occupational Health (suburban area). Six of them were living in central city, four in suburban and six in the rural area.

Table 1. The monitoring was carried out in the heating season, when home windows were not so frequently opened. Outdoor temperatures at each HEAL site during the monitoring were in the range -5.6 - 1.5°C in Stockholm, 4.2 - 15.1°C in Yokohama in daily average of hourly temperatures, and -3 - 15°C in Beijing, -2 - 22°C in Zagreb in hourly temperatures.

2.1.2 Sampling strategy

Sampling for each subject was carried out for seven consecutive days. The sampling in Stockholm and Zagreb was implemented sequentially for small sub-groups of subjects. Therefore, the total sampling period in these cities exceeded one week. The sampling in Beijing and Yokohama was carried out in one week for all subjects.

Table 1. Sampling scheme for exposure monitoring of NO_2

Sampling	Days of sampling 1	2	3	4	5	6	7
Personal samples[a]	--	--	--	--	--	--	--
Indoor samples[b*]	----------			----------			
Outdoor samples[c]	----------			----------			

[a] 24-hour period samples.

[b] Samples collected in the kitchen, living room, bedroom and workplace.

[c] Samples collected outside the home and workplace.

[*] Measurement of peak NO_2 exposure levels in a few homes was optionally recommended.

2.1.3. Collection of personal information

For the interpretation of monitoring data, the following personal information was collected from each subject by using questionnaires:

1. Information on NO_2 emission sources inside and outside homes and workplaces.

2. Time schedule (activity record) during the monitoring.

2.1.4. Analysis of samples

All samples collected were analyzed by the monitoring laboratory for each HEAL. Detailed analytical procedures for badge samples, which are given in Appendix 1, were prepared by the TCC. Analysis of the samples by each monitoring laboratory was carried out after the confirmation of satisfactory analytical performance of the laboratory by the TCC in the training phase, where two-step analytical quality assurance was implemented (see section 2.3.). Analytical quality assurance was also implemented using NO_2 gas exposed badge samplers during the pilot exposure monitoring study.

2.2. Environmental monitoring

The main purposes of the environmental NO_2 monitoring in the HEAL project were to assess long-term trends of NO_2 concentration in the atmosphere on a regional and international scale and to provide a first-level assessment of human exposure to NO_2 on an international basis. Concentration of NO_2 in ambient air, especially in the non-heating season, largely affects NO_2 concentration indoors. Therefore, the data obtained in the environmental monitoring may be useful for the approximate estimation of NO_2 exposure level of the general population.

In the pilot phase, however, the main effort was directed to exposure monitoring. For this reason, environmental NO_2 data were collected at fixed environmental monitoring stations in high and low pollution areas at each HEAL site during the period of exposure monitoring. The environmental data were evaluated through comparison with the outdoor NO_2 data obtained at homes and workplaces in the exposure monitoring.

2.3. Quality assurance

2.3.1. Quality assurance programme

To ensure the reliability and comparability of the monitoring data, an analytical quality assurance programme was implemented. Two analytical quality assurances were carried out in the training phase. They were (1) analytical quality assurance using $NaNO_2$ standard solutions (AQA1) and (2) analytical quality assurance using NO_2 gas exposed badge samples (AQA2). Analysis of actual samples collected in the pilot monitoring phase commenced after the achievement of satisfactory analytical performance in the training phase.

Analytical quality assurance using NO_2 gas exposed badge samples (AQA3) was also carried out during the pilot exposure monitoring to evaluate reliability and comparability of the monitoring data.

Duplicate samples were collected and analyzed in the exposure monitoring for quality assurance throughout all procedures from sampling to analysis.

All analytical quality assurances were designed with due consideration of the principles recommended by UNEP/WHO (1986b) which require the analysis of 4-6 quality control (QC) samples. However, analytical performance evaluation was carried out through modified procedures as mentioned in section 2.3.3.

2.3.2. Quality control samples for analytical quality assurance

Sodium nitrite standard solutions were used as QC samples in AQA1. QC samples for AQA1 consisted of:

1. Two internal QC samples (IQC-1 and IQC-2); $NaNO_2$ concentrations were known to participating institutions.

2. Two series of five external QC samples (EQC I-1, 2, ... 5, EQC II-1, 2, ... 5); $NaNO_2$ concentrations were unknown to participating institutions. The concentrations were in the range 8.21 - 171µg/ml (see Table 3). Amounts of $NaNO_2$ in 0.25ml of EQC samples correspond to the amounts of NO_2 collected by 24 hour exposure of badge sampler to 6 - 126 ppb of NO_2.

Analytical procedures for these QC samples were specified to be similar to those for the analysis of NO_2 badge samples. That is, AQA1 was designed to check the reliability of colourimetry which was used for the analysis of NO_2 badge samples. Details of the analysis of QC samples in AQA1 are given in Appendix 2. (U.S.A. analyzed the QC samples by a Technicon Auto Analyzer II after 100 times dilution.)

NO_2 gas exposed badge samples were used as the QC samples in AQA2 and AQA3. The QC samples consisted of*:

1. Two sets of four same internal QC samples (IQC I-1, 2, ... 4, IQC II-1, 2, ... 4); Amounts of NO_2 collected by the samplers were known to participating institutions.

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* A part of the QC samples were used in AQA2, and the remaining QC samples were used in AQA3, e.g., EQC I-1, 2, .. 5 were used in AQA2, and EQC II-1, 2, .. 5, EQC III-1, 2, .. 5 were used in AQA3.

2. Three series of 5 EQC samples (EQC I-1, 2, ... 5, EQC II-1, 2, ... 5, EQC III-1, 2, ... 5); Amounts of NO_2 collected by the samplers were unknown to participating institutions. The amounts of NO_2 were in the range 37.2 - 421 nmole (see Table 5), which correspond to the amounts of NO_2 collected by 24-hour exposure of badge sampler to 8 - 86 ppb of NO_2.

Details of the analysis of badge samples is given in Appendix 1.

2.3.3. Evaluation of analytical performance

Maximum allowable deviation (MAD) intervals were set to be $y=x \pm (1.5\mu g/ml + 0.1x)$ for AQA1 and $y=x \pm (10nmole + 0.1x)$ for AQA2 considering the data quality expected in the pilot monitoring (y = reported value, x = target value). Errors of 1.5µg/ml and 10 nmole correspond to 1 ppb and 2 ppb, respectively, in the measurement of NO_2 concentration in air by 24-hour exposure of badge sampler.

Analytical performance of the participating institution was evaluated by the following two criteria:

1. All reported values should be in the MAD interval.

2. Calculated regression line (target value vs. reported value) should be in the MAD interval.

Evaluation using the regression line was carried out statistically as shown in Figure 1. That is, the evaluation was done through checking whether the 90% confidence interval of the regression line is in the MAD interval or not throughout the tested range.

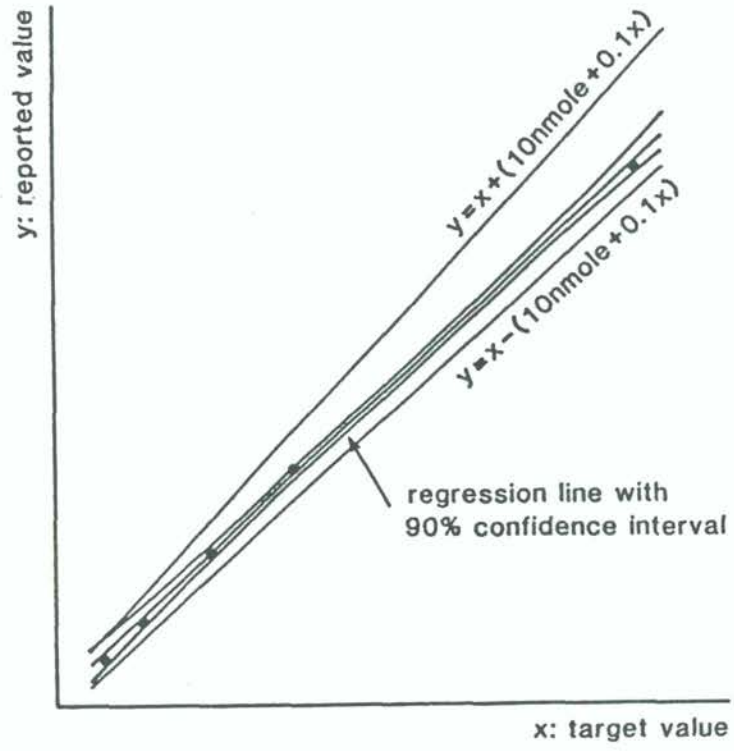
Regression line with 90% confidence interval is expressed by the equation

$$y = A + Bx \pm t_{n-2}\,(0.1)\sqrt{\left(\frac{SSy-(SSxy)^2/SSx}{n-2}\right)\left(\frac{1}{n}+\frac{(x-\bar{x})^2}{SSx}\right)}$$

where, $t_{n-2}\,(0.1)$ is t-value at p = 0.1 and degree of freedom = n-2, SSx is $\Sigma x^2-(\Sigma x)^2/n$, SSy is $\Sigma y_2-(\Sigma y)^2/n$ and SSxy is $\Sigma xy-\Sigma x\Sigma y/n$.

It was expected that the sensitivity and precision of the analytical methods would be good enough to meet these criteria at the settled MAD intervals.

Figure 1. Maximum allowable deviation interval test for analytical quality assurance. MAD lines in this figure are those for AQA2 and AQA3.



The acceptance interval method for statistical evaluation, which was recommended by UNEP/WHO (1986b), was not used in this study because of the following reasons:

1. Empirical normal standard error of the regression line* is necessary to set the acceptance interval. However, the empirical normal standard errors for the present analytical quality assurances were not known because of shortage of experimental data.

2. The acceptance interval method is based on the assumption that all regression lines have the same standard error equal to the empirical normal value. Even if the regression line was rejected when it had a standard error larger than twice the normal value, the standard error of the regression line could be 2 to 3 times different among the institutions (see Tables 3, 4, 6, and 8).

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* standard error of a regression line = $\sqrt{\Sigma d_i^2/(n-2)}$, where, d_i = difference between reported y value and estimated y value by the regression line, n = number of data.

3. RESULTS AND DISCUSSIONS

3.1 Analytical training

3.1.1. Analytical quality assurance using $NaNO_2$ standard solutions (AQA1)

The eight institutions in six countries (refer Institutions and Investigators) participated in the AQA1 in training phase.

Reported values for AQA1 were shown in Table 2. The standard deviation of reported values was larger at higher concentrations as well as at lower concentrations. The large coefficient of variation at higher concentrations may be due to non-linearity of colourimetry in the high optical density region.

Table 3 shows the results of MAD interval test on the regression line (2nd criterion). For institutions B, C, F and G, analytical performance was unsatisfactory since 90% confidence interval of the regression line was out of the interval at the lower end of the tested concentration range.

Table 4 shows the results of MAD interval test on the regression line, where reported values at the two top concentrations were excluded from the test considering the non-linearity of colourimetry at high optical density. In this case, (1) regression lines were closer to y=x than those in Table 3, and (2) standard errors of the regression lines were smaller than those in Table 3 (This means 90% confidence interval of the regression line became narrower). Only one institution's regression line (90% confidence interval) was slightly out of the MAD interval at the lowest concentration tested.

According to the principle of linear regression, the MAD interval test on the regression line will be applied correctly only when the data have even random error. However, errors in AQA1 were largely different by sample concentration as can be seen in Table 2. In the case of Table 3, large errors at higher concentrations, which may be due to non-linearity of colourimetry, largely affected the regression line and standard error. Even in the case of Table 4, the 90% confidence interval of the regression line would be overestimated at the lower end of the tested range. Given due consideration of these facts, the TCC decided that analytical performance in colourimetry was satisfactory for all 8 institutions. Furthermore, based on the results of AQA1, the upper limit of optical density for colourimetry was reduced from 1.0 to 0.6 absorbence in the later studies.

3.1.2. Analytical quality assurance using NO_2 gas exposed badge samples (AQA2)

Five institutions in five countries participated in AQA2 in the training phase. They were the Beijing Sanitary and Anti-epidemic Station, Beijing; National Institute of Occupational Health, Ahmedabad; Yokohama City Institute of Health, Yokohama; Karolinska Institute, Institute of Environmental Medicine and Department of Environmental Hygiene, Stockholm; Institute for Medical Research and Occupational Health, Zagreb.

Reported values and results of the MAD interval test on regression lines are shown in Tables 5 and 6, respectively. Asterisks on the reported value in Table 5 and that on 90% confidence interval of the regression line in Table 6 indicate that those data are out of $y=x \pm (10\text{nmole} + 0.1x)$ interval.

As can be seen in Table 5, the standard deviation of reported values was large for badge samples which absorbed large amounts of NO_2. On the other hand, the co-efficient of variation was large for samples which absorbed small amounts of NO_2.

In the first run of AQA2, analytical performance of institutions C and E were satisfactory. For institutions A, B, and D, the performances were judged to be unsatisfactory by both criteria. There was a common trend among three institutions, i.e., slope of the regression line was smaller than 1.

After checking analytical procedures, the temperature for extraction and colour-development was specified to be 25-30 °C, and the 2nd run of AQA2 was carried out in institutions A, B and D as a part of the training phase. The results are also given in Tables 5 and 6 (A2, B2 and D2).

Table 2. Reported values for analytical quality assurance using $NaNO_2$ standard solutions.

Target Value	Reported Value µg/ml A	B	C	D	E	F	G	H	Average	S.D.	cv(%)	Maximum	Minimum	n
171	181.9 /177.2	177.1	165.76	173.50	184.31	159.06 /158.19	165.4	170	171.24	9.09	5.3	184.31	158.19	10
149	158.5 /154.0	144.9	146.06	153.40	156.21	128.57*/129.97*	153.5	148	151.82	4.92	3.2	158.5	144.9	8
94.1	96.8 / 95.4	93.97	93.60	96.60	95.59	94.77 / 96.34	97.2	93.5	95.38	1.37	1.4	97.2	93.5	10
82.1	88.0 / 84.8	82.60	81.82	83.06	83.33	82.40 / 82.23	86.0	82.0	83.62	2.03	2.4	88.0	81.82	10
44.5	44.9 / 45.3	46.15	45.59	47.37	46.73	44.60 / 44.94	49.3	44.0	45.89	1.57	3.4	49.3	44.0	10
38.8	40.16 / 39.22	40.23	42.35	41.43	41.50	39.72 / 40.07	42.8	39.0	40.65	1.30	3.2	42.8	39.0	10
20.5	21.5 / 21.3	21.75	21.59	22.35	21.73	21.60 / 21.43	21.5	20.3	21.51	0.51	2.4	22.35	20.3	10
17.9	18.3 / 19.0	18.73	19.54	18.87	19.12	18.64 / 18.64	17.4	17.8	18.60	0.63	3.4	19.54	17.4	10
9.41	10.30 / 9.54	9.425	9.98	10.25	10.29	9.76 / 9.93	8.6	9.2	9.73	0.55	5.6	10.30	8.6	10
8.21	8.0 / 8.6	8.890	8.85	9.02	8.82	8.36 / 8.71	6.3	8.0	8.36	0.81	9.7	9.02	6.3	10

A, B, C, D, E, F, G and H: institution
* out of $x \pm (1.5 + 0.1x)$ interval, excluded from statistical calculations in this table

Table 3. Results of maximum allowable deviation (MAD) interval test on analytical quality assurance for eight institutions using $NaNO_2$ standard solutions.

Institution	n	Regression Line	Interval of slope**	S.E. (μg/ml)	Interval of y at x=8.21	Estimate (μg/ml)** at x=171
A	20	y = -0.544 μg/ml +1.048x	1.037 - 1.059	1.55	7.22 - 8.91	177.4 - 180.0
B	10	y = 0.619 +1.002x	0.974 - 1.031	2.69	6.62 - 11.08*	168.6 - 175.4
C	10	y = 2.304 +0.962x	0.948 - 0.976	1.30	9.12 - 11.28*	165.2 - 168.5
D	10	y = 1.229 +1.012x	1.002 - 1.021	0.893	8.79 - 10.28	173.1 - 175.4
E	10	y = -0.479 +1.057x	1.033 - 1.081	2.24	6.34 - 10.06	177.4 - 183.1
F	20	y = 3.76 +0.898x	0.868 - 0.928	4.36	8.75 - 13.51*	153.7 - 161.0
	(16	y = 0.521 +1.004x	0.996 - 1.012	0.540	8.42 - 9.10	94.51 - 95.48)***
G	10	y = 1.500 +0.995x	0.957 - 1.033	3.59	6.69 - 12.65*	167.1 - 176.2
H	10	y = 0.006 +0.993x	0.990 - 0.996	0.276	7.93 - 8.39	169.5 - 170.2

* out of MAD interval (MAD intervals are 5.89 - 10.53 at x=8.21 and 152.4 - 189.6 at x=171)

** 90% confidence interval

*** Four data which had apparent bias error were excluded from evaluation. MAD interval is 83.2 - 105.0 at x=94.1.

Table 4. Results of maximum allowable deviation (MAD) interval test on analytical quality assurance using $NaNO_2$ standard solutions (data at two top concentrations were excluded.

Institution	n	Regression Line	Interval of slope**	S.E. (µg/ml)	Interval of y at x=8.21	Estimate (µg/ml)** at x=94.1
A	16	y = -0.033 µg/ml +1.032x	1.017 - 1.048	1.09	7.76 - 9.13	96.1 - 98.1
B	8	y = 0.986 +0.994x	0.979 - 1.009	0.666	8.50 - 9.80	93.6 - 95.5
C	8	y = 1.641 +0.983x	0.956 - 1.010	1.21	8.52 - 10.90*	92.4 - 95.8
D	8	y = 1.229 +1.011x	0.991 - 1.031	0.878	8.67 - 10.39	95.1 - 97.6
E	8	y = 1.185 +1.006x	0.990 - 1.022	0.713	8.75 - 10.15	94.9 - 96.9
F	16	y = 0.521 +1.004x	0.996 - 1.012	0.540	8.42 - 9.10	94.5 - 95.5
G	8	y = -0.582 +1.057x	1.015 - 1.100	1.89	6.25 - 9.95	96.3 - 101.6
H	8	y = -0.116 +0.997x	0.992 - 1.002	0.234	7.84 - 8.30	93.4 - 94.0

* out of MAD interval (MAD intervals are 5.89 - 10.53 at x=8.21 and 83.2 - 105.0 at x=94.1)

** 90% confidence interval

Table 5. Reported values for analytical quality assurance using NO_2 gas exposed badge samples.

Target Value	Reported Value 100nmole													
	A1	A2	B1	B2	C1	D1	D2**		E1	Average	S.D.	cv(%)	maximum	minimum	n
0.372	0.330	0.3187	0.432 0.420 0.375	0.420	0.353	0.347	0.431 0.514*	(0.495) (0.578*)	0.4220	0.385	0.045	11.7	0.432	0.3187	10
0.700	0.604	0.6300	0.793 0.787 0.702	0.707	0.711		0.797 0.694	(0.861) (0.758)	0.7051	0.713	0.065	9.1	0.797	0.604	10
1.25	1.156	1.1536	1.385 1.285 1.229	1.451	1.328	1.294	1.253 1.349	(1.317) (1.413)	1.2380	1.284	0.092	7.1	1.451	1.1536	11
1.93	1.660	1.7489	1.723 1.776 1.656	1.815	1.796	1.747 1.858 1.753	1.799 1.735	(1.863) (1.799)	1.9068	1.767	0.071	4.0	1.9068	1.656	13
4.21	3.627*	3.9943	3.682* 3.943 3.711	3.836	4.202	3.592*	3.964 4.157	(4.028) (4.221)	3.9680	3.972	0.158	4.0	4.202	3.711	8

A, B, C, D and E: institution
1 and 2: run number
* out of x ± (10nmole + 0.1x) interval, excluded from statistical calculations in this table
** Values in parentheses are original values which have apparent bias error

Table 6. Results of maximum allowable deviation (MAD) interval test on analytical quality assurance using NO_2 gas exposed badge samples.

Institution and run	n	Regression Line	Interval of slope**	S.E. (10^{-7}mole)	Interval of y at x=0.372	Estimate (10^{-7}mole)** at x=4.21
A - 1	5	y = 0.0263 x 10^{-7}mole +0.856x	0.826 - 0.886	0.0391	0.288 - 0.402	3.55* - 3.72
2	5	y = -0.0500 +0.957x	0.933 - 0.981	0.0309	0.261 - 0.351	3.91 - 4.05
B - 1	15	y = 0.131 +0.864x	0.832 - 0.896	0.0954	0.392 - 0.513*	3.68* - 3.86
2	5	y = 0.158 +0.879x	0.781 - 0.977	0.127	0.299 - 0.671*	3.58* - 4.14
C - 1	5	y = -0.0013 +0.992x	0.925 - 1.060	0.0875	0.240 - 0.496	3.98 - 4.37
D - 1	6	y = 0.162 +0.828x	0.755 - 0.900	0.0968	0.328 - 0.611*	3.46* - 3.83
2	10	y = 0.0887 +0.934x	0.892 - 0.976	0.0967	0.357 - 0.515*	3.90 - 4.14
	(10	y = 0.153 +0.934x	0.892 - 0.976	0.0967	0.421 - 0.579*	3.97 - 4.20)***
E - 1	5	y = 0.0792 +0.927x	0.906 - 0.948	0.0271	0.384 - 0.464	3.92 - 4.04

* out of MAD interval (MAD intervals are 0.235 - 0.509 at x=0.372 and 3.69 - 4.73 at x=4.21)

** 90% confidence interval

*** Original data which had apparent bias error were used for evaluation

Analytical performance of institution A was satisfactory in the 2nd run of AQA2. For institution D, recalculated values by the TCC were used for the evaluation, since a blank sample different from that for QC samples seemed to be analyzed in D2. One data was slightly out of the MAD interval (see Table 6). The 90% confidence interval of the regression line was also slightly out of the interval at the lower end of the tested range (see Table 7). The TCC had judged that analytical performance of institution D to be satisfactory when considering that (1) slope of the regression line was largely improved, (2) in the MAD interval test on the regression line, 90% confidence interval of the line was overestimated at the lower end of the tested range (see section 3.1.1.). For institution B, the 1st criterion was satisfied in the 2nd run. However, the regression line including large standard error was out of the MAD interval again. The TCC had decided to accept the results because of the following reasons: (1) since the 1st criterion was satisfied, reliability and comparability of their monitoring data would not be so poor, (2) implementation of the pilot monitoring should be accelerated.

3.2. Quality assurance in pilot monitoring

3.2.1. Analytical quality assurance (AQA3)

Results of AQA3 in the pilot exposure monitoring are shown in Tables 7 and 8. Analytical performances were satisfactory for institutions C, D, and E. For institution A, two data were largely different from target values, so that the evaluation was carried out excluding these. For the 1st criterion, 1 data was slightly out of the MAD interval. Ninety percent confidence interval of the regression line was also slightly out of the interval at the higher end of the tested range. Monitoring data from institution A may include slightly larger error than others. However, the error was not so large that it would affect the reliability and comparability of their data.

3.2.2. Quality assurance by duplicate samples

Duplicate samples were collected in the pilot exposure monitoring in Beijing and Stockholm. Analytical results were shown in Tables 9 and 10. There were a couple of data set in which a fairly big difference was observed. However, in general, duplicate data agreed well with each other.

Table 7. Reported values for analytical quality assurance in pilot monitoring phase.

Target Value	Reported Value 100 nmole A	C	D	E	Average	S.D.	cv(%)	maximum	minimum	n
0.372	0.364	0.440	0.446	0.3717	0.393	0.042	10.7	0.446	0.3453	7
	0.519*	0.359	0.423	0.3453						
0.700	0.682	0.679	0.747	0.7189	0.681	0.046	6.8	0.747	0.593	9
	0.643	0.593	0.721	0.6585						
			0.687							
1.25	1.189	1.266	1.300	1.4023	1.293	0.072	5.6	1.4023	1.189	8
	1.279	1.228	1.384	1.2939						
1.93	1.488*	1.701		2.0512	1.852	0.152	8.2	2.0512	1.701	5
	1.736	1.799		1.9704						
4.21	1.999*	4.004	3.887	4.0493	3.928	0.147	3.7	4.0792	3.715	7
	3.715	3.748	4.013	4.0792						

A, C, D and E: institution
* out of x ± (10nmole + 0.1x) interval, excluded from statistical calculations in this table

Table 8. Results of maximum allowable deviation (MAD) interval test on analytical qu l t_ surance using NO_2 gas exposed badge samples in pilot monitoring phase.

Institution and run	n	Regression Line	Interval of slope**	S.E. (10^{-7}mole)	Interval of y at x=0.372	Estimate (10^{-7}mole)** at x=4.21
A - 3	10	y = 0.310 x 10^{-7}mole +0.621x	0.427 - 0.816	0.451	0.172* - 0.910*	2.37* - 3.48
	(8	y = 0.110 +0.857x	0.818 - 0.897	0.0683	0.369 - 0.490	3.60* - 3.84)***
C - 2	10	y = 0.0446 +0.908x	0.870 - 0.947	0.0890	0.310 - 0.455	3.76 - 3.98
D - 3	9	y = 0.112 +0.915x	0.886 - 0.945	0.0686	0.398 - 0.508	3.87 - 4.06
E - 2	10	y = 0.0674 +0.961x	0.924 - 0.998	0.0859	0.355 - 0.495	4.01 - 4.22

* out of MAD interval (MAD intervals are 0.235 - 0.509 at x=0.372 and 3.69 - 4.73 at x=4.21)

** 90% confidence interval

*** Two data which had large error were excluded from evaluation

Table 9. Quality assurance by duplicate samples in Beijing.

Sample	Date	Data 1 (ppb)	Data 2 (ppb)	Difference (%)
Subject 8	3/22-25	29.80	29.80	0.00
Kitchen	3/25-29	29.51	30.70	3.95
Subject 8	3/22-25	48.37	46.87	3.15
Living Room	3/25-29	48.51	47.91	1.24
Subject 8	3/22-25	9.94	9.74	2.03
Bedroom	3/25-29	10.31	10.94	5.93
Subject 8	3/22-25	17.44	17.64	1.14
Home Outdoor	3/25-29	16.04	15.85	1.19
Subject 8	3/22-25	10.22	9.81	4.09
Workplace	3/25-29	10.82	11.32	4.52
Subject 8	3/22-25	5.59	5.43	2.90
Workplace Outdoor	3/25-29	6.17	6.20	0.49
Subject 15	3/22-25	19.27	19.66	2.00
Kitchen	3/25-29	19.56	21.10	7.58
Subject 15	3/22-25	11.52	11.39	1.13
Living Room	3/25-29	10.61	10.67	0.56
Subject 15	3/22-25	9.84	9.75	0.92
Bedroom	3/25-29	9.26	9.10	1.74
Subject 15	3/22-25	13.42	20.07	39.71
Home Outdoor	3/25-29	11.09	8.23	29.61
Subject 15	3/22-25	20.09	19.99	0.50
Workplace	3/25-29	18.38	18.68	1.62
Subject 15	3/22-25	20.48	18.97	7.66
Workplace Outdoor	3/25-29	19.56	19.48	0.41
			av.	5.17
			s.d.	9.45
			max.	39.71
			min.	0.00
			n	24

Table 10. Quality assurance by duplicate samples in Stockholm.

Sample	Date	Data 1 (ppb)	Data 2 (ppb)	Difference (%)
Subject 5	3/10-14	10.0	9.8	2.0
Living/Bedroom	3/14-17	12.6	11.9	5.7
Subject 18	3/10-14	13.5	14.3	5.8
Living/Bedroom	3/14-17	15.8	14.5	8.6
Outdoor	3/10-14	11.1	11.1	0.0
wp (LTD)*	3/10-14	11.3	10.4	8.3
Outdoor	3/10-14	11.9	11.2	6.1
wp (HTD)*	3/10-14	11.8	12.4	5.0
Outdoor	3/14-17	15.7	15.3	2.6
wp (LTD)	3/14-17	16.5	14.8	10.9
Outdoor	3/14-17	17.3	16.1	7.2
wp (HTD)	3/14-17	17.9	16.9	5.7
Outdoor	2/29-3/3	15.5	13.4	14.5
wp (HTD)	2/29-3/3	15.2	15.4	1.3
Outdoor	3/3-7	19.0	16.3	15.3
wp (HTD)	3/3-7	14.3	13.2	8.0
			av.	6.7
			s.d.	4.3
			max.	15.3
			min.	0.0
			n	16

* Workplaces (wp) were located in high (HTD) and low (LTD) traffic density areas.

3.3. Pilot monitoring

The main purpose of the pilot monitoring is to establish exposure monitoring methodologies through small scale study and also to obtain preliminary information about NO_2 exposure levels at different HEAL sites. Therefore, in this part of the report, HEAL methodologies will be demonstrated/evaluated through the presentation and interpretation of data obtained in the pilot monitoring.

3.3.1. Exposure monitoring

3.3.1.1. Personal exposure to NO_2

Personal exposure values for NO_2 observed at 4 HEAL sites are shown in Table 11. The personal exposure levels varied largely both by subject and sampling date.

Table 11. Personal NO_2 exposure levels at four HEAL sites (unit: ppb).

		Beijing	Stockholm	Yokohama	Zagreb
Statistics for all data	av.	18.28	11.44	40.53	17.43
	s.d.	6.24	4.68	24.18	7.17
	max.	37.78	27.30	110.72	39.50
	min.	5.66	1.50	15.79	2.50
	n	105	105	70	112
Average data for each subject	av.	18.28	11.44	40.53	17.43
	s.d.	4.49	2.97	21.57	5.21
	max.	27.12	17.57	87.59	29.94
	min.	9.93	7.99	22.22	10.83
	n	15	15	10	16
Average data for each sampling date	av.	18.28	11.39	40.53	
	s.d.	1.71	2.90	6.70	
	max.	20.51	15.68	52.59	N/A
	min.	15.83	6.69	33.39	
	n	7	14	7	

In Beijing, the daily personal exposure value for NO_2 ranged from 6 to 38 ppb (average = 18.28 ± 6.24 ppb). Standard deviation of weekly averaged data for each subject (personal variation in the exposure level) was larger than that of averaged data for each sampling date (daily variation)*.

In Stockholm, the daily personal NO_2 exposure values were in the range of 1 - 27 ppb (average = 11.44 ± 4.68 ppb). There was no statistically significant difference between the personal and daily variations.

In Yokohama, the daily personal exposure values ranged from 16 to 111 ppb (average = 40.53 ± 24.18 ppb). The personal variation was larger than the daily variation ($p < 0.1$).

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* $p < 0.1$

In Zagreb, the daily personal exposure values were in the range of 2 to 39 ppb (average = 17.43 ± 7.17 ppb). The difference between personal and daily variations could not be evaluated statistically because of the complicated sampling schedule. However, the data obtained suggested that the difference may be small.

Detailed data on personal NO_2 exposure at 4 HEAL sites are given Appendix 3.

3.3.1.2. Indoor/outdoor NO_2 concentrations

Tables 12-15 show weekly averaged indoor/outdoor NO_2 concentrations at 4 HEAL sites. These tables also show personal NO_2 exposure data, information on indoor emission sources and residing area for each subject.

In Beijing (see Table 12), averaged indoor/outdoor NO_2 concentrations at home were in the following order:

living room
kitchen
outdoor bedroom

** $p < 0.01$

Homes of all subjects except one (subject 13) had indoor emission sources, namely, unvented gas-cookers in their kitchens. Therefore, the NO_2 concentration in the kitchen was higher than those in other rooms and outdoor air except for subject 13. NO_2 concentrations in the living room and bedroom were lower than that in home outdoor air except for one case (subject 11).

NO_2 concentration in the workplace was also lower than that in the outdoor air. These observations agree well with the generally accepted information, i.e., indoor NO_2 concentration is lower than that in outdoor air when there is no indoor emission source. NO_2 concentrations at home outdoors as well as those at workplace indoors and outdoors were not so different by subject compared with NO_2 concentrations at home indoors. This may be due to the fact that all subjects were living in an urban area of the city and working in the same institution.

Table 16 lists correlation co-efficients among NO_2 concentrations at home indoors/outdoors. A poor correlation between NO_2 concentrations indoors and that in outdoor air was observed in Beijing, Yokohama and Zagreb. This observation demonstrates clearly the large influence of indoor emission sources on indoor NO_2 level. In general, a correlation between indoor and outdoor NO_2 concentrations was lower in homes, where windows are usually closed and unvented cookers and heaters are used. A high correlation between NO_2 concentrations in the kitchen and living room at all HEAL sites suggests the diffusion of indoor pollutants from the kitchen to the living room at these homes.

Table 12. Indoor/outdoor NO_2 concentrations (weekly data, unit:ppb) in Beijing.

Subject Number	Personal Exposure	Home				Workplace		Fuel		Residing Area
		Kitchen	Living Room	Bedroom	Outdoor	Indoor	Outdoor	Cooking	Heating	
1	16.86	45.63	-	8.12	19.84	16.94	20.46	G**		urban
2	16.00	66.81	-	14.43	21.86	7.43	18.97	G**		urban
3	17.77	29.87	-	8.05	19.81	10.30	21.57	G**		urban
4	9.93	28.05		6.13	21.59	10.26	17.65	G**	C	urban
5	18.13	41.79		9.09	19.84	12.64	20.36	G**		urban
6	14.78	61.01		8.68	24.05	10.22	17.65	G**	C	urban
7	27.12	50.45	14.89	16.51	20.53	13.36	19.79	G**		urban
8	15.20	29.97	10.29	10.62	19.96	9.44	19.18	G**		urban
9	21.90	41.07	12.95	11.31	23.07	11.20	21.57	G**		urban
10	21.48	31.48		15.12	21.05	12.11	18.47	G**		urban
11	20.15	49.42		25.68	22.63	13.26	19.81	G**		urban
12	20.74	29.52		20.44	25.69	13.04	20.21	G**	C	urban
13	11.53	14.47		5.71	26.70	11.05	20.11	C	C	urban
14	19.25	28.25		18.22	23.36	12.71	19.27	G**	C	urban
15	23.34	47.96	16.63	5.90	10.99	12.70	19.61	G**		urban
av.	18.28	39.72	13.69	12.27	21.40	11.89	19.65			
s.d.	4.49	14.08	2.72	5.95	3.59	2.24	0.98			
max.	27.12	66.81	16.63	25.68	26.70	16.94	21.57			
min.	9.93	14.47	10.29	5.71	10.99	7.43	17.65			
n	15	15	4	15	15	14*	13*			

fuel: G = gas, C = coal

- not measured

* Overlapping data were treated as one data. All subjects were working in Beijing Sanitary and Anti-epidemic Station

** unvented equipment

Table 13. Indoor/outdoor NO_2 concentrations (weekly data, unit:ppb) in Stockholm.

Subject Number	Personal Exposure	Home				Workplace		Fuel		Residing Area
		Kitchen	Living Room	Bedroom	Outdoor	Indoor	Outdoor	Cooking	Heating	
1	15.57	15.77	13.84	11.29	13.39	10.16	12.99 / 12.90	G**		suburban
2	12.30	7.30	7.45	7.05	10.05	17.80	16.00 / 14.55	E		urban
3	10.94	2.46	3.16	3.06	3.76	17.99	16.23 / 14.44	E	E	rural
4	9.27	3.97	4.14	4.23	9.53	17.20	16.23 / 14.44	E		suburban
5	17.57	15.86		10.91	12.66	14.80	13.79 / 14.37	G**	O	urban
6	16.07	10.16	6.07	6.31	15.49	14.80	13.79 / 14.37	G**	O	urban
7	10.86	3.03	3.99	2.89	5.54	17.99	16.23 / 14.44	E	E/W	suburban
8	9.50	2.76	2.87	2.37	7.83	14.80	13.79 / 14.37	E	E	suburban
9	10.20	2.39	2.60	1.57	6.79	17.99	16.23 / 14.44	E	E	suburban
10	7.99	1.96	1.84	1.71	5.01	13.76	13.79 / 14.37	E	E	suburban
11	10.00	2.20	2.86	2.17	4.94	17.20	16.23 / 14.44	E	E/W	rural
12	13.50	11.59	11.37	13.73	14.14	10.16	12.99 / 12.90	G**	O	suburban
13	10.46	3.17	2.44	2.14	6.61	17.99	16.23 / 14.44	E	E	rural
14	8.97	2.89	2.70	2.49	7.67	10.16	12.99 / 12.90	E	E	suburban
15	8.24	3.19	4.00	3.61	10.34	13.76	13.79 / 14.37	E	O	suburban
av.	11.43	5.91	4.95	5.04	8.92	14.78	14.12			
s.d.	2.96	4.99	3.60	3.96	3.68	3.10	1.20			
max.	17.57	15.86	13.84	13.73	15.49	17.99	15.33			
min.	7.99	1.96	1.84	1.57	3.76	10.16	12.94			
n	15	15	14	15	15	5*	3*			

fuel: G = gas, E = electricity, O = oil, W = wood

* Duplicate data were averaged. Overlapping data were treated as one data. All subjects were working in Karolinska Institute area

** unvented equipment

Table 14. Indoor/outdoor NO_2 concentrations (weekly data, unit:ppb) in Yokohama.

Subject Number	Personal Exposure	Home				Workplace		Fuel			Residing area
		Kitchen	Living Room	Bedroom	Outdoor	Indoor	Outdoor	Cooking	Boiler***	Heating	
1	22.22	37.38	28.91	14.31	30.80	-	-	G**	G**	E	urban
2	23.02	28.49	22.62	10.47	17.85	-	-	G**	G	G or O	urban
3	55.62	70.60	46.85	24.72	21.67			G**	G**	G**	urban
4	51.82	73.31	15.09	31.09	25.52			G**	G**	G**	urban
5	22.64	19.61	16.03	15.52	25.57	16.78	21.81	G**	G**	G or O	urban
6	33.62	79.17	53.83	6.07	21.01	16.78	21.81	G**	G**	G or O	urban
7	55.41	97.95	106.17	17.63	20.89	21.62	25.96	G**	G	O**	urban
8	28.32	41.07	17.85	8.97	18.12			G**	G	G or O	urban
9	87.59	102.13	24.49	29.88	23.51			G**	G	O**	urban
10	25.08	29.35	22.42	9.99	21.61	16.78	21.81	G**	G	G**	urban
av.	40.53	57.91	35.43	16.87	22.66	19.20	23.89				
s.d.	21.57	30.30	28.02	8.88	3.88	3.42	2.93				
max.	87.59	102.13	106.17	31.09	30.80	21.62	25.96				
min.	22.22	19.61	15.09	6.07	17.85	16.78	21.81				
n	10	10	10	10	10	2*	2*				

fuel: G = gas, E = electricity, O = oil

- not measured

* Overlapping data were treated as one data

** unvented equipment

*** boiler for hot water

Table 15. Indoor/outdoor NO_2 concentrations (weekly data, unit:ppb) in Zagreb.

Subject Number	Personal Exposure	Home				Workplace		Fuel			Residing area
		Kitchen	Living Room	Bedroom	Outdoor	Indoor	Outdoor	Cooking	Boiler***	Heating	
1	10.83	4.24		4.63	11.10	10.57	13.80	E	E	E	urban
2	15.94	25.29	21.73	4.27	7.10	12.43	13.80	G**	E	W/C	rural
3	25.40	37.91	35.87	26.91	9.54	10.51	13.10	G**	G	G	urban
4	18.49	8.74		3.04	5.17	17.21	13.10	G**/W	G	E/W	rural
5	29.94	47.79	20.47	18.80	9.46	27.57	13.63	G**	G	G	suburban
6	22.03	47.24	16.74	11.65	22.96	12.61	13.57	G**	E	D	urban
7	14.14	14.21	13.10	12.97	18.79	9.60	19.26	E	D	D	urban
8	20.33	23.60	19.13	15.84	20.47	10.40	13.63	G**	G	D	urban
9	11.86	15.54	5.56	2.29	7.91	9.91	10.97	G**/W/C	E	E/W/C	rural
10	16.83	18.91	8.21	8.39	5.09	5.40	10.00	G**	O	O	rural
11	10.84	5.46	6.59	6.37	9.80	7.70	8.74	E/W	E	E/W	suburban
12	13.19	7.14	5.54	2.31	6.14	33.39	11.90	E/W	E	W	rural
13	15.30	16.96	17.59	14.33	12.07	9.49	8.84	G**	D	D	suburban
14	17.40	11.79	7.24	4.73	6.94	10.63	8.37	G**	O	O	rural
15	18.76	11.69	13.21	9.46	19.71	9.41	11.23	E	O	E/O	suburban
16	17.53	19.20	10.40	8.66	15.31	14.31	12.31	G**	G	G	urban
av.	17.43	19.73	14.38	9.67	11.72	13.20	12.04				
s.d.	5.21	13.73	8.39	6.83	5.89	7.32	3.10				
max.	29.94	47.79	35.87	26.91	22.96	33.39	19.26				
min.	10.83	4.24	5.54	2.29	5.09	5.40	8.37				
n	16	16	14	16	16	16*	11*				

fuel: G = gas, E = electricity, O = oil, W = wood, C = coal, D = district
- Measurement was carried out half of a week
* All subjects were working in Institute for Medical Research and Occupational Health
** unvented equipment
*** boiler for hot water

A correlation between NO_2 concentration in the kitchen and the time spent for cooking (see Appendix 4) was not observed (correlation co-efficients were 0.077 in Beijing, 0.328 in Stockholm, 0.418 in Yokohama and -0.218 in Zagreb, respectively). This may be partly due to that cooking time was not equal to the working time of gas-cookers.

In Stockholm (see Table 13), there was no significant difference between average NO_2 concentrations in the kitchen, living room and bedroom. They were significantly lower than the home outdoors ($p < 0.1$). Four subjects had unvented gas-cookers. NO_2 concentrations in the kitchens of these subjects were high compared with the corresponding values for other subjects who had used electric-cookers. However, the concentrations were not so high and nearly at the same level with values outdoors. This can be explained partly by the short time for cooking with these subjects (0.51 - 4.08% of the monitoring period). Outdoor NO_2 concentration at home (4 - 15 ppb) largely varied with the residence location.

In Stockholm, even in winter, a high correlation was observed between NO_2 concentrations at home indoors and that outdoors, as well as among those at home indoors (see Table 17). This observation can be explained by the following facts: (1) There was no significant influence of indoor emission sources on indoor NO_2 pollution, and the main source of the indoor pollution was suggested to be outdoor pollutant penetration into the rooms. (2) Outdoor NO_2 concentrations were largely determined by the location of the residence.

In Yokohama (see Table 15), average indoor/outdoor NO_2 concentrations at home were in the following order:

```
            >*  living room >*
kitchen                          bedroom
            >** outdoor      >*
```

* $p < 0.1$, ** $p < 0.01$

The influence of indoor emission sources was especially large in kitchens. The significantly high NO_2 concentrations in kitchens can be explained by the following facts: (1) all subjects had unvented gas-cookers, (2) five of the ten subjects had small unvented gas-boilers for preparing hot water quickly, (3) cooking time was fairly long. The influence of unvented heaters was also observed in NO_2 concentrations in living rooms and bedrooms. However, the averaged NO_2 concentration in the bedroom was significantly lower than that of home outdoors since only two subjects (subjects 4 and 9) used unvented heaters in their bedrooms. NO_2 levels in outdoor air did not show a big difference by subject since all homes and workplaces of the subjects were located in an urban area of the city.

As was expected from the existence of indoor air pollution, there was no correlation between NO_2 concentrations at home indoors and that outdoors (see Table 16). A correlation between NO_2 concentration in the kitchen and those in other rooms suggested that the latter rooms were partly polluted through diffusion of indoor NO_2 pollution from the kitchen.

Table 16. Correlation between indoor/outdoor NO_2 concentrations.

	Kitchen	Living Room	Bedroom
Beijing			
Living Room	0.926**		
Bedroom	0.164	-0.207	
Outdoor	-0.274	-0.650	0.332
Stockholm			
Living Room	0.960***		
Bedroom	0.927***	0.961***	
Outdoor	0.833***	0.760***	0.811***
Yokohama			
Living Room	0.588*		
Bedroom	0.544*	-0.060	
Outdoor	-0.064	-0.193	0.354
Zagreb			
Living Room	0.683**		
Bedroom	0.676**	0.841***	
Outdoor	0.286	0.174	0.359

* p = 0.1
** p = 0.01
*** p = 0.001

In Zagreb (see Table 15), average indoor/outdoor NO_2 concentrations at home were in the following order:

Kitchen >* Living Room >** Bedroom
Kitchen >* Outdoor

* $p < 0.1$, ** $p < 0.01$

Table 17. Summary of activity records of subjects.

		Beijing		Stockholm		Yokohama		Zagreb	
Kitchen	av.		6.79		11.28	12.21*	28.25**		12.02
(%)	s.d.		2.96		4.83	5.76*	3.81**		5.90
	max.		13.39		20.19	21.83*	31.65**		23.40
	min.		2.08		3.54	5.26*	24.80**		3.17
	n		15		15	6*	4**		16
Living	av.		21.29''		10.77''	18.96*	17.26**		15.65''
Room	s.d.		7.13''		5.46''	1.68*	12.09**		9.32''
(%)	max.	N/A	26.79''	N/A	20.18''	21.03*	28.27**	N/A	34.62''
	min.		5.75''		4.12''	16.82*	3.07**		0.00''
	n		7''		14''	6*	4**		14''
Bedroom	av.	54.86'	38.11''	51.04'	34.51''	33.14*	36.64**	48.76'	34.19''
(%)	s.d.	6.70'	5.75''	-	6.00''	5.48*	10.09**	-	2.68''
	max.	62.60'	46.83''	-	44.09''	43.15*	51.63**	55.94'	39.15''
	min.	41.96'	32.94''	-	20.35''	27.40*	30.16**	41.57'	29.22''
	n	8'	7''	1'	14''	6*	4**	2'	14''
Workplace	av.		25.22		23.41	19.74			18.32
(%)	s.d.		5.66		5.37	9.18			3.20
	max.		37.80		34.71	27.08	N/A		22.43
	min.		16.67		12.17	3.47			11.71
	n		15		15	6			16
Other	av.		3.23		9.93	8.34*	8.47**		9.97
Indoor	s.d.		4.01		9.00	4.80*	3.24**		3.05
(%)	max.		13.79		33.04	13.25*	10.88**		17.21
	min.		0.00		0.60	0.00*	3.77**		5.05
	n		15		15	6*	4**		16
Transport	av.				5.78	3.66*	2.93**		7.83
(%)	s.d.	N/A			2.08	2.29*	3.08**		3.51
	max.	see Appendix 4			8.72	7.69*	6.07**		14.29
	min.				1.09	1.48*	0.00**		2.97
	n				15	6*	4**		16
Outdoor	av.		7.78		3.94	3.95*	6.46**		2.15
(%)	s.d.		3.63		2.09	2.72*	3.10**		1.77
	max.		17.86		10.12	7.42*	9.47**		6.73
	min.		3.67		1.49	1.19*	2.18**		0.00
	n		15		15	6*	4**		16
Cooking	av.		6.80		2.14	6.47*	10.61**		5.75
(%)	s.d.		2.96		1.27	1.87*	3.68**		3.59
	max.		13.39		4.08	9.52*	15.39**		12.98
	min.		2.08		0.40	4.37*	6.45**		0.00
	n		15		15	6*	4**		16
Kitchen+	av.		88.99		80.35		83.29		80.05
Living+	s.d.		5.69		9.52		5.76		5.21
Bedroom+	max.		94.15		91.05		95.33		90.79
Workplace	min.		74.40		53.98		74.16		70.78
(%)	n		15		15		10		16

* calculations for the group of subjects who had jobs outside home

** calculations for the group of housewives

' one room was used as living room and bedroom for these subjects

'' living room and bedroom were separate rooms for these subjects

Table 18. Estimation of personal exposure level from fractional exposures in micro- n i) vents.

		Beijing				Stockholm				Yokohama				Zagreb			
		(ppb)	(%)	(ppb)	(%)	(ppb)	(%)	(ppb)	(%)	(ppb)	(%)	(ppb)	(%)	(ppb)	(%)	(ppb)	(%)
Kitchen	av.			2.74	(14.7)			0.52	(4.5)	5.36	(18.5)*	20.25	(37.4)**			2.13	(11.8)
	s.d.			1.57	(8.2)			0.28	(1.8)	3.38	(10.8)*	8.24	(6.1)**			1.50	(6.3)
	max.			6.11	(36.2)			1.04	(7.8)	11.38	(33.9)*	32.10	(45.9)**			6.60	(22.0)
	min.			0.58	(4.4)			0.18	(1.8)	2.00	(8.8)*	13.00	(31.5)**			0.30	(2.0)
	n			15				15		6*		4**				16	
Living Room	av.			3.26	(15.2)''			0.47	(4.2)''	8.13	(23.9)*	4.98	(8.9)**			2.58	(13.2)''
	s.d.			0.71	(2.9)''			0.25	(2.2)''	6.89	(10.3)*	5.12	(9.3)**			2.24	(9.6)''
	max.	N/A		4.12	(17.7)''	N/A		0.80	(7.4)''	20.75	(37.4)*	12.32	(22.1)**	N/A		7.62	(30.0)''
	min.			2.45	(11.1)''			0.10	(1.2)''	2.70	(11.9)*	0.55	(1.9)**			0.00	(0.0)''
	n			4''				14''		6*		4**				14''	
Bedroom	av.	7.74	(42.8)'	4.01	(21.3)''	5.57	(31.7)'	1.51	(12.9)''	4.02	(14.5)*	7.91	(15.0)**	1.93	(15.4)'	3.56	(18.8)''
	s.d.	4.59	(18.4)'	1.22	(8.3)''	-		1.18	(7.0)''	1.30	(5.8)*	2.25	(3.2)**	-		2.42	(9.2)''
	max.	14.83	(73.6)'	5.50	(31.9)''	-		4.90	(31.5)''	5.81	(20.0)*	9.38	(18.1)**	2.59	(23.9)'	9.31	(36.6)''
	min.	2.40	(20.8)'	2.11	(9.0)''	-		0.63	(6.8)''	2.09	(6.2)*	4.63	(10.7)**	1.26	(6.8)'	0.88	(6.6)''
	n	8'		7''		1'		14''		6*		4**		2'		14''	
Workplace	av.			2.95	(17.4)			3.58	(32.3)	4.56	(14.3)*					2.33	(14.1)
	s.d.			0.78	(7.9)			1.12	(10.8)	0.88	(3.2)*					1.00	(6.4)
	max.			4.35	(39.0)			5.14	(46.7)	5.85	(17.4)*	N/A				4.83	(32.4)
	min.			1.57	(9.9)			1.24	(13.8)	3.93	(10.6)*					0.63	(3.8)
	n			15				15		4*						16	
Kitchen+ Living+ Bedroom+ Workplace	av.			13.14	(70.9)			6.32	(54.8)	23.34	(66.6)*	33.14	(61.3)**			10.07	(55.8)
	s.d.			4.11	(14.5)			2.03	(9.0)	12.04	(14.4)*	11.15	(7.7)**			5.08	(13.5)
	max.			20.73	(102.9)			10.47	(70.6)	37.56	(86.5)*	44.25	(68.5)**			21.35	(84.0)
	min.			6.17	(53.5)			2.69	(30.0)	12.88	(55.1)*	18.18	(50.5)**			5.21	(33.4)
	n			12				15		4*		4**				16	
Observed personal exposure	av.			18.28				11.43		30.33*		55.84**				17.43	
	s.d.			4.49				2.96		13.01*		24.37**				5.21	
	max.			27.12				17.57		55.41*		87.59**				29.94	
	min.			9.93				7.99		22.22*		28.32**				10.83	
	n			15				15		6*		4**				16	

* calculations for the group of subjects who had jobs outside home
** calculations for the group of housewives
' one room was used as living room and bedroom for these subjects
'' living room and bedroom were separate rooms for these subjects

The average NO_2 concentration in the kitchen was higher than the values in the other rooms and outdoor air, because of the use of unvented gas-cookers in 11 homes. For five subjects, who did not have unvented gas-cookers, NO_2 concentration in the kitchen was lower than, or nearly the same as, that in outdoor air. It should be also noted that NO_2 concentrations in workplaces of subjects 5 and 12, where unvented gas-boilers were used, were higher than the others. Outdoor NO_2 concentration at home was largely determined by location of the home.

NO_2 concentration at home outdoors did not give any significant correlation with that indoors (see Table 16). On the other hand, a high correlation was observed among indoor NO_2 levels. Here, indoor emission sources in 11 homes produced a large effect on the correlation among NO_2 concentrations in the micro-environments.

3.3.1.3. Estimation of personal exposure level to NO_2

Mean level of personal NO_2 exposure can be defined as the time-weighted average of NO_2 concentrations in various environments in which the subject spends his/her time and is exposed to the pollutant. We live in various types of indoor environment, that is micro-environments, for 80-90% of a day. These micro-environments are generally polluted by NO_2 as described in section 3.3.1.2. Therefore, personal NO_2 exposure levels can be estimated by the combined use of NO_2 concentrations and time spent in these micro-environments. This approach will also estimate the contribution of each micro-environment to the personal NO_2 exposure, and be of value when planning a reduction in exposure.

Time schedule study for each subject was carried out for the same period of the exposure monitoring study at each HEAL site. Summary of activity records of the subjects at the sites is shown in Table 17. Average values of the total time spent in the micro-environments, i.e., kitchen, living room, bedroom and workplace, accounted for 80-89% of a day. The time spent in the kitchen, where NO_2 concentration is generally considered to be high, accounted for 7-12% of a day for the working women and for 28% of a day for the housewives in this study. However, it should be noted that the number of housewives was very small. Detailed information on activity records of the subjects at each site is given in Appendix 4.

Table 18 shows averages of estimated fractional exposure values in micro-environments and estimated NO_2 personal exposure values (total of the fractional values) together with the observed personal NO_2 exposure values. The estimated values were calculated for each subject from NO_2 concentrations and time spent in kitchen, living room, bedroom and workplace. The estimated personal exposure values were smaller than the observed values at all 4 sites. The ratio of the estimated value to the observed value ranged from 55 to 71% which were lower than the percentage of total time spent in these micro-environments each day. This may be mainly due to the under-estimation of exposures in kitchens, where

extremely high exposures often occur during cooking. For example, an average NO_2 concentration of 320ppb/10hr was observed in a kitchen where intensive cooking was carried out for 10 hours (4 + 6 hours, subject 18 in Zagreb). Such under-estimation may also occur in the workplace and living room where NO_2 concentrations during working/staying times of a subject are sometimes higher than the measured 3-day or 4-day average values. It is expected that the under-estimation will be minimized when the exposure monitoring methodology is improved so as to measure NO_2 concentrations only during working/staying times of each subject in these micro-environments.

A correlation between the estimated personal exposure values and the observed values at each HEAL site is shown in Figure 2a. Correlation co-efficients were 0.72, 0.90, 0.92 and 0.89 at Beijing, Stockholm, Yokohama, and Zagreb, respectively. Figure 2b shows the correlation between the estimated and observed values for all subjects at the 4 sites. A fairly good correlation ($r = 0.95$) was observed.

A significant correlation was also observed between the observed personal exposure values and the estimated fractional exposure values in the following micro-environments:

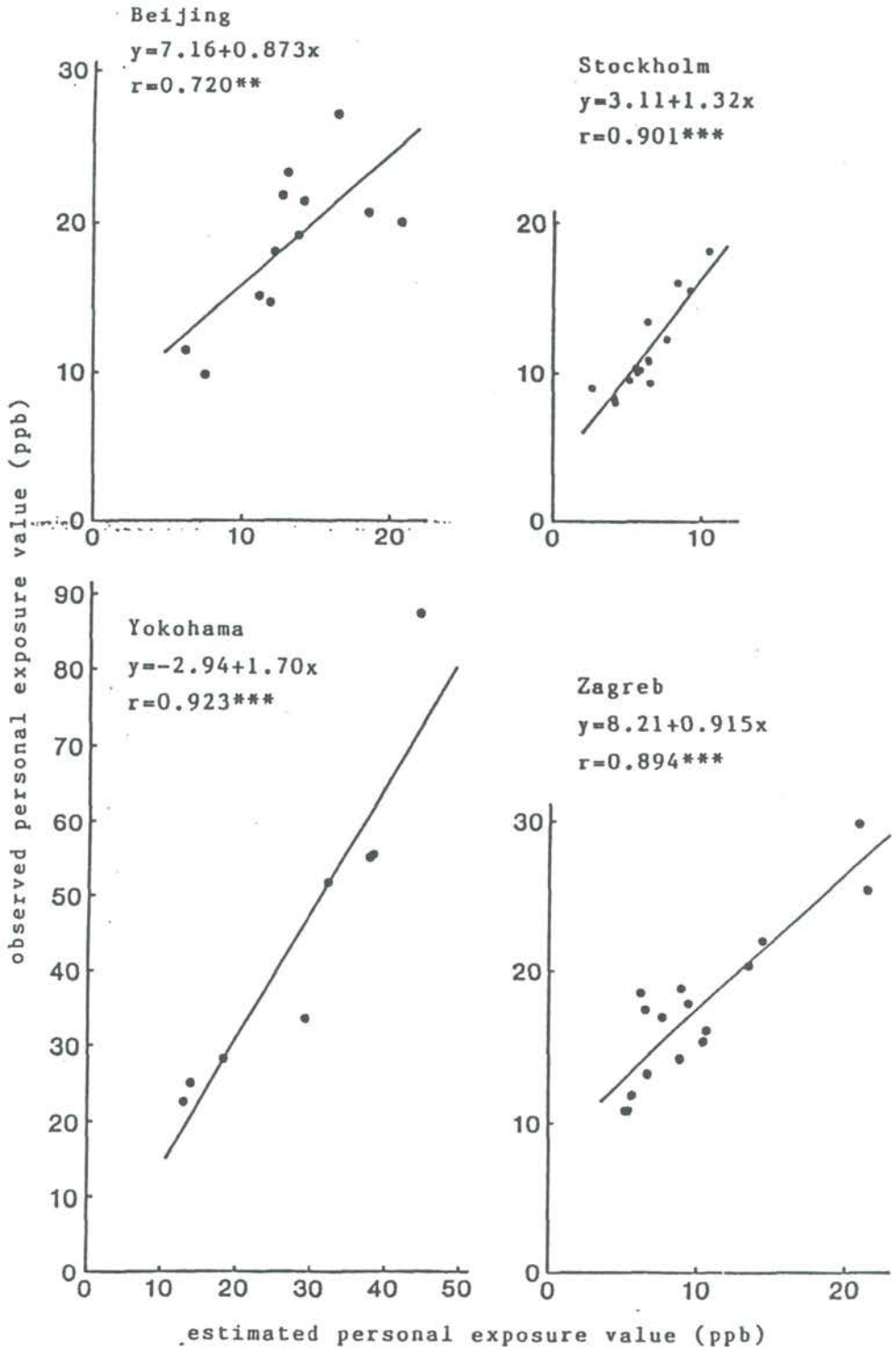
Beijing	kitchen*
Stockholm	kitchen*** and bedroom***
Yokohama	kitchen*** and bedroom**
Zagreb	kitchen**, living room*** and bedroom**

* $p < 0.1$
** $p < 0.01$
*** $p < 0.001$

On the other hand, except for the case in Stockholm, the estimate of personal exposure from home outdoor NO_2 concentration did not give any correlation with the observed personal exposure value. In Stockholm, there was almost no indoor emission source in the home, and high correlation was observed between NO_2 concentrations indoors and outdoors. Furthermore, the average value of observed personal exposures was not so different from the average of NO_2 concentrations at home outdoors as shown in Table 13. These observations in Stockholm suggest that a better estimation of personal exposure level from indoor and outdoor NO_2 concentrations may be achievable in the non-heating season when the penetration of outdoor NO_2 into the rooms is the most important factor for indoor NO_2 pollution.

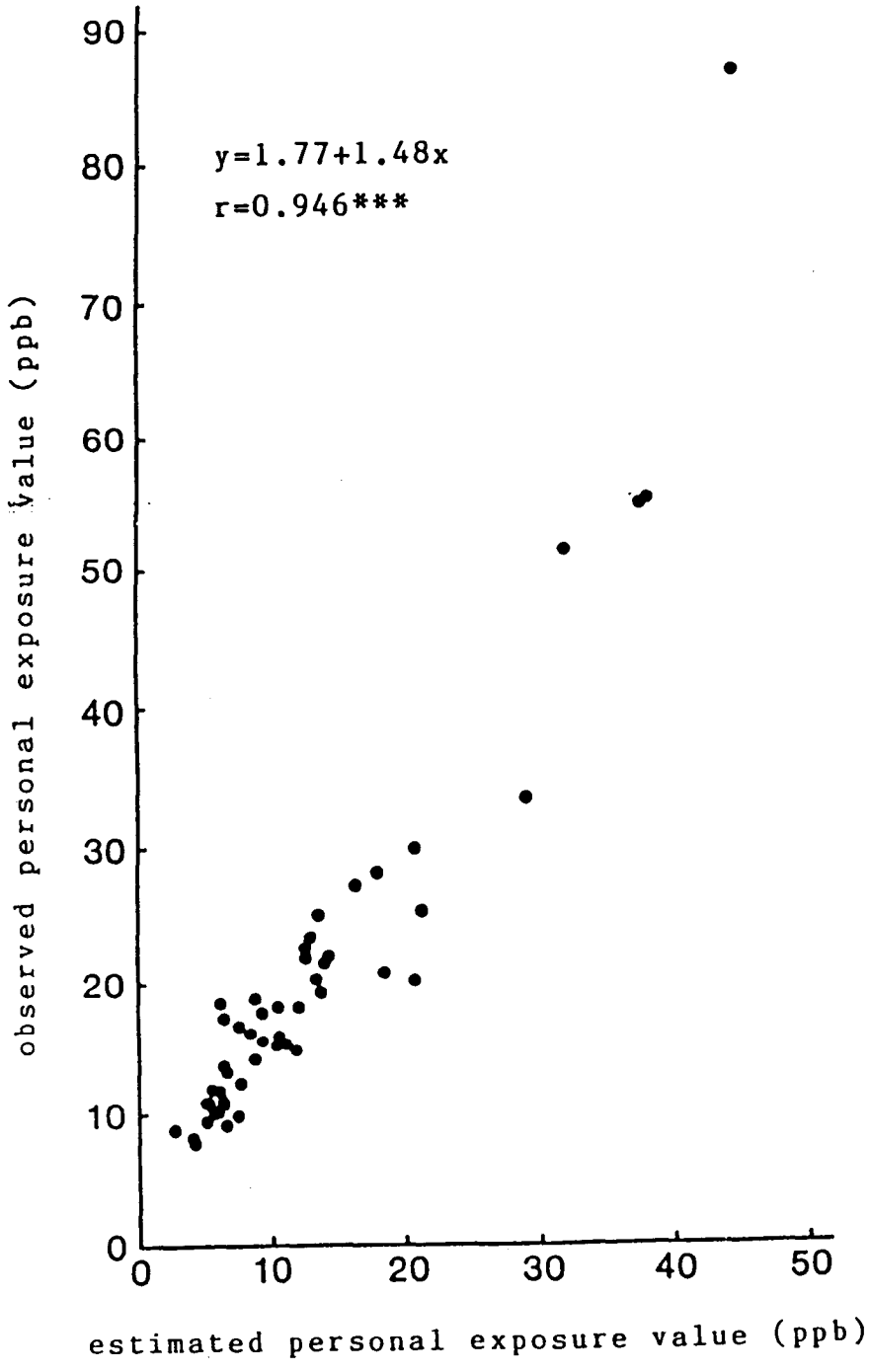
Detailed data on the estimation of personal exposure level from fractional exposures in various micro-environments are given in Appendix 5.

Figure 2a. Correlation between estimated personal exposure values and observed personal exposure values at each HEAL site.



** p = 0.01
*** p = 0.001

Figure 2b. Correlation between estimated personal exposure values and observed personal exposure values for all subjects at 4 HEAL sites.



*** p = 0.001

The following conclusions may be suggested from the results in the pilot exposure monitoring:

1. The personal NO_2 badge method is useful for the measurement of the personal exposure level of the individual.

2. A predominant factor affecting personal NO_2 exposure level is indoor pollution. The weighting of indoor pollution on the personal exposure level will increase in the heating season at each site, when air ventilation rates in the home decreases and the influence of indoor emission sources increases when compared with those in the non-heating season.

3. There was a significant correlation between the observed personal exposure values and the estimated personal exposure values calculated from NO_2 concentrations/times spent indoors may be useful for the first-level estimation of NO_2 exposure of a general population.

4. Improvements of methodology are required for an accurate estimation of personal exposure level from NO_2 concentrations/times spent in various micro-environments. The improvement includes the development of more sensitive NO_2 analysis/sampling methodology by which NO_2 concentrations indoors only during working/staying time of a subject can be measured.

5. The first-level estimation of personal exposure level of a general population may be performed with the present methodologies when the outdoor pollutant is the most important factor for the indoor pollution of the homes of the target population.

3.3.2. Environmental monitoring

Environmental NO_2 data obtained at fixed monitoring stations in each HEAL site are shown in Table 20 together with corresponding personal exposure data and home outdoor NO_2 data obtained in the exposure monitoring. Detailed results of the environmental monitoring are given in Appendix 6.

Environmental NO_2 data did not agree well with the home outdoor NO_2 data obtained in the exposure monitoring. Environmental data measured by the chemiluminescence method were higher than home outdoor data measured by the NO_2 badge method. However, environmental data measured by the badge method were also higher than home outdoor data (Zagreb). The TCC had confirmed the validity of the badge method through a comparison study with the chemiluminescence method prior to the pilot monitoring.

Table 19. Environmental NO_2 concentrations at each HEAL site.

	Date	Environment Urban* (ppb)	Averaged Personal** (ppb,n=15)	Outdoor Home*** (ppb)
Beijing	3/22-23	27.4	18.05	
	3/23-24	26.6	16.31	
	3/24-25	40.7	19.52	
	3/25-26	24.6	18.46	
	3/26-27	46.3	19.27	
	3/27-28	24.5	20.51	
	3/28-29	26.6	15.83	
	av.	31.0		21.40
	s.d.	8.8		3.59
	max.	46.3		26.70
	min.	24.5		10.99
	n	7		15
Stockholm	2/29-3/1	13.5	10.21	
	3/1-2	15.6	10.53	
	3/2-3	29.9	14.80	
	3/3-4	13.1	14.01	
	3/4-5	10.7	7.86	
	3/5-6	9.6	6.69	
	3/6-7	18.6	10.14	
	av.	15.9		10.05
	s.d.	6.9		-
	max.	29.9		-
	min.	9.6		-
	n	7		1
	3/10-11	13.61	10.89	
	3/11-12	10.69	7.56	
	3/12-13	15.29	10.09	
	3/13-14	22.16	14.70	
	3/14-15	20.68	13.40	
	3/15-16	31.48	15.68	
	3/16-17	15.38	12.93	
	av.	18.5		14.08
	s.d.	7.0		-
	max.	31.5		-
	min.	10.7		-
	n	7		2

* chemiluminescence method, hourly data were averaged in the time period of personal exposure monitoring.

** personal exposure data were averaged for each sampling date.

*** NO_2 concentrations home outdoors in urban area.

Table 19 continued.

	Date	Environmental* Urban High (ppb)	Urban Low (ppb)	Averaged Personal** (ppb,n=10)	Outdoor Home*** (ppb)
Yokohama	3/11-12	28.5	6.3	33.39	
	3/12-13	29.1	17.1	34.42	
	3/13-14	58.9	32.1	39.69	
	3/14-15	32.5	26.5	36.57	
	3/15-16	26.0	16.4	42.79	
	3/16-17	50.5	37.0	52.59	
	3/17-18	33.2	27.3	44.28	
	av.	37.0	23.2		22.6
	s.d.	12.6	10.5		3.88
	max.	58.9	37.0		30.80
	min.	26.0	6.3		17.85
	n	7	7		10

	Date	Environmental Urban (ppb)	Suburban (ppb)	Outdoor Home Urban (ppb) (n)	Suburban (ppb) (n)
Zagreb	3/24-28	19.7	16.3	10.6 (2)	
	3/28-31	34.9	18.0	15.0 (5)	13.4 (1)
	3/31-4/4	23.2	20.2	21.7 (4)	6.5 (1)
	4/4-7	27.4	11.1	10.0 (1)	14.6 (1)
	4/7-11	20.1	12.5		8.6 (2)
	4/11-14	19.9	15.0		15.8 (2)
	4/14-18	26.4	15.2		21.0 (1)
	av.	24.51	15.47		
	s.d.	5.56	3.10		
	max.	34.9	20.2		
	min.	19.7	11.1		
	n	7	7		

* chemiluminescence method, hourly data were averaged in the time period of personal exposure monitoring.

** personal exposure data were averaged for each sampling date.

*** NO_2 concentrations home outdoors in urban area.

Significant correlation was observed between environmental NO_2 concentration and daily averaged NO_2 personal exposure level in some areas. Correlation co-efficients were 0.356 in Beijing, 0.720' (2/19-3/7) and 0.880" (3/10-17) in Stockholm, 0.459 (urban high exposure area) and 0.718' (urban low exposure area) in Yokohama. The correlation suggests that environmental NO_2 concentration may be affecting the personal exposure level of people in these areas, although personal exposure estimation from home outdoor NO_2 concentrations was not successful (see Section 3.3.2.3). The importance of environmental data as basic information for exposure estimation for the general population will be larger in the non-heating season than the studied heating season.

3.3.3. International comparison

Although the subjects in the pilot monitoring study were selected from women employed at, or relating to, each monitoring laboratory participating in the HEAL project it may be useful to compare the results. The selection was done so that some of the subjects were living in relatively high polluted areas and some in relatively low polluted areas. Subjects were also selected that had, and some did not have, indoor NO_2 emission sources in their homes. Therefore, the subjects chosen may not represent the general population in each HEAL site. However, with these caveats since the selected subjects had common characteristics through the four HEAL sites, the results obtained in the pilot study may be used for a preliminary international comparison.

Table 20 shows the results of the international comparison. Average personal NO_2 exposure levels at 4 HEAL sites were in the following order:

```
          >*   Beijing
Yokohama                  >** Stockholm
          >**  Zagreb
```

Here, weekly average data for each subject were used for the comparison. Average indoor/outdoor NO_2 concentrations at the 4 sites were in the following orders:

Kitchen	Yokohama, Beijing >** Zagreb >** Stockholm
Living Room	Yokohama >* Beijing, Zagreb >** Stockholm
Bedroom	Yokohama, Beijing >** Stockholm Zagreb >* Yokohama >* Zagreb
Home outdoors	Yokohama, Beijing >** Zagreb, Stockholm

' $p < 0.05$, " $p < 0.01$
* $p < 0.1$, ** $p < 0.01$

Difference in NO_2 concentrations at home outdoors may be mainly due to the size of the city and the residence area of the subjects. In urban areas in Stockholm and Zagreb, averages of NO_2 concentrations at home outdoors were 13 and 16 ppb, respectively, which were higher than the averages for all subjects (see Tables 13 and 15). In Yokohama, in addition to the higher outdoor NO_2 concentration, the influence of indoor emission sources, such as unvented gas-cookers, hot water gas-boilers and heaters, was large, so that indoor NO_2 concentrations became highest among those at four sites. This highest indoor pollution was directly reflected by the highest personal exposure level in the present study. Thus, it was indicated that lifestyle, as well as environmental NO_2 level, is one of the most important factors which affect the personal exposure level at each HEAL site.

Table 20. International comparison of personal NO_2 exposure levels and indoor/outdoor NO_2 concentrations (unit: ppb).

		Beijing	Stockholm	Yokohama	Zagreb
Personal Exposure	av.	18.28	11.43	40.53	17.43
	s.d.	4.49	2.96	21.57	5.21
	max.	27.12	17.57	87.59	29.94
	min.	9.93	7.99	22.22	10.83
	n	15	15	10	16
Kitchen	av.	39.72	5.91	57.91	19.73
	s.d.	14.08	4.99	30.30	13.73
	max.	66.81	15.86	102.13	47.79
	min.	14.47	1.96	19.61	4.24
	n	15	15	10	16
Living Room	av.	13.69	4.95	35.43	14.38
	s.d.	2.72	3.60	28.02	8.39
	max.	16.63	13.84	106.17	35.87
	min.	10.29	1.84	15.09	5.54
	n	4	14	10	14
Bedroom	av.	12.27	5.04	16.87	9.67
	s.d.	5.95	3.96	8.88	6.83
	max.	25.68	13.73	31.09	26.91
	min.	5.71	1.57	6.07	2.29
	n	15	15	10	16
Home Outdoors	av.	21.40	8.92	22.66	11.72
	s.d.	3.59	3.68	3.88	5.89
	max.	26.70	15.49	30.80	22.96
	min.	10.99	3.76	17.85	5.09
	n	15	15	10	16
Workplace	av.	11.89	14.78	19.20	13.20
	s.d.	2.24	3.10	3.42	7.32
	max.	16.94	17.99	21.62	33.39
	min.	7.43	10.16	16.78	5.40
	n	14	5	2	16
Workplace Outdoors	av.	19.65	14.12	23.89	12.04
	s.d.	0.98	1.20	2.93	3.10
	max.	21.57	15.33	25.96	19.26
	min.	17.65	12.94	21.81	8.37
	n	13	3	2	11

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ANALYSIS OF NO_2 BADGE SAMPLERS: QC 2 AND REAL SAMPLE ANALYSIS

REAGENTS

All reagents should be prepared from analytical-grade chemicals (or higher grade chemicals) with nitrite-free water, which can be prepared, if necessary, by redistilling distilled water in an all-glass still after adding a crystal each of potassium permanganate ($KMnO_4$) and barium hydroxide ($Ba[OH]_2$). Most of the reagents are stable for several months if kept well-stoppered in brown glass bottles in a refrigerator. All reagents should be allowed to warm to room temperature before use. Great care should be paid to minimize the contact of reagents with ambient air throughout preparation and storage, which will cause absorption of NO_2 and an unexpected high reagent blank.

1. 0.1 % N-(1-naphthyl)-ethylenediamine dihydrochloride stock solution: Dissolve 0.1 g of the reagent in 100 ml of water. The stock solution can be stored in a refrigerator for up to 3 months.

2. Colour developing reagent: Dissolve 5 g of sulfanilic acid (or 5.52 g of monohydrate) in approximately 800 ml of water in a 1,000 ml volumetric flask. The mixture should be stirred mechanically, preferably by ultrasonification (warming is permissible to speed up the process). To the cooled mixture, add 50 ml of 85% phosphoric acid and 50 ml of the 0.1% stock solution of N-(1-naphthyl)-ethylenediamine dihydrochloride, and made to 1,000 ml. A faint pink colour is commonly observed due to the presence of minute amounts of dissolved nitrite, which usually can be ignored. However, if an intense colour appears due to nitrite impurities in the reagents, or high NO_2 levels in the laboratory, the colour developing reagent should be prepared again. The reagent can be stored in a refrigerator for up to 1 month.

3. Standard sodium nitrite solution, 1.0 g/litre stock solution: A stock solution should be prepared by dissolving accurately weighed 1.0 g of reagent grade sodium nitrite granular crystal in water and made to 1,000 ml in a volumetric flask. Drying of the sodium nitrite is unnecessary. The stock solution should be stable for 3 months.

4. Standard sodium nitrite solution, 0.05 g/litre and 0.006 g/litre working standards: Pipette 5 ml of the stock solution into a 100 ml volumetric flask and make to the mark with water (0.05 g/litre working standard). Pipette 12 ml of the 0.05 g/litre standard solution into a 100 ml volumetric flask and dilute to the mark with water. it is permissible to store these working standard solutions in a refrigerator for up to 1 month.

Concentrations of the working standards have been changed to obtain a calibration curve in the absorbance range of 0-0.6 Abs. QC 1 using NO_2- standard solutions revealed that the calibration curve is not linear at higher absorbances, e.g., at 0.8 Abs. All absorbance measurements are recommended to be carried out at absorbances lower than 0.6 Abs.

CALIBRATION CURVE

Add graduated amounts of working sodium nitrite standard solutions (0.25, 0.5 and 1 ml of 0.006 g/litre standard, 0.25 and 0.5 ml of 0.05 g/litre standard) to a series of 25 ml volumetric flasks, and fill up to the marks with the colour developing reagent. Good results can be obtained with these small volumes of standard solutions if they are carefully pipetted. If preferred, however, larger volumes may be used with correspondingly larger volumetric flasks. Mix well and allow 15 minutes for complete colour development (at room temperature). Measure the absorbances of the coloured solutions and the colour developing reagent blank at 545 nm using distilled water as a reference.

Plot the absorbances against the sodium nitrite concentrations in the coloured solutions. The sodium nitrite concentration in each coloured solution is calculated from:

$$NaNO_2(\mu g/ml) = \frac{W\ g}{1{,}000\ ml} \times \frac{5\ ml}{100\ ml} \times \frac{X\ ml}{25\ ml} \times 10^6$$

$$= 2(W)(X)$$

for the coloured solutions prepared with the 0.05 g/litre standard solution and

$$NaNO_2(\mu g/ml) = \frac{W\ g}{1{,}000\ ml} \times \frac{5\ ml}{100\ ml} \times \frac{12\ ml}{100\ ml} \times \frac{X\ ml}{25\ ml} \times 10^6$$

$$= 0.24(W)(X)$$

for the coloured solutions prepared with the 0.006 g/litre standard solution where:

W = weight of sodium nitrite dissolved to prepare the stock standard solution (g)

X = volume of working standard solution used for the colour development (ml)

Following the Beer's Law, draw the straight line giving the best fit, preferably by linear least squares method, to determine the slope (absorbance per µg/ml of sodium nitrite) and the Y-intercept of the calibration curve.

Figure 1. Structure of the NO_2 badge sampler.

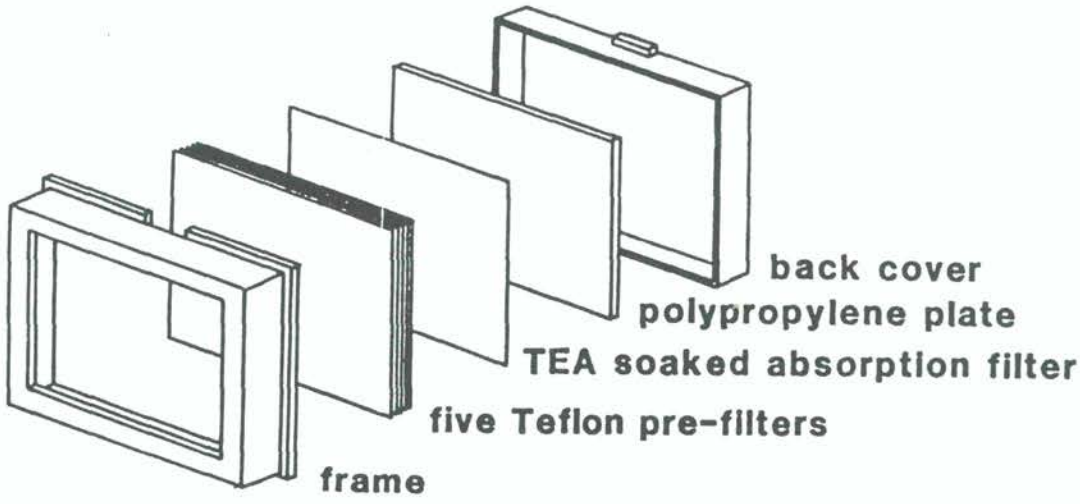
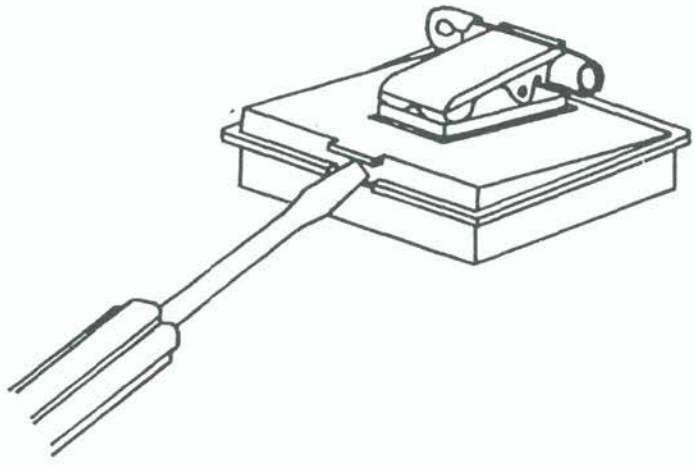


Figure 2. How to open the back cover of the NO_2 badge sampler.



Appendix 1

ANALYTICAL PROCEDURE

Figure 1 shows the structure of NO_2 badge sampler. The sampler has a filter paper soaked with triethanolamine (TEA) for absorption of NO_2 and five Teflon pre-filters for protection of the absorption filter and for minimizing effects of wind on the NO_2 absorption efficiency. Determination of the amount of NO_2 collected on the absorption filter is carried out colorimetrically using a calibration curve with $NaNO_2$ standard solutions. Procedures for the determination are as follows:

(1) Take out a NO_2 badge sampler from the airtightly sealed plastic bag.

(2) Open the back cover of the sampler using a screwdriver or backside of a spatula (Figure 2).

(3) Take out the absorption filter using clean forcepts and put it into a test tube with stopper. At this time, the filter should be pushed deep into the test tube. The filter should be fully dipped in colour developing reagent in procedure (5).

(4) Go back to the procedure (1).

Procedures (1) - (3) should be carried out sample by sample for all samples in one batch of analysis. It may be possible to analyze about 30 samples including blank samples in one batch.

(5) Add a proper volume (10, 20 or 30 ml) or colour developing reagent to each test tube using a volumetric pipette both for extraction of NO_2 in the form of NO_2^- and colour development.

Procedures (1) - (5) should be carried out as quickly as possible to minimize the contact of filters and colour developing reagent to ambient air.

(6) Turn test tubes upside down two or three times for extraction of NO_2, mixing and colour development. This procedure should be repeated several times at intervals of 10 minutes (it takes about 40 minutes to complete the extraction, mixing and colour development). Vigorous shaking will loosen fibres of the filter which will interfere the absorbance measurement.

(7) Measure the absorbance of the coloured sample solutions at 545 nm using distilled water as a reference.

The volume of colour developing reagent in procedure (5) should be changed sample by sample based on an estimated amount of NO_2 collected by each sampler. This is necessary to adjust absorbances of the coloured sample solutions lower than 0.6 Abs. For your convenience, approximate absorbances of the solutions are shown in Table 1 for various sampling and analytical conditions. When the absorbance of a sample solution largely exceed 0.6 Abs. (you can judge by eye), add 10 ml of colour developing reagent (in the case that the total solution volume is 10 or 20 ml), or pipette 10 ml of the coloured sample solution to another test tube and add 10 ml of colour developing reagent (in the case that the total solution volume is 30 ml). If necessary, repeat this dilution procedure. The dilution procedure should be carried out using volumetric pipettes.

The amount of NO_2 collected by each NO_2 badge sampler is calculated by the following equation using the calibration curve obtained through the procedures mentioned in the above section.

$$NO_2\ (10^{-7}\ \text{mole}) = \frac{(A - R) \times V_s - (B - R) \times V_b}{S} \times \frac{10^{-6}}{69.00\ g}$$

where:

A = absorbance of the coloured sample solution (Abs.)

B = absorbance of the coloured blank sample solution (Abs.)

R = absorbance of the colour developing reagent (Abs.)

V_s = volume (ml) of colour developing reagent used for extration and colour development of the sample (dilution factor should be taken into account when the sample solution was diluted for the measurement)

V_b = volume (ml) of colour developing reagent used for extraction and colour development of the blank sample (usually 10 ml, i.e., minimum volume)

S = slope of the calibration curve (Abs.·ml/μg)

69.00 = molecular weight of $NaNO_2$

Appendix 1

Table 1. Approximate absorbances of coloured sample solutions.

Average NO_2 conc. in air (ppb)	Period of sampling (day)	Amount of NO_2 collected (10^{-7} mole)	Volume of colour developing reagent (ml)	Absorbance of coloured sample soln. (Abs.)
10	3	1.5	10	0.6
20	1	1	10	0.4
20	2	2	20	0.4
50	1	2.5	20	0.5
100	1	5	30	0.6

Appendix 1

Calculation of the average NO_2 concentration in air during the sampling

There is a relationship between the amount of NO_2 collected by the NO_2 badge sampler and the average NO_2 concentration in air during the sampling as follows:

$$M = CNO_2 \times K'_{OG} \times A \times t \times \frac{1}{RT} \times 10^{-9}$$

where:

M	=	amount of NO_2 collected by the NO_2 badge sampler (mole)
K'_{OG}	=	collection efficiency (cm/sec)
A	=	area of absorption surface (cm^2)
t	=	period of the sampling (sec)
R	=	gas constant ($cm^3 \cdot atm/mole \cdot K$)
T	=	temperature
CNO_2	=	average NO_2 concentration in air during the sampling (ppb)

This equation can be transformed to:

$$CNO_2 = M \times \frac{RT}{K'_{OG} \cdot A \cdot t} \times 10^9$$

$$= M \times \frac{T}{K'_{OG} \cdot t} \times 8.31 \times 10^9$$

K'_{OG} was determined to be 0.1405 by a NO_2 standard gas exposure experiment by the TCC (temperature = 25°C, relative humidity = 65%). This value agrees well with the reported value (0.14) by Yanagisawa et al.) (temperature = 20°C, relative humidity = 60%, wind velocity = 2 m/sec). K'_{OG} can be assumed constant when the temperature and relative humidity during the sampling are not extremely different from 25°C and 65%, respectively (for most personal and indoor samples, these conditions may be satisfied). However, under some extreme outdoor conditions, e.g., temperature of -5°C, there is a possibility that K'_{OG} is different from 0.1405. Therefore, the TCC requested to carry out (1) measurements of outdoor highest, lowest and average temperatures and (2) a comparative study between the NO_2 badge sampler and well calibrated NO_2 metres under extreme outdoor conditions, when there is a possibility of extreme conditions during the sampling (it is, of course, preferable to carry out these measurements and study in all HEALs).

Reference

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Appendix 2

QUALITY ASSURANCE USING $NaNO_2$ SOLUTIONS

REAGENTS

The general comments on Reagents (1) preparation of 0.1% N-(1-naphthyl)-ethylenediamine dihydrochloride stock solution and (2) colour developing reagent are prepared as in Appendix 1.

(3) Standard sodium nitrite solution, 1.6 g/litre stock solution: A stock solution should be prepared by dissolving accurately weighed 1.6 g of reagent grade sodium nitrite granular crystal to 1,000 ml of water in a volumetric flask. Drying of the sodium nitrite is unnecessary. The stock solution should be stable for 3 months.

(4) Standard sodium nitrite solution, 0.08 g/litre and 0.0096 g/litre working standards: Pipette 5 ml of the stock solution into a 100-ml volumetric flask and fill up to the mark with water (0.08 g/litre working standard). Pipette 12 ml of the 0.08 g/litre standard solution into a 100-ml volumetric flask and dilute to the mark with water. It is permissible to store these working standard solutions in a refrigerator for up to 1 month.

CALIBRATION PROCEDURE

Add graduated amounts of working sodium nitrite standard solutions (0.25, 0.5 and 1 ml of 0.0096 g/litre standard, 0.25 and 0.5 ml of 0.08 g/litre standard) to a series of 25-ml volumetric flasks, and fill up to the marks with the colour developing reagent. Good results can be obtained with these small volumes of standard solutions if they are carefully pipetted. If preferred, however, larger volumes may be used with correspondigly larger volumetric flasks. Mix well and allow 15 minutes for complete colour development (at ordinary temperature). Measure the absorbances of the coloured solutions and the colour developing reagent blank at 545 nm using distilled water as a reference.

Plot the absorbances against the sodium nitrite concentrations in the coloured solutions. The sodium nitrite concentration in each colour developed standard solution is calculated from:

$$NaNO_2(\mu g/ml) = \frac{W\ g}{1{,}000\ ml} \times \frac{5\ ml}{100\ ml} \times \frac{10^6\ \mu g}{g} \times \frac{X\ ml}{25\ ml}$$

$$= 2(W)(X)$$

for the 0.08 g/litre standard solution and

$$NaNO_2(\mu g/ml) = \frac{W\ g}{1{,}000\ ml} \times \frac{5\ ml}{100\ ml} \times \frac{12\ ml}{100\ ml} \times \frac{10^6\ \mu g}{g} \times \frac{X\ ml}{25\ ml}$$

$$= 0.24(W)(X)$$

for the 0.0096 g/litre standard solution where:

W = weight of sodium nitrite dissolved to prepare the stock standard solution (g)

X = volume of working standard solution used for the colour development (ml)

Following the Beer's Law, draw the straight line giving the best fit, preferably by linear least squares method, to determine the slope (absorbance per µg/ml of sodium nitrite) and the Y fraction of the calibration curve.

ANALYTICAL PROCEDURE

Pipette 0.25 ml of a series of $NaNO_2$ solutions for QC to a series of 25-ml volumetric flasks, and fill up to the mark with the colour developing reagent. Mix well and allow 15 minutes for complete colour development. Measure the absorbances of the coloured solutions and the colour developing reagent blank at 545 nm using distilled water as a reference. Determine the $NaNO_2$ concentrations in the coloured solutions using the calibration curve obtained through the procedure mentioned in the above section. The concentration of sodium nitrite in each QC sample is calculated from:

$$NaNO_2\ (\mu g/ml) = \frac{A - B}{S} \times \frac{25\ ml}{0.25\ ml} = \frac{100(A - B)}{S}$$

where:

A = absorbance of the coloured solution
B = absorbance of the colour developing reagent blank
S = slope of the calibration curve (Abs. x ml/µg)

If the absorbance of coloured solution exceeds the linear range of the calibration curve, dilute the solution accurately with the colour developing reagent and measure the absorbance again.

Personal NO_2 exposure levels in Beijing (unit:ppb)

Date	Subj. 1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	a.v.	s.d.	max.	min.	n
3/22-23	15.46	22.49	16.70	10.17	19.57	15.92	18.32	16.02	19.46	16.14	21.78	23.88	9.23	21.16	24.39	18.05	4.49	24.39	9.23	15
23-24	15.27	15.77	20.14	7.66	11.10	15.62	22.42	12.21	17.83	19.18	17.13	17.45	9.28	21.04	22.49	16.31	4.59	22.49	7.66	15
24-25	20.50	15.27	17.83	11.41	7.58	13.36	32.17	16.83	24.93	21.97	18.77	26.81	16.70	21.26	27.46	19.52	6.54	32.17	7.58	15
25-26	15.09	16.95	14.50	13.43	37.78	12.09	27.93	11.97	21.06	21.89	24.31	17.51	9.37	10.41	22.57	18.46	7.64	37.78	9.37	15
26-27	20.94	9.35	21.32	10.79	14.83	26.07	30.04	19.81	27.65	21.43	12.72	24.27	12.09	12.67	25.09	19.27	6.72	30.04	9.35	15
27-28	16.31	19.57	17.89	10.41	17.83	8.95	32.66	17.39	22.65	31.45	29.85	18.32	12.09	31.61	20.69	20.51	7.73	32.66	8.95	15
28-29	14.43	12.59	16.02	5.66	18.20	11.45	26.30	12.17	19.69	18.27	16.51	16.95	11.97	16.57	20.69	15.83	4.82	26.30	5.66	15
av.	16.86	16.00	17.77	9.93	18.13	14.78	27.12	15.20	21.90	21.48	20.15	20.74	11.53	19.25	23.34					
s.d.	2.70	4.32	2.35	2.54	9.66	5.53	5.25	3.11	3.44	4.90	5.68	4.10	2.66	6.98	2.47					
max.	20.94	22.49	21.32	13.43	37.78	26.07	32.66	19.81	27.65	31.45	29.85	26.81	16.70	31.61	27.46					
min.	14.43	9.35	14.50	5.66	7.58	8.95	18.32	11.97	17.83	16.14	12.72	16.95	9.23	10.41	20.69					
n	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7					

Appendix 3

Personal NO_2 exposure levels in Stockholm (unit:ppb)

Date	Subj. 1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	a.v.	s.d.	max.	min.	n
2/29-3/1		10.60	11.10	10.40			10.30		9.10		8.80		11.20			10.21	0.93	11.20	8.80	7
3/1-2		11.50	10.40	9.90			10.80		9.70		11.60		9.80			10.53	0.80	11.60	9.70	7
3/2-3		18.30	15.00	15.40			15.70		13.30		14.00		11.90			14.80	2.03	18.30	11.90	7
3/3-4		14.30	17.90	8.60			12.70		14.40		16.10		14.10			14.01	2.91	17.90	8.60	7
3/4-5		9.60	8.00	4.00			12.10		8.00		7.50		5.80			7.86	2.59	12.10	4.00	7
3/5-6		9.50	3.60	7.00			5.00		8.70		3.70		9.30			6.69	2.59	9.50	3.60	7
3/6-7		13.80	10.60	9.60			9.40		8.20		8.30		11.10			10.14	1.94	13.80	8.20	7
3/10-11	10.20				12.60	21.40		10.50		8.50		9.50		7.10	7.30	10.89	4.61	21.40	7.10	8
3/11-12	13.10				12.80	11.70		3.80		2.60		8.30		3.20	5.00	7.56	4.47	13.10	2.60	8
3/12-13	12.90				13.70	17.50		2.90		1.50		13.80		14.70	3.70	10.09	6.29	17.50	1.50	8
3/13-14	27.30				14.40	13.80		14.20		10.60		17.60		8.90	10.80	14.70	5.78	27.30	8.90	8
3/14-15	16.30				21.00	16.40		11.50		10.10		12.30		10.10	9.50	13.40	4.09	21.00	9.50	8
3/15-16	15.80				25.80	16.50		12.30		11.20		22.00		10.30	11.50	15.68	5.62	25.80	10.30	8
3/16-17	13.40				22.70	15.20		11.30		11.40		11.00		8.50	9.90	12.93	4.45	22.70	8.50	8
av.	15.57	12.51	10.94	9.27	17.57	16.07	10.86	9.50	10.20	7.99	10.00	13.50	10.46	8.97	8.24					
s.d.	5.55	3.18	4.62	3.48	5.45	3.05	3.29	4.36	2.58	4.18	4.20	4.83	2.57	3.49	2.99					
max.	27.30	18.30	17.90	15.40	25.80	21.40	15.70	14.20	14.40	11.40	16.10	22.00	14.10	14.70	11.50					
min.	10.20	9.50	3.60	4.00	12.60	11.70	5.00	2.90	8.00	1.50	3.70	8.30	5.80	3.20	3.70					
n	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7					

Appendix 3

Personal NO_2 exposure levels in Yokohama (unit:ppb)

Date	Subj. 1	2	3	4	5	6	7	8	9	10	av.	s.d.	max.	min.	n
3/11-12	25.27	18.60	49.20	35.92	16.76	28.97	30.22	20.17	82.04	26.78	33.39	19.53	82.04	16.76	10
3/12-13	17.01	25.27	42.95	31.63	15.79	55.74	50.74	31.33	55.47	18.27	34.42	15.81	55.74	15.79	10
3/13-14	27.57	25.31	61.95	35.28	25.24	30.91	56.18	30.15	74.55	29.79	39.69	17.73	74.55	25.24	10
3/14-15	25.87	21.50	54.79	36.60	23.56	30.00	40.02	31.44	73.05	28.89	36.57	16.03	73.05	21.50	10
3/15-16	18.00	29.58	44.80	56.17	18.96	22.61	76.34	25.17	110.72	25.51	42.79	30.36	110.72	18.00	10
3/16-17	23.18	24.08	85.18	100.41	23.40	39.24	74.57	28.74	107.32	19.82	52.59	35.24	107.32	19.82	10
3/17-18	18.66	16.83	50.44	66.70	34.79	27.88	59.80	31.27	109.96	26.50	44.28	28.58	109.96	16.83	10
av.	22.22	23.02	55.62	51.82	22.64	33.62	55.41	28.32	87.59	25.08					
s.d.	4.28	4.37	14.49	25.06	6.47	10.93	16.92	4.23	21.87	4.39					
max.	27.57	29.58	85.18	100.41	34.79	55.74	76.34	31.44	110.72	29.79					
min.	17.01	16.83	42.95	31.63	15.79	22.61	30.22	20.17	55.47	18.27					
n	7	7	7	7	7	7	7	7	7	7					

Appendix 3

Personal NO_2 exposure levels in Zagreb (unit:ppb)

Date	Subj. 1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	a.v.	s.d.	max.	min.	n
3/24-25	19.30	23.60	19.30	15.30													19.38	3.39	23.60	15.30	4
3/25-26	8.70	13.80	30.20	14.40													16.78	9.31	30.20	8.70	4
3/26-27	4.80	14.50	30.60	6.00													13.98	11.89	30.60	4.80	4
3/27-28	6.40	9.50	23.10	14.90													13.48	7.31	23.10	6.40	4
3/28-29	11.30	13.10	20.30	22.00	21.80	15.60	7.70	17.60									16.18	5.21	22.00	7.70	8
3/29-30	14.30	22.00	24.50	25.70	27.50	24.60	16.80	22.60									22.25	4.52	27.50	14.30	8
3/30-31	11.00	15.10	29.80	31.10	28.80	17.00	14.40	15.80									20.38	8.09	31.10	11.00	8
3/31-4/1					33.30	20.50	17.60	23.90	18.40	24.10							22.97	5.74	33.30	17.60	6
4/1-2					34.00	38.60	17.10	20.50	14.20	23.00							24.57	9.68	38.60	14.20	6
4/2-3					39.50	21.20	9.90	19.90	2.50	10.20							17.20	12.95	39.50	2.50	6
4/3-4					24.70	16.70	15.50	22.00	11.90	14.30							17.52	4.86	24.70	11.90	6
4/4-5									12.30	24.50	14.00	18.80	13.60				16.64	5.04	24.50	12.30	5
4/5-6									12.60	12.70	19.00	9.50	17.00				14.16	3.80	19.00	9.50	5
4/6-7									11.10	13.90	18.30	10.40	13.60				13.46	3.11	18.30	10.40	5
4/7-8											16.60	11.00	24.20	15.10			16.73	5.52	24.20	11.00	4
4/8-9											17.80	13.20	9.90	12.90			13.45	3.26	17.80	9.90	4
4/9-10											13.50	4.50	3.90	14.20			9.03	5.58	14.20	3.90	4
4/10-11											18.60	8.50	10.10	14.50			12.93	4.56	18.60	8.50	4
4/11-12														15.70	24.00	24.70	21.47	5.01	24.70	15.70	3
4/12-13														17.90	20.90	22.80	20.53	2.47	22.80	17.90	3
4/13-14														16.80	9.60	17.30	14.57	4.31	17.30	9.60	3
4/14-15															14.80	18.20	16.50	2.40	18.20	14.80	2
4/15-16															13.40	20.30	16.85	4.88	20.30	13.40	2
4/16-17															8.50	11.70	10.10	2.26	11.70	8.50	2
4/17-18															30.60	16.30	23.45	10.11	30.60	16.30	2
av.	10.83	15.94	25.40	18.49	29.94	22.03	14.14	20.33	11.86	17.53	16.83	10.84	13.19	15.30	17.40	18.76					
s.d.	4.91	5.04	4.81	8.36	6.06	7.95	3.86	2.85	4.78	6.09	2.24	4.41	6.36	1.67	8.09	4.33					
max.	19.30	23.60	30.60	31.10	39.50	38.60	17.60	23.90	18.40	24.50	19.00	18.80	24.20	17.90	30.60	24.70					
min.	4.80	9.50	19.30	6.00	21.80	15.60	7.70	15.80	2.50	10.20	13.50	4.50	3.90	12.90	8.50	11.70					
n	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7					

Appendix 4

Summary of activity records of subjects in Beijing

Subject Number	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport/Outdoor (%)***	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
1	13.39	5.75	46.83**	25.69	2.28	6.05	13.39	91.66
2	3.08	23.91	35.32**	30.95	2.78	3.97	3.08	93.26
3	5.95	23.81	36.90**	22.22	0.89	10.22	5.95	88.88
4	2.08		49.31*	37.80	0.00	10.81	2.08	89.19
5	7.34		54.27*	32.54	0.00	5.85	7.34	94.15
6	6.55		61.51*	24.11	0.00	7.84	6.55	92.17
7	10.22	20.14	33.33**	20.04	8.53	7.74	10.22	83.73
8	7.44	23.81	45.73**	16.67	0.10	6.25	7.44	93.65
9	7.34	26.79	32.94**	21.73	1.59	9.62	7.44	88.80
10	6.55		54.66*	30.75	4.37	3.67	6.55	91.96
11	5.26		57.74*	24.90	1.88	10.22	5.26	87.90
12	9.82		62.60*	21.63	0.00	5.95	9.82	94.05
13	5.65		41.96*	26.79	7.74	17.86	5.65	74.40
14	2.98		56.85*	19.84	13.79	6.55	2.98	79.67
15	8.13	24.80	35.71**	22.72	4.46	4.17	8.23	91.36
av.	6.79	21.29	54.86*	38.11**	25.22	3.23	7.78	6.80	88.99
s.d.	2.96	7.13	6.70*	5.75**	5.66	4.01	3.63	2.96	5.69
max.	13.39	26.79	62.60*	46.83**	37.80	13.79	17.86	13.39	94.15
min.	2.08	5.75	41.96*	32.94**	16.67	0.00	3.67	2.08	74.40
n	15	7	8*	7**	15	15	15	15	15

* one room was used as living room and bedroom for these subjects.

** living room and bedroom were separate rooms for these subjects.

*** All subjects moved by bicycle or on foot except for subject 12. Subjects 12 did not specify the time riding on a bus/car.

Appendix 4

Summary of activity records of subjects in Stockholm

Subject Number	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport (%)	Outdoor (%)	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
1	5.85	5.46	43.45	26.39	7.64	1.09	10.12	1.09	81.15
2	8.19	10.79	31.12	23.12	15.90	8.00	2.89	0.58	73.22
3	8.87	15.25	36.49	25.22	3.79	6.28	4.09	2.59	85.83
4	14.83	16.57	38.47	21.18	1.74	3.89	3.32	2.35	91.05
5	6.53		51.04*	26.11	10.68	3.86	1.78	4.08	83.68
6	10.00	4.12	31.18	34.71	12.65	4.71	2.65	2.35	80.01
7	8.65	20.18	33.60	24.45	0.60	7.65	4.87	2.68	86.88
8	17.59	17.30	29.82	23.66	1.59	8.55	1.49	2.24	88.37
9	12.51	6.55	44.09	26.51	1.79	5.96	2.58	0.89	89.66
10	17.13	5.18	36.55	22.31	10.06	5.88	2.89	3.98	81.17
11	8.12	14.74	33.08	25.56	5.89	8.72	3.89	1.50	81.50
12	3.54	5.90	20.35	24.19	33.04	7.08	5.90	0.51	53.98
13	14.70	13.01	33.57	22.54	5.66	6.01	4.52	0.40	83.82
14	12.46	4.75	38.87	12.17	22.55	4.45	4.75	2.97	68.25
15	20.19	10.94	32.52	13.03	15.42	4.53	3.38	3.88	76.68
av.	11.28	10.77	51.04* 34.51	23.41	9.93	5.78	3.94	2.14	80.35
s.d.	4.83	5.46	- 6.00	5.37	9.00	2.08	2.09	1.27	9.52
max.	20.19	20.18	- 44.09	34.71	33.04	8.72	10.12	4.08	91.05
min.	3.54	4.12	- 20.35	12.17	0.60	1.09	1.49	0.40	53.98
n	15	14	1* 14	15	15	15	15	15	15

* one room was used as living room and bedroom for this subject.

Summary of activity records of subjects in Yokohama

Subject Number	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport (%)	Outdoor (%)	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
1	13.29	20.24	31.05	14.19	12.70	1.49	7.04	5.75	78.77
2	21.83	17.16	43.15	3.47	6.85	3.47	4.07	7.64	85.61
3	24.80	26.29	33.43		8.93	0.00	6.55	6.45	84.52
4	25.10	28.27	30.16		3.77	5.06	7.64	9.92	83.53
5	10.19	16.82	27.40	23.44	13.25	1.48	7.42	6.43	77.85
6	14.38	21.03	34.42	25.50	0.00	3.47	1.19	9.52	95.33
7	5.26	19.54	32.94	27.08	8.83	4.37	1.98	4.37	84.82
8	31.65	3.07	51.63		10.88	0.59	2.18	10.68	86.35
9	31.43	11.40	31.33		10.31	6.07	9.47	15.39	74.16
10	8.29	18.98	29.87	24.78	8.39	7.69	2.00	5.09	81.92
av.	18.62	18.28	34.54	19.74	8.39	3.37	4.95	8.12	83.29
s.d.	9.59	7.15	7.34	9.18	4.04	2.49	3.00	3.32	5.76
max.	31.65	28.27	51.63	27.08	13.25	7.69	9.47	15.39	95.33
min.	5.26	3.07	27.40	3.47	0.00	0.00	1.19	4.37	74.16
n	10	10	10	6	10	10	10	10	10

av.	12.21*	28.25**	18.96*	17.26**	33.14*	36.64**			8.34*	8.47**	3.66*	2.93**	3.95*	6.46**	6.47*	10.61**
s.d.	5.76*	3.81**	1.68*	12.09**	5.48*	10.09**			4.80*	3.24**	2.29*	3.08**	2.72*	3.10**	1.87*	3.68**
max.	21.83*	31.65**	21.03*	28.27**	43.15*	51.63**			13.25*	10.88**	7.69*	6.07**	7.42*	9.47**	9.52*	15.39**
min.	5.26*	24.80**	16.82*	3.07**	27.40*	30.16**			0.00*	3.77**	1.48*	0.00**	1.19*	2.18**	4.37*	6.45**
n	6*	4**	6*	4**	6*	4**			6*	4**	6*	4**	6*	4**	6*	4**

* calculations for the group of subjects who had jobs outside home (subjects 1, 2, 5, 6, 7 and 10)
** calculations for the group of housewives (subjects 3, 4, 8 and 9)

Appendix 4

Summary of activity records of subjects in Zagreb

Subject Number	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport (%)	Outdoor (%)	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
1	7.06		55.94*	22.43	8.22	5.19	1.16	4.44	85.43
2	13.39	15.89	35.03	18.69	9.62	6.16	1.22	9.16	83.00
3	6.53	21.23	34.59	18.52	7.18	5.22	6.73	5.62	80.87
4	23.40		41.57*	16.71	7.85	8.30	2.16	12.98	81.68
5	13.80	10.98	37.83	17.51	5.79	12.91	1.19	10.09	80.12
6	5.20	34.62	31.80	19.17	5.05	2.97	1.19	2.13	90.79
7	12.75	7.74	33.33	16.96	17.21	11.11	0.89	6.85	70.78
8	3.85	24.89	34.67	21.63	9.93	5.04	0.00	3.85	85.04
9	15.24	2.22	39.15	22.09	8.04	13.12	0.15	2.12	78.70
10	16.77	13.59	32.84	11.71	11.11	9.38	4.61	5.21	74.91
11	11.21	13.74	33.33	20.39	12.25	6.99	2.08	8.04	78.67
12	21.48	0.00	37.95	12.80	10.52	14.29	2.98	0.00	72.23
13	10.42	15.03	29.22	19.15	13.79	9.28	3.13	7.44	73.82
14	15.97	13.13	32.34	20.00	10.80	5.37	2.39	7.46	81.44
15	3.17	26.19	32.44	21.13	10.07	6.15	0.84	0.00	82.93
16	12.13	19.85	34.11	14.31	12.03	3.81	3.76	6.58	80.40
av.	12.02	15.65	34.19	48.76*	18.32	9.97	7.83	2.15	5.75	80.05
s.d.	5.90	9.32	2.68	-	3.20	3.05	3.51	1.77	3.59	5.21
max.	23.40	34.62	39.15	55.94*	22.43	17.21	14.29	6.73	12.98	90.79
min.	3.17	0.00	29.22	41.57*	11.71	5.05	2.97	0.00	0.00	70.78
n	16	14	14	2*	16	16	16	16	16	16

* one room was used as living room and bedroom for these subjects

Appendix 5

Estimation of personal exposure level from fractional exposures in micro-environments (Beijing)

Subject Number	Kitchen (ppb)	(%)	Living Room (ppb)	(%)	Bedroom (ppb)	(%)	Workplace (ppb)	(%)	Kitchen + Living + Bedroom + Workplace (ppb)	(%)	Personal Exposure (ppb)
1	6.11	(36.2)	-		3.80	(22.6)**	4.35	(25.8)	14.27	(84.6)'	16.86
2	2.06	(12.9)	-		5.10	(31.9)**	2.30	(14.4)	9.46	(59.1)'	16.00
3	1.78	(10.0)	-		2.97	(16.7)**	2.29	(12.9)	7.03	(39.6)'	17.77
4	0.58	(5.9)			3.02	(30.5)*	3.88	(39.0)	7.49	(75.4)	9.93
5	3.07	(16.9)			4.93	(27.2)*	4.11	(22.7)	12.11	(66.8)	18.13
6	4.00	(27.0)			5.34	(36.1)*	2.46	(16.7)	11.80	(79.8)	14.78
7	5.16	(19.0)	3.00	(11.1)	5.50	(20.3)**	2.68	(9.9)	16.34	(60.2)	27.12
8	2.23	(14.7)	2.45	(16.1)	4.86	(31.9)**	1.57	(10.4)	11.11	(73.1)	15.20
9	3.01	(13.8)	3.47	(15.8)	3.73	(17.0)**	2.43	(11.1)	12.64	(57.7)	21.90
10	2.06	(9.6)			8.27	(38.5)*	3.72	(17.3)	14.05	(65.4)	21.48
11	2.60	(12.9)			14.83	(73.6)*	3.30	(16.4)	20.73	(102.9)	20.15
12	2.90	(14.0)			12.79	(61.7)*	2.82	(13.6)	18.51	(89.3)	20.74
13	0.82	(7.1)			2.40	(20.8)*	2.96	(25.7)	6.17	(53.5)	11.53
14	0.84	(4.4)			10.36	(53.8)*	2.52	(13.1)	13.72	(71.3)	19.25
15	3.90	(16.7)	4.12	(17.7)	2.11	(9.0)**	2.88	(12.4)	13.01	(55.8)	23.34
av.	2.74	(14.7)	3.26	(15.2)	7.74	(42.8)*	4.01	(21.3)**	2.95	(17.4)	13.14	(70.9)	18.28
s.d.	1.57	(8.2)	0.71	(2.9)	4.59	(18.4)*	1.22	(8.3)**	0.78	(7.9)	4.11	(14.5)	4.49
max.	6.11	(36.2)	4.12	(17.7)	14.83	(73.6)*	5.50	(31.9)**	4.35	(39.0)	20.73	(102.9)	27.12
min.	0.58	(4.4)	2.45	(11.1)	2.40	(20.8)*	2.11	(9.0)**	1.57	(9.9)	6.17	(53.5)	9.93
n	15		4		8*		7**		15		12		15

- NO_2 concentrations were not measured
' excluded from calculations below
* one room was used as living room and bedroom for these subjects
** living room and bedroom were separate rooms for these subjects

Appendix 5

Estimation of personal exposure level from fractional exposures in micro-environments (Stockholm)

Subject Number	Kitchen (ppb)	(%)	Living Room (ppb)	(%)	Bedroom (ppb)	(%)	Workplace (ppb)	(%)	Kitchen + Living + Bedroom + Workplace (ppb)	(%)	Personal Exposure (ppb)
1	0.92	(5.9)	0.76	(4.9)	4.90	(31.5)	2.68	(17.2)	9.26	(59.5)	15.57
2	0.60	(4.9)	0.80	(6.5)	2.19	(17.8)	4.12	(33.5)	7.71	(62.7)	12.30
3	0.22	(2.0)	0.48	(4.4)	1.12	(10.2)	4.54	(41.5)	6.35	(58.0)	10.94
4	0.59	(6.4)	0.69	(7.4)	1.63	(17.5)	3.64	(39.3)	6.55	(70.6)	9.27
5	1.04	(5.9)			5.57	(31.7)*	3.86	(22.0)	10.47	(59.6)	17.57
6	1.02	(6.3)	0.25	(1.6)	1.97	(12.3)	5.14	(32.0)	8.37	(52.1)	16.07
7	0.26	(2.4)	0.80	(7.4)	0.97	(8.9)	4.40	(40.5)	6.43	(59.3)	10.86
8	0.48	(5.1)	0.50	(5.2)	0.71	(7.4)	3.50	(36.9)	5.19	(54.6)	9.50
9	0.30	(2.9)	0.17	(1.7)	0.69	(6.8)	4.77	(46.7)	5.93	(58.1)	10.20
10	0.34	(4.2)	0.10	(1.2)	0.63	(7.8)	3.07	(38.4)	4.13	(51.7)	7.99
11	0.18	(1.8)	0.42	(4.2)	0.72	(7.2)	4.40	(44.0)	5.71	(57.1)	10.00
12	0.41	(3.0)	0.67	(5.0)	2.79	(20.7)	2.46	(18.2)	6.33	(46.9)	13.50
13	0.47	(4.5)	0.32	(3.0)	0.72	(6.9)	4.05	(38.8)	5.56	(53.1)	10.46
14	0.36	(4.0)	0.13	(1.4)	0.97	(10.8)	1.24	(13.8)	2.69	(30.0)	8.97
15	0.64	(7.8)	0.44	(5.3)	1.18	(14.3)	1.79	(21.7)	4.05	(49.1)	8.24
av.	0.52	(4.5)	0.47	(4.2)	5.57	(31.7)*	1.51	(12.9)	3.58	(32.3)	6.32	(54.8)	11.43
s.d.	0.28	(1.8)	0.25	(2.2)	-		1.18	(7.0)	1.12	(10.8)	2.03	(9.0)	2.96
max.	1.04	(7.8)	0.80	(7.4)	-		4.90	(31.5)	5.14	(46.7)	10.47	(70.6)	17.57
min.	0.18	(1.8)	0.10	(1.2)	-		0.63	(6.8)	1.24	(13.8)	2.69	(30.0)	7.99
n	15		14		1*		14		15		15		15

* one room was used as living room and bedroom for this subject

Appendix 5

Estimation of personal exposure level from fractional exposures in micro-environments (Yokohama)

Subject Number	Kitchen (ppb)	(%)	Living Room (ppb)	(%)	Bedroom (ppb)	(%)	Workplace (ppb)	(%)	Kitchen + Living + Bedroom + workplace (ppb)	(%)	Personal Exposure (ppb)
1	4.97	(22.4)	5.85	(26.3)	4.44	(20.0)	-		15.26	(68.7)'	22.22
2	6.22	(27.0)	3.88	(16.9)	4.52	(19.6)	-		14.62	(63.5)'	23.02
3	17.51	(31.5)	12.32	(22.1)	8.26	(14.9)			38.09	(68.5)	55.62
4	18.40	(35.5)	4.27	(8.2)	9.38	(18.1)			32.04	(61.8)	51.82
5	2.00	(8.8)	2.70	(11.9)	4.25	(18.8)	3.93	(17.4)	12.88	(56.9)	22.64
6	11.38	(33.9)	11.32	(33.7)	2.09	(6.2)	4.28	(12.7)	29.07	(86.5)	33.62
7	5.15	(9.3)	20.75	(37.4)	5.81	(10.5)	5.85	(10.6)	37.56	(67.8)	55.41
8	13.00	(45.9)	0.55	(1.9)	4.63	(16.4)			18.18	(64.2)	28.32
9	32.10	(36.6)	2.79	(3.2)	9.36	(10.7)			44.25	(50.5)	87.59
10	2.43	(9.7)	4.26	(17.0)	2.98	(11.9)	4.16	(16.6)	13.83	(55.1)	25.08
av.	11.32	(26.1)	6.87	(17.9)	5.57	(14.7)	4.56	(14.3)	28.24	(63.9)	40.53
s.d.	9.39	(13.1)	6.14	(12.1)	2.58	(4.7)	0.88	(3.2)	11.95	(11.1)	21.57
max.	32.10	(45.9)	20.75	(37.4)	9.38	(20.0)	5.85	(17.4)	44.25	(86.5)	87.59
min.	2.00	(8.8)	0.55	(1.9)	2.09	(6.2)	3.93	(10.6)	12.88	(50.5)	22.22
n	10		10		10		4		8		10

av.	5.36	(18.5)*	20.25	(37.4)**	8.13	(23.9)*	4.98	(8.9)**	4.02	(14.5)*	7.91	(15.0)**	23.34	(66.6)*	33.14	(61.3)**	30.33*	55.84**
s.d.	3.38	(10.8)*	8.24	(6.1)**	6.89	(10.3)*	5.12	(9.3)**	1.30	(5.8)*	2.25	(3.2)**	12.04	(14.4)*	11.15	(7.7)**	13.01*	24.37**
max.	11.38	(33.9)*	32.10	(45.9)**	20.75	(37.4)*	12.32	(22.1)**	5.81	(20.0)*	9.38	(18.1)**	37.56	(86.5)*	44.25	(68.5)**	55.41*	87.59**
min.	2.00	(8.8)*	13.00	(31.5)**	2.70	(11.9)*	0.55	(1.9)**	2.09	(6.2)*	4.63	(10.7)**	12.88	(55.1)*	18.18	(50.5)**	22.22*	28.32**
n	6*		4**		6*		4**		6*		4**		4*		4**		6*	4**

- NO_2 concentrations were not measured
' excluded from calculations below
* calculations for the group of subjects who had jobs outside home (subjects 1, 2, 5, 6, 7 and 10)
** calculations for the group of housewives (subjects 3, 4, 8 and 9)

Appendix 5

Estimation of personal exposure level from fractional exposures in micro-environments (Zagreb)

Subject Number	Kitchen (ppb)	(%)	Living Room (ppb)	(%)	Bedroom (ppb)	(%)	Workplace (ppb)	(%)	Kitchen + Living + Bedroom + Workplace (ppb)	(%)	Personal Exposure (ppb)
1	0.30	(2.8)			2.59	(23.9)*	2.37	(21.9)	5.26	(48.6)	10.83
2	3.39	(21.2)	3.45	(21.7)	1.50	(9.4)	2.32	(14.6)	10.66	(66.9)	15.94
3	2.48	(9.7)	7.62	(30.0)	9.31	(36.6)	1.95	(7.7)	21.35	(84.0)	25.40
4	2.05	(11.1)			1.26	(6.8)*	2.88	(15.6)	6.18	(33.4)	18.49
5	6.60	(22.0)	2.25	(7.5)	7.11	(23.8)	4.83	(16.1)	20.78	(69.4)	29.94
6	2.46	(11.2)	5.80	(26.3)	3.70	(16.8)	2.42	(11.0)	14.37	(65.2)	22.03
7	1.81	(12.8)	1.01	(7.2)	4.32	(30.6)	1.63	(11.5)	8.78	(62.1)	14.14
8	0.91	(4.5)	4.76	(23.4)	5.49	(27.0)	2.25	(11.1)	13.41	(66.0)	20.33
9	2.37	(20.0)	0.12	(1.0)	0.90	(7.6)	2.19	(18.5)	5.58	(47.0)	11.86
10	3.17	(18.8)	1.12	(6.6)	2.76	(16.4)	0.63	(3.8)	7.67	(45.6)	16.83
11	0.61	(5.6)	0.91	(8.4)	2.12	(19.6)	1.57	(14.5)	5.21	(48.1)	10.84
12	1.53	(11.6)	0.00	(0.0)	0.88	(6.6)	4.27	(32.4)	6.68	(50.7)	13.19
13	1.77	(11.6)	2.64	(17.3)	4.19	(27.4)	1.82	(11.9)	10.42	(68.1)	15.30
14	1.88	(10.8)	0.95	(5.5)	1.53	(8.8)	2.13	(12.2)	6.49	(37.3)	17.40
15	0.37	(2.0)	3.46	(18.4)	3.07	(16.4)	1.99	(10.6)	8.89	(47.4)	18.76
16	2.33	(13.3)	2.06	(11.8)	2.95	(16.9)	2.05	(11.7)	9.40	(53.6)	17.53
av.	2.13	(11.8)	2.58	(13.2)	1.93	(15.4)*	3.56	(18.8)	2.33	(14.1)	10.07	(55.8)	17.43
s.d.	1.50	(6.3)	2.24	(9.6)	-		2.42	(9.2)	1.00	(6.4)	5.08	(13.5)	5.21
max.	6.60	(22.0)	7.62	(30.0)	2.59	(23.9)*	9.31	(36.6)	4.83	(32.4)	21.35	(84.0)	29.94
min.	0.30	(2.0)	0.00	(0.0)	1.26	(6.8)*	0.88	(6.6)	0.63	(3.8)	5.21	(33.4)	10.83
n	16		14		2*		14		16		16		16

* one room was used as living room and bedroom for these subjects

Appendix 6

Environmental monitoring data in Beijing (high exposure area, chemiluminescence)

Date	NO_2 (ppb)*	NO_2 (ppb)**	NO (ppb)*	NO (ppb)**	Temp. (°C)	wind (m/s)*
3/22-23	28.54	27.42	12.71	13.37	-3 - 5	3.4
3/23-24	27.71	26.56	11.69	10.21	0 - 10	3.3
3/24-25	35.60	40.70	25.99	47.80	-1 - 6	3.3
3/25-26	28.58	24.62	34.18	16.63	0 - 12	3.3
3/26-27	40.41	46.31	31.59	42.30	3 - 15	3.3
3/27-28	32.24	24.51	28.23	13.04	0 - 13	3.4
3/28-29	27.21	26.57	8.83	9.75	3 - 12	3.4
3/29-30	30.11		15.15		3 - 10	3.4
3/30-31	42.15		35.87		3 - 11	3.4
av.	32.51	30.95	22.69	21.87		
s.d.	5.62	8.79	10.58	16.07		
max.	42.15	46.31	35.87	47.80		
min.	27.21	24.51	8.83	9.75		
n	9	7	9	7		

* averages of hourly data from 1 a.m. to 23 p.m.
** averages of hourly data from 8 a.m. to 7 a.m. (corresponding to the time period of exposure monitoring)

Appendix 6

Environmental monitoring data in Stockholm (high exposure area, chemiluminescence)

Date	NO_2 (ppb)*	NO_2 (ppb)**	NO (ppb)*	Temp. (°C)*	Wind (m/s)*
2/29-3/1	14	13.5	7.3	-1.7	5.0
3/1-2	15	15.6	9.0	-4.4	3.8
3/2-3	23	29.9	24.7	-5.6	3.1
3/3-4	24	13.1	19.4	-4.1	3.4
3/4-5	15	10.7	5.0	0.6	4.8
3/5-6	12	9.6	4.3	1.5	3.7
3/6-7	12	18.6	5.2	1.1	2.4
3/7	19		12.0	0.4	2.6
3/10-11	20	13.6	8.2	-1.1	3.7
3/11-12	14	10.7	6.7	1.0	4.3
3/12-13	13	15.3	5.2	-2.1	2.9
3/13-14	19	22.2	7.4	-3.6	2.0
3/14-15	24	20.7	14.2	-4.0	3.1
3/15-16	27	31.5	20.2	-4.5	3.0
3/16-17	27	15.4	31.4	-3.8	3.3
3/17	16		6.6	-3.4	3.8
av.	18.4	17.2	11.7		
s.d.	5.3	6.8	8.1		
max.	27.0	31.5	31.4		
min.	12.0	9.6	4.3		
n	16	14	16		

* averages of hourly data from 0 a.m. to 23 p.m.
** averages of hourly data from 3 p.m. to 2 p.m.
(corresponding to the time period of exposure monitoring)

Appendix 6

Environmental monitoring data in Yokohama (chemiluminescence)

Date	Urban High Exposure Area NO_2 (ppb)*	NO_2 (ppb)**	wind (m/s)*	Urban Low Exposure Area NO_2 (ppb)*	NO_2 (ppb)**	Temp. (°C)*	wind (m/s)*
3/11-12	42	28.5	4.3	17	6.3	11.6	4.6
3/12-13	28	29.1	3.8	7	17.1	15.1	5.3
3/13-14	32	58.9	2.3	19	32.1	11.5	2.4
3/14-15	56	32.5	2.1	31	26.5	13.3	2.7
3/15-16	32	26.0	3.8	27	16.4	9.9	3.9
3/16-17	28	50.5	4.0	16	37.0	6.8	4.3
3/17-18	50	33.2	3.0	39	27.3	4.2	3.9
3/18	35		3.2	30		7.0	4.1
av.	37.9	37.0		23.3	23.2		
s.d.	10.5	12.6		10.3	10.5		
max.	56	58.9		39	37.0		
min.	28	26.0		7	6.3		
n	8	7		8	7		

* averages of hourly data from 0 a.m. to 23 p.m.
** averages of hourly data from 22 p.m. to 21 p.m.
(corresponding to the time period of exposure monitoring)

Appendix 6

Environmental monitoring data in Zagreb (badge method)

Date	NO_2 (ppb)*	NO_2 (ppb)**	temp. (°C)	wind (m/s)
3/24-28	19.7	16.3	-1 - 16	1 - 5
3/28-31	34.9	18.0	-1 - 17	1 - 9
3/31-4/4	23.2	20.2	2 - 18	1 - 7
4/4-7	27.4	11.1	9 - 14	1 - 9
4/7-11	20.1	12.5	-2 - 18	1 - 8
4/11-14	19.9	15.0	2 - 22	1 - 9
4/14-18	26.4	15.2	-1 - 18	1 - 7
av.	24.5	15.5		
s.d.	5.6	3.1		
max.	34.9	20.2		
min.	19.7	11.1		
n	7	7		

* high exposure area (urban, commercial/residential), from 2:30 p.m. to 2:30 p.m.
** low exposure area (suburban, residential), from 1:30 p.m. to 1:30 p.m.

EXPOSURE MONITORING OF NITROGEN DIOXIDE IN INDIA

An international pilot study within the WHO/UNEP Human Exposure Assessment Locations (HEAL) Programme

SECTION II

Technical report edited by

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2. MATERIALS AND METHODS

2.1 Exposure monitoring

Bombay, a commercial capital of India, is one of the HEAL sites for this project, with a population of 8.2 million and having numbers of small and large scale industries and high traffic density. Exposure monitoring was carried out with the help of passive monitor, i.e., NO_2 badges. The sampling scheme is given in Table 1 in Section I.

2.1.1 Selection of subjects

To estimate personal exposures and environmental levels of NO_2 prevailing in Bombay, 17 subjects and four sites were selected. The subjects were selected randomly to carry personal monitors. The only criteria used for their selection in the study were; (1) person should reside in the vicinity of our monitoring site, and (2) person should be non-smoker. The subjects were healthy adults, 24-50 years of age. Out of 17 subjects, three were men working and others were either women working or housewives.

2.2 Environmental monitoring

The four air monitoring sites were selected as follows: two sites were located in city and two sites were located in suburbs. This high and low exposure sites of city and suburbs were covered.

Although all four sites were selected, two in city and two in suburbs, there is not much difference between city and suburbs. However, in the city, pollution is mostly due to automobiles and other commercial activities, where in the suburbs, it is an industrial pollution, as a large petrochemical complex is located in suburbs. Environmental monitoring is carried out manually by impingement technique using TGS-ANSA procedure. The four-hourly data were collected for seven consecutive days.

2.3 Quality assurance

Prior to the implementation of the pilot monitoring, analytical quality assurances were carried out in collaboration with TCC (see Section I - Exposure Monitoring of NO_2 in China, Japan, Sweden and Yugoslavia). Quality assurance by cross check with the TCC using duplicate samples was also carried out to ensure reliability/comparability of the monitoring data (Appendix 1).

A trial study on the performance of badges with respect to TGS-ANSA method was carried out for one week at a highly polluted site in suburbs. NO_2 was monitored by badges for one day, two days, three days, ... seven days, and NO_2 was monitored by TGS-ANSA method for seven days continuously collecting four-hourly data (see Table 1).

Table 1. NO_2 levels measured by two different methods at ambient air monitoring site Maravli Chembur.

Sample Number	NO_2 badge method		TGS-ANSA method
	period of exposure	NO_2 conc. (ppb)	NO_2 conc. (ppb)
1	24 hours	30.3	37.2
2	48 hours	28.8	43.1
3	72 hours	31.4	50.5
4	96 hours	33.4	59.1
5	120 hours	33.1	58.5
6	144 hours	33.9	58.5
7	168 hours	34.5	58.0

On evaluating the performances of these two methods, it was observed that (1) TGS-ANSA method recorded higher levels of NO_2, (2) the correlation between the data by two methods was insignificant at 90% probability.

3. RESULTS AND DISCUSSIONS

3.1 Personal exposure to NO_2

Personal exposure levels to NO_2 observed in Bombay are shown in Table 2. In Bombay, daily personal exposure level to NO_2 ranged from 8 to 64 ppb (average = 29.61 ± 12.95 ppb). The difference between personal and daily variations could not be evaluated statistically because of the complicated sampling schedule as can be seen from Appendix 2. However, the data obtained suggested that the difference may be small. Detailed data on personal NO_2 exposure at Bombay site are given in Appendix 2.

3.2 Indoor/outdoor NO_2 concentrations

Table 3 shows weekly averaged indoor/outdoor NO_2 concentrations at Bombay site. This table also shows personal NO_2 exposure data, information on indoor emission sources and residing area for each subject. In Bombay, averaged indoor/outdoor NO_2 concentrations at home were in the following order:

Kitchen >** Outdoor, Living Room >** Bedroom

** $p < 0.01$

Table 2. Personal NO_2 exposure levels of HEAL site, Bombay, (unit:ppb).

Statistics for all data	av.	29.61
	s.d.	12.95
	max.	63.97
	min.	8.19
	n	118
Average data for each subject	av.	29.55
	s.d.	11.72
	max.	50.38
	min.	12.14
	n	17
Average data for each sampling date	av.	29.62
	s.d.	2.37
	max.	32.59
	min.	25.19
	n	7

The averaged NO_2 concentration in kitchen was higher than those in the other rooms and outdoor air because of the use of unvented gas/kerosene cookers in 16 homes. For subject 5, the average concentration of NO_2 in kitchen was lower than that in outdoor air. This may be attributed to the fact that polluted outdoor air is restricted to flow in at this residence. The NO_2 concentrations in workplaces of subject 1, 4 and 5 were lower than those in kitchens, living rooms, bedrooms and home outdoor air, where as for subject 2, NO_2 concentration in workplace was lower than that in kitchen, living room, home outdoor air but higher than that in bedroom, and for subject 3, NO_2 concentration in workplace was lower than that in kitchen only but higher than those in living room, bedroom and home outdoor air. NO_2 concentration in workplace was also lower than that in outdoor air for all the subjects except for subject 3, where NO_2 concentration in workplace was higher than that in workplace outdoor air, since his workplace is autogarage.

Table 4 lists correlations coefficients between NO_2 concentrations at home indoors/outdoors. Significant correlation between NO_2 concentrations indoors and outdoors was observed. This observation clearly demonstrates the influence of outdoor NO_2 concentration on indoor NO_2 level as open doors and windows is a common practice in a tropical region. Significant correlation between NO_2 concentrations in kitchen and living room suggests that the diffusion of indoor pollutants from kitchen to living room occurred. Thus, significant correlations were observed between all indoor NO_2 levels.

Table 3. Indoor/outdoor NO_2 concentrations (weekly data, unit:ppb) in Bombay.

Subject Number	Personal Exposure	Home				Workplace		Fuel		Residing Area
		Kitchen	Living Room	Bedroom	Outdoor	Indoor	Outdoor	Cooking	Heating	
1	19.80	66.90	33.20	30.30	34.40	21.20	22.20	G	Nil	suburban
2	24.74	76.65	32.39	22.80	30.56	22.10	35.40	G	Nil	suburban
3	23.10	66.30	18.20	25.00	28.30	42.90	24.30	G	Nil	suburban
4	17.10	36.20	26.00	19.40	27.45	15.90	25.61	G	Nil	suburban
5	17.50	28.20	18.30	18.33	35.70	15.19	24.79	G	Nil	suburban
6	33.90	41.30	45.50*	-	27.10	-	-	G	Nil	suburban
7	28.63	25.34	24.05*	-	24.87	-	-	G/K	Nil	suburban
8	12.14	46.83	18.19	10.32	19.46	-	-	G	Nil	suburban
9	24.73	26.82	22.10	29.12	25.81	-	-	G	Nil	suburban
10	25.80	36.40	23.10	28.40	25.60	-	-	G	Nil	suburban
11	50.29	82.06	51.98*	-	61.08	-	-	G/K	Nil	urban
12	33.76	46.01	41.05	35.86	50.88	-	-	G/K	Nil	urban
13	43.89	72.58	43.81	46.20	39.08	-	-	G/K	Nil	urban
14	50.38	72.53	52.30	42.88	39.50	-	-	G	Nil	urban
15	45.50	53.72	38.02	49.38	40.73	-	-	G/K	Nil	urban
16	26.62	45.82	31.59*	-	27.05	-	-	G	Nil	urban
17	24.44	46.06**	-	-	25.53	-	-	G/K	Nil	urban
av.	29.55	51.16	32.49	29.83	33.12	23.46	26.46			
s.d.	11.72	18.42	11.82	11.89	10.60	11.30	5.15			
max.	50.38	82.06	52.30	49.38	61.08	42.90	35.40			
min.	12.14	25.34	18.20	10.32	19.46	15.19	22.20			
n	17	17	16	12	17	5	5			

fuel: G = gas, K = kerosene

* Living room and bedroom were same for these subjects.

** Kitchen, living room and bedroom were one and same for this subject.

Table 4. Correlation between indoor/outdoor NO_2 concentrations.

	Kitchen	Living Room	Bedroom	Outdoor
Bombay				
Living Room	0.59**			
Bedroom	0.46*	0.81***		
Outdoor	0.53*	0.61**	0.59**	
Personal exposure	0.51*	0.79***	0.85***	0.71***

* $p < 0.1$
** $p < 0.01$
*** $p < 0.001$

3.3 Estimation of personal exposure level to NO_2

The mean level of personal NO_2 exposure can be defined as the time-weighted average of NO_2 concentrations in various environments in which the subject spends his time and is exposed to pollutants including NO_2. We live in various kinds of indoor environments, that is micro-environments, for 70-95% of a day. These micro-environments are generally polluted by NO_2 as described in section 3.2. Therefore, personal NO_2 exposure levels can be estimated approximately by the combination of NO_2 concentrations and spending times in these micro-environments. This approach for the estimation will also provide the estimated value on the contribution of each micro-environment to the personal NO_2 exposure, which is important to find out the exposure characteristics and also to make a countermeasure for personal NO_2 exposure.

The time schedule study for each subject was carried out for the same period of the exposure monitoring study. Summary of activity records of the subjects at the Bombay site is shown in Table 5. Average value of the total spending time in the micro-environments, i.e., kitchen, living room, bedroom and workplace, accounted for 70-95% of a day. The time spending in the kitchen, where NO_2 concentration is generally considered to be high, accounted for 8-21% of a day for the working women and 8-37% of a day for the housewives in this study. Detailed information on activity records of the subjects at Bombay site is given in Appendix 3.

Table 5. Summary of activity records of subjects at HEAL site in Bombay.

	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport (%)	Outdoor (%)	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
av.	21.92	24.25	42.75	35.00	6.20	5.56	5.45	12.15	87.39
s.d.	20.99	21.68	32.57	10.46	3.10	4.09	1.98	7.92	6.52
max.	91.70	75.00	62.50	50.01	12.40	12.50	8.33	29.10	95.80
min.	2.08	6.30	25.00	20.83	2.10	2.08	2.10	4.10	70.82
n	17	14	14	5	15	6	13	14	17

Table 6. Estimation of personal exposure level from fractional exposures in micro-environments.

		Subject Group I*		Subject Group II**	
		(ppb)	(%)	(ppb)	(%)
Kitchen	av.	16.75	51.01	3.36	11.86
	s.d.	13.70	47.09	4.36	22.08
	max.	46.48	190.18	11.15	56.31
	min.	3.44	10.15	1.18	5.97
	n	12		5	
Living Room	av.	12.66	38.23	3.50	16.94
	s.d.	11.52	30.00	2.49	11.61
	max.	34.13	10.00	6.91	34.90
	min.	3.72	7.38	1.52	6.38
	n	10		5	
Bedroom	av.	19.12	53.27	7.02	32.65
	s.d.	10.96	13.13	2.15	11.15
	max.	35.66	78.37	10.10	51.00
	min.	4.40	36.24	4.85	23.00
	n	7		5	
Workplace	av.			9.16	42.76
	s.d.			7.16	28.60
	max.	N.A.		21.45	92.86
	min.			4.42	22.30
	n			5	
Kitchen + Living + Bedroom + Workplace	av.	39.64	114.15	23.05	105.21
	s.d.	18.86	34.85	9.38	35.52
	max.	66.93	190.18	32.68	164.51
	min.	13.64	51.24	13.68	79.53
	n	12		5	
Observed personal exposure value	av.	33.35		20.46	
	s.d.	11.94		3.37	
	max.	50.49		24.74	
	min.	12.14		17.10	
	n	12		5	

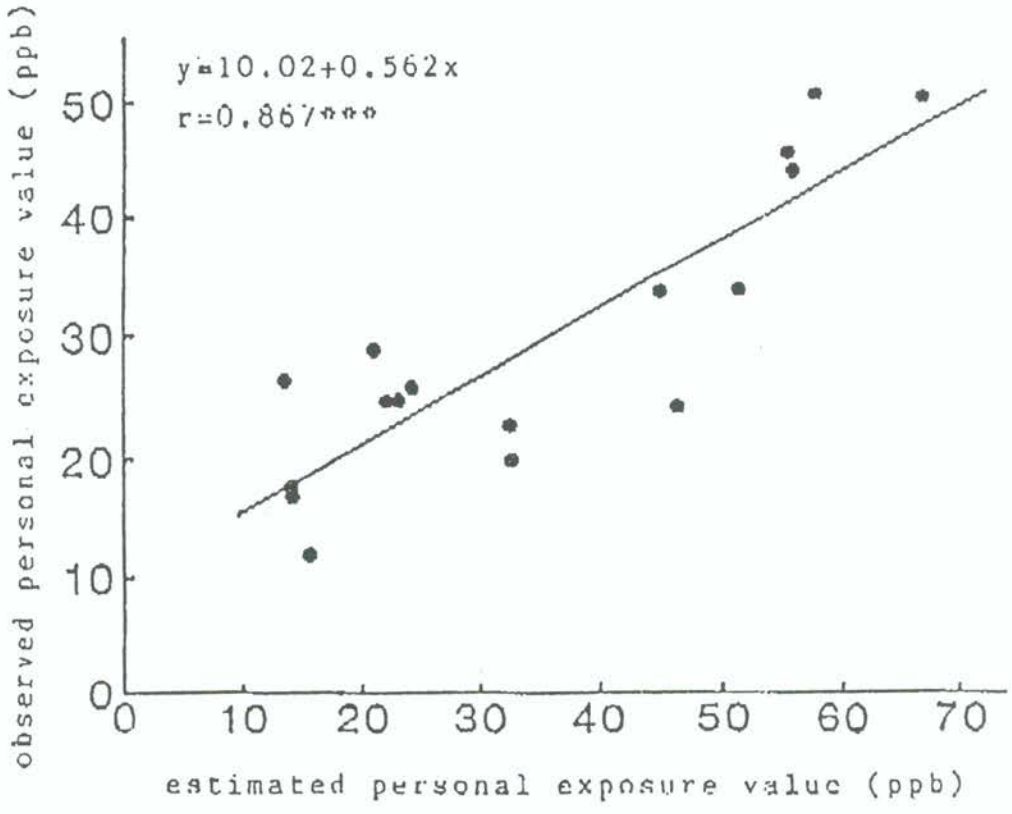
* Calculations for the subjects who did not have jobs outside home.

** Calculations for the subjects who had jobs outside home.

Table 6 shows averages of estimated fractional exposure values in micro-environments and estimated NO_2 personal exposure value (total of the fractional values) together with those of observed personal NO_2 exposure values. The estimated values were calculated for each subject from NO_2 concentrations and spending times in kitchen, living room, bedroom and workplace. The estimated personal exposure values were higher than observed values for 10 subjects, and for 7 subjects, it was vice versa, i.e., some were overestimated and some were underestimated.

Correlation between the estimated personal exposure values and the observed values at Bombay was significant. The correlation coefficient was 0.87 and p value was less than 0.001 (see Figure 1).

Figure 1. *Correlation between estimated personal exposure values and observed personal exposure values at HEAL site in Bombay.*



*** $p < 0.001$

A significant correlation was observed between the observed personal exposure values and estimated fractional exposure values in the following micro-environments:

Kitchen ($p < 0.1$)
Bedroom ($p < 0.001$)

In Bombay, significant correlation between the observed personal exposure values and NO_2 concentrations indoors/outdoors was also observed (see Table 4).

Detailed data on the estimation of personal exposure level from fractional exposures in various micro-environments are given in Appendix 4.

Following conclusions may be suggested from the results in the pilot exposure monitoring in Bombay:

1. The personal NO_2 badge method is useful for the estimation of personal exposure level of individual in the HEAL project.

2. The predominant factor affecting personal NO_2 exposure level is indoor pollution. The weight of indoor pollution to the personal exposure level may not vary as per the seasons when considering the small variations in climate in Bombay. Perhaps during summer rains, indoor pollution may exert a greater influence on personal exposure.

3. There was significant correlation between the observed personal exposure values and the personal exposure values estimated from NO_2 concentrations/spending times indoors may be useful for the first-level estimation of NO_2 exposure level of the general population concerned.

4. It is desirable that better methodology be developed for accurate estimation of personal exposure level from NO_2 concentrations/spending times in various micro-environments. Such improvements would include the development of more sensitive monitoring, sampling and recording methods to evaluate NO_2 exposure in each micro-environment.

5. The personal badge has been used on randomly selected subjects to monitor NO_2 exposures, and it has proved successful. The method can be adopted to monitor general population exposure to NO_2 irrespective of the sources of NO_2 to the target population.

3.4 Environmental monitoring

Environmental NO_2 data obtained at fixed monitoring stations in Bombay are shown in Table 7 together with corresponding personal exposure data and outdoor NO_2 data obtained in the exposure monitoring. Detailed data on the environmental monitoring are given in Appendix 5.

Table 7A. Environmental NO_2 concentrations at four monitoring sites (unit: ppb) in Bombay.

	Urban		Suburban	
	High	Low	High	Low
av.	38.48	27.20	60.10	32.48
s.d.	6.86	7.93	10.84	5.42
max.	53.00	40.00	75.40	39.6
min.	32.20	14.70	43.30	21.3
n	8	8	8	8

Table 7b. Averaged personal NO_2 exposure levels in four different polluted areas (unit: ppb) in Bombay.

	Urban		Suburban	
	High	Low	High	Low
av.	44.79	25.20	20.40	25.03
s.d.	9.28	4.23	5.92	9.99
max.	63.97	32.31	35.85	47.66
min.	27.85	16.19	9.33	8.19
n	35	14	34	35

Table 7c. NO_2 concentrations home outdoors in four different polluted areas (unit: ppb) in Bombay.

	Urban		Suburban	
	High	Low	High	Low
av.	46.25	26.29	31.28	24.57
s.d.	15.52	3.18	4.66	2.90
max.	87.25	29.21	35.83	28.47
min.	40.53	21.85	27.35	19.06
n	10	4	10	10

Environmental NO_2 data did not agree well with home outdoor NO_2 data obtained in the exposure monitoring. Environmental data measured by TGS-ANSA method were higher than home outdoor data measured by NO_2 badge method. In the pilot monitoring, environmental NO_2 was not measured by the badge method simultaneously due to shortage of badges. However, TGS-ANSA method recorded higher levels than the badge method in a trial study as mentioned in Section 2.

Significant correlation was observed between environmental NO_2 personnel exposure level in urban low exposure area and suburban high exposure area. Correlation coefficients were 0.84** and 0.73**, respectively. The correlation suggests that environmental NO_2 concentration is affecting the personal exposure level of people in these areas as is suggested by correlation between personal NO_2 exposures and home outdoor NO_2 concentrations. In urban high exposure area and suburban low exposure area, the correlation was insignificant.

3.5 International comparison

The subjects in the pilot monitoring study were randomly selected. The only criteria used for their selection in the study were: (1) person should reside in the vicinity of our monitoring site, and (2) person should be a non-smoker. The subjects were healthy adults, 24-50 years of age. Out of 17 subjects, three were men working, two were women working and other 12 were housewives. Out of these subjects, 10 were residing in high exposure area.

Table 8 shows the results of international comparison. Averaged personal NO_2 exposure levels at five HEAL sites were in the following order:

Yokohama >' Beijing
Yokohama >'' Zagreb
Yokohama >'' Stockholm

Bombay >'' Beijing, Zagreb

Here, weekly averaged data for each subject was used for the comparison. Averaged indoor/outdoor NO_2 concentrations at HEAL sites were in the following orders:

Kitchen

Yokohama, Bombay, Beijing >'' Zagreb >'' Stockholm
Bombay >' Beijing

** $p < 0.01$
' $p < 0.1$, '' $p < 0.01$

Living Room

Yokohama >'
Zagreb, Beijing >'' Stockholm
Bombay >''

Bedroom

Yokohama, Beijing >''
Bombay >'' Stockholm
Zagreb >'

Yokohama >' Zagreb

Home Outdoors

Bombay >'' Yokohama, Beijing >'' Zagreb, Stockholm

Difference in NO_2 concentrations at home outdoors may be mainly due to the size of the city. In Bombay, in addition o the higher outdoor NO_2 concentration, influence of indoor emission sources, such as unvented gas and kerosene cookers was large, so that indoor NO_2 concentrations became highest among those at the five HEAL sites. This highest indoor pollution was directly reflected in the second highest personal exposure level in the pilot study. Thus, it was indicated that lifestyle as well as environmental NO_2 level is one of the most important factors which affect the personal exposure level at each HEAL site.

' $p < 0.1$, '' $p < 0.01$

Table 8. International comparison of personal NO_2 exposure levels and indoor/outdoor NO_2 (unit: ppb) concentrations.

		Beijing	Stockholm	Yokohama	Zagreb	Bombay
Personal Exposure	av.	18.28	11.43	40.53	17.43	29.55
	s.d.	4.49	2.96	21.57	5.21	11.72
	max.	27.12	17.57	87.59	29.94	50.38
	min.	9.93	7.99	22.22	10.83	12.14
	n	15	15	10	16	17
Kitchen	av.	39.72	5.91	57.91	19.73	51.16
	s.d.	14.08	4.99	30.30	13.73	18.42
	max.	66.81	15.86	102.13	47.79	82.06
	min.	14.47	1.96	19.61	4.24	25.34
	n	15	15	10	16	17
Living Room	av.	13.69	4.95	35.43	14.38	32.49
	s.d.	2.72	3.60	28.02	8.39	11.82
	max.	16.63	13.84	106.17	35.87	52.30
	min.	10.29	1.84	15.09	5.54	18.20
	n	4	14	10	14	16
Bedroom	av.	12.27	5.04	16.87	9.67	29.83
	s.d.	5.95	3.96	8.88	6.83	11.89
	max.	25.68	13.73	31.09	26.91	49.38
	min.	5.71	1.57	6.07	2.29	10.32
	n	15	15	10	16	12
Home Outdoors	av.	21.40	8.92	22.66	11.72	33.12
	s.d.	3.59	3.68	3.88	5.89	10.60
	max.	26.70	15.49	30.80	22.96	61.08
	min.	10.99	3.76	17.85	5.09	19.46
	n	15	15	10	16	17
Workplace	av.	11.89	14.78	19.20	13.20	23.46
	s.d.	2.24	3.10	3.42	7.32	11.30
	max.	16.94	17.99	21.62	33.39	42.90
	min.	7.43	10.16	16.78	5.40	15.19
	n	14	5	2	16	5
Workplace Outdoors	av.	19.65	14.12	23.89	12.04	26.46
	s.d.	0.98	1.20	2.93	3.10	5.15
	max.	21.57	15.33	25.96	19.26	35.40
	min.	17.65	12.94	21.81	8.37	22.20
	n	13	3	2	11	5

RESULTS OF DUPLICATE SAMPLE ANALYSIS

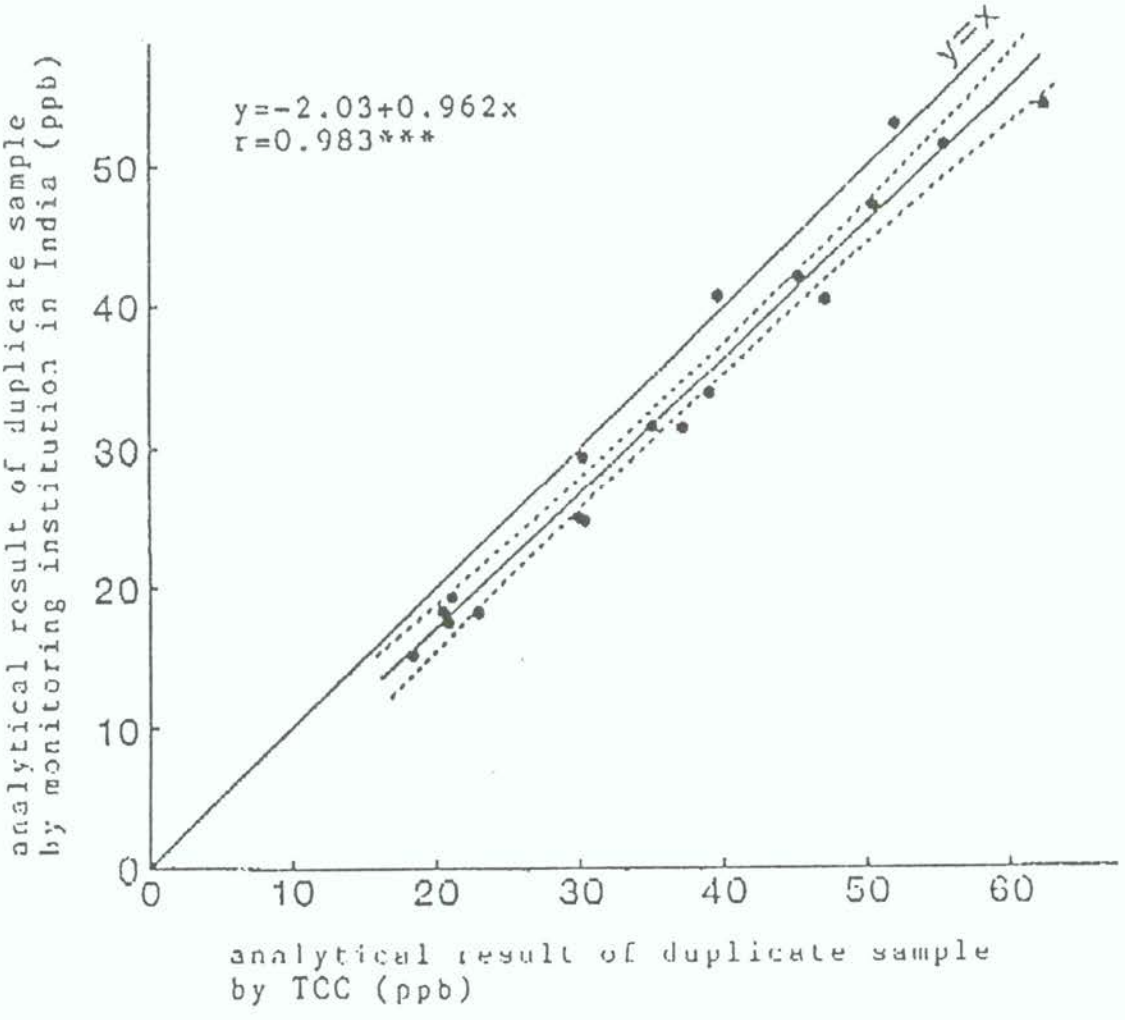
Eighteen pairs of duplicate samples were collected for quality assurance by cross check with the TCC. Analytical results of the duplicate samples are shown in Table 1. The TCC did not have a blank sample, so that the blank data was not subtracted from the analytical results by the TCC. The results by the TCC, therefore, were systematically higher than those by the monitoring institution in India.

Analytical performance evaluation was carried out by linear regression (see Figure 1). Analytical results by two institutions showed good correlation. The value of y intercept of the regression line was reasonable as a badge blank. The regression line was in the MAD intervals of $y=x \pm (2 \text{ ppb} + 0.1x)$ at the probability higher than 90% when considering that the results by the TCC included badge blank which may be equal to the y intercept. The monitoring data in India, therefore, may be reliable/comparable.

Table 1. *Analytical results of duplicate samples.*

Sample Number	Subject Number	Sampling Place	Analytical Result (ppb)	
			India	TCC
1	15	Kitchen	54.37	62.4
2		Kitchen	53.08	51.9
3		Living Room	42.02	45.1
4		Living Room	34.05	39.0
5		Bedroom	51.57	55.3
6		Bedroom	47.20	50.3
7		Home Outdoor	40.53	47.0
8		Home Outdoor	40.80	39.6
9	5	Kitchen	24.91	29.8
10		Kitchen	31.43	35.0
11		Living Room	18.39	20.3
12		Living Room	18.23	22.8
13		Bedroom	17.52	20.6
14		Bedroom	19.28	20.9
15		Workplace	15.20	18.1
16		Home Outdoor	29.31	30.1
17		Home Outdoor	31.43	37.1
18		Workplace Outdoor	24.8	30.2

Figure 1. Correlation between analytical results of duplicate samples by monitoring institution in India and those by TCC.



*** $p < 0.001$
Dotted lines: 90% confidence intervals of the regression line

Appendix 2

Personal NO_2 exposure levels (unit:ppb) in Bombay.

Date	Subj. 1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	a.v	s.d.	max.	min.	n
16/1/89	19.36	19.77	35.85	16.60	21.70													22.66	7.60	35.85	16.60	5
17/1/89	16.67	28.07	30.70	14.41	14.38													20.85	7.91	30.70	14.38	5
18/1/89	18.62	23.78	21.76	15.27	17.98													19.48	3.33	23.78	15.27	5
19/1/89	20.83	18.39	15.79	18.06	18.98													18.41	1.81	20.83	15.79	5
20/1/89	16.42	32.39	18.95	23.52	17.31													22.72	7.25	32.39	16.42	5
21/1/89	28.10	28.52	-	17.82	13.62													22.00	7.47	28.52	13.62	4
22/1/89	18.54	22.24	15.75	9.33	18.95													16.96	4.84	22.24	9.33	5
27/1/89						29.64	29.92	11.98	29.16	25.74								25.29	7.63	29.92	11.98	5
28/1/89						35.40	28.43	9.00	23.08	27.69								24.72	9.83	35.40	9.00	5
29/1/89						36.47	25.85	11.66	25.51	21.00								24.10	8.98	36.47	11.66	5
30/1/89						40.71	24.85	20.76	30.82	31.97	55.04	39.58	45.04	-	53.63			38.04	12.00	55.04	20.76	9
31/1/89						41.38	21.32	14.67	34.96	24.65	52.90	30.17	41.95	49.64	47.72			35.94	12.94	52.90	14.67	10
1/2/89						37.22	22.35	8.75	17.51	17.51	52.70	27.85	38.04	52.02	44.30			31.83	15.33	52.70	8.75	10
2/2/89						16.41	47.66	8.19	12.09	31.73	58.90	28.00	54.38	59.03	43.25			35.96	19.41	59.03	8.19	10
3/2/89											51.90	39.39	46.61	63.97	49.70			50.31	8.98	63.97	39.39	5
4/2/89											42.80	38.76	48.03	53.62	46.33			45.91	5.59	53.62	38.76	5
5/2/89											37.80	32.56	33.19	38.73	33.59	23.37	16.19	30.78	8.14	37.73	16.19	7
6/2/89														36.45	-	25.84	24.09	28.79	6.69	36.45	24.09	3
7/2/89																28.48	23.47	25.98	3.54	28.48	23.47	2
8/2/89																22.84	20.59	21.72	1.59	22.84	20.59	2
9/2/89																29.86	32.31	31.09	1.73	32.31	29.86	2
10/2/89																27.24	29.50	28.37	1.60	29.50	27.24	2
11/2/89																28.73	24.92	26.83	2.69	28.73	24.92	2
av.	19.80	24.74	23.10	17.10	17.56	33.90	28.63	12.14	24.73	25.80	50.29	33.76	43.89	50.49	45.50	26.62	24.44					
s.d.	3.97	5.10	8.33	5.82	2.80	8.62	8.93	4.43	7.92	5.32	7.35	5.37	6.93	10.02	6.29	2.72	5.35					
max.	28.10	32.39	35.85	28.52	21.70	41.38	47.66	20.76	34.96	31.97	58.90	39.58	54.38	63.97	54.63	29.86	32.31					
min.	16.42	18.39	15.75	9.33	13.62	16.41	21.32	8.19	12.09	17.51	37.80	27.85	33.19	36.45	33.59	22.86	16.19					
n	7	7	6	7	7	7	7	7	7	7	7	7	7	7	7	7	7					

Summary of activity records of subjects in Bombay.

Subject Number	Kitchen (%)	Living Room (%)	Bedroom (%)	Workplace (%)	Other Indoor (%)	Transport (%)	Outdoor (%)	Cooking (%)	Kitchen + Living + Bedroom + Workplace (%)
1	16.67	20.83	33.33	20.83	4.17	4.17	-	8.33	91.66
2	2.08	16.67	25.00	37.50	8.33	4.17	6.25	-	81.25
3	2.08	8.33	33.33	50.01	4.17	2.08	-	-	93.75
4	4.17	8.32	25.00	33.33	4.17	12.50	4.17	4.17	70.82
5	4.17	8.33	33.33	33.33	4.18	8.33	8.33	-	79.16
6	8.30	75.00*	-	-	4.20	-	4.20	8.30	83.30
7	20.90	66.66*	-	-	8.30	-	4.20	4.20	87.56
8	10.20	33.30	42.00	-	8.30	2.10	4.10	4.10	85.50
9	25.00	20.70	42.00	-	12.30	-	-	8.30	87.70
10	16.60	20.90	45.90	-	12.40	-	4.20	4.20	83.40
11	37.50	-	52.10*	-	6.20	-	4.20	18.75	89.60
12	25.00	33.40	37.40	-	4.20	-	-	12.50	95.80
13	29.20	12.40	45.80	-	6.30	-	6.20	14.60	87.40
14	29.10	6.30	58.30	-	-	-	6.30	29.10	93.70
15	25.00	8.30	62.50	-	2.30	-	2.10	12.50	95.80
16	25.00	-	62.50*	-	4.20	-	8.30	16.00	87.50
17	91.70**	-	-	-	-	-	8.30	25.00	91.70
av.	21.92	24.25	42.75	35.00	6.20	5.56	5.45	12.15	87.39
s.d.	20.99	21.68	32.57	10.46	3.10	4.09	1.98	7.92	6.52
max.	91.70	75.00	62.50	50.01	12.40	12.50	8.33	29.10	95.80
min.	2.08	6.30	25.00	20.83	2.10	2.08	2.10	4.10	70.82
n	17	14	14	5	15	6	13	14	16

* Living room and bedroom were same for these subjects.

** Kitchen, living room and bedroom were one and same for this subject.

Appendix 4

Estimation of personal exposure level from fractional exposures in micro-environments (Bombay).

Subject	Kitchen (ppb)	Kitchen (%)	Living Room (ppb)	Living Room (%)	Bedroom (ppb)	Bedroom (%)	Workplace (ppb)	Workplace (%)	Kitchen + Living Room + Bedroom + Workplace (ppb)	Kitchen + Living Room + Bedroom + Workplace (%)	Personal Exposure (ppb)
1	11.15	56.31	6.91	34.90	10.10	51.00	4.42	22.30	32.58	164.51	19.80
2	1.60	6.46	5.39	21.80	5.69	23.00	9.52	38.49	22.23	89.75	24.74
3	1.38	5.97	1.52	6.58	8.33	36.00	21.45	92.86	32.68	111.41	23.10
4	1.51	8.83	2.17	12.69	4.85	28.36	5.30	30.99	13.83	80.87	17.10
5	1.18	6.74	1.53	8.74	6.13	34.91	5.12	29.14	13.96	79.53	17.50
6	3.44	10.15	34.13	100.00	-	-	-	-	51.49	110.15	33.90
7	5.19	18.12	15.76	55.05	-	-	-	-	20.95	73.17	28.63
8	4.99	41.06	6.20	51.04	4.36	36.24	-	-	15.59	128.34	12.14
9	6.64	26.85	4.55	18.40	11.95	48.32	-	-	23.14	93.57	24.73
10	6.07	23.53	4.81	18.60	13.00	50.39	-	-	23.88	94.52	25.80
11	35.70	70.99	31.23	62.09	-	-	-	-	66.93	133.08	50.29
12	13.40	39.69	15.92	47.16	15.48	45.85	-	-	44.80	132.70	33.76
13	24.66	56.19	6.38	14.54	24.74	56.37	-	-	55.78	127.10	43.89
14	25.17	49.86	3.73	7.38	28.95	57.34	-	-	57.85	114.58	50.49
15	15.59	34.27	3.87	8.50	35.66	78.37	-	-	55.12	121.14	45.50
16	13.64	51.24	-	-	-	-	-	-	13.64	51.24	26.62
17	46.48	190.18	-	-	-	-	-	-	46.48	190.18	24.44
av.	12.82	40.97	9.62	31.15	14.11	45.51	9.16	42.76	34.77	111.52	29.58
s.d.	13.17	43.63	10.35	26.93	10.33	15.04	7.16	28.60	18.10	34.18	11.74
max.	46.48	190.18	34.13	100.00	35.79	78.37	21.45	92.86	66.93	190.18	50.38
min.	1.18	5.97	1.52	6.58	4.36	23.00	4.42	22.30	13.64	51.24	12.03
n	17		15		12		5		17		17

av.	16.75	(3.36)	51.01	(16.86)	12.66	(3.50)	38.23	(16.94)	19.12	(7.02)	53.27	(32.65)	39.64	(23.05)	114.15	(105.21)	33.35	(20.46)
s.d.	13.70	(4.36)	47.09	(22.08)	11.51	(2.49)	30.00	(11.61)	10.96	(2.15)	13.13	(11.15)	18.86	(9.38)	34.85	(35.52)	11.94	(3.37)
max.	46.48	(11.15)	190.18	(56.31)	34.13	(6.91)	100.00	(34.90)	35.66	(10.10)	78.37	(51.00)	66.93	(32.68)	190.18	(164.51)	50.49	(24.74)
min.	3.44	(1.18)	10.15	(5.97)	3.73	(1.52)	7.38	(6.38)	4.40	(4.85)	36.24	(23.00)	13.64	(13.83)	51.24	(79.53)	12.14	(17.10)
n	12''	(5)'	12''	(5)'	10''	(5)'	10''	(5)'	7''	(5)'	7''	(5)'	12''	(5)'	12''	(5)'	12''	(5)'

' Calculations for the group of subjects (1 to 5) who had jobs outside home.

'' Calculations for the group of housewives (subjects 6 to 17).

Environmental monitoring data in Bombay.

	Suburban high			Suburban low			Urban high			Urban low		
Date	NO_2 (ppb)	Temp (°C)	Wind (m/s)	NO_2 (ppb)	Temp (°C)	Wind (m/s)	NO_2 (ppb)	Temp (°C)	Wind (m/s)	NO_2 (ppb)	Temp (°C)	Wind (m/s)
16/1/89	71.0	24	1.2									
17/1/89	62.1	19	1.5									
18/1/89	54.1	25	1.2									
19/1/89	57.1	25	1.3									
20/1/89	50.8	24	1.1									
21/1/89	75.4	26	0.7									
22/1/89	67.3	24	2.4									
23/1/89	43.3	24	0.0									
27/1/89				29.6	26	1.9						
28/1/89				33.3	29	1.2						
29/1/89				39.6	29	1.1						
30/1/89				36.4	28	1.2	32.6	29	-			
31/1/89				21.3	31	0.9	32.2	29	-			
1/2/89				32.2	26	1.4	35.1	28	-			
2/2/89				34.9	29	1.1	38.0	28	-			
3/2/89				32.6	29	0.0	53.0	29	-			
4/2/89							43.3	29	-			
5/2/89							35.4	27	-	14.7	29	1.4
6/2/89							38.3	27	-	24.2	26	1.3
7/2/89										21.3	26	1.8
8/2/89										28.0	27	1.4
9/2/89										23.6	25	1.1
10/2/89										32.0	25	0.8
11/2/89										33.5	26	0.8
12/2/89										40.0	26	0.8
av.	60.1			32.5			38.5			27.2		
s.d.	10.8			5.4			6.9			7.9		
max.	75.4			39.6			53.0			40.0		
min.	43.3			21.3			32.2			14.7		
n	8			8			8			8		

EXPOSURE MONITORING OF NITROGEN DIOXIDE IN THE LOS ANGELES BASIN, USA

An international pilot study within the
WHO/UNEP Human Exposure Assessment Locations (HEAL) Programme

SECTION III

Technical report edited by

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TABLE OF CONTENTS

1. INTRODUCTION

The survey was designed to obtain a random selection of individuals in the Los Angeles and Orange County area of California (USA) willing to participate in a two-day monitoring study. The sampling design called for a two-stage random selection which involved (1) the random selection of a household, and (2) the random selection of an individual over the age of eight within the household. Household data included characteristics of the residential dwelling and the heating and cooking facilities within the dwelling; individual data included the targeted respondent's exposure for the previous day to cooking inside the home and travel outside the home. In addition, information about major activities (e.g., employment, hobbies) in which the target respondent was engaged for at least 20 hours per week was obtained.

The sample used was a basic random digit dial (RDD) sample for Orange County and Los Angeles County. Target selection within the household involved a random selection among all household members eight years of age or older. The survey was conducted using computer-assisted telephone interviewing (CATI) software developed by the Computer-assisted Survey Methods Program at the University of California, Berkeley.

The workplan called for seven days of personal and residential monitoring for nitrogen dioxide using female volunteers who are not employed outside the home. The study followed as closely as possible the details contained in the report by Matsushita and Tanabe (1990). Certain deviations from the WHO protocol were necessitated by constraints due to the need to complete field work before the end of the 1988 winter season, the protocol used by the larger University of California, Irvine (UC Irvine)/Harvard Exposure Study within which this study was accommodated, and the wishes of the US Environment Protection Agency (in the case of the sample size).

This study has demonstrated that it is feasible to collect reliable personal and indoor residential exposure data from a random sample of the public. With carefully developed and simple questionnaires and sampling instructions, relatively complicated tasks can be completed by human volunteers. While data of high quality were generated in this study, the sample of nine individuals is too small to conduct multivariate statistical analyses of factors influencing personal exposure and indoor concentrations of nitrogen dioxide.

2. MATERIALS AND METHODS

2.1 Subject Selection

The nine subjects participating in the WHO/EPA HEAL NO_2 Pilot Study were selected from among participants of a concurrent study conducted by UC Irvine and Harvard University on personal exposure to nitrogen dioxide. That study was

conducted during 1987 and 1988 in Los Angeles and Orange Counties of California, USA and was sponsored by the Gas Research Institute (GRI). The two counties surveyed account for a substantial majority of the population in the Los Angeles Basin and this geographic region contains a wide range of ambient concentrations of nitrogen dioxide. The area is one of the few jurisdictions in the USA that is still in violation of the USEPA Federal Health Standard of 50 ppb (annual average).

The sample for the survey was a random digit dialling sample (RDD) selected in proportion to the total number of residential telephone numbers for Orange County and Los Angeles County. The RDD sample was obtained from Survey Sampling, Inc. (Connecticut), a company which specializes in the production of telephone samples. Survey Sampling created a stratified random sample in which all telephone households within each county had an equal chance of being chosen to participate. Briefly, the RDD sample used in this survey was prepared using the following procedures.

First, telephone working blocks were determined for each county. A telephone block is made up of 100 telephone numbers based on the first two numbers after the exchange. For example, 554-1200 to 554-1299 makes up one telephone block. A block is classified as a working block when three telephone numbers are found to be residences in the telephone directory. Second, the computer generated telephone numbers are compared with the business numbers from telephone directories in order to remove all listed business numbers. Next, the proportion of telephone households was determined for each county by multiplying the estimated percentage of telephone households by the number of households in the county. This determines the portion of the sample telephone numbers from each county required for the sample.

A sampling interval was then determined for each county by dividing the sample required into the total estimated number of telephone households for that county. This interval was used to choose the sample for each county from all generated telephone numbers. Approximately 79% of the phone numbers were from Los Angeles County and 21% from Orange County.

A random digit dialling (RDD) sample of 3,500 telephone numbers for Los Angeles County and Orange County was purchased from Survey Sampling, Inc. in February, 1987. Of the 3,500 numbers, 190 numbers were used in the pretest and 3,310 numbers were used for the GRI Study. A second sample of 1,500 numbers for the same areas was purchased in March, 1988. All 4,310 numbers were subsequently utilized for the GRI survey. Documentation from Survey Sampling, Inc. regarding the sample is attached to this report as Appendix A.

To acquire a randomly selected target for the GRI monitoring study an individual was chosen from the household by randomly assigning two numbers to each telephone number - one number to select the sex of the target and the other

number to choose age position within the household. The first random number (for sex of respondent) was programmed in CATI to ask either for the number of males in the home over the age of eight or the number of females in the home over the age of eight. If the household did not contain any individuals of the randomly chosen sex, the programme then pathed to a question which asked for the number of individuals of the only sex in the household.

Once the total number of persons in the household of the same sex was entered, the programme utilized this value with the second random number to randomly choose one of five positions in the household: oldest, next to the oldest, middle, next to the youngest, or youngest. This person then became the target respondent.

For the WHO/EPA HEAL study we used only female target respondents from the GRI study. For the HEAL programme we adopted the additional criteria that the subjects should be non-smokers and should not have employment outside the home. While the HEAL programme called for sampling working and non-working women in homes with and without sources of NO_2, we sought to reduce exposure variation that we could not expect to assess in the course of this pilot study. Since the sample is so small in this study we sought to reduce the number of potential sources of NO_2 variation.

If at all possible, the information on the target individual was obtained from the initial respondent. If the information could not be obtained by this method, the interviewer asked if the target individual was unavailable, the interview was placed in a callback situation and a second contact to the target was made.

At the completion of the interview, the target was asked to participate in the monitoring portion of the study. If an individual under the age of 16 was targeted, the adult responsible for the child was asked to give initial consent. Actual acceptance of the monitoring, however, came from the target individual even if the targeted individual was under the age of 16.

Those accepting to participate in the UC Irvine/Harvard study, and meeting the criteria outlined here for joining the WHO/EPA HEAL Pilot Study for NO_2, were invited to participate in this additional investigation.

Interviewing was conducted by the Centre for Survey Research (CSR) in the Public Policy Research Organization (PPRO) of the UC Irvine.

2.2 Interviewer Training

Interviewer training consisted of two parts: (1) an informal lecture and practice session, and (2) a hands-on learning session with the CATI software on personal computers (PC-XT's). The first part of the initial training was dedicated to general interviewing skills, information about

the study, and the survey instrument. The CSR manual containing in-depth interviewing practices was discussed in an open format and tape recordings of interviews were used to illustrate techniques. Among the points emphasized were interview bias, techniques to obtain respondent co-operation, and general interviewing practices such as reading questions in order and verbatim. The second half of the initial training session consisted of an in-depth discussion of the GRI questionnaire. The questionnaire discussion centered on respondent selection, question intent, probes, skip patterns, and question wording.

The second training session took place in the CSR interviewing room which is equipped with 7 PC-XT's programmed for CATI. During this session, interviewers learned the CATI commands and practiced the questionnaire. CATI training consisted of reading the CATI manual written by CSR staff, and going through a "learn" questionnaire which teaches the CATI commands in an actual questionnaire format. The interviewer then went through the questionnaire several times trying the various options and finally rehearsing the actual interview by practicing on other interviewers and the supervisor. The study director and a trained supervisor monitored the practice sessions, giving suggestions when needed.

2.3 Questionnaire Design

The initial telephone questionnaire centered on the type of residence, type of heating system and fuel, cooking fuel, microwave and range vent use, range pilot light, whether or not the range is used to heat the home, target occupation, and target cooking and travel experiences.

The CATI questionnaire design involved two distinctive parts to the interview. In the initial contact with the household, an interview was conducted with an adult over the age of 18 who was responsible for the home. Household characteristics, specifically the type and age of the dwelling and the type of heating and cooking facilities, were obtained during this portion of the interview. Relevant branching of questionnaire items with respect to the type of fuel used in the household as well as the type of cooking facilities within the household was included.

Approximately half-way through the interview, the random selection of the target individual was performed. Questions regarding principle activities of the target, for example, employment outside of the home, as well as cooking and travelling time (in minutes) performed by the target respondent for the previous day were then posed.

A copy of the eight page CATI questionnaire can be obtained from WHO, Geneva or the editor of the report. Both English and Spanish versions of the questionnaire were developed and administered in the GRI study. For the WHO/EPA HEAL study, only English speaking respondents were selected.

Prior to raw data output of the completed cases from the CATI system, all completed cases were checked for any problems regarding unanswered questions by using the CATI cleaning programme. all completed cases were then written to raw data output. Additional consistency checking of the responses that was not covered by the CATI programme was then performed. A detailed codebook was prepared containing the exact wording of each question and the column and record allocation for each variable.

2.4 Field Protocol

As specified by Dr. Tanabe, personal sampling was conducted over a seven day period (refer Section I, Table 1). All NO_2 measurements were made with passive diffusion-type sampling badges assembled in Japan and analyzed by the Environmental Research Centre at the University of Nevada, Las Vegas (UN Las Vegas). Personal badges were deployed for seven consecutive 24-hour periods. Indoor residential samples were taken in the kitchen, bedroom of the subject, and living room. Additional residential samplers were located outside the home to measure ambient NO_2 concentrations.

On the first day of sampling, two sets of indoor and outdoor samplers were deployed, one for two days and the other for three days. At the end of three days, the four-day indoor and outdoor samplers were set out. These samplers were located at each of the three indoor sites and outside the home. Additional samplers were allocated as duplicate field samplers and analyzed by the UN Las Vegas laboratory as "blind" samples. The two-day set was co-located with the UC Irvine/Harvard samplers and provides a potential data set for an independent quality assurance check on the concentrations generated in this study.

All samplers were labelled in advance with a unique identification number and delivered to the home by our trained field technician. A copy of the card used to assign samplers to each volunteer can be obtained from WHO, Geneva or the editor of the report. With a field blank and duplicate field samples, at least 21 NO_2 badges were assigned to each home.

The technician explained the study protocol to the subject and left a simple one-page checklist for the subject to follow when sealing three day sampling badges and opening four day badges. The technician also completed housing and personal characteristics questionnaires, and reviewed an example of the activity diary used in this study.

Sampling for Day 1 began as the field technician left the home. Forty-eight hours later the technician returned to pick up the two-day samples and to review the diary records for consistency and completeness. The technician inspected the sealing of the day one personal badges and if necessary reviewed proper sealing of the sampler bags with the subject. A pre-stamped return mailer was provided for the subject to send the remaining samplers back to our laboratory at the end of the seven day period.

2.5 Analytical Procedures

At our laboratory all diaries were reviewed as received and any inconsistencies were identified and the subject promptly called to attempt to clarity entries. Sampler bags were checked to insure proper sealing. Any problems were noted in the laboratory notebook. All samplers were mailed to ERC at UN Las Vegas for analysis in bags of approximately 50 samplers. An opened sacrificial sampler was placed with each bag. Duplicate samples and quality assurance samples were not identifiable by the laboratory.

Laboratory methods are described in Appendix B. ERC reported values in terms of the absolute number of nanograms of NO_2 determined on the filters. Nitrogen dioxide concentrations were calculated using the formula:

$$CNO_2 = M \times \frac{T}{K'_{OG}} \times \frac{}{x\ t} \times 8.31 \times 10^9$$

CNO_2 = NO_2 concentration, ppb
M = net moles of NO_2 collected on filter
T = temperature, degrees kelvin
K'_{OG} = collection efficiency (cm/sec)
t = sampling time, seconds

K'_{OG} was determined to be 0.1405 at 25°C, 65% RH. Assuming an average sampling temperature of 25°C, we simplify:

$$CNO_2 = \frac{M}{t} \times \frac{298.16}{0.1405} \times 8.31 \times 10^9$$

$$= \frac{M}{t} \times 1.76 \times 10^{13}$$

Expressing t in minutes and M in nanograms of NO_2:

$$CNO_2 = \frac{M}{t} \times 6.389$$

To produce NO_2 concentrations in ppb, the total amount of NO_2 (expressed in nanograms) determined on the filter by UN Las Vegas ERC was divided by the time the badge was open (expressed in seconds) and multiplied by the scalar value 6.389.

2.5.1. Quality Assurance

Data quality was evaluated using eight field blank and ten blind duplicate samples. In one test home, nine sets of

duplicate samples was deployed for 77 additional quality control samples. In addition, an interlaboratory comparison is possible, but was not undertaken in this analysis, between the two-day samples analyzed at UN Las Vegas and paired samples produced and analyzed at Harvard. Each home had at least one blank field blank filter that was opened at the site and sealed in the manner that samplers are sealed following use. This allows evaluation of any gain of NO_2 due to opening and storage of resealed samplers. Twenty-five unopened blank filters were kept at the laboratory for comparison.

2.5.2. Record Keeping

The initial recruitment interview was conducted by telephone with responses logged directly into a personal computer. More detailed residential and activity questionnaires conducted at home were made in hard copy and computer coded.

Since weather stations were not operated for this pilot study, detailed weather information for the sampling days was extracted from the weather page of the *Los Angeles Times*. Temperature extremes are not generally experienced in this region, particularly during the end of the winter season.

3. RESULTS

3.1 Survey Interview

Telephone interviews were conducted from 5:00 pm to 9:00 pm on weekdays. 10:00 am to 1:00 pm on Saturdays. The study director supervised the entire interviewing effort.

The survey consisted of approximately 40 closed-ended questions, taking an average of 13 minutes to administer. During the interview a target person was randomly chosen from among all persons eight years old or over living in the household. If the target agreed to participate in the additional study (or the respondent consented for the target) the interviewer obtained the target's last name and address.

Table 1 displays the outcome for the 4,310 telephone numbers used in the survey. Thirty-six percent (1,536) of the telephone numbers were ineligible numbers. One-quarter (1,083) of these telephone numbers were found to be non-working numbers (including disconnected numbers, computer lines, and circuit problems in dialling). Approximately 10% (424) of the numbers were for non-households, both businesses and group quarters, e.g., retirement home, hospital, etc. An additional 1% (29) were wrong numbers, i.e., the person answering the phone indicated that the interviewer had reached a wrong number.

Table 1. Outcome of the Sample.

	Sub-total Number	Sub-total Percent	Total Number	Total Percent
Ineligible			1,536	36%
Non-working numbers (disconnected, other problems)	1,083	25%		
Non-households (group quarters, businesses)	424	10%		
Wrong numbers	29	1%		
Unable to Contact			870	20%
No answers/busy signals	641	15%		
Answering machines	229	5%		
Active Sample			1,906	44%
Callbacks still pending at completion of survey period	276	6%		
Partial interviews still pending at completion of survey period	49	1%		
Refusals	265	6%		
Completed interviews	1,316	31%		
Total			4,312	100%

There was a further 20% (870) of the telephone numbers which were consistently unanswered. Consistently 'unable to contact' telephone numbers are defined as those which are attempted at least six times on six different days. In 15% (641) of the cases, there was an answering machine.

At the completion of the survey period, 44% (1,906) of the telephone numbers were still active of which 6% (276) were numbers in which someone had answered the telephone, but had requested the interviewer to call back at a later date, and 1% (49) were partial interviews in which the respondent refused to continue during the substantive interview. There were a

total of 1,316 (31%) completed interviews and a total of 265 (6%) cases in which the respondent had refused twice to participate in the survey.

At the completion of the survey period there were a total of 1,906 eligible phone numbers in the sample. The response rate, therefore, for the survey was 69% (1,316/1,906). The completion rate (which does not include the 276 callbacks still pending at the end of the survey) was 81% (1,316/1,630).

In Table 2 is displayed the acceptance rate for the monitoring portion of the survey. Approximately 61% of those respondents who completed the survey indicated a willingness to participate in the monitoring portion of the study. An additional 3% of the respondents requested further information be sent to them before making a decision. In 2% of the cases, the target was not at home or the respondent/target was undecided and further attempts to contact the target were unsuccessful. Finally, in 34% of the cases, the target refused to participate in the monitoring study.

Table 2. *Distribution of Participation in the 2-Day Monitoring Study.*

Willing to participate	Number	Percent
Yes	805	61%
No	443	34%
Need more information	42	3%
Person not available	20	2%
Don't know	6	1%
Total	1,316	101%

3.2 Quality Control

Results of paired field samples are presented in Figure 1. The co-efficient of determination (R^2) for these ten samples is 0.96, indicating very good repeatability of badge measurements. The quality of the NO_2 results is reinforced with the duplicate samples deployed in the test home and presented in Table 3. The co-efficients of variation for these samples averages around 2%, permitting precise determination of NO_2 concentration.

Blank badge results, also presented in Table 3, are not quite as reassuring. Since the total amount on NO_2 on the blank filters is small it is expected that the co-efficients of variation will increase. The field blanks, which are opened in the field and resealed, have an acceptable co-efficient of variation of approximately 13%. The unopened blanks, however, have a co-efficient of variation of nearly 33%. On closer inspection of the individual values, it is

clear that the majority of the error is due to two samples that have between two and three times the normal background levels of NO_2. The individual sample values are given in Appendix C. Since samples have considerably more NO_2 than blanks, the high variability of blanks can be put in perspective; however, precision could be increased with less blank variability.

Figure 1. Duplicate field samples.

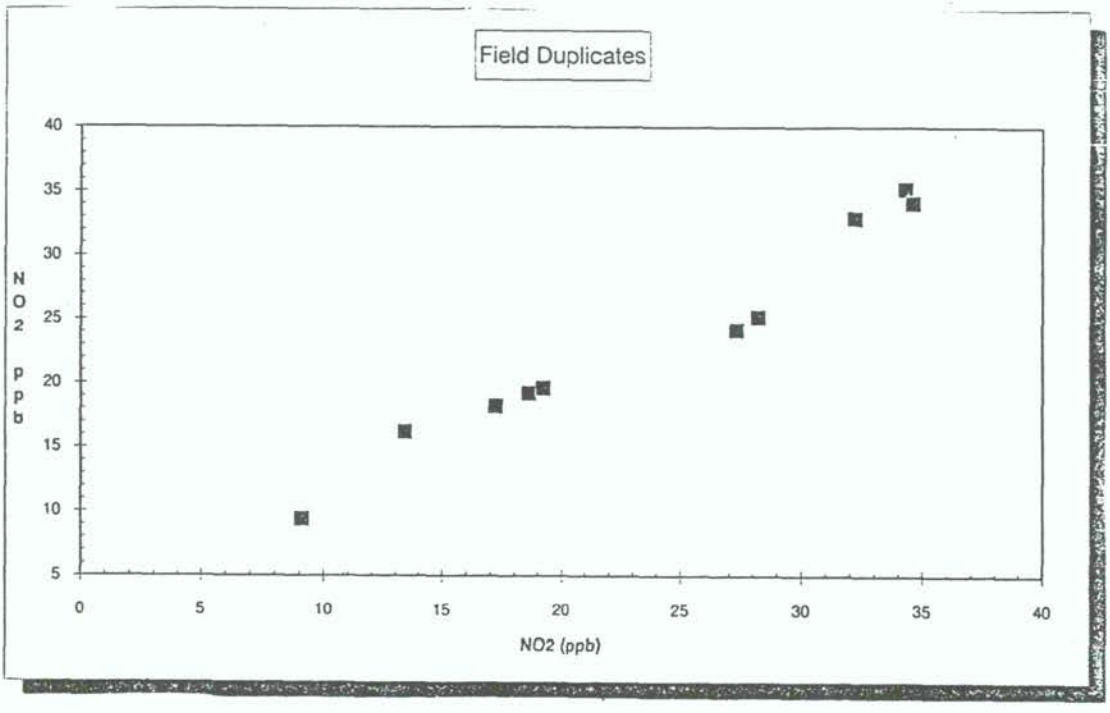

Table 3. Variability of duplicate samples. Variation expressed as co-efficient of variation (mean/standard deviation) x 100.

Field Samples	Co-efficient of Variation (%)	NO_2 (ppb)	Number of Filters
Master bedroom	1.9%	14.1	8
Bedroom £2	1.5%	13.8	10
Family room	3.3%	24.0	10
Dining room	1.2%	16.6	9
Garage	1.5%	16.3	10
Kitchen, range hood	2.7%	49.7	8
Kitchen, bookcase	5.9%	26.3	11
Kitchen, refrigerator	1.4%	21.5	11

Blank Filters	Co-efficient of Variation (%)	NO_2 (ng)	Number of Filters
Field blanks	12.7%	631.3	8
Unopened blanks	32.7%	838.0	25
Blanks (outliers removed)	10.0%	583.5	23

3.3 Nitrogen Dioxide Results

With only nine subjects it is not possible to conduct a detailed statistical evaluation of factors that are related to exposure and indoor concentration of NO_2. Accordingly, we focus on descriptive presentation of the data and comparisons that are reasonable given the small data set.

In Figure 2 the average of the seven 24-hour personal NO_2 badge samplers is shown. Average one-week personal exposures range from approximately 30 ppb to 15 ppb with standard deviations of the one-week means ranging from approximately 2 to 15 ppb.

In this report we focus on the two-day fixed location samples since these were opened and closed by our technician. A visual inspection of the data indicates that the two day technician data are of high quality. In Figure 3 we present the two-day indoor and outdoor concentrations for all homes. As we have found in our other studies in Southern California, there appears to be a relationship between outdoor NO_2 concentrations and the concentration measured at all other indoor locations. Bedroom concentrations tend to be lowest and living room concentrations intermediate. Kitchen or outdoor concentrations tend to be the highest.

Subjects 3, 5, and 9 had electric ranges. All other subjects had gas-fuelled ranges with continuously-burning pilot lights. While the homes with electric ranges tend to have lower indoor concentrations NO_2 they are also frequently located in less densely populated areas of Southern California with lower ambient NO_2 concentrations.

Table 4 presents the results of bivariate regressions among the various fixed-location two-day monitors. The sample size for each of these regressions is the nine pairs derived from the nine subjects. All regressions are statistically significant. Outdoor concentrations are correlated with all indoor locations with coefficients of determination ranging from 0.4 to approximately 0.7. This implies that 40-70% of the variation of indoor concentrations depends on outdoor levels. Figure 4 illustrates this more clearly by removing the living room and kitchen values from Figure 3. In turn, each of the indoor rooms is related with the other rooms. The highest relationship is observed between the living room and kitchen. Given the physical proximity of these room, that observation makes intuitive sense and is consistent with other results obtained by our laboratory.

3.3.1 Time Activity Diaries

Time and activity diaries were carefully completed by all subjects, permitting summary of the patterns in this group of subjects. Figure 5 presents a pie chart of the average use of time in the study sample. Almost 78% of this sample's time is spent at home. Less than 8% of their time is spent outdoors, including while travelling by motorized vehicle. From this pilot study we have demonstrated that it is feasible to

collect accurate activity diaries that may be associated, in a larger probability sample, with exposure to NO_2. With the small sample selected for this pilot study it would not make sense to attempt to relate specific activities to NO_2 exposure.

Figure 2. Average of seven daily NO_2 personal samples.

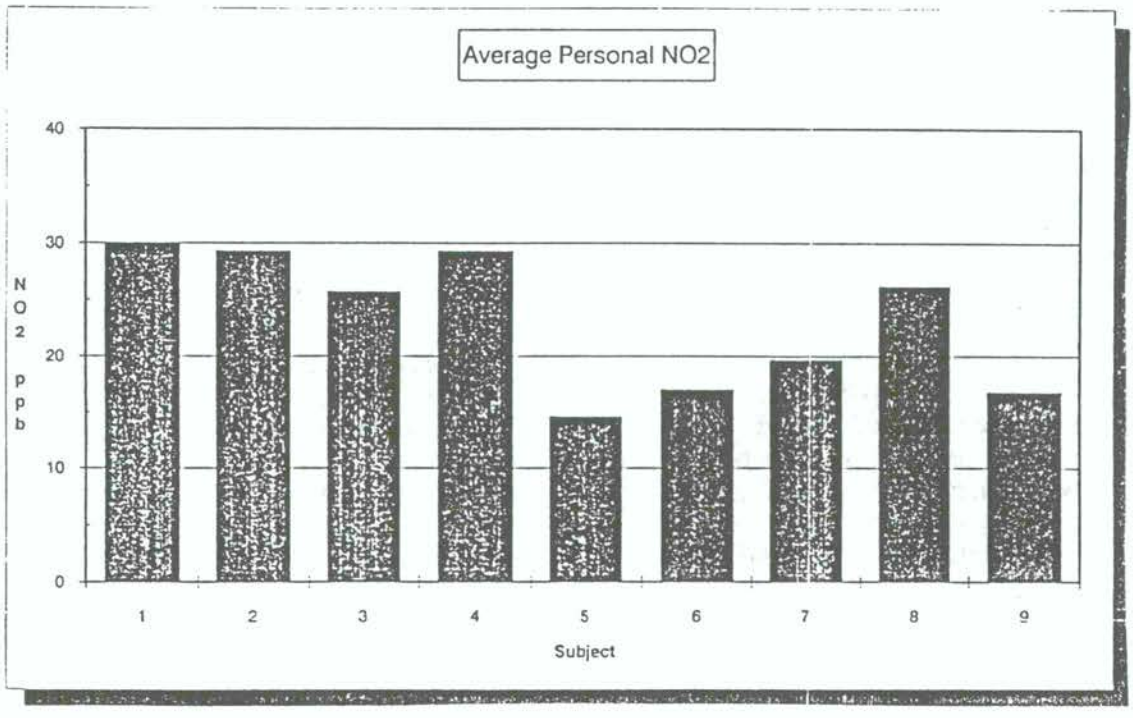

Figure 3. Summary of two-day fixed-location samples.

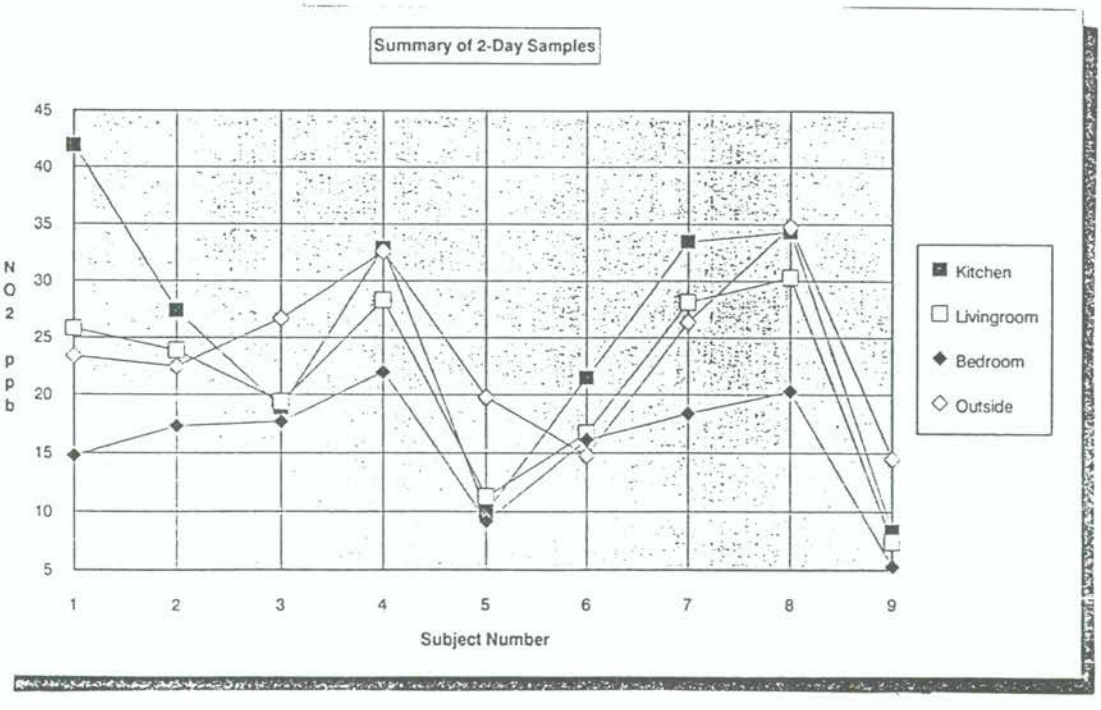

Figure 4. Bedroom versus outside NO_2 by subject.

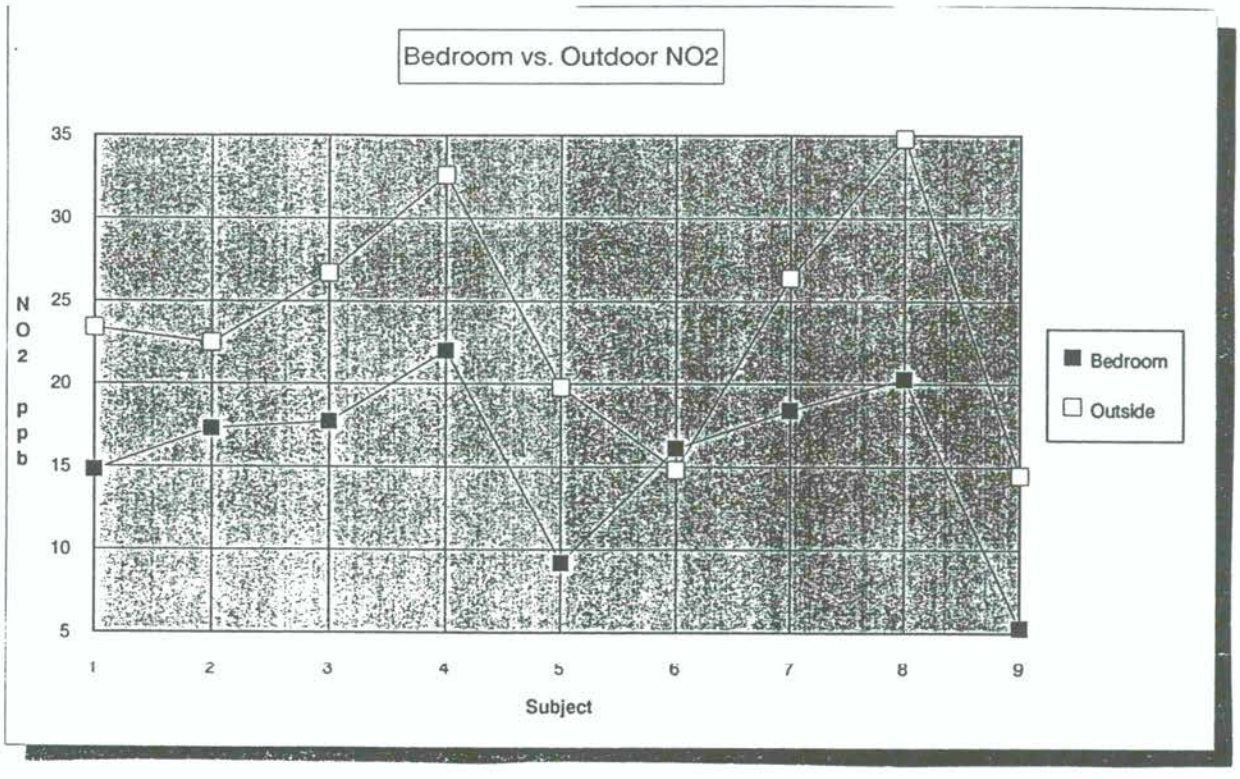

Figure 5. Average use of time by subjects.

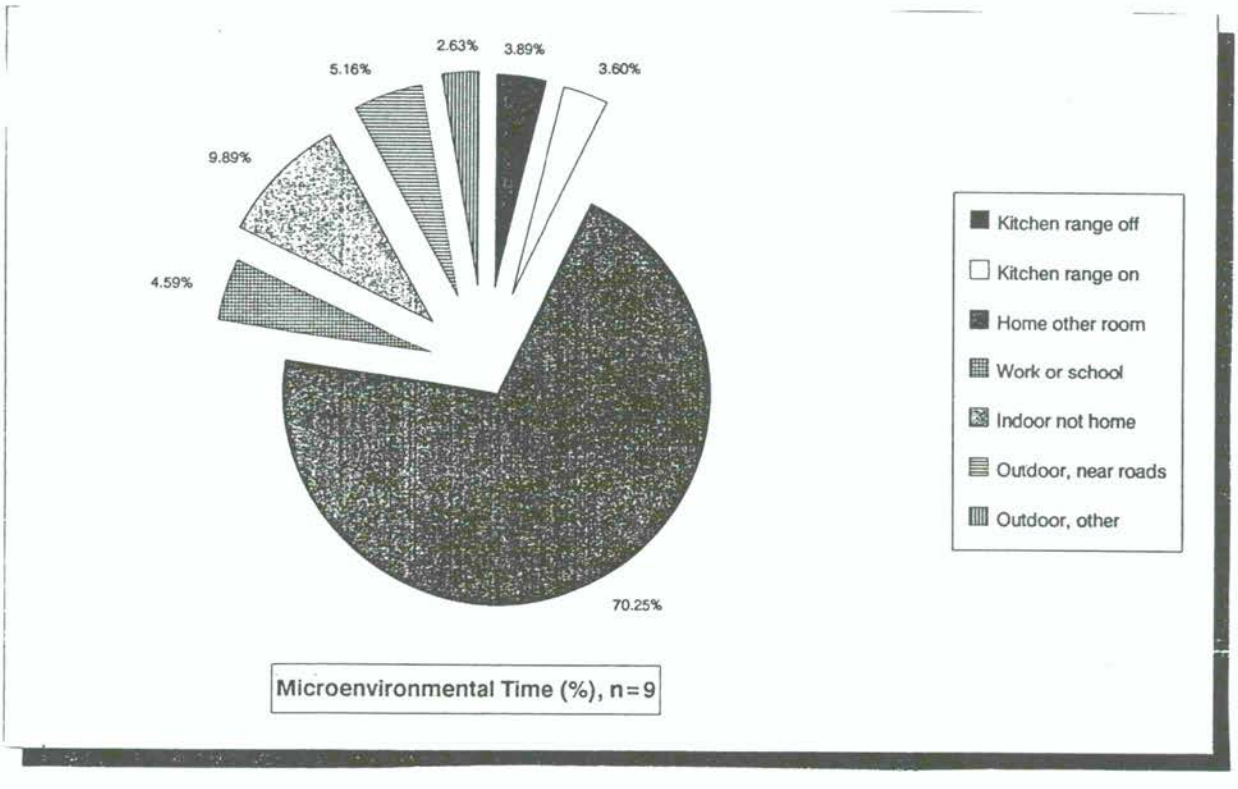

Table 4. Regression comparison among fixed-location samplers.

Dependent Variable Independent Variable	Bedroom Kitchen	Bedroom Outdoor	Kitchen Outdoor	Living Room Outside	Living Room Kitchen	Bedroom Living Room
R^2	0.54	0.62	0.40	0.69	0.86	0.80
S.E. of Y Estimate (ppb)	3.8	3.5	9.6	4.8	3.3	2.5
Constant	7.1	1.5	0.6	-1.5	4.8	3.1
X Coefficient	0.34	0.59	1.04	0.95	0.65	0.59
S.E. of Coef.	0.12	0.18	0.48	0.24	0.10	0.11

STATISTICAL ANALYSIS OF SAMPLE
SURVEY SAMPLING, INC.

Sample date: 1 March 1988

Total Counties: 2 (Orange and L.A.)

Average Household Effective Buying Income: $38,932

Total Persons in Sample Area: 10,489,600

	Total	% All Households	% Telephone Households
Households in Sample Area	3,819,800		
Listed Telephone Households	1,706,470	44.7	47.5
Est. Telephone Households	3,592,600	94.1	100.0

In the following sample analysis, the percentages are defined as follows:

Sample - % of total sample which fell in this county.
HHLD's - % of total households in universe from this county.
EstPhone - % of total telephone households in universe from this county.
W/Phone - % of telephone households in this county.
W/LPhone - % of telephone households in this county which are listed.

COUNTY	Sample	HHLDS	EstPhone	W/Phone	W/LPhone
Los Angeles CA	79.2	79.7	79.2	93.4	45.2
Orange CA	20.8	20.3	20.8	96.6	56.3

NITROGEN DIOXIDE (NITRITE) ANALYSIS OF EXPOSED AIR FILTERS

306 samples were received for analysis. The filters were analyzed using the following procedure and concentrations expressed in the absolute number of nanograms of NO_2^- on the filter. Each filter was quickly removed from its filter holder and placed into a 15 ml polystyrene test tube which was then immediately capped shut. After an entire batch of samples was processed in this manner, 10 ml of 15 mega-ohm resistivity water was pipetted into each tube. The entire run (usually 60 test tubes) was placed on an apparatus that inverted the tubes once every 2 seconds to facilitate NO_2^- extraction. The filters were subjected to this procedure for 10 minutes. Afterward, the extract was poured into a 4 ml polystyrene sampling cup.

Analysis was undertaken with a Technicon Autoanalyzer using Technicon colourimetric method 100-70W. Data were recorded on a chart recorder and comparison of peak height between standards and samples was used to assess sample concentration. The detection limit for the analysis was 5ng/ml. The samples with the lowest concentrations (i.e. blanks) were approximately 50 ng/ml. Samples greater than 2000 ng/ml were diluted and reanalyzed. The concentration of NO_2^- in 10 ml of water was then multiplied by ten to give the absolute amount of NO_2^- on the filter.

The quality control for the analysis included the analysis of an in-house quality control (QC) sample on every batch run and a duplicate analysis of every tenth sample. A water blank and a medium concentration standard were run every tenth sample in order to monitor instrumental drift. A blank test tube containing 10 ml of water without filter was "extracted" and analyzed with each batch of filters.

The results of the analysis of in-house QC samples (1000 ng/ml) were always within 5% of the expected value. Duplicate samples were always within 1% of its previous value. The batch "extracted" blank test tube always showed less than 5 ng/ml NO_2^-. Through analysis of a blank and standard every tenth sample it was concluded that instrumental drift was virtually non-existent.

Appendix C

WHO NO_2 Quality Control Samples.

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Master Bedroom			
Collection period	278	21970	13.78
7 days, 4:10 pm on	271	22920	14.38
9/5/88 to 5:55 pm on	258	22920	14.38
16/5/88 (10185 minutes)	255	22770	14.28
	276	22920	14.38
	272	22190	13.92
	259	22120	13.88
	290	22040	13.83
Mean ± S.D. 14.10 ± 0.27 ppb			
Range hood, left to right from first to last sample			
Collection period	228	82350	51.84
7 days, 4:20 pm on	270	80150	50.45
9/5/88 to 5:40 pm on	283	81620	51.38
16/5/88 (10150 minutes)	269	77570	48.83
	280	77570	48.83
	266	77940	49.06
	240	77570	48.83
	243	76840	48.37
Mean ± S.D. 49.70 ± 1.33 ppb			

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Family Room 8 feet off floor			
Collection period	223	37780	23.55
7 days, 3:00 pm on	277	37220	23.20
9/5/88 - 5:50 pm on	287	40830	25.45
10/5/88 (10250 minutes)	273	38330	23.89
	289	38890	24.24
	222	37220	23.20
	275	38890	24.24
	221	36670	22.86
	274	39440	24.59
	288	39170	24.42
Mean ± S.D. 23.96 ± 0.78 ppb			
Bedroom No. 2, 6 feet off ground			
Collection period	307	21620	13.64
7 days, 4:40 pm on	310	21620	13.64
9/5/88 to 5:30 pm on	356	21910	13.82
16/5/88 (10130 minutes)	360	21690	13.68
	358	21760	13.73
	357	21760	13.73
	359	21690	13.68
	309	22500	14.19
	308	22500	14.19
	306	21910	13.82
Mean ± S.D. 13.81 ± 0.21 ppb			

WHO NO_2 Quality Control Samples cont.

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Kitchen bookcase, 6 feet above ground			
Collection period	261	41670	26.13
7 days, 3:30 pm on	262	40830	25.60
9/5/88 to 5:20 pm on	263	41670	26.13
16/5/88 (10190 minutes)	232	47780	29.96
	227	42220	26.47
	241	37780	23.69
	233	41670	26.13
	224	42780	26.82
	231	43610	27.34
	264	40000	25.08
	235	41110	25.78
Mean ± S.D. 26.29 ± 1.55 ppb			
Garage, 5 feet above ground			
Collection period	284	25290	15.94
7 days, 4:05 pm on	285	25220	15.89
9/5/88 to 5:05 pm on	281	26250	16.54
16/5/88 (10140 minutes)	286	25740	16.22
	267	26030	16.40
	246	26030	16.40
	282	25510	16.07
	245	26320	16.58
	244	25590	16.12
	225	25880	16.31
Mean ± S.D. 16.25 ± 0.24 ppb			

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Dining room, 4 feet above ground on hutch			
Collection period	247	27010	16.99
7 days, 3:50 pm on	234	26420	16.62
9/5/88 to 5:10 pm on	229	26060	16.39
16/5/88 (10160 minutes)	239	26280	16.53
	236	26790	16.85
	237	26280	16.53
	254	26130	16.43
	238	26200	16.48
	230	26200	16.48
Mean ± S.D. 16.59 ± 0.20 ppb			
Field blanks, opened-then sealed			
Collection period	265	580	
4:25 pm on 9/5/88	279	580	
	363	580	
	362	590	
	365	810	
	361	590	
	364	660	
	260	660	
Mean ± S.D. 631.25 ± 80.08 ng			

WHO NO_2 Quality Control Samples cont.

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Kitchen, on top of refrigerator, 5.5 feet above ground			
Collection period	268	34200	21.55
7 days, 3:45 pm on	250	33910	21.37
9/5/88 to 4:45 pm on	248	33910	21.37
16/5/88 (10140 minutes)	257	34200	21.55
	256	34780	21.92
	242	34490	21.73
	251	34200	21.55
	226	34780	21.92
	253	33040	20.82
	252	34200	21.55
	249	34200	21.55
Mean ± S.D.	21.53 ± 0.30 ppb		

Location	Sample id	NO_2 (ng)	NO_2 (ppb)
Unopened blanks			
	291 na	580	
	331 na	720	
	333 na	580	
	337 na	560	
	340 na	580	
	297 na	580	
	318 na	510	
	319 na	580	
	330 na	580	
	348 na	510	
	344 na	1500	
	347 na	590	
	349 na	740	
	352 na	590	
	317 na	510	
	302 na	560	
	326 na	630	
	342 na	630	
	346 na	560	
	353 na	630	
	299 na	510	
	320 na	1030	
	328 na	590	
	329 na	590	
	367 na	510	
Mean ± S.D.	631.25 ± 80.08 ng		