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An international survey of indoor air quality, ventilation, and smoking activity in restaurants: a pilot study

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During a pilot study of indoor air quality in restaurants, a survey was performed in 34 medium-priced restaurants in six countries in Asia, Europe, and North America using a uniform protocol. The concentration of selected constituents of environmental tobacco smoke (ETS) present in occupied areas was determined during lunch and dinner periods by measuring the levels of four particulate-phase markers and two gas-phase markers. The particulate-phase markers determined were respirable suspended particles, ultraviolet particulate matter, fluorescing particulate matter, and solanesol particulate matter. The gas-phase markers were nicotine and 3-ethenylpyridine (3-EP). Correlation between the markers was investigated to explore an improved monitoring approach. It was concluded that at least one marker in each phase was necessary to describe adequately the ETS load. An assessment was made of the ventilation system in each restaurant, and effective ventilation rates were determined based on CO_2 measurements. Smoking activity was also monitored. These data were used to model nicotine and 3-EP concentrations that resulted in a satisfactory prediction of their levels, especially at the higher concentrations. A total number of 1370 questionnaires were returned by the restaurant patrons in five countries. In some countries, dissatisfaction rates above 20% were observed for draft, freshness of air, and noise. The dissatisfaction rates related to tobacco smoke were less than 20%, which is lower than would be predicted based on measured ETS levels. Based on the results of this international pilot study, recommendations are given for future studies of this type.
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Introduction

A number of studies have been conducted that assess the impact of environmental tobacco smoke (ETS) in public facilities including restaurants. Some of these studies have focused on chemical measurements (Jenkins et al., 2000), ventilation measurements (e.g., Bohanon et al., 1998), occupant questionnaires pertaining to air quality acceptability (e.g., Moschandreas and Vuilleumier, 1999), and observation of smoking activity (e.g., Ott, 1999). Results have been used to propose various indoor air quality (IAQ) models (Bohanon and Cole, 1997). However, only a few investigations have included data for all of these parameters within any single investigation (e.g., Hodgson et al., 1996; Daisey et al., 1998). To construct and test IAQ models, all parameters that impact on the results should be measured simultaneously.

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We report the results of an international study that incorporated simultaneous measurements of the parameters described above. As this was a pilot investigation, it was not designed to yield representative data for the individual countries. However, the data yielded valuable information on the experimental protocol, methodology, ETS concentration, and occupant perception of the indoor environment. Recommendations for improved study design and implementation are based on these results.

This project was designed as a pilot for a better understanding of issues associated with a comprehensive indoor environment investigation using the following six steps:

1. To survey restaurants in several countries.
2. To determine concentrations of selected ETS constituents present in occupied spaces in restaurants.
3. To determine acceptability of indoor environmental conditions as judged by occupants of the restaurant space.
4. To appraise the ventilation system including operation and maintenance of heating, ventilating, and air conditioning (HVAC) systems, and to estimate ventilation rates.
5. To determine smoking rates simultaneous with the other measurements.

6. To investigate correlation among ETS constituent concentrations, smoking rates and ventilation rates including the potential for modeling, and occupant perceptions of the indoor environment.

The study was based on a uniform protocol and conducted in 34 medium-priced restaurants where smoking was permitted that were located in six countries (France, Korea, Japan, Switzerland, United Kingdom, and United States). Air samples and questionnaires were obtained during the lunch and/or dinner period on high occupancy days. Minor modifications of the protocol for the study were necessary to accommodate circumstances in individual countries.

Components of the investigation

This investigation consisted of three principal parts: area sampling and chemical analysis, questionnaires completed by occupants, and assessment of the ventilation system. Each of these will be described below.

Area Sampling and Chemical Analysis

As the results obtained by different teams in different countries were to be consolidated, an experimental protocol was established and distributed to the participants to ensure that consistent methodology would be used in sampling and analytical determinations. The participating laboratories agreed to abide by this protocol whenever possible. However, some deviations were observed, reflecting local practices, facilities, and analytical procedures available to the participants. The protocol relied on area monitoring to obtain quantitative information on the environmental conditions within the restaurants. Respirable suspended particulate matter (RSP), although not specific to ETS, was determined because many investigators use it as a reference criterion. Three additional markers were measured to assess the ETS contribution to the aerosol particulate matter. Two of them, ultraviolet particulate matter (UVPM) and fluorescing particulate matter (FPM), reflect the airborne levels of combustion aerosols (Jenkins et al., 2000). These markers can be determined simultaneously (Ogden et al., 1989; Conner et al., 1990) and have been used extensively in similar studies to provide a more selective assessment of ETS contribution to RSP (Jenkins et al., 2000). The third marker, solanesol particulate matter (Sol-PM), has become increasingly used in recent studies. It is based on the measurement of the concentration of solanesol, a nonvolatile trisesquiterpenoid alcohol that can be found only in aerosols that result from the combustion of tobacco. Therefore, it is specific for the assessment of the ETS contribution to RSP (Ogden and Maiolo, 1989; Jenkins et al., 2000; Douce et al., 2001). The use of all the three markers rests on the application of factors to derive the particulate matter concentrations from the measured data (Nelson et al., 1997, 1998). As solanesol is in itself descriptive of the ETS level in the environment, its concentration values were reported in addition to the Sol-PM levels.

The gas-phase markers for ETS determined in this study were nicotine and 3-ethenylpyridine (3-EP), the latter being a product of the thermal decomposition of nicotine (Heavner et al., 1995). Both compounds are specific to tobacco smoke. In addition to markers associated with ETS, measurements were made for carbon monoxide and carbon dioxide levels. Carbon dioxide was monitored in real time and used to estimate the effective ventilation rate as a function of time and occupancy. The temperature and relative humidity (RH) in the occupied areas were also recorded.

Questionnaires

Restaurant owners are motivated by business reasons to strive for customer satisfaction. However, little guidance on how to provide acceptable indoor environmental conditions for restaurants is available. Some national or international ventilation standards or guidelines provide general information (e.g. CEC, 1992; CEN, 1998; ASHRAE, 1999) based on laboratory environment studies (Cain et al., 1983) or field tests in some workplace environments (Fanger, 1988). Only a few studies have tested the applicability of this information to the hospitality environment (Moschandreas and Vuilleumier, 1999). In this study, questionnaires were used to evaluate occupant perceptions of the indoor environment, and these were combined with chemical analysis and ventilation assessment to provide relations between all three components in restaurants. This approach is helpful in the evaluation of the applicability of published guidelines to restaurant indoor air environments.

Ventilation Assessment

In the simplest terms under steady-state conditions, the concentration of a given indoor air contaminant is the result of the source generation rate divided by the contaminant removal rate, which requires an assessment of ventilation to determine. A variety of ways to estimate ventilation rates have been used (Turner et al., 1995). Direct measures of airflow are reliable only if all of the air exchange is controlled through a mechanical system. A tracer gas such as sulfur hexafluoride (SF_6) can be introduced into the area and its concentration measured as a function of time. A similar method uses carbon dioxide, naturally generated by the people in the space, as a tracer gas, where the generation rate is estimated as a function of the number of people present (Seppänen et al., 1999).

Each of these methods offers advantages and disadvantages. Mechanical air exchange measurements are not well suited to restaurants. Lack of control of normal events such as door and window openings, variable use of kitchen ventilator fans, open-hearth fires, and other factors can make this approach very inaccurate in describing a dynamic ventilation situation. Use of SF_6 as a tracer gas is a reliable method. However, it must be well mixed in the space and some have concerns about introducing it into occupied spaces. In addition, the needed instrumentation is costly and not readily available.

The carbon dioxide method was used in this investigation. When measurements of carbon dioxide concentration are combined with a single-compartment model, estimates for air exchange rate may be obtained. Simultaneous measurements of both indoor and outdoor CO_2 levels are required at regular intervals over the time period of concern. Additional data requirements include the volume of the room(s) and an accurate time-based count of the people present in the tested area including patrons, employees, and investigators. Selection of the sampling points is an important consideration. The carbon dioxide method is best applied to single rooms, but may also be applied to multiroom or multifloor facilities. In the case of low occupancy, the indoor CO_2 levels are close to the background threshold and the calculations are imprecise. As a result, the carbon dioxide method is most accurate in single rooms with high occupancy.

Methods

Experimental Procedure

Sampling Location and Duration Two sampling locations were identified in each restaurant. If the restaurant had smoking and nonsmoking sections, measurements were made in each section. The locations for ETS sampling equipment were chosen to be unobtrusive; at least 50 cm from any wall, and located approximately at head height of a seated person. Care was taken to assure that sampling would not be influenced by fans or ventilation systems, or by direct exposure to sidestream or exhaled mainstream smoke plumes. At each location, the protocol recommended that sampling was to be performed in duplicate over a 3–4-h period during the busy serving hours, and was scheduled for an active rather than a quiet day of the week. In most cases, the same restaurant was investigated over two consecutive days. The instruments for CO_2 measurement were placed near the exhaust.

Air Sampling Methods Particulate matter samples were collected using 37-mm filters at a flow rate of 2 l/min with a cyclone vortex assembly establishing a cutoff at an aerodynamic diameter of 3.5–4 μm. Opaque filter holders (SKC Inc., Eighty-Four PA) were recommended for the sampling to ensure the stability of solanesol collected on the filter (Ogden and Richardson, 1998). Owing to unacceptably high detection limits, French investigators did not report solanesol data. US investigators encountered an early batch of opaque holders that caused highly irregular weight increases by transfer of an oily residue from the holders to the filters. The RSP values were rejected, but determination of the other markers (UVPM, FPM, and Sol-PM) was unaffected.

Gas-phase samples were collected with XAD-4 cartridges (SKC Inc., Eighty-Four PA) using an airflow of approximately 1 l/min. Battery-operated membrane pumps with timers and electronically regulated flow (SKC Inc., Eighty-Four PA, or equivalent) were used for sampling. Flow rates were checked before and after each sampling, and an averaged value was reported, unless the difference was larger than 10% in which case the sample was considered invalid. At each session, field blanks were prepared, handled, and analyzed in the same manner as the samples. Analytical results for a nonzero blank were subtracted from the corresponding raw results.

Analytical Determinations The analytical procedures for the measurement of ETS markers have been reviewed (Jenkins et al., 2000) and documented in ASTM methods D 5075-96 (1996) (Nicotine and 3-EP), ASTM D 5955-96 (1996) (UVPM and FPM), and ASTM D 6271-98 (1998) (Sol-PM), and are summarized as follows.

Particulate matter. Air concentrations of RSP were determined by gravimetry. Fluoropore-TM filters (Millipore Corp., Bedford MA, FALP037000) were weighed in triplicate on an electro balance with $\pm 1\,\mu$g resolution after overnight equilibration under controlled humidity (60% RH) and temperature (22°C). After sampling, the same weighing procedure was repeated and the weight difference was reported as RSP. The two estimates of the air concentration of combustion aerosols (UVPM, FPM) were obtained simultaneously from a methanol extract of the filters that sampled the RSP (ASTM, D 5955-96, 1996). The extract was injected into an HPLC equipped with a UV detector and a fluorescence detector in series, and connected directly to an autosampler by short tubing. The readings were expressed as equivalent concentrations of surrogate standards (2,2′,4,4′-tetrahydroxybenzophenone for UV and scopoletin for fluorescence), and converted to UVPM and FPM concentrations using conversion factors (Conner et al., 1990; Nelson et al.,1997, 1998; ASTM, D 5955-96, 1996). The air concentration of solanesol was determined by HPLC analysis of the same extract (Ogden et al., 1989). In addition to reporting solanesol levels, these results were used to derive a selective estimate of the concentration of particulate matter originating from ETS (Sol-PM) using conversion factors (Nelson et al., 1997, 1998; ASTM, D 6271-98, 1998).

Gas-phase components. The XAD-4 tubes were extracted with ethyl acetate containing 0.01% of triethylamine and quinoline as internal standards. Nicotine and 3-EP were determined simultaneously in the extract by capillary gas chromatography with NPD detection, according to ASTM

Table 1. Limits of detection (μg/m^3) for the ETS constituents reported by the participating laboratories.

LOD (μg/m^3)	Nicotine	3-EP	RSP	UVPM	FPM	Sol-PM
France	0.30	0.15	28	2.9	1.41	NA
Japan	0.25	0.25	21	2.2	NA	0.41
Korea	0.60	0.30	9	0.37	1.8	0.50
Switzerland	0.08	0.05	15	0.42	0.38	6.5[a]
USA	0.03	0.01	21	0.78	0.41	0.50
UK	0.42	0.42	20	2.1	0.48	0.97

[a]The large LOD resulted from a low resolution of the HPLC column and smaller injected volume.

method D 5075-96. Measurement of carbon dioxide was performed using nondispersive IR detectors.

Limits of detection. The limit of detection (LOD) of the analytical methods, expressed in units of μg/m^3 in air, for each country is shown in Table 1. These values are those reported by the laboratory conducting the work. Although there may be some variation in the method used to estimate the LOD, they are generally based on the premise that the signal must be greater than three times the standard deviation of the background or blank. The data show some variability reflecting the exact air sample volumes that were used, the analytical method and instrumentation employed, as well as the statistical criteria used to establish the LOD.

Questionnaire The Pilot Study Protocol provided a questionnaire written in English that sought basic information from the occupants. The questionnaire also assessed their perception concerning a number of indoor environmental conditions including noise, temperature, draft, odor, humidity, freshness, environmental tobacco smoke, and overall IAQ and/or indoor environmental quality (IEQ). The questions were translated and adapted to each local language. In some cases, questions were added locally to the original questionnaire.

Questionnaires were completed simultaneously with indoor air measurements. Normally, the wait staff administered questionnaires after they took orders for the meal. The completed questionnaires were returned to the wait staff. No reliable estimates were obtained for the percentage of questionnaires returned. The wait staff received a small gratuity for the additional workload of administering and returning the questionnaires.

The perceptions of the occupants were classified as satisfied or dissatisfied with a particular item. A dissatisfied occupant was defined as a person marking one of the two lower ratings of a 5-point scale. The evaluation was slightly different in Switzerland in that acceptability towards IAQ was assessed on a +5 to −5 scale, so as to follow the method used in the European Building Audit (Bluyssen et al., 1995). In analyzing the Swiss data, the occupants who voted below the border between just acceptable and just not acceptable were taken as being dissatisfied. The percentages of occupants dissatisfied with IAQ and with IEQ were averaged and used to obtain the inverse estimate of percent "Overall Acceptance."

The number of questionnaires returned and the variability of the ventilation rate during a given sampling period placed the restaurants in two categories with substantial gaps between the two groups. Those restaurants ($n=14$) that had either few questionnaires returned ($n=10$) and/or high variation in ventilation rate during the course of a measurement period ($n=10$) were excluded from the correlation analysis. Consequently, the percent of Overall Acceptance data were analyzed for 15 restaurants, each of which had at least 20 questionnaires returned and less than a factor of two variation in ventilation rate.

Assessment of Ventilation System The protocol used to assess ventilation addressed both natural and mechanical ventilation systems. Professional HVAC engineers evaluated the adequacy, cleanliness, and structural integrity of the mechanical HVAC systems in use. Two methods were employed to determine the rate of outside air supply to the test space. These were: (a) a test and balance (T&B) engineering consultant estimated air exchange rates from the available mechanical ventilation parameters and in-duct airflow measurements, and (b) CO_2 measurements combined with counts of people in the space were used to estimate air exchange rates. The latter method is applicable in the presence of smoking, because the exhalation rate of CO_2 by humans is about 0.3 l/min or approximately 30–35 g/h (ASHRAE, 1999), whereas emission from cigarettes is about 500 mg/cigarette.

Estimation of Ventilation Rates The use of carbon dioxide to estimate restaurant ventilation rates is based on CO_2 exhalation at a rate of 0.3 l/min-person (ASHRAE, 1999). If CO_2 is removed by ventilation from outside air at a constant rate and the room air is well mixed, the difference between measured indoor and outdoor CO_2 concentration provides a means to estimate the rate of ventilation. The concentration of CO_2 in the indoor air at any time may be estimated by (Ishizu, 1980)

$$C(t) = \left(C_0 - \frac{G + C_i Q_i}{Q_i}\right) \exp\left(-\frac{Q_i t}{V}\right) + \frac{G + C_i Q_i}{Q_i} \quad (1)$$

where $C(t)$ is the CO_2 concentration after time t, G the generation rate of CO_2 from people, C_i the outside CO_2 concentration, t the time, C_0 the initial CO_2 concentration, Q_i the estimated ventilation rate, and V is the total room volume.

The parameters C_0, G, V, and an initial estimate of the ventilation rate Q_i were entered into a Microsoft Excel for Windows® spreadsheet. Columns of time intervals and number of persons present during those time intervals were used with a column containing Eq. (1) to compute estimated $C(t)$ values. Since these values were based on an initial estimate of Q_i, they were not expected to be accurate. Then, Microsoft Excel Solver was used to vary the value of Q_i to minimize the mean square error between the calculated $C(t)$ values and those measured at the same time intervals during the experiment. This method yielded an estimate of the ventilation rate that fit the model to the experimental results.

Determination of Occupancy and Smoking Activity The number of people in each room, whether in smoking or nonsmoking areas, was estimated from an average count every 30 min. Two methods to determine smoking activity were recommended in the study protocol. The first method was collection and counting the cigarette butts about every 30 min. The second method was frequent periodic visual observation and tabulation of occupant smoking. Regardless of the method used, the number of cigarettes smoked per hour was estimated.

Results and discussion

Results from the Analytical Measurements

Investigations in 34 restaurants in six countries generated a large amount of analytical data and reflected the diversity of situations encountered in such locations. Because of the many sources of variability, the raw data were screened for aberrant results. Estimates of tobacco smoke contribution to RSP (UVPM, FPM, and Sol-PM) were compared to the corresponding gravimetric determination of total RSP. When one (or both) estimate based on combustion by-products (UVPM and FPM) was substantially higher than the corresponding gravimetric determination, it was rejected if the Sol-PM was compatible with the total RSP. When both estimates of tobacco smoke contribution, combustion-based and solanesol-based, were substantially higher than the corresponding gravimetric determination, the latter was rejected. When replicate determinations were obtained, individual results among the set of replicates that deviated by more than 25% from the mean were also rejected following the recommendations made in Woolfenden and McClenny (1999). The median, arithmetic mean, geometric mean, standard deviation, and the levels corresponding to the 80th and 95th percentiles in each country for each analyte after elimination of the outliers are shown in Table 2.

The results in Table 2 are comparable to other reported area-monitoring data for the levels of ETS markers in restaurants where smoking was permitted. A summary of these data is available in a review by Jenkins et al. (2000). Furthermore, the data can be compared by geographic region and by gas- and particulate-phase constituents. Carbon monoxide does not appear in Table 2 because the data did not correlate to any other ETS marker, in accordance with previous reports (Jenkins et al., 2000).

Nicotine Results

Nicotine arithmetic mean levels between 2.8 and 8.4 $\mu g/m^3$ were measured during the 1990–1993 period in US restaurants where smoking was allowed (Jenkins et al., 2000). Means of 3.1–4.5 $\mu g/m^3$ (Moschandreas and Vuilleumier, 1999) and 6.0 $\mu g/m^3$ (Maskarinec et al., 2000) were reported in smoking sections of US restaurants. Mean nicotine levels of 14.8 $\mu g/m^3$ (Muramatsu et al., 1987) and 4.8 $\mu g/m^3$ (Baek et al., 1997) were reported for Asian restaurants. In the case of European restaurants, reports of a mean of 15 $\mu g/m^3$ (Kirk et al., 1988), and ranges of 5.0–90 $\mu g/m^3$ averaged at 32 $\mu g/m^3$ (Löfroth, 1993), 2.2–33.5 $\mu g/m^3$ averaged at 12.3 $\mu g/m^3$ (Meger et al., 2000), and 0.4–46 $\mu g/m^3$ averaged at 7.0 $\mu g/m^3$ (Hyvärinen et al., 2000) have been published. Nicotine levels shown in Table 2 are within the range of previously reported results in the comparable geographic regions.

Particulate Matter Results

Particulate matter levels in restaurants where smoking is allowed are often reported only as total RSP. However, several studies report particulate matter results from area monitoring that are more specific to combustion including ETS (UVPM, FPM, and Sol-PM). Mean values for combustion-derived particulate matter between 26 and 67 $\mu g/m^3$ have been reported in US studies (Crouse et al., 1988; Crouse and Carson, 1989; Ogden et al., 1990). Mean values of 29.4 $\mu g/m^3$ for combustion aerosol and 20.4 $\mu g/m^3$ for Sol-PM were reported in US restaurants (Maskarinec et al., 2000). The mean UVPM and FPM values found in this study are similar to those reported by Maskarinec, but the Sol-PM is somewhat lower. In Asia, mean levels of 45.6, 22.5, and 25.4 $\mu g/m^3$ were reported for UVPM, FPM, and Sol-PM, respectively (Baek et al., 1997). Korean results shown in Table 2 are comparable to those values for UVPM but somewhat higher for FPM and Sol-PM.

Comparison of ETS Levels in Smoking and Nonsmoking Areas

Three of the restaurants investigated in the US and three of the restaurants investigated in the UK were divided into smoking and nonsmoking sections. A comparison of the levels measured in these settings is shown in Table 3. These results indicate that a greater difference exists between the means of the measured levels of ETS-specific markers (nicotine and Sol-PM) in smoking and nonsmoking areas than those that are less specific (UVPM and FPM). The ratio of the means of nonsmoking to smoking levels for nicotine matches well that of Sol-PM.

Table 2. Summary of the compiled analytical results ($\mu g/m^3$).

Country	Statistic	Nicotine	3-EP	Solanesol	RSP	UVPM	FPM	Sol-PM
France	n =	15	15	0	15	15	15	0
	Arithmetic mean	30.3	3.08	NA	188	116	90.5	NA
	SD	21.1	2.20	NA	75.5	61.9	59.5	NA
	Min	0.00	0.00	NA	56.0	34.0	6.00	NA
	Max	71.6	7.80	NA	312	223	199	NA
	Median	24.1	2.40	NA	194	118	101	NA
	Geom. mean	19.7	2.22	NA	170	99.0	65.9	NA
	Percentile 80th	44.2	3.96	NA	243	156	123	NA
	Percentile 95th	69.2	7.52	NA	292	221	188	NA
Japan	n =	16	16	16	16	16	0	16
	Arithmetic mean	11.7	2.67	2.16	242	108	NA	77.9
	SD	5.35	1.54	2.33	175	70.2	NA	83.9
	Min	3.37	0.44	0.00	0.00	29.5	NA	0.00
	Max	22.4	4.86	7.67	611	291	NA	276
	Median	11.1	3.06	1.03	194	87.7	NA	36.9
	Geom. mean	10.5	2.13	1.25	172	91.4	NA	45.0
	Percentile 80th	14.8	3.80	3.82	423	133	NA	138
	Percentile 95th	20.9	4.85	6.94	558	258	NA	250
Korea	n =	47	47	50	50	49	47	50
	Arithmetic mean	5.72	1.66	1.14	109	41.4	49.8	37.6
	SD	4.13	0.85	0.65	29.9	16.2	20.1	21.4
	Min	1.60	0.10	0.02	54.0	14.3	17.3	0.65
	Max	18.8	3.50	3.05	172	87.8	102	101
	Median	3.95	1.80	1.20	107	39.3	45.4	40.0
	Geom. mean	4.63	1.33	0.86	105	38.2	46.0	28.3
	Percentile 80th	7.50	2.37	1.60	138	53.6	66.2	52.9
	Percentile 95th	13.5	2.89	2.16	159	66.3	89.2	71.4
Switzerland	n =	32	35	31	31	36	33	31
	Arithmetic mean	7.81	2.54	1.52	92.0	57.0	73.3	50.1
	SD	10.7	2.57	1.73	67.9	58.1	71.8	57.0
	Min	0.08	0.16	0.00	0.00	0.00	0.00	0.00
	Max	39.6	10.5	6.06	277	231	286	200
	Median	3.98	1.59	0.75	74.6	36.2	52.3	24.8
	Geom. mean	3.79	1.70	0.71	67.9	32.8	41.6	23.6
	Percentile 80th	7.04	3.70	2.69	126	92.8	126	88.7
	Percentile 95th	34.6	7.91	5.28	219	178	218	174
UK	n =	20	18	15	12	16	16	15
	Arithmetic mean	9.78	2.73	3.12	195	123	87.3	103
	SD	6.92	1.36	1.92	84.3	75.1	48.7	63.5
	Min	0.80	0.71	0.36	62.1	25.5	14.4	11.8
	Max	27.6	6.38	7.60	391	316	197	250
	Median	10.1	2.57	2.81	201	97.6	81.0	92.8
	Geom. mean	6.88	2.37	2.47	177	102	72.8	81.4
	Percentile 80th	15.3	3.63	4.75	233	173	134	157
	Percentile 95th	20.0	4.35	5.72	311	243	157	189
USA	n =	18	20	19	0	20	20	19
	Arithmetic mean	3.03	1.27	0.47	NA	31.8	26.2	15.6
	SD	3.36	1.22	0.59	NA	26.3	22.8	19.5
	Min	0.04	0.11	0.00	NA	1.43	0.78	0.00
	Max	9.43	3.72	1.61	NA	81.4	69.5	53.0
	Median	1.52	0.87	0.20	NA	243	18.3	6.67
	Geom. mean	1.33	0.77	0.11	NA	18.8	14.0	3.61
	Percentile 80th	6.81	2.04	1.10	NA	62.2	53.7	36.4
	Percentile 95th	9.10	3.51	1.47	NA	75.8	64.2	48.4

NA: not available.

Table 3. Mean values of nicotine, Sol-PM, and the mean of UVPM and FPM (combustion PM) in restaurants with separate smoking and nonsmoking sections in the USA and the UK.

Country	Nicotine (μg/m^3) Nonsmoking/smoking	Sol-PM (μg/m^3) Nonsmoking/smoking	Combustion PM (μg/m^3) Nonsmoking/smoking
USA	0.44/2.9 ratio = 15%	3.1/16.7 ratio = 18%	14.8/24.9 ratio = 59%
UK	1.12/7.6 ratio = 13%	11.8/93.1 ratio = 13%	41.8/104 ratio = 40%

Note: LOD was used when the sample yielded a result below the detection limit.

Table 4. Correlation table (Spearman above to the right of the diagonal and Pearson below to the left of the diagonal for six ETS markers in each country).

	Nicotine	3-EP	RSP	UVPM	FPM	Sol-PM
USA						
Nicotine	1	0.946	NA	0.831	0.810	0.862
3-EP	0.988	1	NA	0.893	0.871	0.858
RSP	NA	NA	1	NA	NA	NA
UVPM	0.954	0.955	NA	1	0.993	0.955
FPM	0.929	0.929	NA	0.995	1	0.960
Sol-PM	0.908	0.886	NA	0.956	0.967	1
UK						
Nicotine	1	0.971	−0.063	0.791	0.674	0.804
3-EP	0.969	1	−0.018	0.771	0.613	0.785
RSP	−0.150	−0.037	1	0.322	0.329	0.418
UVPM	0.770	0.802	0.347	1	0.941	0.989
FPM	0.752	0.732	0.299	0.969	1	0.932
Sol-PM	0.844	0.848	0.623	0.971	0.970	1
Switzerland						
Nicotine	1	0.977	0.923	0.979	0.825	0.927
3-EP	0.987	1	0.907	0.959	0.796	0.923
RSP	0.930	0.946	1	0.897	0.785	0.939
UVPM	0.982	0.973	0.943	1	0.882	0.878
FPM	0.972	0.973	0.964	0.989	1	0.793
Sol-PM	0.974	0.957	0.898	0.981	0.970	1
Korea						
Nicotine	1	0.103	0.100	0.420	0.354	0.406
3-EP	0.022	1	0.296	0.235	0.276	0.291
RSP	0.134	0.305	1	0.444	0.503	0.461
UVPM	0.371	0.186	0.433	1	0.589	0.774
FPM	0.354	0.275	0.555	0.543	1	0.533
Sol-PM	0.348	0.233	0.437	0.778	0.494	1
Japan						
Nicotine	1	0.671	−0.294	0.024	NA	−0.302
3-EP	0.726	1	−0.688	−0.388	NA	−0.526
RSP	−0.260	−0.698	1	0.641	NA	0.655
UVPM	−0.034	−0.463	0.793	1	NA	0.834
FPM	NA	NA	NA	NA	1	NA
Sol-PM	−0.261	−0.623	0.856	0.961	NA	1
France						
Nicotine	1	0.670	−0.002	0.418	0.489	NA
3-EP	0.931	1	0.303	0.637	0.587	NA
RSP	0.054	0.111	1	0.552	0.571	NA
UVPM	0.592	0.547	0.519	1	0.968	NA
FPM	0.630	0.575	0.521	0.986	1	NA
Sol-PM	NA	NA	NA	NA	NA	1

NA: not available.

Correlation between the Marker Levels

The protocol called for the measurement of a number of ETS markers using methods that have been well assessed in field studies. An objective was to explore whether, based on correlation between ETS marker concentrations, comparison of the different results obtained in a single sampling can be an efficient way to improve the reliability of the results. This approach provided an insight into the composition of ETS that serves as an investigative tool, and that might lead to the ability to conduct studies, while measuring a reduced set of analytes. Therefore, the relations between the levels of RSP, UVPM, FPM, Sol-PM, nicotine, and 3-EP were investigated. A correlation table was constructed from the ETS data from each country. The results are shown in Table 4, with the Spearman rank correlation coefficients given above and to the right of the diagonal and the Pearson correlation coefficients below and to the left of the diagonal. They suggest that a better correlation exists between the markers present in the same phase, whether gas or particulate. Figure 1 shows the correlation between the raw data obtained in Switzerland (80 data points including replicates) for the ETS markers investigated.

The results shown in Figure 1, as well as scatter plots generated using data from other countries, and the correlation matrix in Table 4 suggest the following conclusions:

- There is a good correlation between the two spectrometric estimations of particulate-phase concentration (UVPM and FPM). This is expected because both determinations are made from the same sample. Therefore, any outlying point in the correlation should be investigated for possible analytical error or interference in one of the determinations.
- The solanesol concentration (or the derived Sol-PM estimation) is well correlated with UVPM or FPM. The ratio of Sol-PM to UVPM *versus* UVPM was plotted in Figure 2. This plot shows that at higher smoke levels, the ratio was close to one. There were, however, some data points that show lower Sol-PM than UVPM or FPM at lower smoke levels, suggesting that the contribution of combustion aerosols other than from tobacco smoke was substantial in those cases. Where a ratio larger than one is found, they occur at trace ETS levels and are likely a result of conducting measurements close to the LOD.
- Over the entire data set, it appears that the RSP level does not correlate well with levels of particulate matter that are highly specific to tobacco smoke. Based on the medians, combustion aerosols were found to contribute about 50% of the RSP and Sol-PM about 34%. In order to simplify some parts of further discussions, a single estimate of the ETS contribution to RSP was calculated from the different available markers. This contribution was determined by first calculating the average of the values of UVPM and FPM that represent all the contributions from combustion sources. This result, usually an overestimate, was averaged with the value for tobacco-specific Sol-PM. The resulting value, labeled ETS-RSP, was used throughout the remainder of this study as the indicator of ETS levels. At very high ETS levels, ETS–RSP and the total RSP may be almost equal. At lower ETS levels, ETS–RSP may be a fraction of the total RSP.
- The nicotine concentrations are well correlated with the 3-EP concentrations. There is a trend towards lower nicotine to 3-EP ratio at lower smoke levels as shown in Figure 3. As nicotine exhibits larger sorption effects, this ratio will be higher in fresh ETS. This is more likely to happen in those situations where ETS levels are highest (Guerin et al., 1992; Eatough, 1993).

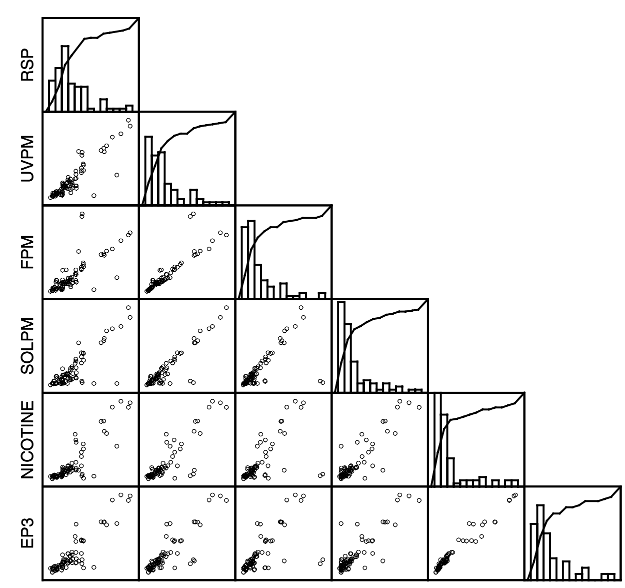

Figure 1. Correlation plots between the ETS markers and plot of their distribution (Swiss restaurant data, $n = 80$). (The right-most chart in each row shows a cumulative frequency plot overlayed on a concentration histogram for the respective ETS marker.).

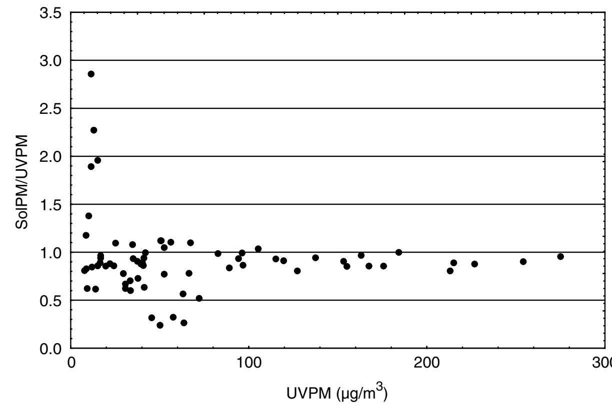

Figure 2. Ratio of Sol-PM to UVPM *versus* UVPM (Swiss data).

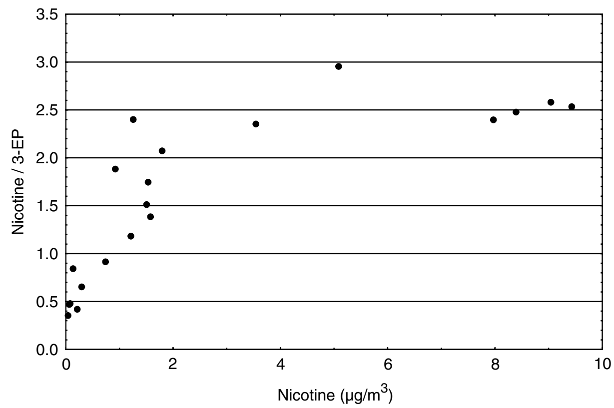

Figure 3. Ratio of nicotine to 3-EP *versus* nicotine level (US data).

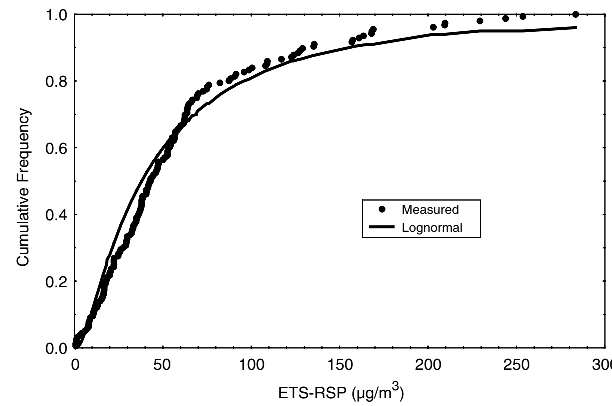

Figure 5. Distribution of ETS–RSP results from all the restaurants surveyed ($n = 156$).

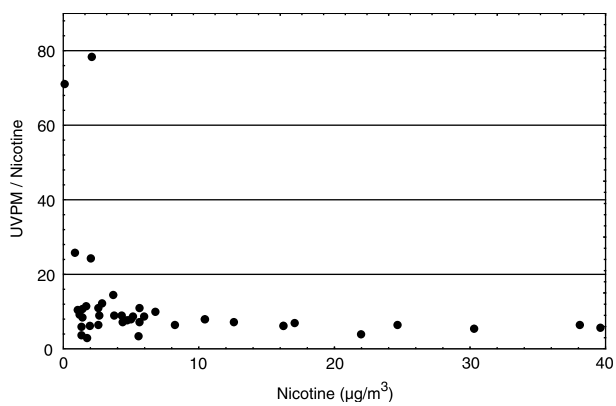

Figure 4. Ratio of UVPM to nicotine *versus* nicotine level (Swiss data).

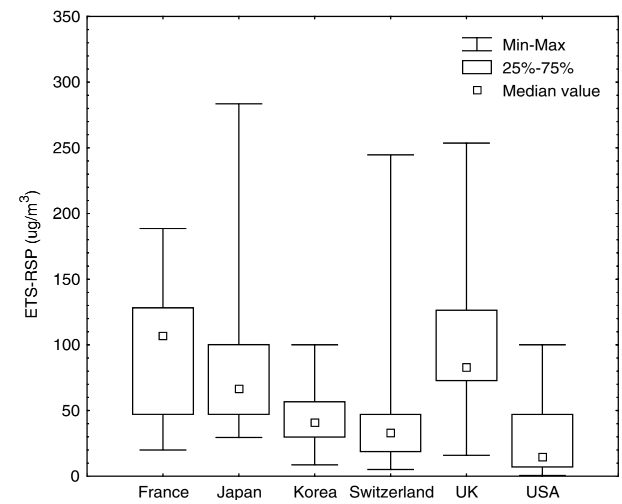

Figure 6. Box and Whisker plots for ETS–RSP median values, 25–75% quartiles and range by country.

The concentration of the particulate matter markers can also be correlated to the gas-phase data. As an example, the ratio of UVPM and nicotine levels was computed and plotted as a function of the nicotine concentration in Figure 4. At higher nicotine concentration, the ratio is more constant as the impact of sorption effects is less important. In this case, the ratio of ETS–RSP to nicotine is close to 5; comparable to the values found in experimental settings, or when fresh smoke data are included in the evaluations (Van Loy et al., 1998). It can be observed that ratios between the levels of gas-phase and particulate-phase compounds become extremely variable at low ETS concentrations, typically below nicotine concentrations of 5–10 $\mu g/m^3$ according to Figure 4. At these low levels, using nicotine data to predict the ETS–RSP levels appear to be inappropriate.

Distribution of the Levels of the Measured ETS Constituents

Figure 5 shows the distribution of the estimates for ETS–RSP concentrations, while Figure 6 shows a plot of these concentrations by country. Overall, the data follow a lognormal distribution, although the central values and ranges of ETS–RSP differ by country. This suggests that comparing arithmetic means is not the ideal way to summarize distributions that have consistently been reported to be lognormally distributed (Jenkins et al., 2000), or to follow some other distribution than normal (Sofuoglu and Moschandreas, 1999). This is especially important when the results contain a large percentage of values below the analytical detection limit (Kirk et al., 1988). In cases of lognormal or other skewed distributions, a small number of large values will result in an arithmetic mean that is much larger than the median or geometric mean, thereby misrepresenting the distribution of values.

Consideration of frequency distributions provided another way of assessing the ratio of particulate matter to nicotine. Figure 7 shows the cumulative frequency of the ratios of nicotine to the levels of each of the three markers UVPM, FPM, and Sol-PM, as well as ETS-RSP and RSP. The spread of the ratios encompasses more than an order of

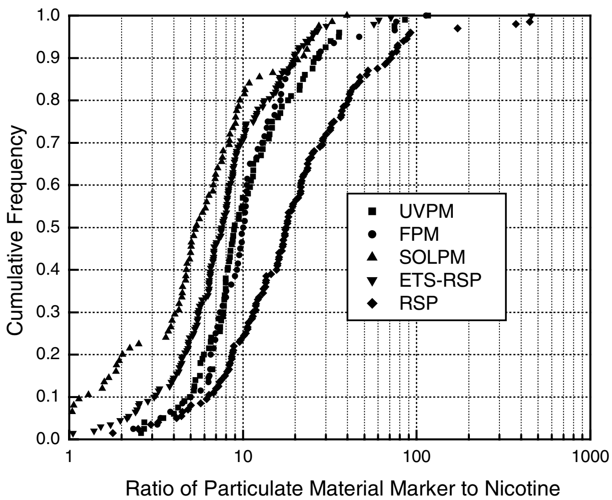

Figure 7. Cumulative frequency of the ratios to nicotine of the levels of UVPM, FPM, Sol-PM, ETS-RSP, and RSP.

magnitude. It confirms that UVPM and FPM give overall equivalent results. Their respective medians differ slightly and correspond to ratios of 9.0 and 10.0 for UVPM and FPM, respectively, which is compatible with the estimations proposed by Van Loy et al. (1998). Sol-PM is more specific for ETS, and its median corresponds to a ratio of 5.4. The median of ratio ETS-RSP to nicotine was found to be 7.6.

Methodology Assessment

There were no consistent problems associated with the methodology used in this study. It was confirmed that it is important to perform two or more replicate determinations at each location, which helps to detect outliers. For example, an inordinately high value may indicate that the sample was taken too close to a smoke source, and that the result is not representative of the assessed environment. Based on the criteria described earlier, about 5% of the data were considered as outliers and rejected. Additionally, information can be derived from the correlation of the concentration of ETS markers. For instance, a nonzero intercept in the regression line of UVPM *versus* FPM occurred in one data set, which suggests an erroneous blank correction.

In many of the cases in which very low values were found for solanesol compared to the spectrometric markers (UVPM or FPM), the nicotine levels were also very low, suggesting there may be alternate sources of these markers. In the Swiss data, two sampling locations that yielded highly elevated results for UVPM and FPM relative to Sol-PM could be traced to the presence of an open fireplace in the room. In this case, the solanesol still provided a reliable estimation of ETS–RSP.

A main conclusion regarding the methodological part of this study was that there were substantial benefits in adhering to the published procedures. Performing duplicate determinations was found extremely important in ensuring a quality audit of the data that could be substantiated. The study results could be used to make a recommendation regarding the markers to be measured in order to assess ETS level with a minimal workload. Markers for both the gas- and particulate-phase are required.

Nicotine and 3-EP are two viable gas-phase candidates. Nicotine concentrations are influenced particularly at low concentrations by their sorption property, which speaks for the use of 3-EP. In this study, the nicotine/3-EP ratio was found to decrease significantly below about $2\,\mu g/m^3$ of nicotine, which puts some limits to its use at low smoke levels. Yet, nicotine has become widely accepted as an ETS marker, and the existence of a large database for this compound makes its determination valuable for comparative purposes.

For the particulate phase, it is important to select a marker that is as specific as possible for ETS. RSP does not meet this criterion. UVPM and FPM are combustion-related particulate matters, but not tobacco specific. As Sol-PM is specific for tobacco smoke, it is recommended for the particulate phase. Determinations of UVPM, FPM, and Sol-PM require the use of a conversion factor. This factor may vary when different cigarettes are smoked, and averaged values have been published for a series of countries that reflect the specific sales distribution of the brands sold on the respective markets (Nelson et al., 1997). The relevant factors proposed by these authors were used in the determination of UVPM, FPM, and Sol-PM reported in this paper. In contrast, measurement of the compound solanesol is a direct analytical determination of a specific compound found in the particulate phase that does not require a conversion factor. We also concluded that performing correlation of the marker concentrations is a useful quality-assurance technique by making individual outlier results more apparent.

Results from IAQ Questionnaire

Restaurant patrons in five countries returned a total of 1370 questionnaires. Table 5 shows the combined percent of occupants dissatisfied by country for each indoor environmental parameter included on the questionnaire. Although most categories are below 10%, dissatisfaction rates above 20% are seen for draft, freshness, and noise. Dissatisfaction rates for IAQ and IEQ are generally below 10% and are not significantly different from each other when both were recorded. No restaurant had more than 20% dissatisfied. The mean and standard deviation for the percent of Overall Dissatisfied for the combined IAQ and IEQ results were 6.1 and 3.8, respectively.

Figure 8 shows the percent of Overall Acceptance (IAQ and IEQ) categories by country for all restaurants, suggesting that a low proportion of occupants were dissatisfied with the indoor environment, although some country-specific differences cannot be excluded. Indoor environmental design guidelines and standards provide for "guidance values" of

Table 5. Summary of the percent of occupants dissatisfied in the category shown and based on occupant response to questionnaires.

Country (# restaurants)	*N* (# questionnaires)	Temp. (%)	Draft (%)	Humid. (%)	Fresh. (%)	Tobacco smoke (%)	Noise (%)	Odor (%)	IAQ (%)	IEQ (%)
France (5)	424	5.7	8.7	5.0	37.9	17.5	27.0	10.5	7.8	6.9
Japan (4)	92	4.3	17.4	6.5	19.6	12.0	31.5	2.2	ND	12.0
Switzerland (5)	264	3.9	38.5	12.0	15.8	14.2	15.8	19.1	3.1	2.7
USA (5)	369	8.0	15.8	9.2	7.7	12.9	17.2	8.1	ND	3.0
UK (10)	221	7.2	11.8	ND	ND	2.7	ND	ND	5.0	ND

ND: not determined.

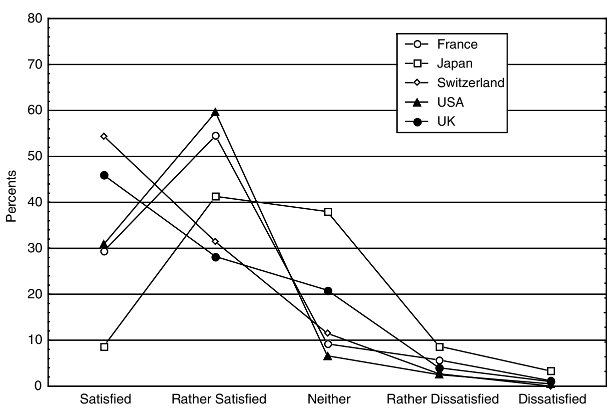

Figure 8. Overall Acceptance categories by country (all restaurants).

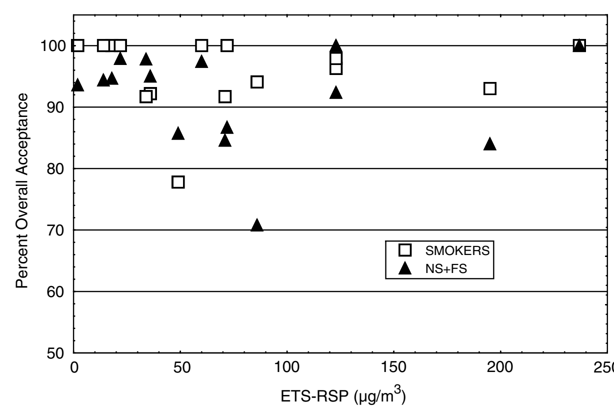

Figure 9. Percent Overall Acceptance (IAQ and IEQ) of smokers and nonsmokers in 15 restaurants *versus* ETS–RSP (NS + FS: nonsmokers and former smokers).

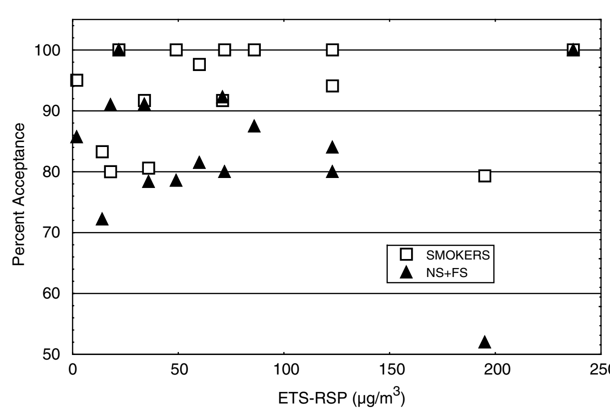

Figure 10. Percent acceptance towards tobacco smoke by smokers and nonsmokers in 15 restaurants *versus* ETS–RSP (NS + FS: nonsmokers and former smokers).

dissatisfaction in the range of 10–30% (CEC, 1992) or for more than 80% acceptance (ASHRAE, 1999). A CEN Report on *Design Criteria for the Indoor Environment* specifies different "levels of expectation" (CEN, 1998). The data presented above indicate that physical stresses such as noise and draft challenge those limits. On the other hand, expectations of occupants toward IAQ appeared to have been met in restaurants in this survey.

Figure 9 shows the ratings for Overall Acceptance separating the data into smoker and nonsmoker responses for those restaurants having more than 20 questionnaires returned, and which did not have highly variable ventilation rates ($n = 15$). In general, nonsmokers tend to be slightly more dissatisfied than are smokers. However, these results show little evidence for a relation between ETS–RSP levels and Overall Acceptance, as perceived by the patrons in a real-world environment. As shown in Figure 10, acceptance for tobacco smoke was almost as high as with Overall Acceptance, with a tendency of the nonsmokers clustering around the 80% acceptance level at any measured ETS levels.

Subjective responses from smokers and nonsmokers for spaces containing ETS have been investigated in experimental settings (Cain et al., 1983, 1987; Leaderer et al., 1984; Clausen et al., 1987; Gunnarsen and Fanger 1991; Straub et al., 1992; Walker et al., 1997). At a given ETS level, smokers tend to give higher acceptability ratings than nonsmokers. Among nonsmokers, 80% acceptability of air quality is generally achieved at ETS levels corresponding to an ETS–RSP concentration range of 60–100 $\mu g/m^3$ (Walker et al., 1997). The acceptance rate observed in this study is indeed substantially higher than what would have been predicted from the results of the above studies, including the most recent by Walker et al. (1997).

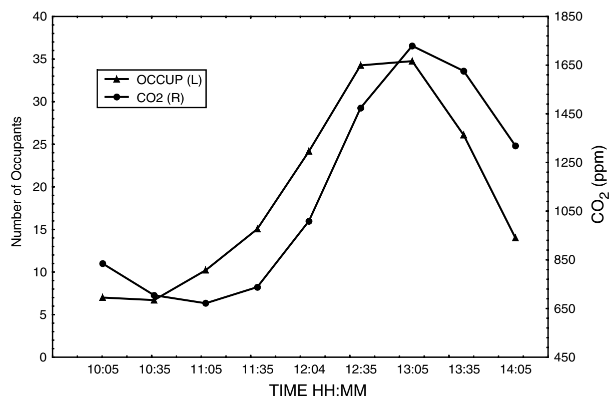

Figure 11. Number of occupants and CO_2 concentration *versus* time (Swiss restaurant).

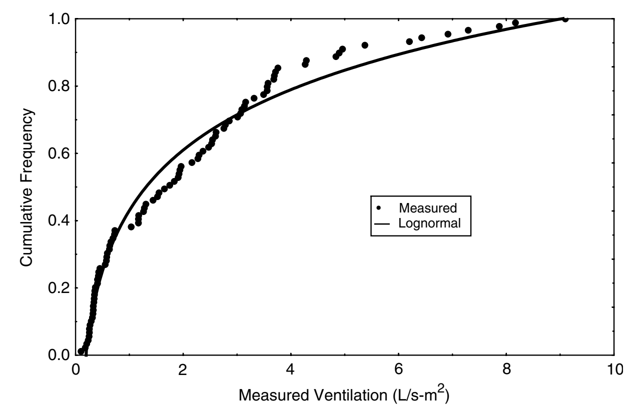

Figure 12. Distribution of measured ventilation rates (l/s m^2) for 33 restaurants assessed ($n = 89$).

Results from Ventilation Assessment

Figure 11 shows a plot of CO_2 concentration and occupancy for a Swiss restaurant *versus* time. Allowing for a phase lag, it is clear that the CO_2 concentration tracks the occupancy well. The standard error of prediction for the model-predicted carbon dioxide concentration and that measured was approximately $\pm$125 ppm CO_2 or about $\pm$10–15% of the mean CO_2 concentration for the restaurant concerned.

In the survey, 89 meal periods were observed in 33 restaurants for which a ventilation rate could be estimated. It is a common practice to normalize ventilation rates to take into account the size of the location. Figure 12 shows a distribution of measured ventilation rates (l/s m^2) for the 33 restaurants assessed. A lognormal distribution seems to fit this data set ($r = 0.91$) with a median of 1.8 l/s m^2 and a standard deviation of 2.1 l/s m^2. The observation that ventilation rates are below recommended rates is consistent with the relatively high CO_2 concentrations seen in some of the restaurants. The European CEN Report CR 1752 requires a ventilation rate of 8 l/s m^2 for hospitality spaces where smoking is not permitted, and 10.6 l/s m^2 where smoking is permitted (CEN, 1998). Figure 12 suggests that none of the restaurants surveyed were ventilated in agreement with this guideline, although the occupant expectations for IAQ had been generally met.

Indoor Air Modeling

A further goal was to determine if an ETS constituent concentration in the indoor air of restaurants could be predicted from cigarette ETS yield values and ventilation rates calculated from carbon dioxide measurements. The experimental design of this study provides the necessary data to perform the calculations, including the measurement of a number of ETS constituents, the number of cigarettes smoked per time interval, and the effective ventilation rates that are described above. ETS yields of constituents are available in the literature (Martin et al., 1997). A simple model was used, which is derived from the basic physical model, (ventilation estimation model) to calculate the steady-state concentration of an indoor air pollutant from the quotient of the rate of generation divided by the rate of removal as

$$C = C_0 + G/Q_i \quad (2)$$

If gas-phase constituents such as nicotine or 3-EP that typically have low concentration in outdoor air are selected, C_0 can be assumed to be negligible and Eq. (2) reduces to

$$C = G/Q_i \quad (3)$$

where G is the generation rate (μg/h) and Q_i is the removal rate (m^3/h).

G, the generation rate for nicotine (μg/h), is found from the product of M, the ETS yield of nicotine (μg/cig) measured experimentally (Martin et al., 1997), and N, the number of cigarettes smoked per hour (cig/h), which yields the expression $G = M(\mu g/cig)N(cig/h) = \mu g/h$. As nicotine participates in removal processes other than ventilation, the removal rate becomes $Q_i{*}F$, where F is a factor to account for these processes. In these cases, $Q_i{*}F$ becomes an effective ventilation rate (Nelson et al., 1992) and Eq. (3) becomes

$$C = G(\mu g/h)/Q_i * F(m^3/h) = \mu g/m^3 \quad (4)$$

ETS yields of nicotine and 3-EP from cigarettes were taken to be 1585 and 334 μg/cig, respectively, which is reported to provide the best predictions for real ETS situations (Martin et al., 1997). For 28 sessions in five Swiss restaurants, nicotine and 3-EP concentrations were calculated using Eq. (4), and the results are summarized in Table 6. Figure 13 shows a plot of predicted *versus* measured nicotine for a value of $F = 1.4$.

Although the results appear to predict the measured concentrations well, especially at the higher concentrations, a set of objective criteria is desirable. Table 7 shows the results of the model compared to a set of criteria for adequate model performance recommended by ASTM (1997). According to these results, the model is found adequate based on all criteria. A number of models for indoor particle concentrations failed to fulfill these criteria (Bohanon and Cole, 1997).

In view of the simplicity and potential uncertainties associated with the input parameters, the model predicts nicotine and 3-EP concentrations surprisingly well. Parameters that would benefit from further investigation include the ETS yields of smoke constituents from the cigarettes actually smoked during the investigation and the processes for nicotine removal in addition to ventilation. Experimental errors in counting the number of cigarettes smoked are a potential factor. This study has demonstrated that there is significant potential for the use of carbon dioxide measurements to estimate ventilation rates and the further use of those results to estimate the concentration of indoor ETS constituents.

Table 6. Results for regression of predicted on measured nicotine and 3-EP concentrations using Eq. (4).

Compound	*F*	Slope	Intercept	*r*
Nicotine	1.0	1.41	0.61	0.945
Nicotine	1.2	1.18	0.51	0.945
Nicotine	1.4	1.01	0.43	0.945
3-EP	1.0	1.24	−0.58	0.959

F: Effective ventilation factor.

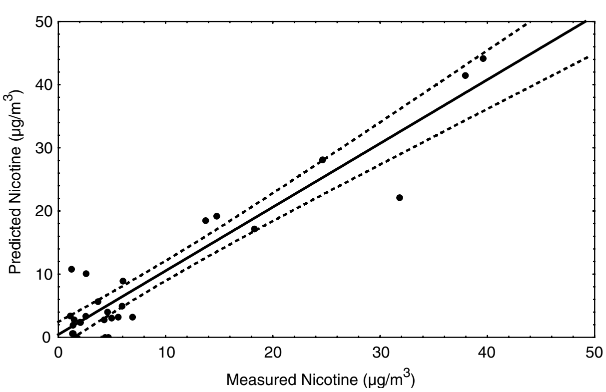

Figure 13. Plot of predicted *versus* measured nicotine using an effective ventilation factor of 1.4 (dashed lines 95% confidence interval).

Conclusions

Cost-effective investigations of ventilation, personal perceptions, smoking activity, and analytical measurements of ETS may be designed using insight gained from this pilot study. There is a synergy obtained from the components of a comprehensive indoor environment investigation, because an important benefit can be gained from utilizing diverse information collected simultaneously. For example, correlation between different determinations enhances the ability to detect data outliers and experimental errors. To develop a general understanding of indoor environments, it is necessary to obtain simultaneously analytical data, information on occupant activities, and to assess the ventilation.

Studies as comprehensive as this one benefit significantly by having a standardized protocol for all participants to follow. This protocol should mandate at least two replicates for each analytical determination. Owing to the strong correlation between the concentrations of various ETS components, it is not necessary to measure a large number of constituents to gain insight into ETS levels in restaurant facilities. However, it is necessary to measure at least one constituent each in the vapor and particulate phases. The most beneficial measurements are perhaps nicotine or 3-EP, and solanesol or Sol-PM. Carbon dioxide generated by occupants was found to be a viable tool to determine ventilation rates. Indications are that these ventilation rates when combined with smoking activity and nicotine ETS yield per cigarette suggest a good potential for this data set to be modeled by an available IAQ model for the prediction of airborne nicotine concentration.

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Table 7. Comparison of model to ASTM criteria for statistical evaluation of IAQ models (ASTM D 5157-97).

	r	Slope	Intercept	NMSE	FB	FS
Range	−1 to 1				0 to 2	0 to 2
Perfect	1	1	0	0	0	0
ASTM criteria	$\|r\|>0.9$	0.75–1.25	<25% of mean	<0.25	<0.25	<0.5
Model ($F=1.4$)	0.945	1.01	19.6%	0.18	0.06	0.19

Terms defined in ASTM D 5157-97.

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