

IMPACT OF VARIOUS AIR EXCHANGE RATES ON THE LEVELS OF ENVIRONMENTAL TOBACCO SMOKE (ETS) COMPONENTS

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SUMMARY

Environmental tobacco smoke (ETS), derived primarily from side-stream cigarette smoke between puffs, is a major contributor to indoor air pollution wherever smoking occurs. In the frame of activities to evaluate human exposure to ETS components in indoor environments, a series of tests were undertaken to investigate the impact of various ventilation rates on the air concentration of ETS-components. The tests were carried out at the European Commission-Joint Research Centre's INDOORTRON facility, a 30 m³ walk-in type environmental chamber.

Preliminary evidence indicates that changes in ventilation rates simulating conditions expected in many residential and commercial environments during smoking do not have a significant influence on the air concentration levels of ETS constituents, e.g. CO, NO_x, aromatic compounds, nicotine. This suggests that efforts to reduce ETS originated indoor air pollution through higher ventilation rates in buildings, including residential areas and hospitality venues, would not lead to a meaningful improvement of indoor air quality. Moreover, the results show that "wind tunnel"-like rates or other high rates of dilution ventilation would be expected to be required to achieve pollutant levels close to ambient air limit values.

KEYWORDS:

Environmental Tobacco Smoke (ETS), Air Exchange Rates, ETS components, INDOORTRON environmental chamber.

INTRODUCTION

Environmental tobacco smoke (ETS) is a complex mixture of thousands of compounds in particulate and vapor phases. The contribution of various environments to personal exposure to ETS components varies with the time-activity pattern of the exposed individuals, e.g. exposure of infants residing in the home of a smoker would be greater for those who do not attend day care. For adults residing with non-smokers, the workplace may be the principal location where exposure takes place. Although

the exposures of nonsmokers are much lower than those of smokers, there is some evidence that secondhand (SH) exposure to tobacco smoke increases the risks of heart disease, lung cancer, asthma and other diseases. The evidence, however, is often conflicting. For example, some studies indicate an increased risk of lung cancer from ETS exposures while others do not. One important reason for the uncertainties in scientific studies is that estimates of the amount of ETS to which nonsmokers are exposed are not quantitative. This makes it more difficult to determine the relationships between ETS exposure and risk of disease.

Many of the vapor-phase chemicals in ETS can be removed from air by sticking or "sorbing" to indoor surfaces (e.g. carpet or furnishings) and then be re-emitted back into the air at a later time. This sorption/desorption behavior is often evident from the odor of cigarette smoke in homes, offices and hospitality venues long after smoking has ceased. This means that the chemical composition of ETS changes over time, but we know very little about these dynamic changes and/or on how these changes relate to measurements of single compounds used to trace exposures (e.g. measurements of nicotine in air). However, nicotine and respirable suspended particulates (RSPs) are often used to quantify exposure as ETS cannot be measured directly as a whole. In the U.S., nicotine concentrations in homes where smoking occurs typically range from less than 1 µg/m³ to over 10 µg/m³. Concentrations in offices where people smoke typically range from near zero to over 30 µg/m³. Levels in restaurants, and especially bars, tend to be even higher, and concentrations in confined spaces such as cars can still be higher. Measurements of ETS-associated RSPs in homes where people smoke range from a few µg/m³ to over 500 µg/m³, while levels in offices are generally less than 100 µg/m³ and levels in restaurants can exceed 1000 µg/m³. In Western societies, with adult smoking prevalence of 30-50%, it is estimated that over 50% of homes are occupied by at least one smoker, resulting in a high prevalence of ETS exposure in children and other non-smokers. Application of high ventilation rates in indoor spaces and/or separation of smokers from non-smokers in public areas have often

been suggested to reduce human exposure to ETS. However, experts in building ventilation have stated that dilution ventilation, used in all mechanically ventilated buildings, will not efficiently control ETS in restaurants, bars etc. They have also stressed the lack of recognized standards for acceptable ETS exposure as well as the lack of information on typical exposure levels [1,2,3 and therein cited literature].

In order to assess the impact of various air exchange rates on the levels of ETS-derived air contaminants, a series of experiments have been carried out using the INDOORTRON facility, a 30m³ walk-in type environmental chamber. The study is part of our investigations on tobacco smoke constituents, including research on tobacco additives [4], human exposure studies to main-stream and side-stream tobacco smoke, and the impact of ETS on indoor air quality. Emphasis is given on the identification and quantification of the main ETS volatile components (many of them known as causing serious health effects) at different ventilation rates, rather than to examine available control technologies for environmental tobacco smoke. A particular feature of our study was the monitoring of the various ETS originated components close (1.5m) to the emission source (cigarette burning) during the smoking period, to evaluate human exposure in the direct proximity of the source.

MATERIALS AND METHODS

The INDOORTRON is a walk-in type environmental chamber of a 30-m³ volume featuring the ability to independently control temperature (15-40 °C), relative humidity (20 - 90%) and air change rates (ach) ranging from 0.1-2 air change rates per hour ("climate mode"). Under non-controlled climatic conditions ("rinsing mode") air change rates can be increased up to 5 ach.

Air exchange rates were determined by using the tracer gas (SF₆) technology according to ASTM E 741-93 standard with a Lagus Applied Technology Autotrac 101 automatic SF₆ analyser (GC-ECD). Homogeneity within the chamber in "climate mode" is close to 100%; in the "rinsing mode" homogeneity drops down to ca.75% (at 5 ach).

After some preliminary test runs two series of experiments were designed and executed:

First series of experiments

Five cigarettes were smoked consecutively with a commercial smoking machine (BORGWALDT, Mod. 20) following the ISO smoking regime in the INDOORTRON facility.

For these experiments the chamber was operated at stagnant air conditions and at three different ventilation rates i.e. 0.2, 0.5 and 1 ach while maintaining the relative humidity (RH) at 50 % and the temperature at 23 °C.

Second series of experiments

Four cigarettes were smoked (BORGWALDT, Mod. 20) simultaneously five times, making a total of twenty cigarettes smoked during each experiment.

The chamber was operated at five different ventilation rates i.e. 0.5, 1, 2, 3.5 and 5 ach while maintaining the relative humidity (RH) at 50% and the temperature at 23°C (at 5 ach relative humidity dropped down to 23%).

During the experiments air samples were taken at distinct time intervals in order to follow changes in concentration of some of the characteristic compounds that are formed during cigarette burning.

The cigarettes used are commercially available with declared nicotine and tar amount of 0.6 and 7.0 mg/ cigarette, respectively.

Smoking conditions:

	a) five cigarettes	b) twenty cigarettes
Puff volume	35 ml	4*35 ml = 140 ml ¹
Puff duration	2.0 sec	3.0 sec
Puff intermission	60.0 sec	60.0 sec
Butt length	35 mm	Approx 35 mm ¹¹

¹Puff volume set on the smoking machine 140 ml resulting from 35 ml for each cigarette in a four cigarette set.

¹¹Approximate value because not under control of the IR detector of the smoking machine.

The following substances were analysed:

Volatile Organic Compounds (VOCs): benzene, toluene, pyridine, m+p-xylene, limonene and nicotine (first and second series of experiments at stagnant air conditions, 0.5, 1 and 2 ach).

Carbonyl compounds: formaldehyde and acetaldehyde (second series of experiments at 0.5, 1 and 2 ach).

Inorganic gases : NO_x (NO+NO₂) and carbon monoxide (CO) [all experiments]

Ozone/Carbon monoxide/NO_x Monitoring

Monitoring of ozone, carbon monoxide and of nitrogen oxides was performed on-line during the entire time period of the experiments. The sampling line was placed as for the other analyzed compounds in the vicinity of the source (cigarette burning).

The automatic analyzers used are:

Ozone: Thermo Environmental Instruments Inc., model 49. Principle of measurement: UV-Photometric.

NO_x analyzer: Thermo Environmental Instruments Inc., model 42C. Principle of measurement: Chemiluminescence.

Carbon monoxide analyzer: Thermo electron Corp., model 48. Principle of measurement: Gas filter correlation/IR-photometric

Analysis of volatile organic compounds (VOC)

VOCs and Nicotine sampling was performed using TENAX TA tubes, analysis was made by thermal desorption and GC/MSD with SIM (78-benzene, 79-pyridine, 91-toluene, ethyl benzene and xylenes, 68-limonene and 84-nicotine).

Temperatures setting for conditioning and desorption according to ISO norms are 280 and 260, respectively.

The volume sampled for VOCs was kept as low as possible in order to avoid saturation and breakthrough phenomena, from a minimum of 250 ml to a maximum of 500 ml at a sampling flow rate of 100 ml/min.

Method for analysis of VOCs:

Thermal desorption:

Perkin Elmer ATD 400 equipped with a TENAX TA cold trap of 100 mg

Desorption temperature: 260 °C

Desorption flow: Helium at 50 ml/min

Cold trap low temperature: - 30 °C

Cold trap high temperature: 260 °C

Valve temperature: 200 °C

Transfer line temperature: 220 °C

Gas Chromatography:

Column: J&W Scientific DB-5-MS 30m*0.25mm, 1µm film.

Oven temperature: Temperature program

from 10°C (2 min) to 220°C at 4 °C/min

from 220 °C to 260 °C (1.5 min) at 15 °C/min

total run time 55 minutes

Transfer line temperature: 280 °C

Carrier: Helium at 15 Psi (constant pressure mode)

Mass Selective Detector:

Acquisition mode: scan

Solvent delay: 1.00 minute

Low mass: 50 amu

High mass: 350 amu

Analysis of carbonyl compounds (Formaldehyde and Acetaldehyde)

Sampling of the carbonyl compounds was performed using Sep-Pak DNPH-Silica cartridges consisting of 2,4-dinitrophenylhydrazine-coated silica. These cartridges trap volatile aldehydes and ketones (reaction with DNPH in the cartridges) to form stable hydrazone derivatives. An ozone scrubber was placed before the Sep-Pak DNPH-Silica cartridge. The collection efficiency is given with a value of >95% for sampling rates up to 2 l/min.

Method for analysis of carbonyl compounds:

Column: Waters Nova-Pak C18, 60A, 4 µm (3.9 x 300) mm

Mobile Phase: A = Acetonitrile/THF/Water (30:10:60)

B = Acetonitrile/Water

Gradient: 0% B for 1 min, linear gradient to 100% B over 10 min

Flow Rate: 1 ml/min

Column Oven: 25 °C

Injection Volume: 25 µl

Wavelength: 360 nm

Volume Sampled:

for five cigarettes: 50 liters (1.7-2.0 l/min)

for twenty cigarettes: 13 liters (1.7 – 2.0 l/min)

Elution: Cartridges are eluted with 5ml acetonitrile and this solution analysed with HPLC (detection at 360 nm).

RESULTS AND DISCUSSION

CO and NO_x monitoring

Figs.1 and 2 show the variation of the CO and NO_x concentrations during consecutive smoking of five cigarettes at stagnant air conditions and by applying different air exchange rates within a period of ca. 100 minutes (duration of the experiments). Within the burning period (ca. 37 minutes after lighting) CO and NO_x concentrations are increased to reach the maximum by the end of the burning period. The measurements clearly indicate that during this time (burning period) changes in the ventilation rate do not have any significant influence on the concentration of the pollutants. Changing the ventilation rate from static conditions to up to one exchange per hour (i.e. 30m³), results in changes to the CO concentrations of up to 25% only, compared to the values obtained at stagnant air conditions. Similar results were found for NO_x too.

After the burning period (lasting ca. 37 min.), an overall reduction of the air concentrations of the pollutants was observed. This can be attributed to air exchange rate variations and the absence of the strong emission source (cigarette burning). At the end of the experiments (after ca. 100 min.), a change of the ventilation rate of up to one exchange per hour, results to ca. 67% and 70% reduction of the CO and NO_x concentrations, respectively (compared to stagnant air conditions).

The formation of carbon monoxide (CO) and of nitrogen oxides (NO_x) (Fig. 3 and 4) during the second series of experiments with twenty smoked cigarettes (and with clearly higher smoke volume produced) follows the same trend, already observed during the first series of experiments (five cigarettes smoked). During the smoking period (ca. 37 min.), peak concentrations up to 30 ppm (CO) and 800 ppb (NO_x) were measured (at 0.5 ach). This corresponds, as expected, to concentrations up to four times higher compared to the concentrations measured during the first series of experiments. Variations of peak concentrations of CO and NO_x during the initial phase of the experiment (smoking period) and at different ventilation rates (0.5, 1, 2 and 3.5 ach) do not exceed 47% despite the large change in air exchange rate. Twenty minutes after the end of the smoking period CO and NO_x concentrations dropped down up to 80% at ventilation rates of up to three and a half exchanges per hour. An increase of the ventilation rate up to five exchanges per

hour leads to a further reduction (up to 25%) of CO and NO_x concentrations compared to those at 3.5 ach.

Another experiment was carried out (at 2 ach) with smoking of ten cigarettes during a period of ca. 20 min, followed by a non-smoking period of 60 min and subsequent smoking of another set of ten cigarettes. Production and

elimination of CO and NO_x is shown in Fig. 5. Maximum concentrations of ca.370 ppb for NO_x and of ca. 12 ppm for CO were measured. During the non-smoking period of one hour the concentrations for both NO_x and CO dropped down to 70 ppb and 3 ppm, respectively. Starting smoking again, NO_x and CO levels reached values slightly higher than those measured during the first smoking period.

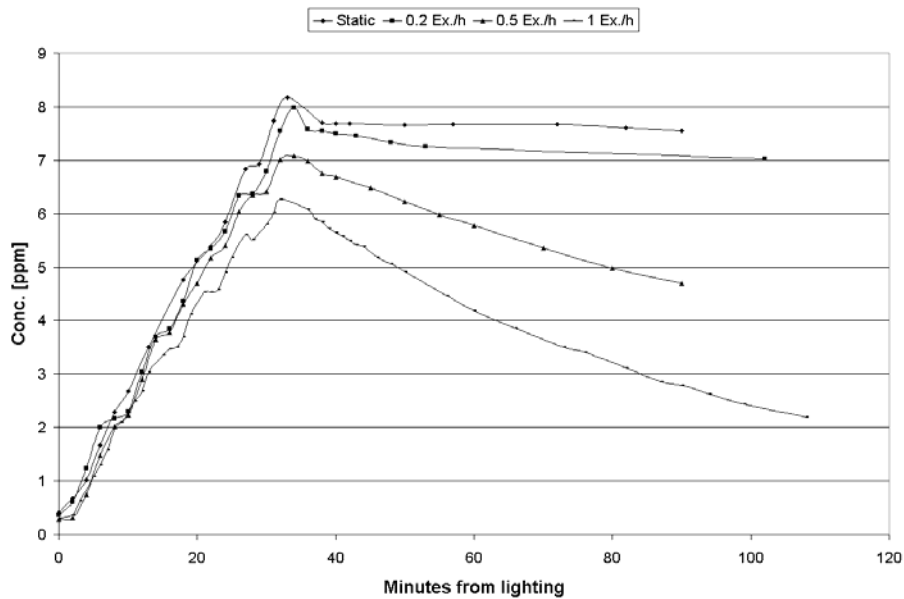


FIGURE 1
Concentration of CO at different air exchange rates (first series of experiments).

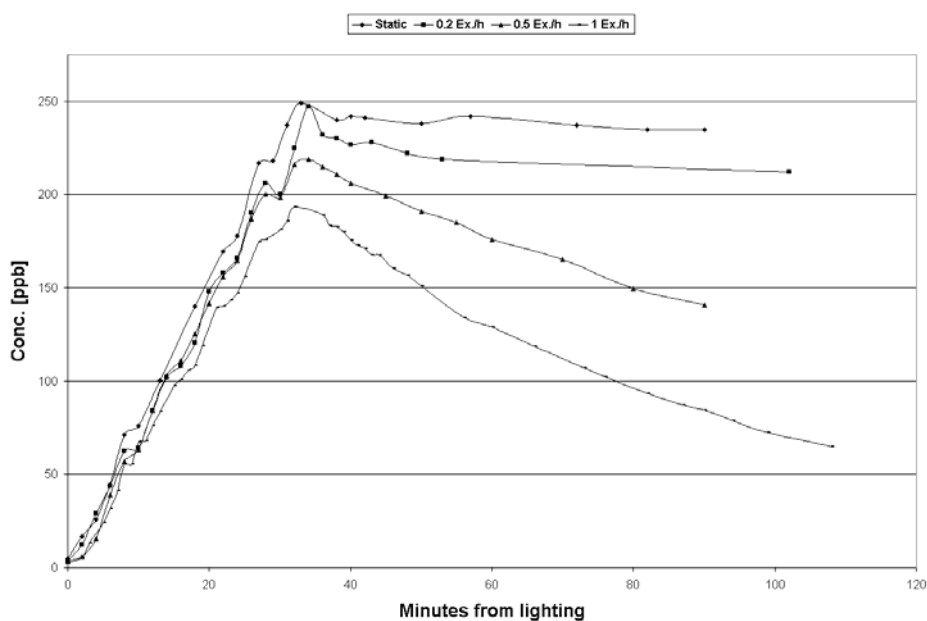


FIGURE 2
Concentration of NO_x at different air exchange rates (first series of experiments).

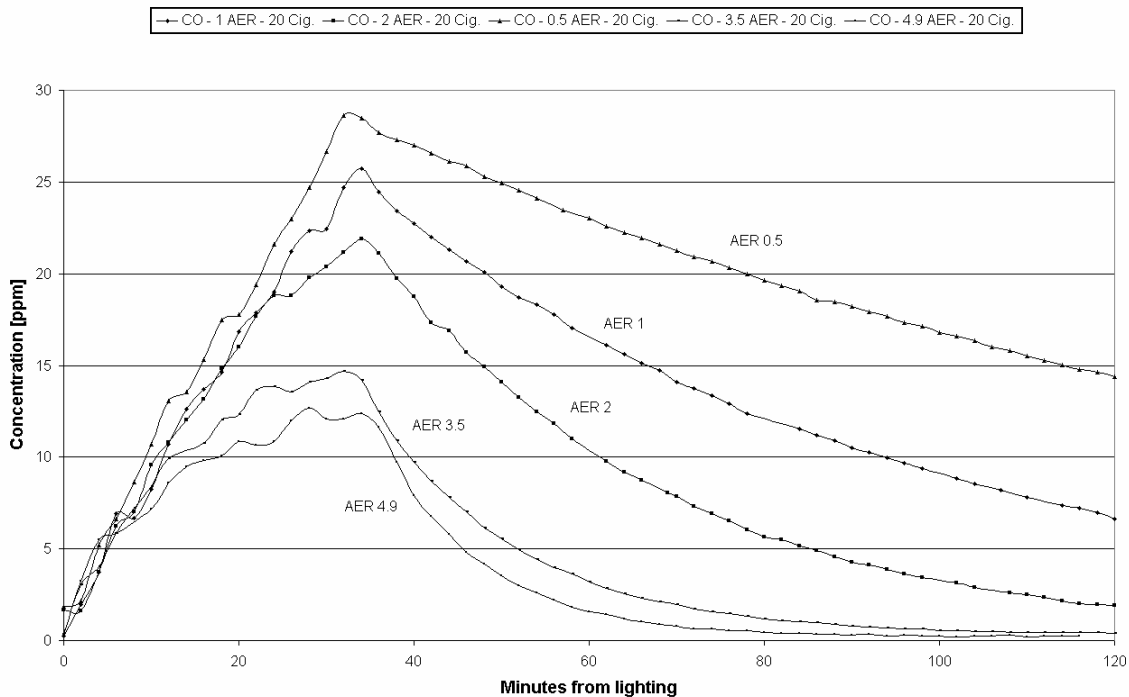


FIGURE 3
Concentration of CO at different air exchange rates (second series of experiments).

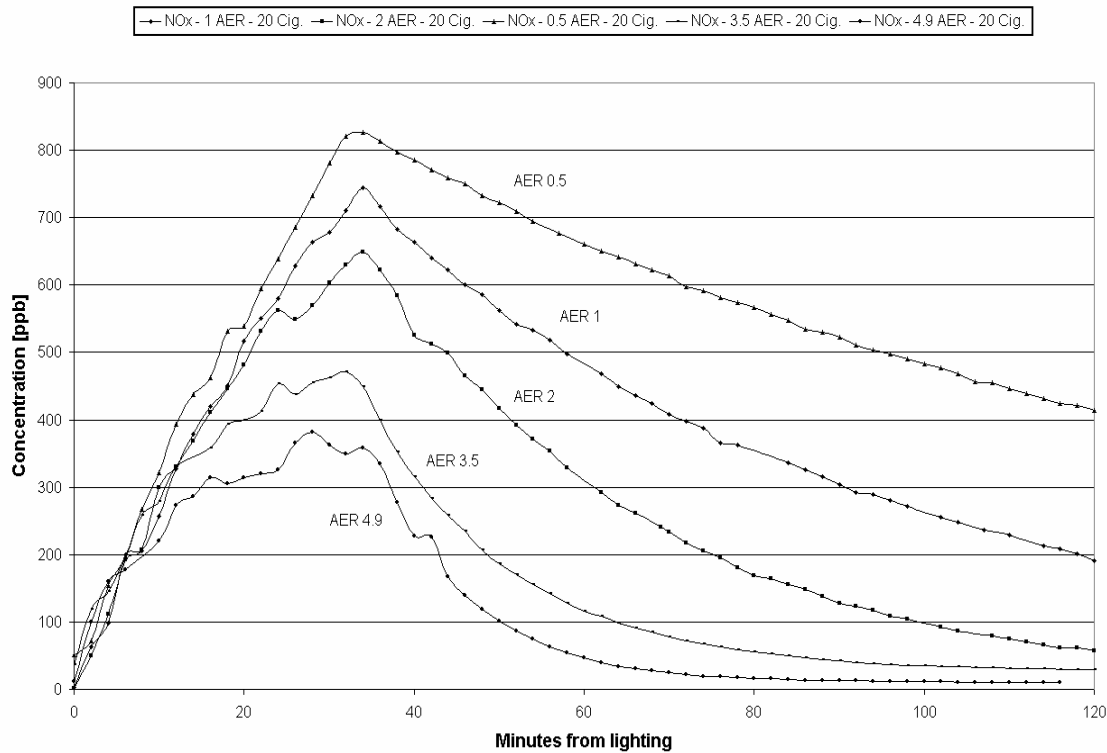


FIGURE 4
Concentration of NO_x at different air exchange rates (second series of experiments).

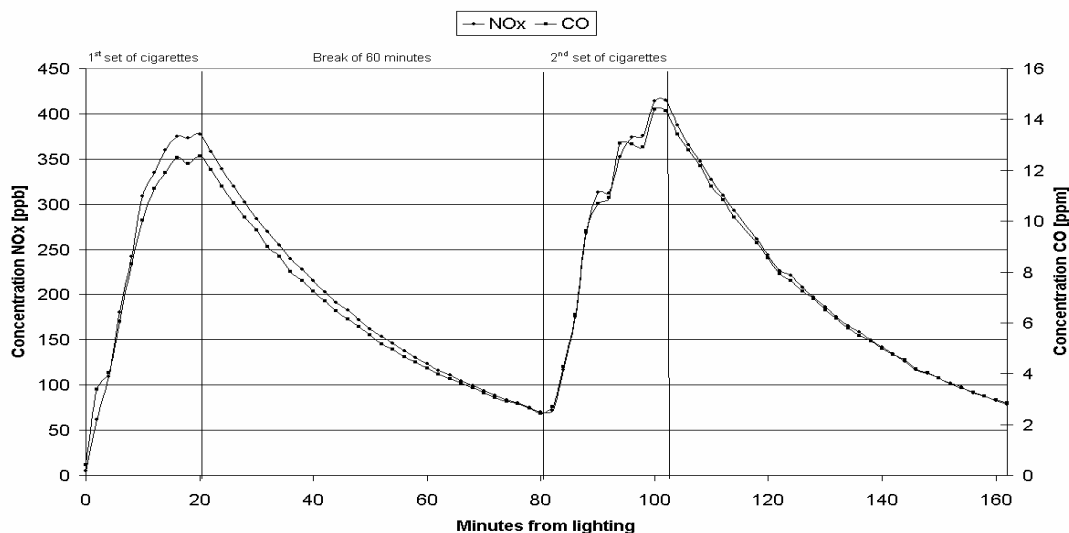


FIGURE 5
Concentration of NO_x and CO smoking ten cigarettes, stopping for one hour and smoking again 10 cigarettes at an air exchange rate of 2 (60 m³/h).

Volatile Organic Compounds

Apart from CO and NO_x some organic compounds produced during cigarette burning were regularly monitored, in particular, during the initial phase of the experiments (smoking period). The results show that peak concentrations of benzene, toluene, m+p-xylene, limonene and pyridine do not change significantly at different ventilation rates (Table 1). For nicotine, the measured concentration at one air exchange rate amounts to ca. 85% of the concentration measured at stagnant air conditions.

In the second series of the experiments the formation of benzene, BTEX (sum of aromatics), pyridine, limonene and nicotine was monitored during the entire duration of the experiment and at different ventilation rates i.e. including measurements beyond (ca. 80 min) the initial

smoking period (ca. 37 min). Peak concentrations up to 210 and 1640 µg/m³ for benzene and nicotine, at 0.5 ach, were measured (Figures 6-10). Under the same conditions, 3-ethenyl-pyridine, a combustion product from nicotine often used as a marker for environmental tobacco smoke reached values up to 275 µg/m³.

Even at elevated air exchange rates (2 ach) the concentrations of 160 µg/m³ for benzene and 1200 µg/m³ for nicotine are (at the end of the smoking period) still high. For both compounds, at the end of the experiment (ca. after 120 min) air concentration dropped down to 14 µg/m³ and 115 µg/m³ for benzene and nicotine, respectively. A similar behavior was observed for the other volatile organic compounds monitored.

TABLE 1
Peak concentrations in µg/m³ of volatile organic compounds at different air exchange rates (first series of experiments).

Air Exchange Rates (first series of experiments)				
AER [h ⁻¹]	0	0.2	0.5	1
Benzene	57.5	55.9	54.1	54.1
Toluene	89.8	89.5	85.0	89.1
m/p-Xylene	40.9	40.5	40.8	39.5
Pyridine	50.5	49.0	50.5	46.6
Limonene	39.7	40.8	36.2	36.8
Nicotine	351.0	340.5	355.5	301.1

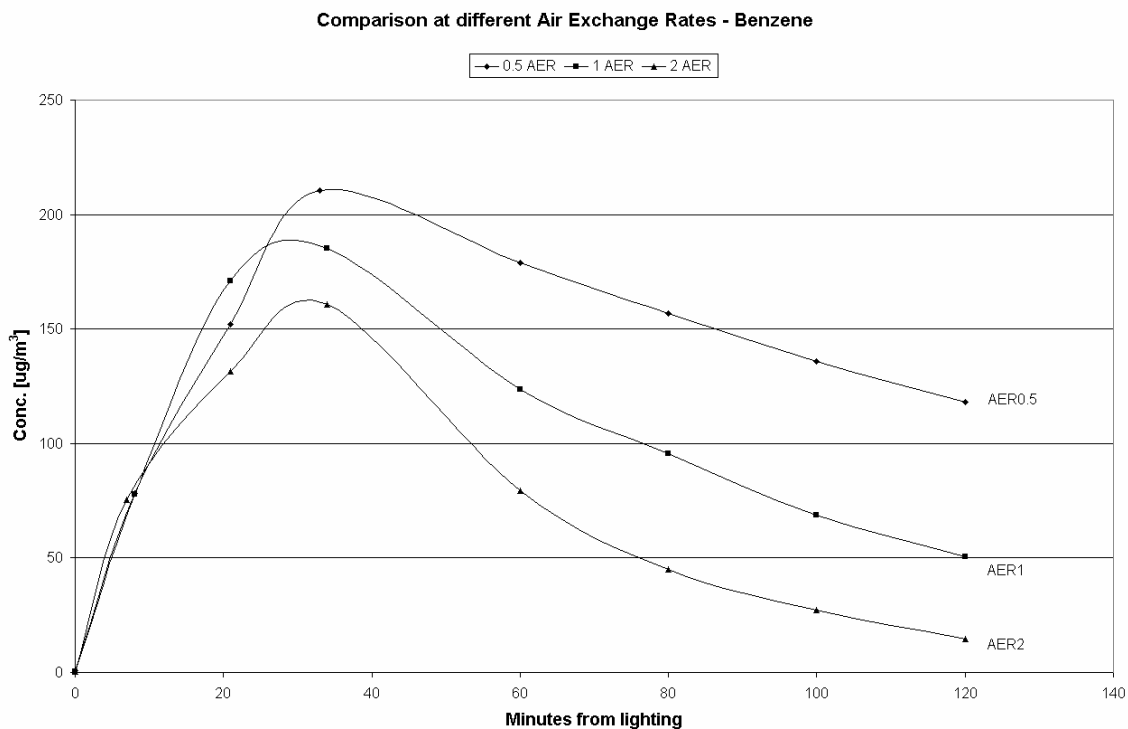


FIGURE 6
Benzene concentrations (second series of experiments).

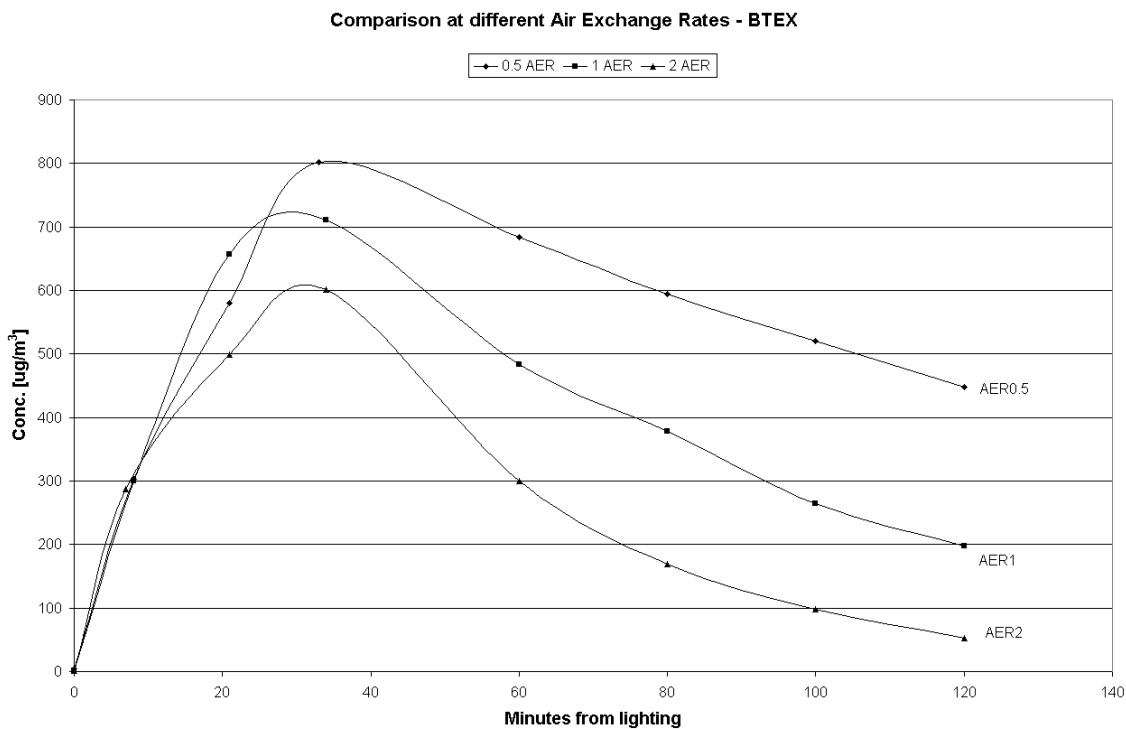


FIGURE 7
BTEX concentrations (second series of experiments).

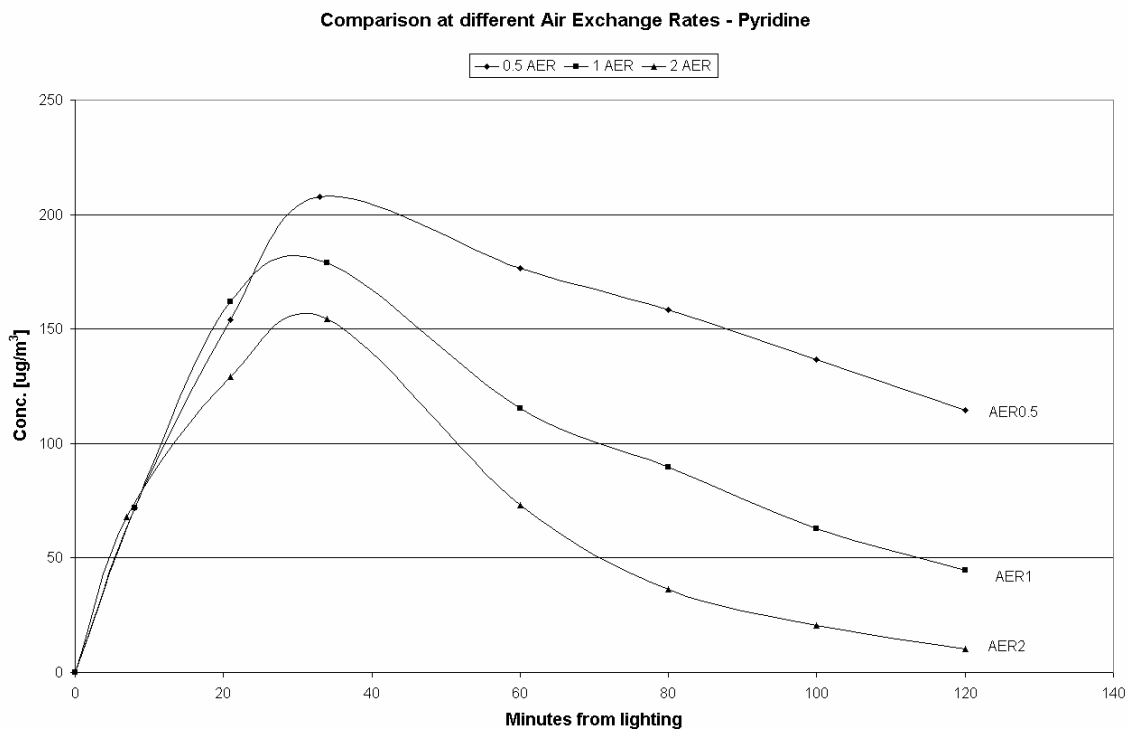


FIGURE 8
Pyridine concentrations (second series of experiments).

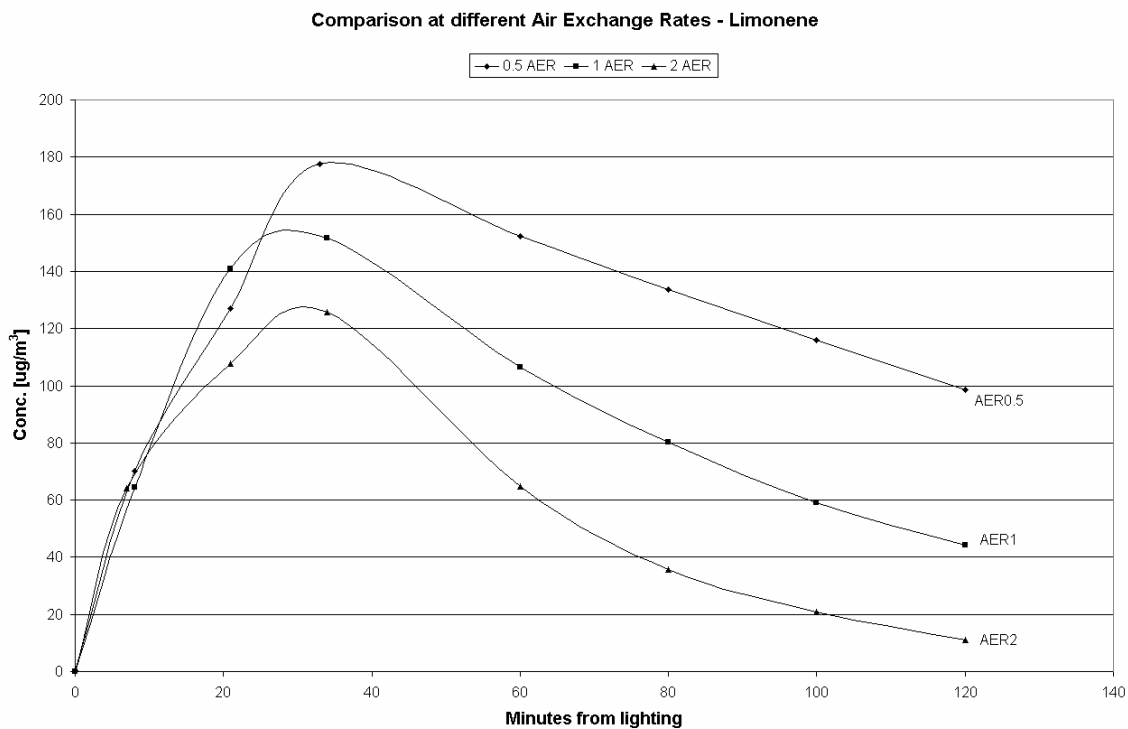


FIGURE 9
Limonene concentrations (second series of experiments).

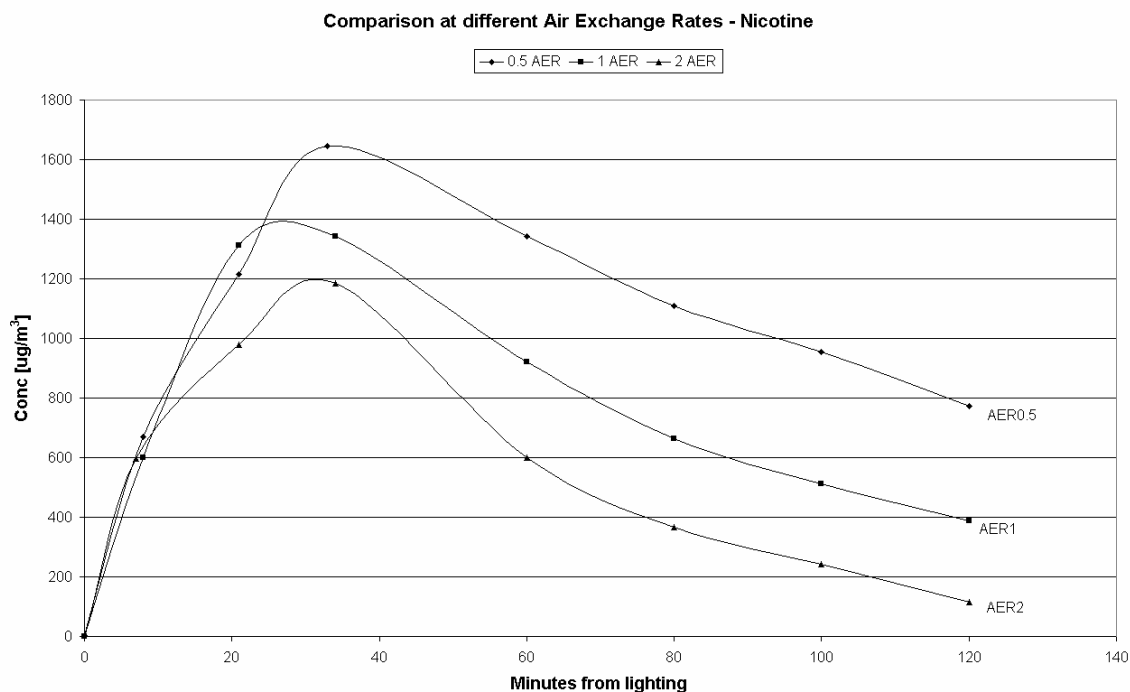


FIGURE 10
Nicotine concentrations (second series of experiments).

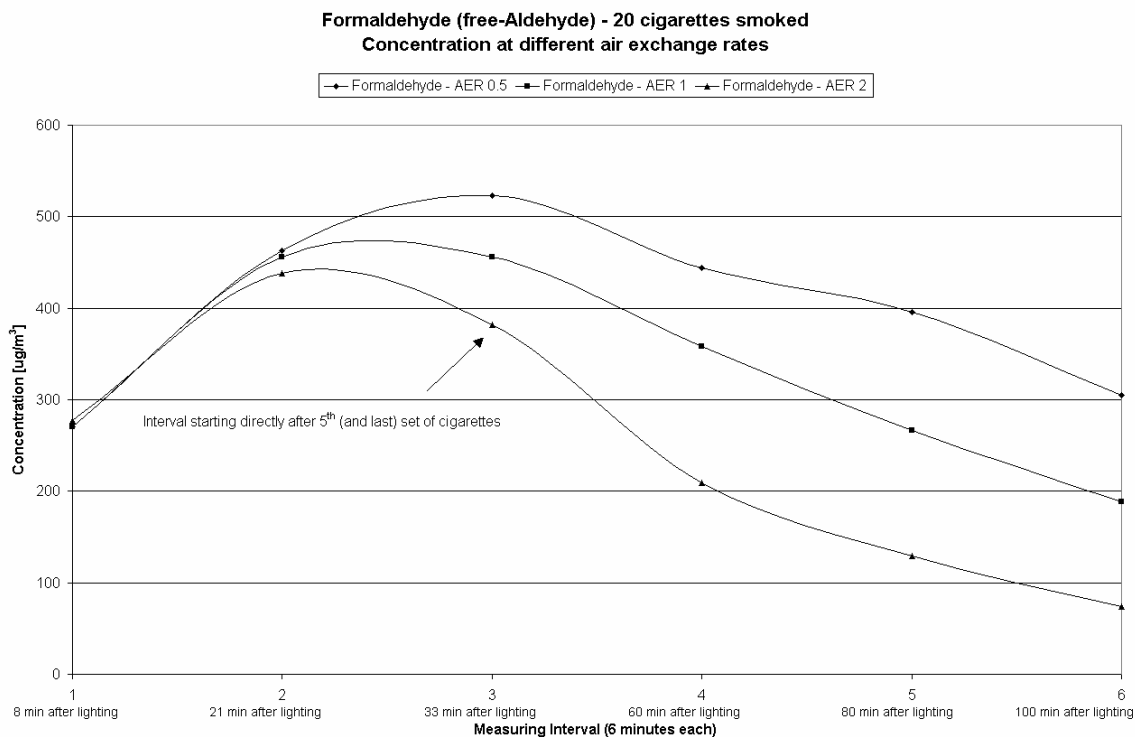


FIGURE 11
Formaldehyde concentrations (second series of experiments).

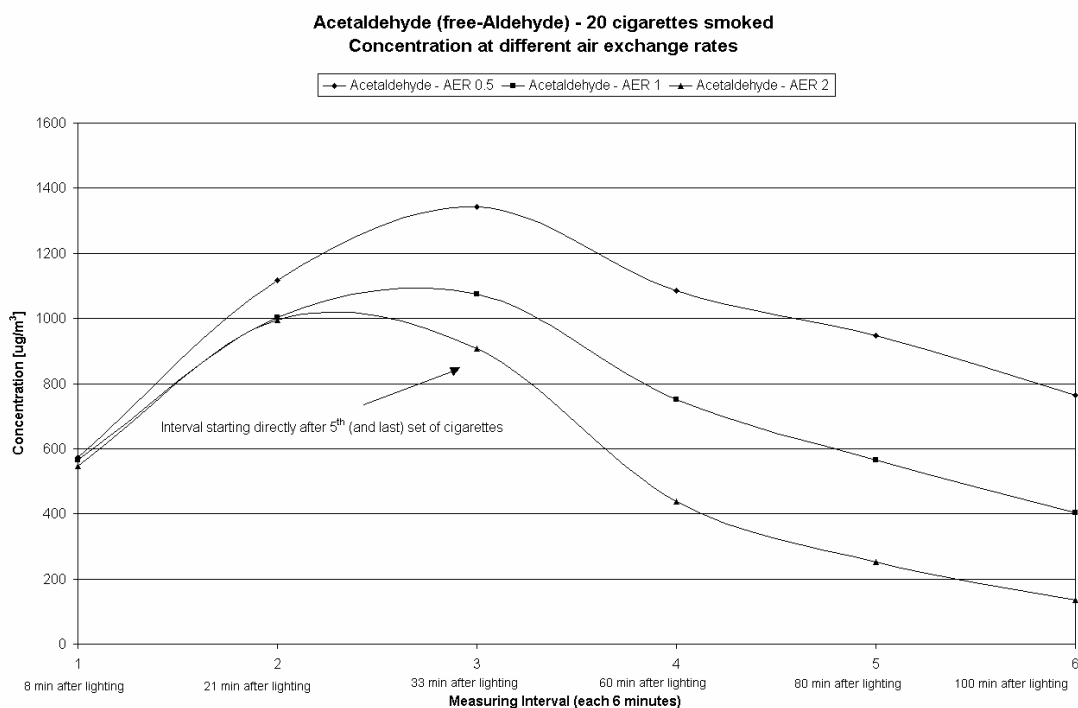


FIGURE 12
Acetaldehyde concentrations (second series of experiments).

Carbonyl compounds

It is well known, that high amounts of carbonyl compounds, e.g., formaldehyde and acetaldehyde, are produced during smoking [4, 5]. During the first series of experiments some few measurements were made to quantify formaldehyde and acetaldehyde produced during cigarette burning under the conditions of our studies. On the basis of these preliminary results, we decided (during the second series of experiments) to monitor the formaldehyde and acetaldehyde production during the entire time of the experiment and at different ventilation rates i.e. to include measurements beyond (ca. 60 min) the initial smoking period (ca. 37 min). Results are reported in the Figures 11 and 12. Peak concentrations up to 1400 and 500 $\mu\text{g}/\text{m}^3$ were measured for acetaldehyde and formaldehyde, respectively. Even at elevated air exchange rates (2 ach) the concentrations of 900 $\mu\text{g}/\text{m}^3$ for acetaldehyde and of ca.400 $\mu\text{g}/\text{m}^3$ for formaldehyde are (at the end of the smoking period) still high. For both compounds, at the end of the experiment (ca. after 100 min), air concentrations dropped down to 180 $\mu\text{g}/\text{m}^3$ and 90 $\mu\text{g}/\text{m}^3$ for acetaldehyde and formaldehyde, respectively.

MODELING

In addition to the experimental activity, modeling work was carried out with the aim to simulate CO and NO_x buildup and decay during the entire period of the

experiments (up to 120 min) at different air exchange rates. Moreover, an attempt was made to calculate at which air exchange rates CO and NO_x concentrations reach levels comparable to those in ambient air (100-150 ppb for NO_x, 3-5 ppm for CO) to which people is frequently exposed in urban areas.

As spatial (chamber air) homogeneity was guaranteed in most of the experiments (*see also comments in the chapter Materials and methods*), a first order, linear ODE (ordinary differential equation) was used to simulate mathematically the experimental setup. The concentration change of NO_x or CO was attributed to:

- emissions from the smoking device,
- removal due to air exchange and,
- introduction of outdoor polluted air into the chamber (for the experiments in “rinsing mode”).

Besides assuming a well-mixed chamber, we considered no other source or sink terms for the two pollutants under consideration as little deposition on the steel walls of the chamber or chemical activity for the specific gases under consideration is expected to occur in such a short time (~2 h).

Expressing mathematically the aforementioned assumptions leads to the following equations, comprising the model:

$$dC/dt = R/V - AER \times C \text{ during smoking} \quad (1)$$

$$dC/dt = -AER \times C \text{ after smoking} \quad (2)$$

where

C [ppb] is the chamber concentration,

R [ppb \times m³/min] is the emission rate,

V [m³] is the chamber volume,

AER [1/min] is the air exchange and

t [min] is the time

Solving analytically the ODEs gives:

$$(1) \Rightarrow C(t) = R/(V \times AER) * [1 - \exp(-AER \times t)] \text{ for } t < \text{smoking duration}$$

$$(2) \Rightarrow C(t) = C_0 \times \exp(-AER \times t) \text{ for } t > \text{smoking duration}$$

where

C_0 [ppb] is the concentration at the end of the smoking event

The static experiment data were used to estimate the emission rate of both NO_x and CO applying a linear regression analysis as emission rate was expected to be constant during the burning of a cigarette. The same emission rate was used to simulate both the first and second series of experiments, multiplied by 4 in the latter case.

Model and experimental data for NO_x and CO agree fairly well confirming all the assumptions made in the model while verifying at the same time the quality of the experimental procedure as well. The correlation coefficient between measured and calculated time series stays above 99% in all cases while the normalized bias is kept below 5% in all but one dataset.

In Figures 13 and 14 are presented the modeled against the experimental data of NO_x and CO for 1 ach and 20 smoked cigarettes.

However the present model underestimates the emissions of nicotine experiments (see Figure 15). This is expected because the model equations (1) and (2) do not account for rapid sorption of nicotine and also neglect any removal due to deposition which may occur for this substance. This is consistent with the findings of Klepeis et al. [6] and Daisey et al. [7].

Consequently, the model successfully reproduces the experimental results of NO_x and CO and thus can be readily and safely applied to give answers when simulating hypothetical cases; it is much less accurate when applied to nicotine, the emissions of which in all experiments has been underestimated.

Such hypothetical cases, similar to the second series of experiments but with higher air exchange rates were simulated and are presented in Figures 16-17.

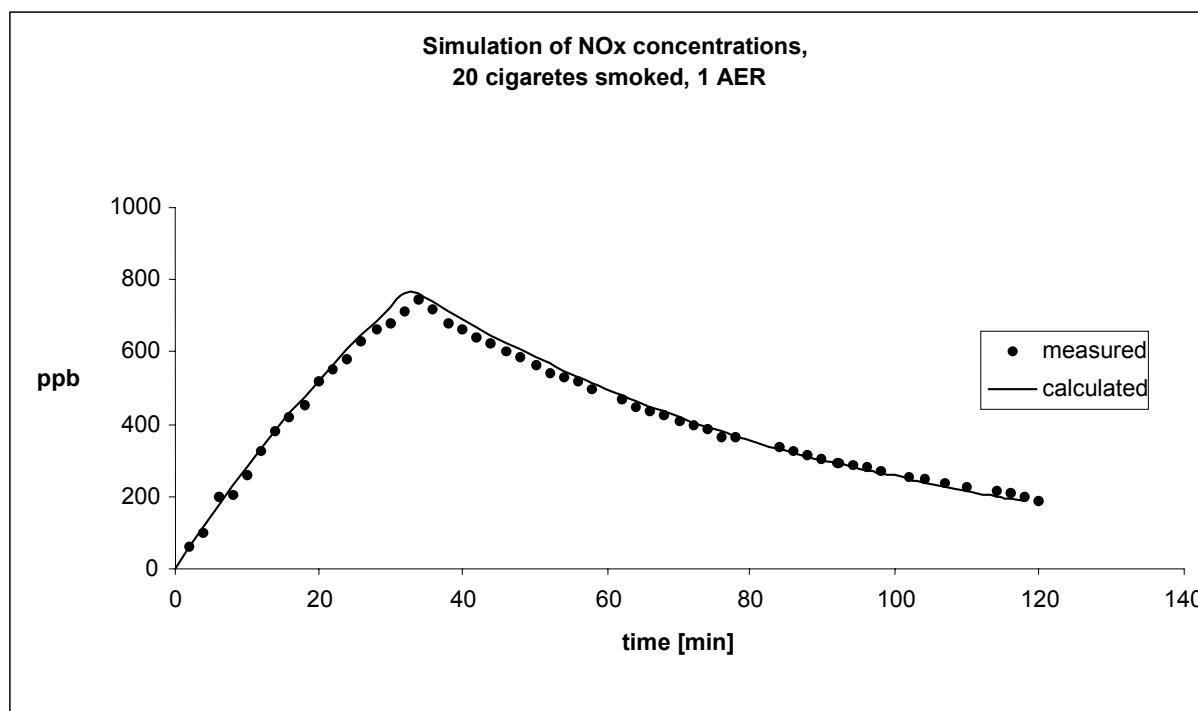


FIGURE 13
Modeled against experimental NO_x data for 1 ach and 20 smoked cigarettes.

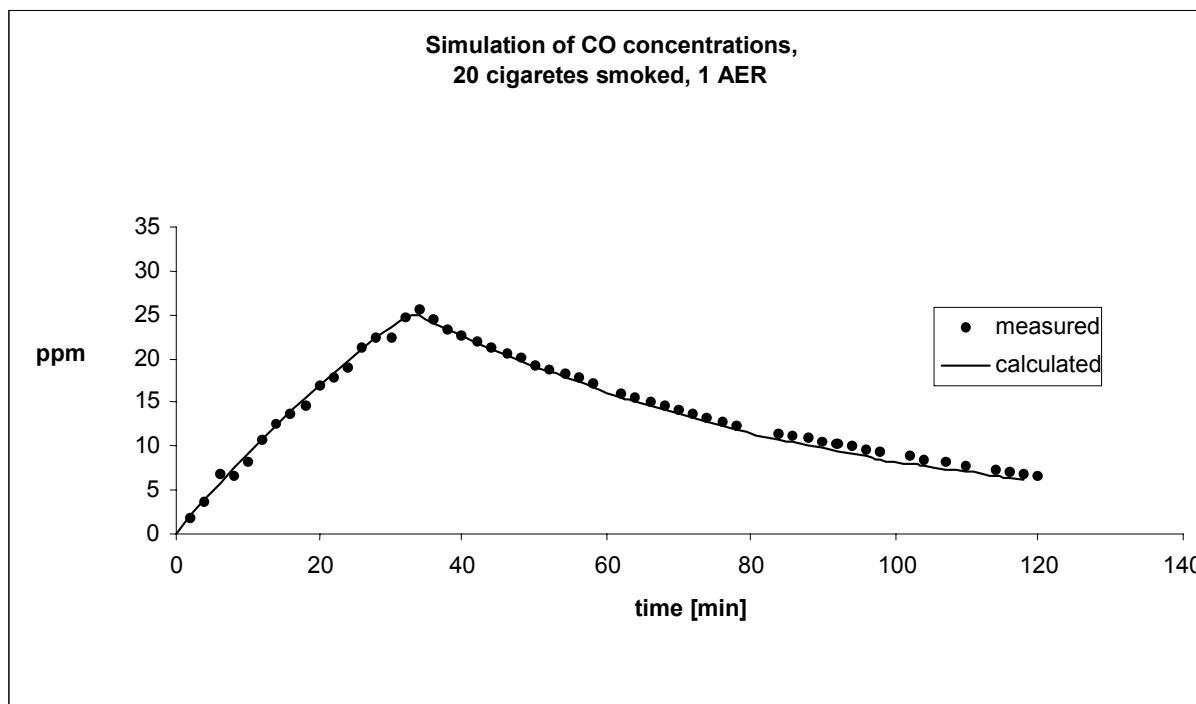


FIGURE 14
Modeled against experimental CO data for 1 ach and 20 smoked cigarettes.

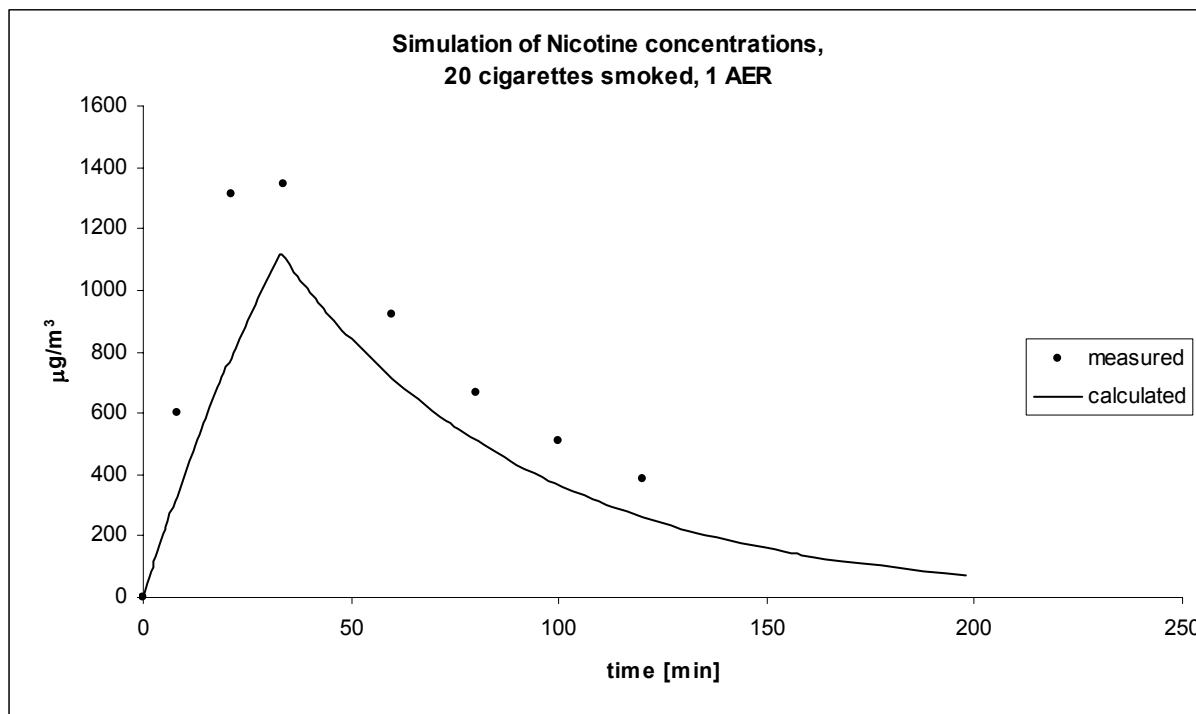


FIGURE 15
Modeled against experimental Nicotine data for 1 ach and 20 smoked cigarettes.

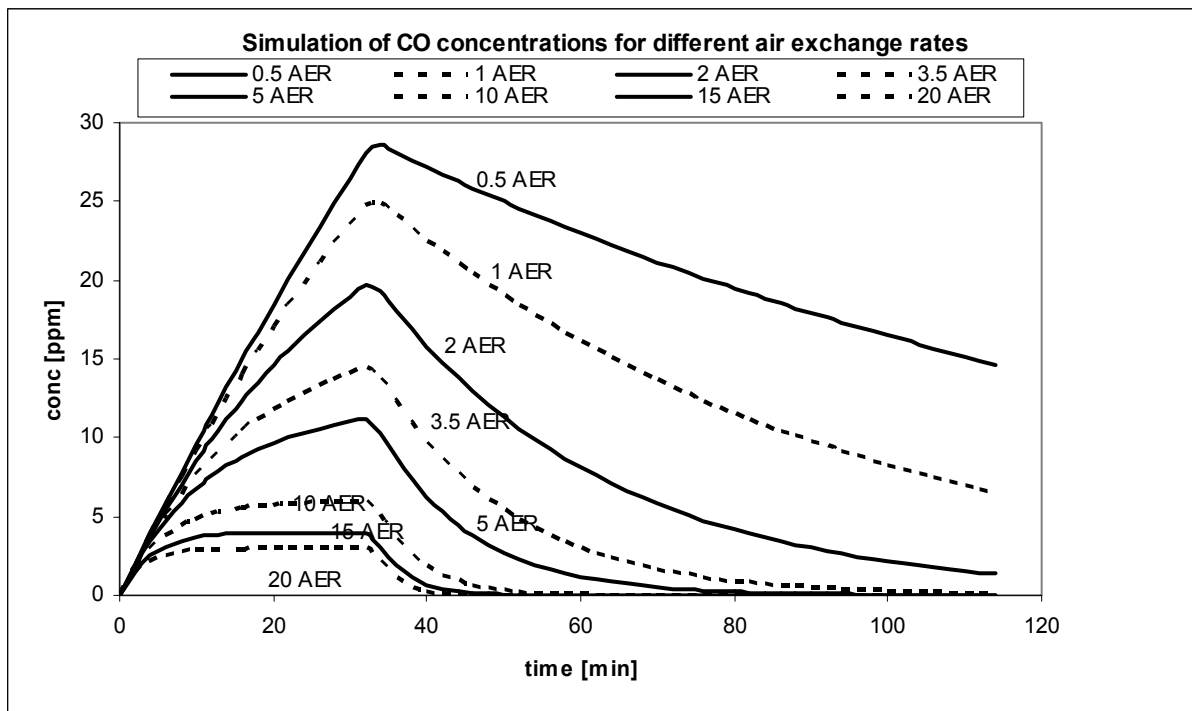


FIGURE 16

Simulation of CO concentrations for different air exchange rates (model parameters according to second series of experiments).

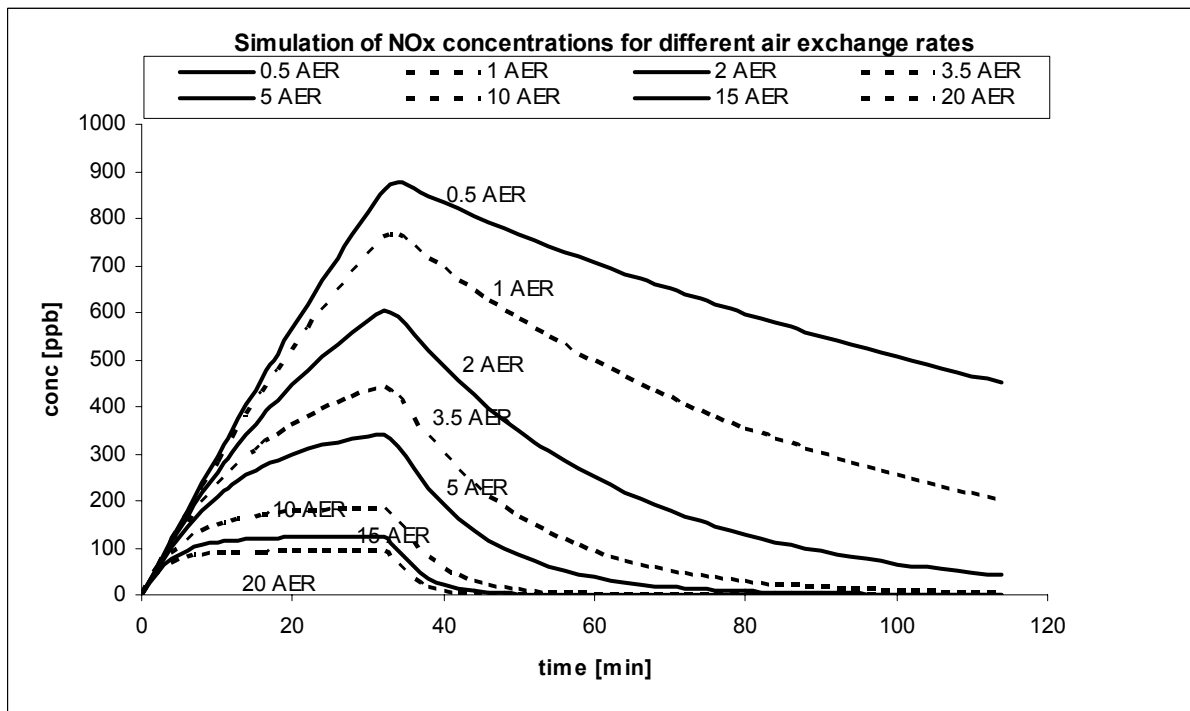


FIGURE 17

Simulation of NO_x concentrations for different air exchange rates (model parameters according to second series of experiments).

CONCLUDING REMARKS

Results obtained from our studies clearly indicate, that cigarette smoking represents a strong source of a large number of chemicals such as: volatile hydrocarbons, carbonyls, polycyclic aromatic hydrocarbons, inorganic gases and particles etc. They are produced at high concentrations during the burning process and are not rapidly and substantially eliminated from the indoor air atmosphere, even when high air exchange rates were applied. Diffusion of the emitted compounds (side-stream compounds and burning products) is relatively slow, so dilution via mixing with new incoming fresh air is not very effective as a control measure.

Moreover, these preliminary results show that “wind tunnel”-like rates or other high rates of dilution ventilation would be expected to be required to achieve pollutant levels close to those frequently occurring in ambient air. Our findings are comparable with the results obtained in studies in the US, carried out at different hospitality venues (restaurants, bars).

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