Using traffic-born aerosols as tracergases for the continous determination of air exchange-rates of buildings in operation.

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Abstract

The developement of new highly sensitive detection techniques for particle bound polycyclic aromatic compounds (PPAH) on the nano-particles of traffic born soot open a new dimension for real time measuring techniques for air exchange rates in buildings in operation. The principles and first measurements are presented and demonstrate the principal applicability of this method.

Key Words

Air exchange rate, PAH-sensor, air hygiene, energy loss of buildings

Introduction

The determination of air exchange rates of rooms or complete buildings has attracted much interest in recent years, mainly because of the strong impact of air infiltration on the energy and power balance of houses. Many different measurement methods (Raatscher 1995) have been developed, from the determination of a „tightness-classification" by means of blower doors (Geissler 1997) up to long term registration of the mean air exchange rate by means of deposited tracer gas micro-sources and receivers (Maas 1997). All the tracer gas methods used however inject tracer gases into the rooms to be measured. For real-time information about air exchange rates, very expensive and sophisticated devices are necessary, whereas the microsources and receivers deliver the long term mean values only after lab processing and are therefore not real time but batch processes.

It is desirable to determine the air leakage of a building in operation, in real time, over several months in order to be able to regulate the air exchange rate effectively; correlate the air leakage to weather data (wind velocity and direction, temperature difference etc.) thus developing a „leakage signature" of a building, so that existing software instruments , as COMERLE etc. (Feustel 1996) can be applied.

The development of extremely sensitive photoemission detectors for particle bound polycyclic aromatic hydrocarbons (PPAH) on the carbon particles of traffic born soot (Burtscher 1992; Siegmann 1997) offers a new dimension for the long term monitoring of air exchange rates. This method principally allows the use of the externally abundant nano-particles as tracer gas. In this publication, the measurement principle will be described and some first real measurements presented and discussed. It is clear however, that many more detailed

questions will have to be answered, before a practically applicable method for routine measurements of air changes can be presented.

Methods

The detection method has extensively been described in relation to air pollution measurements (Burtscher 1992; Siegmann 1997). Only a short summary of the main features will therefore be presented here.

The method is based on the fact, that polycyclic aromatic hydrocarbons (PAH) are produced in almost all burning processes. They condense on soot particles produced (particle bound PAH: PPAH). They can be ionised by illumination with short wavelengths: hf>5 eV (UV-range). Different sources can be differentiated by using excimer lamps of appropriate emission wavelengths, e.g. cigarette smoke and particles emitted by diesel engines (Matter 1995).

The electrons can be eliminated by an electric field and the flow of the charged PPAH particles be measured as a current through a conducting filter. Sensitivities down to 1 ngPPAH/m^3 have been attained. The mass relation between the particles mass and the PPAH part was determined to be in the range of 1: 200-300. Due to the charging process, only nano-particles with diameters in the range of the mean free path in air or less take part in the charge separation process, e.g. nano-particles in the range of 10 - 300 nm diameter. These particles show also the highest abundance in traffic emissions: up to some millions of particles per cm^3 and behave similar to an ideal gas, contrary to the micro-particles of soot. They penetrate the respiratory system down to the alveolar level and are therefore of medical interest (cancerogenic compounds). Penetrating the leaks of the building shell, a penetration factor of almost exactly 1 has been found, e.g. no loss of particles by the diffusion through the leaks (Koutrakis 1992, Thatcher 1995).

The measurement method applied relies on the simultaneous measurement of the outdoor- and the indoor-concentration of the PPAH in air. From these measurements, the air exchange rate of a room can principally be determined with a mass-balance model and the integration of the mass-balance equation.

Mass-balance models describe the relationship between input and output concentrations of substances introduced into a well-mixed compartment. They are often used for indoor air quality studies to describe the indoor air pollution concentrations in terms of spatial averages (Alonzan 1979; Dockery 1990;Switzer 1992). As described by (Dockery 1990), the indoor particle concentration in these models is determined by four processes:

(1) Air is considered as noncompressible and the pressure outside and inside the building is assumed to be the same. Therefore, the volume of air flow into the building is balanced by an equal air flow out of the building.

(2) The air flow through the building envelope might lead to a loss of airborne particles by impacting and/or diffusion. A factor p describes the fraction of particles which penetrates inside the building.

(3) Indoor particles are removed by gravitational and diffusive deposition onto the available surfaces: particle deposition rate.

(4) Generation of indoor particles by sources like smoking, cooking or resuspension.

With the principle of mass conservation the following equation can be derived (Dockery1990),

$$\frac{dC_i(t)}{dt} = p \cdot \phi_v C_o(t) - (\phi_v + \phi_d) \cdot C_i(t) + \frac{g(t)}{V} \qquad (1)$$

where:

$C_i(t)$ is the indoor concentration (mg/m^3),

$C_o(t)$ is the outdoor concentration (mg/m^3),

p is the penetration factor for outside particles,

f_v is the ventilatory air exchange rate (h^{-1}),

f_d is the particle deposition rate (h^{-1}),

g(t) is the indoor generation rate (mg h^{-1}), and

V is the volume of the room.

Defining average values of C_i and C_o and integrating equation (1) over the sampling period t_s leads to the steady state equation for the average indoor particle concentration (16):

$$\bar{C}_i(t) = \frac{p \cdot \phi_v}{(\phi_v + \phi_d)} \cdot \bar{C}_o + \frac{1}{(\phi_v + \phi_d)} \cdot \frac{\bar{g}}{V} \qquad (2)$$

where averaging over the sampling time is denoted by a bar. Equation (2) was used for modelling the indoor particle concentrations (Switzer 1996; Dockery 1981) and for calculation of the ratio of mean indoor and mean outdoor particle concentration (Alonza 1979;Koutrakis 1992, Raunemaa 1989). (Koutrakis 1992; Thatcher 1995) used equation (2) to determine the penetration factor for fine and coarse particles. Both studies found that p = 1 for particles with diameters less than 10μm. This results are in agreement with findings of (Oezkaynak 1996). They determined penetration factors of p =1 for both $PM_{2.5}$ and PM_{10} (mass of particles with aerodynamic diameters less than 2.5μm, respectively 10μm). In the following, particle penetration through the building envelope is therefore assumed to occur without losses and p is considered as unity.

In this work, the mass-balance equation was numerically integrated for the case of absent indoor particle sources. Consequently, the indoor generation rate (g(t)) is considered to be zero. The measured time series of indoor and outdoor PPAH concentrations are smoothed using a Savitzky-Golay filter as described in (Press 1992) in order to preserve the characteristics of the input signal. By filtering with this smoothing algorithm, the derivative of the indoor concentration time series is also obtained. Consequently, the measured indoor and outdoor concentrations and the derivatives of the indoor concentration for every time interval are used to solve equation (1), i.e. to perform a minimisation of least squares:

$$\sum_{j=m}^{n}\left(\frac{dC_{ij}}{dt} + \phi_p \cdot C_{ij} - \phi_v \cdot C_{oj}\right)^2 = \sum_{j=m}^{n} a_j = A = \min \qquad (3)$$

where $\Phi_p = \Phi_v + \Phi_d$

The number of data points is (n-m). Derivation of equation (3) with respect to the model parameters leads to a linear equation system:

$$\sum_{j=m}^{n}\left(\frac{dC_{ij}}{dt} \cdot C_{ij} + \phi_p \cdot C_{ij}^2 - \phi_v \cdot C_{oj} \cdot C_{ij}\right)^2 = 0 \qquad (4a)$$

$$\sum_{j=m}^{n}\left(-\frac{dC_{ij}}{dt} \cdot C_{oj} - \phi_p \cdot C_{ij} \cdot C_{oj} + \phi_v \cdot C_{oj}^2\right)^2 = 0 \qquad (4b)$$

The solution of the above equation system yields values for Φ_p and Φ_v. With the determined values and the use of equation (1), the indoor PPAH concentration is calculated. This is done by solving the following equation (1) with the use of a fourth order Runge-Kutta algorithm. Therefore, the time series of the indoor

PPAH concentration can be modelled with the measured outdoor PPAH concentration, the model parameters as obtained by the least square minimisation and the (measured) indoor PPAH concentration at the starting time t_j.

Results

The stability of the detectors and their sensitivity raise questions about he applicability of this method. As a first check, two such detectors were set up inside and outside of a university building in the city of Zurich together with a meteo station registering the necessary values of the weather (air temperature, wind velocity and direction): Fig.2 (Hüglin,Skillas 1997).

It was found in various cases that the path of the measured PPAH concentration cannot be reproduced accurately with the method described above. For derivation of the mass balance equation it is assumed that the entering outdoor air is instantaneously mixed with the indoor air. Obviously, this is a simplification of the real situation. Outdoor air which enters indoors through holes and cracks of the building envelope is dispersed in the room by diffusion and mainly by convection of the indoor air mass. We therefore introduce as a first approximation a delay time which describes these effects. In our model, this delay time is represented by a constant time lag between the indoor and outdoor signal. The least squares (equation (3)) are therefore minimised for delay times between 0 and 105 minutes with a stepsize of one minute and for the values of ϕ_L and ϕ_P.

As the curves in fig.1 and 3 show, the measured time-evolution of the indoor concentration can be reproduced in a reasonably accurate way. The corresponding mean values for the ventilatory and the deposition rate turned out to be:

(fig.1) $\Phi_v = 0.78$ $\Phi_d = -0.03$

(fig.3) $\Phi_v = 0.61$ $\Phi_d = -0.06$

As the values demonstrate, the method delivers at least reasonable or plausible values. The negative values of ϕ_D can be explained by the existence of internal sources of PAH and/or offset problems with the sensors.

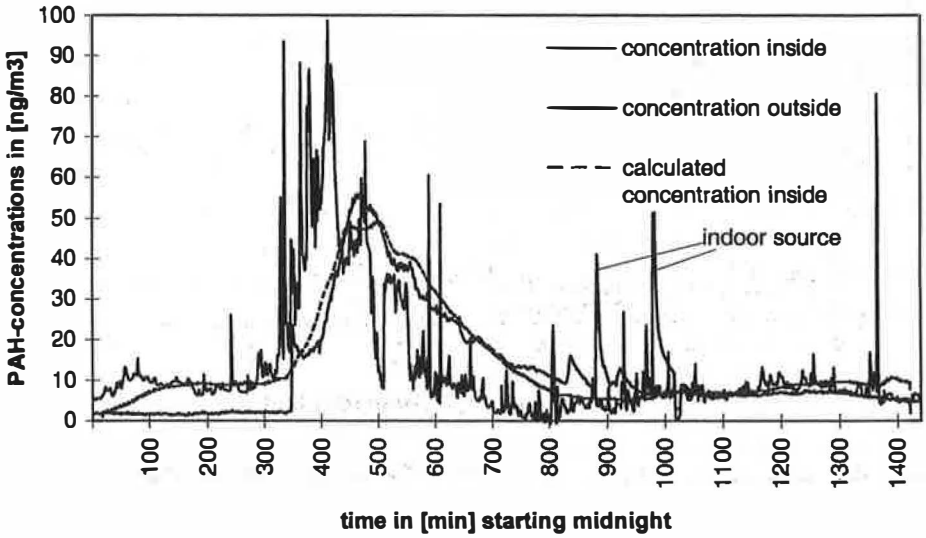

Figure 1: measured and calculated concentrations of PAH on April,14th 1997

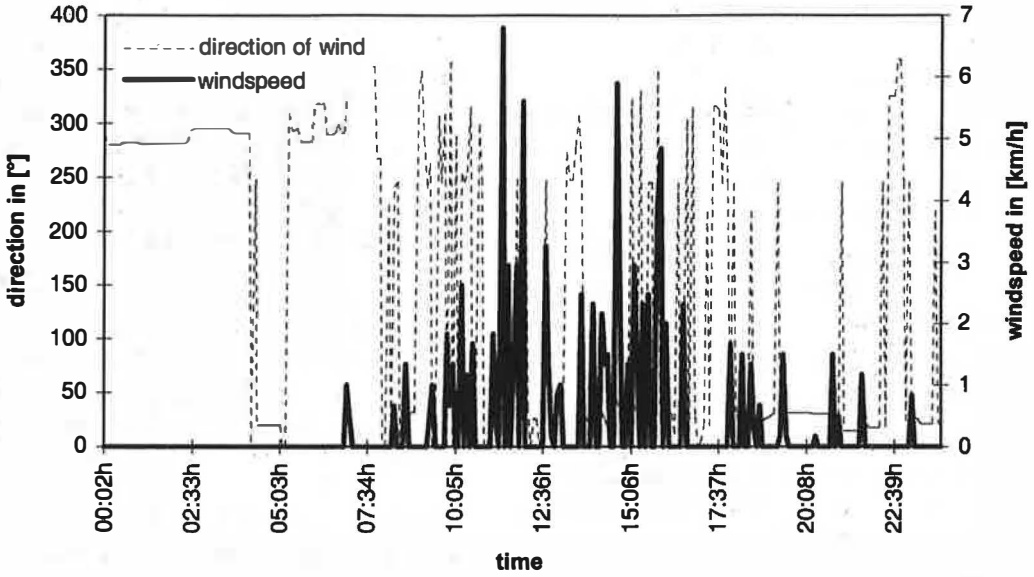

Figure 2: Windspeed and direction on april,14th 1997

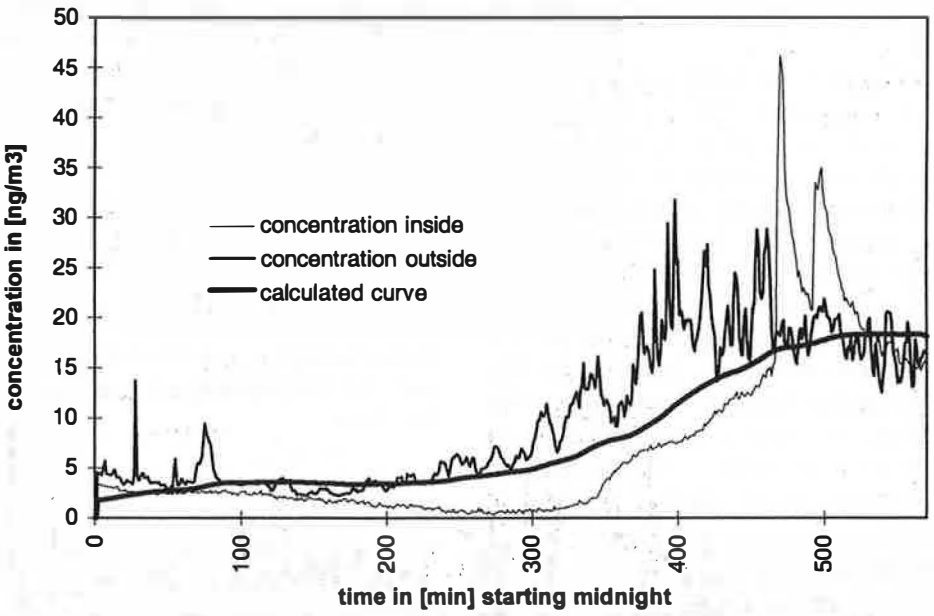

Figure 3: measured and calculated concentrations of PAH on April,15th 1997

5. Discussion

The above presented first tentative results demonstrate the principal applicability of this method. There are many questions left open however:

- the elimination of any kind of sensor drift and the lower limit of detectable concentrations,

- the separate determination of the deposition rate by a suitable test,

- the validity of the „mixing-time" approach, or the question of how much the location of the internal sensor influences the results: indoor air flow fields,

- the sensitivity of the results to the position of the outdoor sensor,

- the detectability of stochastically occurring indoor sources as cigarette smokers or similar.

In view of the interesting potential of this method, the above mentioned points are going to be investigated systematically. Of course also comparisons of this method with simultaneously applied standard tracer gas procedures as well as flow field simulations for the indoor as well as for outdoor situations are planned. The authors hope to present in the near future an easy to use and commercially developed detector assembly with integrated algorithms which allows the direct and real time monitoring of the air change rates as well as the exposure to aerosols of the people indoors .

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