VARIABILITY IN VOC-CONCENTRATIONS OVER TIME IN A LARGE BUILDING AND THEIR RELATIONSHIP WITH SOME CHARACTERISTICS OF THE INDOOR ENVIRONMENT

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ABSTRACT

In a large library building measurements were made during a 5-years period of air humidity, air temperature, carbondioxide (CO_2) and volatile organic compounds (VOC) in intake, supply and exhaust air. Altogether 95 VOC were identified. Eight compounds were classified as indoor-related VOC since their concentrations were much higher indoors than outdoors. There is a positive outdoor/indoor correlation for total VOC (TVOC) but not for indoor-related VOC which are positively correlated with CO_2 in exhaust air. Ventilation by 75% recirculated air cleanses the indoor air from CO_2 over night but not from indoor-related VOC.

INTRODUCTION

In a large library building, measurements were made during a 5-years period of air humidity, air temperature, carbondioxide (CO_2), and volatile organic compounds (VOC) in intake, supply and exhaust air. The number of staff and visitors in the building were counted.

METHOD

Building

The study object was the Stockholm University Library building built in 1983, a six stories, energy-efficient concrete building. It has mechanical ventilation with recirculated air, dimensioned for a supply airflow of 85,000 m³/h. Whithout recirculation of air the outdoor airflow rate is 26-40 L/s,person and with 75% recirculated air 7-10 L/s,person (or in unit of floor area of the building 1.8 and 0.4 L/s, m², respectively). The supply air is particle filtered and optionally preheated and/or humidified. Soon after its opening the library building was classified as 'sick' by the health authorities and, in 1990/91, the ventilation system was remodelled into a 100% outdoor air supply system. The target capacity of the outdoor air supply was marginally increased to 88,000 m³/h, corresponding to 27-41 L/s,person or 1.8 L/s,m².

Chemical and physical measurements

VOC in the air were sampled and determined with the method described in (1). The method has been tested to be appropriate in an international interlaboratory comparison of sampling for organic vapour in indoor air (2). Air samples of 15 L are adsorbed on porous polystyrene fil-

ters, then desorbed by heat and, finally, analyzed in a gas chromatograph, GC (Varian 3700). For representative samples, identification of separated compounds were performed by mass spectrometry, GC/MS (Varian 3700/Finnigan MAT ITD 700). The detection limit for quantification is at least 0.1 μg/m³ and for identification about 0.3 μg/m³. The area of each peak in the chromatograms is integrated and the concentration is determined by using the response factor of *n*-octane. The analysis comprises organic compounds with boiling points between 50-100 °C and 240-260 °C, that is the whole range of VOC (3). TVOC was calculated as the integrated area of the chromatogram. Formaldehyde was sampled and analyzed by a chemosorption method developed by (4). For temperature and air humidity measurements Kane-May, KM 8004, was used and for CO_2-concentration Binos IR gas analyzer.

Study design

The indoor air quality of the library building was investigated in three subsequent studies. The first, Study 1, was conducted when the building was 5-years old during an 8-month period (fall, winter, spring) representing changing climatic loads and two ventilation settings. After a building intervention in which the air handling system was remodelled, two 3-month studies were conducted, one in the same year, Study 2, and the other, one year later, Study 3. During the study periods the mechanical ventilation system was run 24 hours a day, all days of the week, with invariant outdoor air rate and air mixing. In Study 1, two main ventilation settings were used: 75% recirculated air (November-March) or no recirculated air (100% outdoor air, September-October & April-May).

Air sampling was performed in the air handling unit of the building once a week for study periods of 3 weeks. Air temperature, relative humidity and CO_2, were measured every hour from 7 a.m. to 5 p.m., during 21 days in all. For concentrations of VOC two parallel air samples were taken during 20 days in the intake air, the supply air and the exhaust air, once in the morning at 7 a.m., and once in the afternoon at 3 p.m. Formaldehyde was sampled during 5 days, two samples in parallel, at 7 a.m. and 3 p.m. in the intake air, supply air, and exhaust air and, altogether 20 samples were analyzed. In the post-intervention studies, air temperature, relative humidity, and CO_2-concentration were measured every hour from 7 a.m. to 5 p.m. during 2 days in each study. The VOC samplings were performed in intake, supply and exhaust air, in mornings and afternoons for 6 and 7 days, respectively, whereas formaldehyde measurements were excluded.

RESULTS

Temperature and air humidity

The intake air temperatures in Studies 1, 2 and 3 were -3 to +13, -3 to +12 and 0 to +10 °C, respectively. For all three studies the mean temperature in the supply air was 16 °C (SD 1) and in the exhaust air 19 °C (SD 1). The air humidity in Study 1 varied between 1.1 and 6.7 g/kg dry air in the intake air, and it was almost the same in intake, supply and exhaust air in the mornings. In the afternoons, during the period with 75% recirculated air, the air humidity increased by 0.1 to 3.9 g/kg dry air in the supply and exhaust air.

Carbon dioxide, CO_2

The CO_2-concentrations in the exhaust air varied over the day between 350 and 540 ppm during the nonrecirculation period, and between 360 and 990 ppm during the 75% recirculation period. After remodelling the system to nonrecirculation, the variations in CO_2 in exhaust air

were 380 to 550 ppm. In all three studies and all sampling points, morning concentrations were the same as outdoor concentrations.

The daily variation in CO_2-concentration is illustrated for Study 1 in Figure 1. Using the equation: $C = q/nV * (e^{-nt})$ where C = CO_2-concentration in L/m³, q = CO_2 generation rate in L/h, n = number of air exchanges per hour and V = building volume, m³, the theoretical variation in CO_2-concentration can be calculated. With 900 persons in the building, generating 20 L CO_2/ h,person, the variation for different air exchanges was calculated (Fig. 1). The 75% recirculation of air gave an air exchange rate of 0.4 to 0.6 ach and 0% recirculation 1.2 to 2.5 ach. After remodelling the ventilation, the air exchange rate was calculated to be 1.2 to 2.5 ach all the time, independent of outdoor climate.

Formaldehyde and volatile organic compounds, VOC

In Study 1, formaldehyde concentrations were low, in the intake air usually less than the chemical detection limit (0.003-0.004 mg/m³), and in the supply air the 50-percentile was 0.008 and the 90-percentile 0.019 mg/m³. In the exhaust air the 90-percentile and 50-percentile was 0.060 and 0.020 mg/m³, respectively. The highest concentrations were measured in the supply and the exhaust air in the afternoon during the 75% recirculation period.

In all, 95 VOC were identified in the air samples. Compounds with concentrations of 0.1 μg/m³ and above were quantified. Most of these compounds are common in both indoor and outdoor air, like 17 aliphatic and 13 aromatic hydrocarbons and the terpenes α-pinene and limonene. Some compounds are common and characteristic for indoor air, namely the aldehydes hexanal, nonanal and decanal, the ester acetic acid butyl ester and the halocarbone 1,1,1-trichloroethane. Minor quantities of 2-ethyl-hexanol and 2-(2-ethoxyethoxy)ethanol were also detected.

The same VOC appear indoors and outdoors, but in Studies 1, 2 and 3 the mean outdoor TVOC concentrations are less than indoors; in the morning 73, 88 and 62 % of the indoor concentrations, and in the afternoon 47, 53 and 41 %, respectively. However, occasionally the intake or supply air showed higher concentrations of TVOC than the exhaust air. Eight compounds had very low outdoor air concentrations, never exceeding 50% of the indoor concentrations and were, therefore, classified as typical *indoor-related VOC*. These compounds are: octene, α-pinene, carene, limonene, 1-hexanal, 2-ethylhexanol, acetic acid butyl ester, and 1,1,1-trichlorethane. The total indoor concentrations of these were low, never exceeding 40 μg/m³, and over the 5-years time period, no decrease of the total concentrations of these compounds was exhibitied.

Table 1 shows the 10-, 50-, and 90-percentiles of total volatile organic compounds (TVOC) and the sum of indoor-related VOC (peaks) in intake, supply, and exhaust air. Both indoors and outdoors the highest concentrations were found in Study 1 in wintertime (November-March) when 75% return air was recirculated. In Study 2 and 3, VOC concentrations were low, approximately the same levels as for 100% outdoor air supply in Study 1. Concentrations of individual compounds were also low, for toluene and xylenes at most 10-20 μg/m³, whereas half of the remaining compounds were <5 μg/m³, and all others <1 μg/m³.

There is a high positive correlation between outdoor and indoor TVOC concentrations (correlation factors for intake and exhaust air being 0.6 - 0.8 in all three studies), while the

Table 1. Stockholm University Library building, Study 1, 2 and 3: TVOC and sum of indoor-related VOC, $\mu g/m^3$, in the air handling unit.

		90-percentile TVOC	90-percentile 'Indoor VOC'	50-percentile TVOC	50-percentile 'Indoor VOC'	10-percentile TVOC	10-percentile 'Indoor VOC'
Study 1							
75% recirculation	Intake	134	4	97	2	47	1
	Supply	280	38	151	28	96	20
	Exhaust	246	45	144	39	110	27
Study 1							
100% outdoor air	Intake	42	2	36	2	22	1
	Supply	42	2	36	2	28	1
	Exhaust	69	14	61	11	41	8
Study 2							
100% outdoor air	Intake	71	2	63	2	28	1
	Supply	79	2	59	2	29	1
	Exhaust	91	14	73	8	50	6
Study 3							
100% outdoor air	Intake	37	1	21	1	17	1
	Supply	53	2	20	1	16	1
	Exhaust	101	17	49	7	39	5

outdoor/indoor correlation for the 8 indoor-related VOC is low (<0.2 in all three studies). On the other hand, the indoor-related VOC correlate with the CO_2-concentrations in the exhaust air which may indicate that they are related not only to the indoor materials but also to the occupants and their activities.

Depending on outdoor air supply, there is an increase for both CO_2 and indoor-related VOC in the exhaust air over the day. For CO_2 the increase also depends on the number of occupants. In supply and exhaust air CO_2 always decreases to the intake air concentration over night, even during the periods of recirculated air. The indoor-related VOC also decrease over night, but the decrease is less during the period with 25% outdoor airflow compared to the periods with 100% outdoor airflow. That is, when ventilated with 75% recirculated air the system does not 'cleanse' the indoor air fully from pollutants. Benzene, which is a typical outdoor air pollutant, showed no indoor variation from morning to afternoon and the concentration was always the same indoors and outdoors. For the pre-intervention study, Study 1, Figures 2 and 3 show the concentration variations of CO_2 and indoor-related VOC in supply air.

DISCUSSION

The TVOC concentrations in the indoor air are much influenced by the outdoor air concentrations. The latter may vary with a factor of ten or more depending on i.a. the time of the year, whereas the indoor-related VOC are associated with occupancy, or rather the occupants' activities, in the building as well as of the amount of outdoor airflow.

The TVOC concentrations in exhaust air were usually higher than in the intake and supply air. But, in some cases the intake air was more polluted than the supply and/or exhaust air, that is, a portion of the pollutants were 'lost' on their way through the system. This phenomenon could be explained in at least two ways: [1] There is a state of equilibrium between the concentrations of VOC in the indoor air and the amount of compounds adsorbed to the indoor materials. The equlibrium is influenced by i.a. air and surface temperatures, air humidity, surface area and type of materials, and intake air concentrations and flow rate. When air concentrations changes, the equilibrium is displaced so that higher air concentrations cause increasing adsorption

to the materials and lower air concentrations lead to an increasing desorption of VOC from the materials. That is, when ventilating polluted materials with clean intake air, the pollutants are desorbed and the exhaust air will be more polluted than the intake air (cf. 1). [2] In the presence of UV-light and free radicals, chemical reactions lead to a continuous degradation of volatile organic compounds to low molecular polar compounds which are diluted in the atmosphere. Normally, the indoor environment is protected from both UV-light and reactive compounds and this may lead to a preservation of the VOC in the indoor air providing higher indoor concentrations. On the other hand, the presence of, for example, ozone from office machines may cause a more intensive degradation indoors than outdoors, resulting in more polar compounds, especially formaldehyde, and less nonpolar compounds with high molecular weight (5). Polar compounds, like low molecular aldehydes, are discriminated in the method of sampling and analysis applied in this investigation which results in lower TVOC values.

CONCLUSIONS

TVOC have been used as a measure of the amount of air pollution indoors and of the background of outdoor pollutants indoors. It also may be used for assessing part of the adsorption and desorption of pollutants in the indoor environment and the influences of chemical reactions in indoor and outdoor air. But, selective analysis leading to identification of compounds associated with the building, the indoor materials, the occupants, or the activities in the building (i.e., indoor-related VOC), gives more detailed and important information about the indoor environment. Studies of the variation in concentrations of these compounds leads to a more adequate assessment of the ventilation effectiveness (assuming adequate air mixing) than just monitoring the CO_2, which is a nonreactive compound with minor exchange due to adsorption and desorption from or to indoor materials. Selective analysis of indoor-related VOC also gives information about the introduction of new compounds, chemical reactions in indoor materials, and whether or not the number or concentrations of compounds are changing over time.

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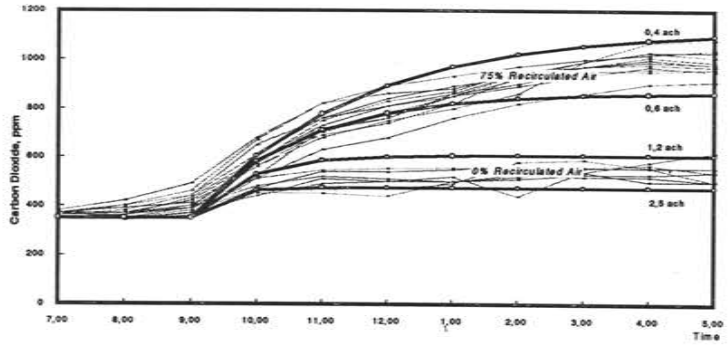

Fig. 1. Stockholm University Library, Study 1: CO_2, ppm, in exhaust air at 75% and 0% recirculated air, compared to predicted concentration for 900 occupants and 0.4, 0.6, 1.2, and 2.5 ach.

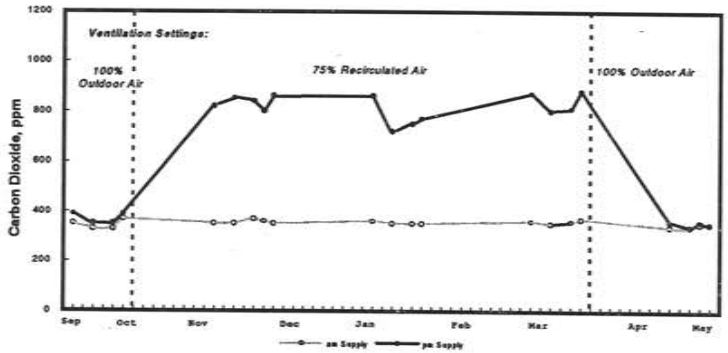

Fig. 2. Stockholm University Library, Study 1: CO_2, ppm, am and pm in supply air at the air handling unit.

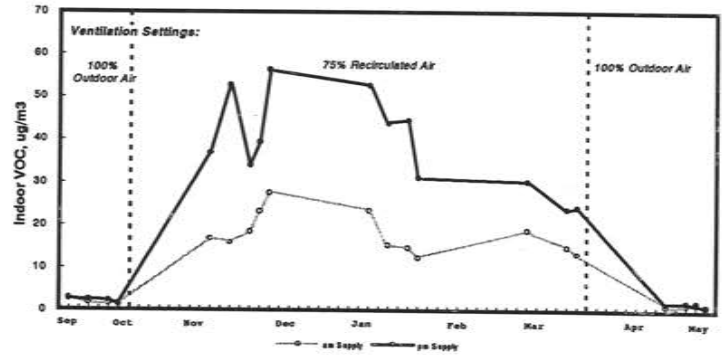

Fig. 3. Stockholm University Library, Study 1: Sum of indoor-related VOC, $\mu g/m^3$, am and pm in supply air at the air handling unit.